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D-Optimal Design with Split Plot Approach for Quadratic Mixture-Amount Experiments (Case Study of Three Components with Composition Constraints)

Rancangan D-Optimal dengan Pendekatan Split Plot pada Eksperimen Mixture-Amount Model Kuadratik (Studi Kasus Tiga Komponen dengan Kendala Komposisi)

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Abstract

The mixture experiments (MAE) were influenced by both the proportions of the components and the total amount. The traditional MAE encompasses classical mixture experiments for each total amount, which complicates the application of complete randomization; therefore, a split-plot design is suggested. In this design, the plot factor represents the total mixture amount, whereas the subplot represents the composition of the materials. Another issue is the increasing number of experiments as the number of materials and total amount increase. This study proposes a split-plot approach using a point-exchange algorithm based on the D-optimal criteria to generate an efficient design. The model used is a quadratic mixture of numbers model, which can capture the linear, quadratic, and interaction effects between material proportions and total mixture numbers. The case study involved three components with proportion constraints and three levels of mixture numbers: high, medium, and low. The results show that the algorithm generates optimal points generally located at the edge of the design region and that increasing the number of experimental units improves the stability of designs involving total mixture numbers.

Keywords: D-optimal; Mixture-amount experiment; Split-plot design.

1. INTRODUCTION

The experimental design comprised a sequence of tests designed to detect and identify variations in response outcomes resulting from alterations in the input variables of the process [7]. Experimental design is extensively utilized across various scientific disciplines, including the industrial sector, especially in product manufacturing. In practice, product formulation is often performed through a trial-and-error approach, which increases production costs and requires considerable time to make the experimental designs more efficient in terms of price and time. One design that is often used to create product formulations is a mixture experiment. In a mixture design, the result is influenced exclusively by the proportion of the components and not by the total amount of the mixture [6]. This principle is illustrated by the creation of food flavors. Mixture design studies the composition of the right mixture to produce the product and whether each ingredient has the same or different proportions to produce a good product. In the mixture design, if the composition of one ingredient increases, the composition of at least one other ingredient decreases, and vice versa[3]. The difference in the proportions of each component in the mixture resulted in different responses.

In certain instances, the results of a mixture design are affected by both the ratios of the mixture components and the total amount of the mixture. For instance, when formulating a specific blend with total mixtures of 50 g and 150 g that consist of three primary ingredients, the ratio of each ingredient in these mixtures may be identical or vary. It is crucial to assess whether the total amount of the mixture influences the product's response. A design that considers the total mixture amount is referred to as the Mixture Amount Experiment (MAE) [11]. In MAE, it was presumed that both the proportions of the components and the total amount of the mixture affected the response. The concept of MAE was initially introduced by Piepel and Cornell in 1987, who developed MAE models and methodologies by creating comprehensive and partial designs that are applicable to both finite and infinite scenarios. This model modifies the Scheffé model by incorporating the total mixture amount as a linear or quadratic factor influencing the response value while preserving the original structure of the mixture model. This method is limited to situations where variations in the total amount affect only the magnitude of the response value, without altering the relative influence of the individual components [10].

In MAE applications, assessing the composition of mixtures for each total amount presents difficulties in achieving complete randomization, requiring further adjustments. A viable solution is to implement a split-plot design, which is suitable when full randomization is impractical because of structural limitations. Within the MAE framework, the primary plot factor is the total number of mixtures, whereas the subplot factors are the mixture components. In addition to the problem of randomization limitations, the total can considerably increase the size of the experimental unit, resulting in elevated production costs. Therefore, an optimal design is crucial to reduce the number of experimental units using the chosen model. Optimal design refers to an experimental design methodology that utilizes computerization to create efficient designs based on specific criteria, such as A-, D-, I-, and G-optimal, each offering distinct benefits depending on whether the emphasis is on estimation or prediction efficiency [9].

Research on Mixture design using split plots has been carried out by Arina et al in 2022, which examines concrete experiments with steel slag mixtures using a Mixture Process Variable (MPV) design with a split-plot approach and D-optimal design. This design performs limited randomization between mixture components consisting of cement, fine sand, coarse gravel, steel slag, and water, with the observed process variable being the steel slag particle size [2]. In addition, research on the application of the mixture-amount experiment was conducted by Sari et al in 2024, who used the A-Optimal Design with a modified divided plot design and applied the Second-Order Scheffé model [11].

This study utilizes a quadratic mixture-amount model that simultaneously accounts for both the proportions of components and the total amount of the mixture, including their interactions [8]. This model enables responses to be affected by nonlinear relationships between the proportions and amounts of ingredients. Originally developed by Pal and Mandal ini 2012, this model introduced a quadratic mixture-amount framework and suggested optimal designs (A-optimal and D-optimal) for parameter estimations. To create the optimal design in MAE, a point-exchange algorithm was employed to develop an efficient design that complies with randomization constraints based on the D-optimal criterion. The point-exchange algorithm aims to improve the initial design by exchanging trial points to satisfy the specific optimal criteria. Implementing this method is anticipated to produce an optimal formulation in terms of the cost, time, and accuracy of the model estimation according to the MAE structure involving various total mixtures. Goos and Vanderbroek (2003) implemented a point-exchange algorithm within a split-plot design framework, applying it to a protein extraction experiment that involved varying numbers and sizes of main plots, all while utilizing the D-optimality criteria[5]. This study uses an algorithm to identify the best formulation from a case study involving three components with varying composition constraints and total mixture amounts.

2. METHODOLOGY

2.1. Mixture Experiment

Mixture design involves creating a design from various combinations to explore and adjust the ratios of two or more components within each mixture, with the assumption that any variations in response are solely due to the proportion of each ingredient in the mix [6]. In this type of design, the response value changes as the proportion of each component are altered. If the mixture consists of q ingredients and x_i represents the proportion of the i-th component, the constraint function of the mixture experiment is $\sum_{i=1}^{q} x_i = 1$ and $x_i \geq 0$, $i = 1,2, \ldots, q$ [3]. The mixed-design model that characterizes the y-response is commonly referred to as the Scheffé mixed model. This model encompasses various degrees of form, including the Linear Model, Quadratic Model, and Cubic Model, which are represented in equation (2.1), (2.2), and (2.3), respectively:

$$y(x) = \sum_{i=1}^{q} \beta_i x_i + \varepsilon \tag{2.1}$$

$$y(x) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} \beta_{ij} x_i x_j + \varepsilon \tag{2.2}$$

$$y(x) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} \beta_{ij} x_i x_j + \sum_{i=1}^{q-2} \sum_{j=i+1}^{q-1} \sum_{k=j+1}^{q} \beta_{ijk} x_i x_j x_k \tag{2.3}$$

In this context, $y(x)$ represents the response variable; β_i denotes the i-th parameter of the mixed-component model; β_{ij} signifies the i-th parameter of the mixed component model; β_{ijk} indicates the i-th parameter of the mixed-component model; x_i is the value of the i-th explanatory variable; x_j is the value of the j-th explanatory variable; x_k is the value of the k-th explanatory variable, and ε represents the error term.

2.2. Mixture amount Experiment

An experiment known as a Mixture Amount Experiment (MAE) involves conducting a mixture design with multiple total amount of the mixture. In these types of experiments, it is presumed that the response is affected not just by the ratios of the mixture components but also by the total amount of the mixture, which influences the quality of the end product. For example, when dealing with two components, their proportions are represented by x_1 and x_2, meeting the condition $\mathrm{x}_1 + \mathrm{x}_2 = 1$, there by creating a factor space shaped like a one-dimensional simplex. The three mixtures tested—namely, the first component, the second component, and the 50%:50% mixture—were assessed at two distinct total amounts, ALow and Ahigh, as illustrated in Figure (1).

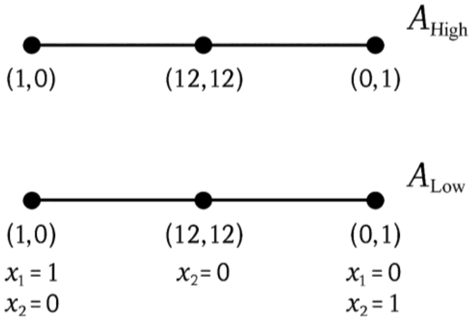


Figure 1. The Completer {2,2} Simplex-Lattice Design at Two Levels, A_{Low} and A_{High}, of Total Amount

The determination of the model in the MAE experiment is influenced by how the effect of the total mixture amount is characterized. If changing the total mixture amount affects only the response value without altering the component proportions, the effect is considered additive, as demonstrated here [1]:

$$y(x, A) = \{mixture\ model\} + [effect\ amount] + \varepsilon \tag{2.4}$$

where A represents the total mixture amount in units; $\{mixture\ model\}$ is the model for the mixture design in equations (1),(2),(3); $[effect\ amount]$ is $\beta_1 A$ (linier) dan $\beta_1 A + \beta_{11} A^2$ (quadratic)

When the total quantity of a mixture affects the proportion of its components and the resulting outcome, the properties of the mixture interact with the total amount. The interactive MAE model was developed using the Scheffé model framework, which outlines the characteristics of the components of the mixture along with the influence of the mixture's total amount:

$$y(x, A) = \{mixture\ model\} + \{mixture\ model\}[effect\ amount] + \varepsilon \quad (2.5)$$

The model in Equation (2.5) separates the effect amount as a linear or quadratic effect on the response value without affecting the structure of the mixture model. This approach is limited to cases where changes in the total amount directly affect the size of the response value and do not change the relative influence of each component.

In 2012, Pal and Mandal proposed a quadratic mixture-amount model. They formulated an optimal design model that simultaneously considered both the proportions of components and total quantity of the mixture, along with their interactions [8]. This model is shown in equation (2.6).

$$y = \beta_{01}A + \beta_{02}A^2 + A\left(\Sigma_{i=1}^{q}\beta_{0i}x_i + \Sigma_{i=1}^{q}\beta_{0i}x_i^2 + \Sigma_{i<j}\,\beta_{ij}x_ix_j\right) \quad (2.6)$$

where A is the total amount of mixture (in units), x_i is the proportion of the i-th component, with In this context, A represents the total quantity of the mixture (measured in units), whereas x_i denotes the fraction of the i-th component, ensuring that $\Sigma_{i=1}^{q}x_i = 1$. The β coefficients reflect the influence of each component and its interactions on the response.

The advantage of this model is that the effect of the total amount is reflected through the direct interaction between and the proportion rather than through a change in the value of the regression coefficient as a function of the amount. As such, it more efficiently represents the complex relationship between ingredient composition and amount, while maintaining a relatively small number of parameters.

2.3. Split Plot Design

A split-plot design is utilized when structural randomization is impractical due to the presence of numerous experimental units or variables that are difficult to manage [2]. Typically, this design includes a main plot variable along with subplot variables. The main plot represents factor that is not easily modified, while the subplots are more adaptable to change [7]. Randomization was performed in two phases in a split-plot design. Initially, the main plot factors were assigned randomly to the experimental units, followed by randomization of the subplot factors within each main plot.

The model of split plots design on the j-th observation ($j = 1, 2, \ldots, k_i$) in the i-th main plot ($i = 1, 2, .., b$) with b being the number of main plots can be written as follows [2]:

$$\boldsymbol{Y}_{ij} = \boldsymbol{f}'\left(\boldsymbol{w}_i, \boldsymbol{s}_{ij}\right)\boldsymbol{\beta} + \gamma_i + \varepsilon_{ij} \quad (2.7)$$

In this context, $\boldsymbol{f}'\left(\boldsymbol{w}_i, \boldsymbol{s}_{ij}\right)$ represents the expansion of the model of the main plot and sub-plot variables. Vector $\boldsymbol{\beta}$, with dimensions $p \times 1$, contains the parameters. The term γ_i accounts for the error in the main plot and ε_{ij} denotes the error in the subplot. When translated into the matrix form, Equation (2.7) can be expressed as follows:

$$\boldsymbol{Y} = \boldsymbol{X\beta} + \boldsymbol{Z\gamma} + \boldsymbol{\varepsilon} \quad (2.8)$$

Consider $\boldsymbol{X}$ as a matrix that includes both main plots (A) and subplots (x) with dimensions $n \times p$. Vector $\boldsymbol{\beta}$, with dimensions $p \times 1$, represents the parameters. Another matrix, $\boldsymbol{Z}$, is composed of 1s and 0s for the main plots, containing n observations with dimensions $n \times b$. The vector $\boldsymbol{\gamma}$ denotes the random effects associated with the main plots and ε is a vector of errors. If $\boldsymbol{Y}$ is arranged according to the main plot, then

$$\boldsymbol{Z} = \boldsymbol{I}_b \otimes \boldsymbol{1}_k = \begin{pmatrix} 1 & 0 & 0 & \cdots & 0 \\ 0 & 1 & 0 & \cdots & 0 \\ 0 & 0 & 1 & \cdots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \cdots & 1_b \end{pmatrix} \otimes \begin{pmatrix} 1 \\ 1 \\ \vdots \\ 1_k \end{pmatrix}$$

Here, b represents the count of main plots, k denotes the number of subplots, and ⊗ signifies the Kronecker multiplication matrix. The assumptions of the model in Equation (2.8) are $\boldsymbol{\gamma} \sim N(\boldsymbol{0}_b, \sigma_\gamma^2 \boldsymbol{I}_b)$, $\boldsymbol{\varepsilon} \sim N(\boldsymbol{0}_n, \sigma_\varepsilon^2 \boldsymbol{I}_n)$, $cov(\boldsymbol{\gamma}, \boldsymbol{\varepsilon}) = \boldsymbol{0}_{nxb}$.. Based on these assumptions, the covariance matrix is given by Equation (2.9).

$$cov(\boldsymbol{y}) = \boldsymbol{V} = \sigma_\varepsilon^2 \boldsymbol{I}_n + \sigma_\gamma^2 \boldsymbol{Z}\boldsymbol{Z}` = \sigma_\varepsilon^2 (I_n + \eta \boldsymbol{Z}\boldsymbol{Z}') \tag{2.9}$$

The variance component ratio, denoted as $\eta = \frac{\sigma_\gamma^2}{\sigma_\varepsilon^2}$, measures the degree of correlation among observations within the same main plot [2]. The unknown parameter vector β in Equations (2.7) and (2.8) is estimated using the generalized least squares (GLS) estimator, i.e.:

$$\widehat{\boldsymbol{\beta}} = (\boldsymbol{X}'\boldsymbol{V}^{-1}\boldsymbol{X})^{-1}\boldsymbol{X}'\boldsymbol{V}^{-1}\boldsymbol{y} \tag{2.10}$$

The variance components $(\sigma_\gamma^2, \sigma_\varepsilon^2)$ and the matrix $\boldsymbol{V}$ are unknown, which necessitates the estimation presented in equation (2.10).

$$\widehat{\boldsymbol{V}} = \hat{\sigma}_\varepsilon^2 (I_n + \eta \boldsymbol{Z}\boldsymbol{Z}') \tag{2.11}$$

Once the variance components have been estimated, they are substituted into equation (2.11), leading to the Feasible Generalized Least Squares (FGLS), as described in:

$$\widehat{\boldsymbol{\beta}} = \left(\boldsymbol{X}'\widehat{\boldsymbol{V}}^{-1}\boldsymbol{X}\right)^{-1}\boldsymbol{X}'\widehat{\boldsymbol{V}}^{-1}\boldsymbol{y} \tag{2.12}$$

with the following courage matrix:

$$var\left(\widehat{\boldsymbol{\beta}}\right) = \left(\boldsymbol{X}'\widehat{\boldsymbol{V}}^{-1}\boldsymbol{X}\right)^{-1} \tag{2.13}$$

2.4. Optimal design

Optimal Design is an aspect of experimental design that aims to estimate unbiased parameters with the least variance, facilitating precise statistical inference at the lowest cost. The main goal of optimal design is to identify a set of n design points or levels that effectively disclose the model's coefficients [1]. The effectiveness of the optimal design depends on the model employed and the desired number of observations, which are determined by specific criteria. A commonly used criterion in optimal design is the D-optimality criterion. This criterion serves as a benchmark for the quality of the parameter estimates, as indicated by the value of $var\left(\widehat{\boldsymbol{\beta}}\right)$ in equation (2.13). The objective of this optimization was to achieve the smallest possible value of $var\left(\widehat{\boldsymbol{\beta}}\right)$ by either maximizing the determinant or minimizing the inverse factor of the information matrix. If X is a matrix of dimensions $n \times p$, where n denotes the number of design points and p represents the number of parameter coefficients, then the information matrix can be expressed as follows:

$$\boldsymbol{M} = \boldsymbol{X}'\widehat{\boldsymbol{V}}^{-1}\boldsymbol{X} = \sigma_\varepsilon^{-2}\, \boldsymbol{X}'(\boldsymbol{I}_n + \eta \boldsymbol{Z}\boldsymbol{Z}')^{-1}\boldsymbol{X} \tag{2.14}$$

The D-optimality criterion involves choosing the design matrix by maximizing the determinant of the information matrix among all potential designs, which can be represented as follows:

$$|\boldsymbol{M}^*| = \max\ |\boldsymbol{M}|$$

2.5. Algoritma *Point-exchange*

An algorithm can effectively identify the optimal design points, with the point exchange algorithm being a commonly employed technique for this purpose. This method improves the information matrix by adding or removing points from the initial design. The usual steps in the point-exchange algorithm involve presenting candidate sets, establishing model assumptions, determining the number of design runs, and specifying optimal criteria [5]. Goss and Vandebroek created a modified version of this algorithm tailored for split-plot designs. This method begins by forming a candidate set using the Cox direction formula, where the first component is defined as and the remaining components as , enabling the calculation of changes in each component using the following formulas: A list of candidate sets is created according to the Cox direction formula [4]:

$$\tilde{x}_i = x_i + \delta \text{ dan } \tilde{x}_j = x_j - \frac{x_j\delta}{1-x_i} \tag{2.15}$$

for $i \neq j$ dan $i = 1,2, \ldots, p$, where p represents the number of model parameters.
Next, the prediction variance for each candidate point is assessed to determine the optimal points. The arrangement of whole plots and subplots was established to mirror the split-plot design structure. With this configuration, the initial design is randomly chosen from the set of candidates. Subsequently, the determinant of the information matrix and the prediction variance of the initial design were calculated to assess design efficiency. In the subsequent phase, the subplot points from the initial design are swapped with those from the candidate set. This involves substituting the point in the initial design that exhibits the lowest prediction variance with the candidate point that exhibits the highest prediction variance. This procedure is repeated iteratively until convergence is reached, indicating that the change in the determinant of the information matrix becomes negligible. After achieving an ideal arrangement of the subplots, the levels of the main plot factors were swapped. This process was repeated until no further significant enhancements in design optimality were detected. The steps, from calculating the prediction variance to exchanging the main plot factor levels, were carried out 1000 times using various initial designs. Of these iterations, the design that produced the highest determinant of the information matrix was chosen as the final optimal design [5].

2.6. Material and Method

The point exchange algorithm was applied to MAE to identify the optimal composition for each total mixture. To assess the algorithm's effectiveness, a case study involving three components was conducted, with composition constraints set at $0.4 \leq x_1 \leq 0.6$, $0.25 \leq x_2 \leq 0.35$, dan $0.15 \leq x_3 \leq 0.35$. These constraints are based on research by Angriany et al. in 2021 [1]. The composition optimization assumes that the total mixture is divided into three levels: high $(+1)$, medium (0), and low (-1). Figure 2 illustrates the design region for this case, where the boundaries represent the maximum possible values for each component.

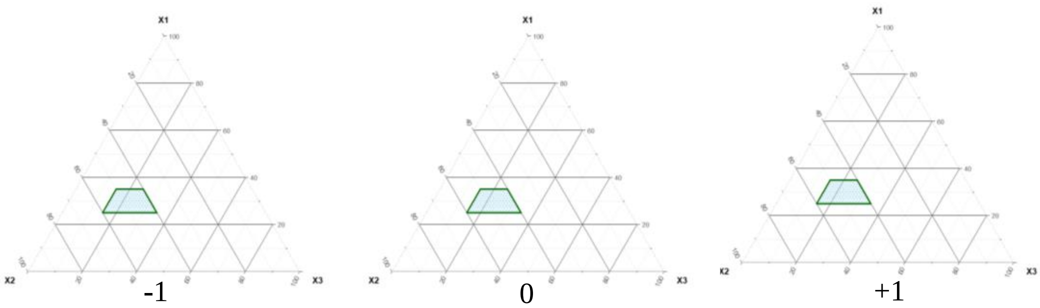


Figure 2. Design Region for Each Total Mixture Amount

The first step in the optimal design process in MAE is to determine the model assumptions. A quadratic model is used. According to Pal and Mandal [8]:

$$y = \beta_{01}A + \beta_{02}A^2 + A(\beta_{01}x_1 + \beta_{02}x_2 + \beta_{03}x_3 + \beta_{11}x_1^2 + \beta_{22}x_2^2 + \beta_{33}x_3^2 \\ + \beta_{12}x_1x_2 + \beta_{13}x_1x_3 + \beta_{23}x_2x_3)$$

because $x_1 + x_2 + x_3 = 1, x_i \geq 0$, and $A \in [-1,0,1]$then the model can be written:

$$y = \beta_{00}A^2 + A(\beta_{01}x_1 + \beta_{02}x_2 + \beta_{03}x_3 + \beta_{11}x_1^2 + \beta_{22}x_2^2 + \beta_{33}x_3^2 \\ + \beta_{12}x_1x_2 + \beta_{13}x_1x_3 + \beta_{23}x_2x_3)$$

where $\beta_{00}A^2$ is the quadratic effect of amount, reflecting the influence of total amount on the response, regardless of mixture composition, $A(\beta_{01}x_1 + \beta_{02}x_2 + \beta_{03}x_3)$ is the linear effect of mixture components, $\beta_{11}x_1^2 + \beta_{22}x_2^2 + \beta_{33}x_3^2$ is the quadratic effect of each component, and $\beta_{12}x_1x_2 + \beta_{13}x_1x_3 + \beta_{23}x_2x_3$ is the interaction effect between components.

In the context of MAE, an optimal design process necessitates the implementation of a split-plot design, which involves determining the ratio between main plot and subplot varieties. This study examines η values of 1, 5, and 10. Following the establishment of the model and variety ratio, the subsequent task is the construction of the point exchange algorithm, as derived from the research conducted by Goss and Vandebroek, as detailed in subchapter 2.5.

3. RESULT AND DISCUSSION

3.1. Candidate Set of Design Point

The first step in obtaining an optimal design involving the total mix variable is to create a candidate set of design points. In each case there are different constraints for each ingredient component. Based on the ingredient constraints and the mix design constraint function, a candidate set of points can be formed based on the cox-direction formula based on equation (2.15). The results of the candidate set are summarized in Table 1.

Table 1. Candidate Set of mixture each level of total amount

No	x_1	x_2	x_3	No	x_1	x_2	x_3
1	0.550	0.300	0.150	22	0.500	0.275	0.225
2	0.475	0.300	0.225	23	0.460	0.300	0.240
3	0.400	0.250	0.350	24	0.455	0.275	0.270
4	0.438	0.325	0.237	25	0.490	0.300	0.210
5	0.600	0.250	0.150	26	0.440	0.300	0.260
6	0.400	0.300	0.300	27	0.520	0.275	0.205
7	0.500	0.350	0.150	28	0.480	0.300	0.220
8	0.488	0.325	0.187	29	0.425	0.275	0.300
9	0.500	0.250	0.250	30	0.530	0.300	0.170
10	0.438	0.275	0.287	31	0.495	0.325	0.180
11	0.400	0.350	0.250	32	0.550	0.275	0.175
12	0.538	0.275	0.187	33	0.410	0.325	0.265
13	0.450	0.350	0.200	34	0.445	0.275	0.280
14	0.420	0.300	0.280	35	0.525	0.300	0.175
15	0.460	0.325	0.215	36	0.465	0.325	0.210
16	0.470	0.300	0.230	37	0.540	0.275	0.185

17	0.445	0.275	0.280	38	0.435	0.325	0.240
18	0.410	0.300	0.290	39	0.500	0.300	0.200
19	0.475	0.325	0.200	40	0.570	0.275	0.155
20	0.430	0.300	0.270	41	0.460	0.300	0.240
21	0.480	0.325	0.195	42	0.485	0.275	0.240

The candidate points can be represented geometrically within the design area. When dealing with composition, the design area is illustrated as a trapezoid, featuring 42 candidate points for each mixture category (High, Medium, Low), resulting in a total of $42 \times 3 = 126$ (N) candidate points. The visual representation of the candidate sets for each mixture level is shown in Figure (3).

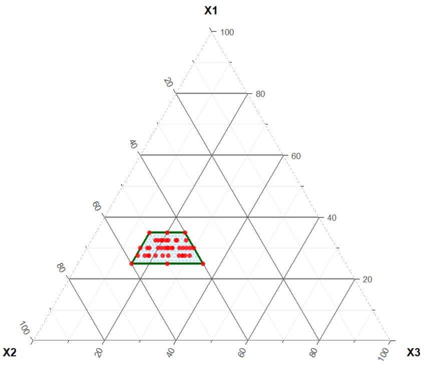


Figure 3. The set of candidate design points at each total mix amount

Based on figure (3), the set of candidate design points involves corner points, edge points, and axial points.

3.2 Efficiency Evaluation of MAE Based on the Number of Experimental Units

A method utilizing the D-Optimality criterion was implemented using the point exchange algorithm to assess the efficiency of the Mixture Amount Experiment (MAE). This evaluation was performed using three distinct configurations for the number of experimental units $n = 12$, $n = 21$, and $n = 32$. The configurations are described in the following subsections.

3.2.1. D-Optimal Design for Mixture Amount Experiment with $n = 12$

In the experiments with $n = 12$, a configuration of 6 main plots and 2 sub-plots was tested. The structure comprising 12 trials using the point-exchange algorithm is outlined in Table 2. The design point values (x_1, x_2, x_3) denote the number of mix proportions for each ingredient, while A signifies the total number of mixes to be employed.

Table 2. D-Optimal results for MAE employing the algorithm with $n = 12$

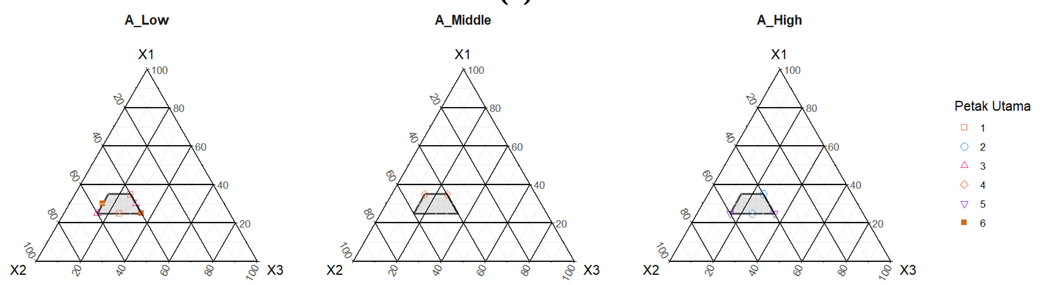
Main-plot	Sub-Plot	$\eta = 1$				$\eta = 5$				$\eta = 10$			
		x_1	x_2	x_3	A	x_1	x_2	x_3	A	x_1	x_2	x_3	A
1	1	0.400	0.350	0.250	1	0.400	0.350	0.250	-1	0.500	0.350	0.150	-1
	2	0.600	0.250	0.150	1	0.500	0.250	0.250	-1	0.400	0.300	0.300	-1
2	1	0.400	0.250	0.350	-1	0.500	0.250	0.250	1	0.400	0.350	0.250	-1
	2	0.438	0.275	0.287	-1	0.400	0.350	0.250	1	0.500	0.250	0.250	-1
3	1	0.500	0.350	0.150	1	0.600	0.250	0.150	-1	0.550	0.300	0.150	-1
	2	0.400	0.250	0.350	1	0.400	0.300	0.300	-1	0.400	0.250	0.350	-1
4	1	0.600	0.250	0.150	-1	0.500	0.350	0.150	0	0.400	0.250	0.350	1
	2	0.400	0.350	0.250	-1	0.400	0.350	0.250	0	0.550	0.300	0.150	1
5	1	0.500	0.250	0.250	-1	0.600	0.250	0.150	1	0.600	0.250	0.150	0

	2	0.500	0.350	0.150	-1	0.400	0.250	0.350	1	0.500	0.350	0.150	0
6	1	0.500	0.250	0.250	0	0.550	0.300	0.150	-1	0.400	0.350	0.250	1
	2	0.550	0.300	0.150	0	0.400	0.250	0.350	-1	0.500	0.250	0.250	1
Determinant		6.87E-20				2.09E-22				2.31E-23			

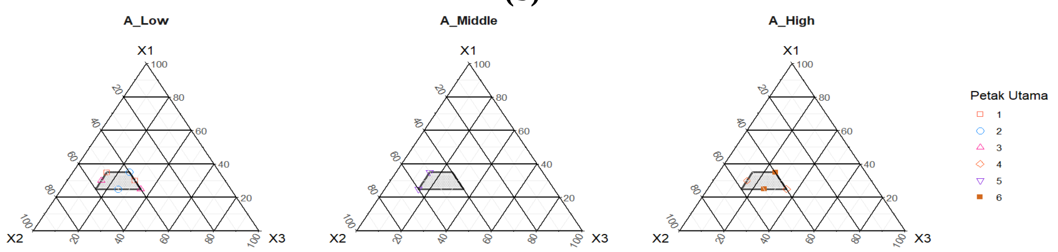
The design results in scenario 1, namely n = 12 with η = 1, obtained that the total amount in level height (A_{Hight}) was in the 1st main plot, and in the 3rd main plot, the total amount ini level medium (A_{Medium}) was in the 2nd and 4th main plots, while for the total in level low (A_{low}) was in the 6th main plot. The optimization results obtained with $\eta = 1$ of the 12 formulations showed that in the level height, there were four optimal points; in the medium total mixture level, there were two optimal points; while in the low total mixture level, there were six optimal points. The results show that the 1st main plot produces two different formulation points with the total amount of mixture at a high level. The first formulation is 40% of ingredient 1, 35% of ingredient 2, and 25% of ingredient 3. The second formulation is 60% of ingredient 1, 25% of ingredient 3, and 60% of ingredient 3. These numbers indicate that if a product is to be developed with a specific total mixture, for example, 50 g (low), 100 g (medium), or 150 g (high), then the optimal points obtained from the design results can be used as a reference to determine the formulation. As an illustration, one of the optimal points at the high total mixture level was 40% for ingredient 1, 35% for ingredient 2, and 25% for ingredient 3. If this formulation is applied to the 150-gram variant, then the measurements of each ingredient are 60 grams of ingredient 1, 52.5 grams of ingredient 2, and 37.5 grams of ingredient 3.

Another Example based on the results in Table 2, to produce a product at a low total mixture level (50 g), one of the optimal points indicates a proportion of 40% of ingredient 1, 25% of ingredient 2, and 35% of ingredient 3, resulting in a composition of 20 g of ingredient 1, 12.5 grams of ingredient 2, and 17.5 grams of ingredient 3. It should be noted that at each high mixture level (A_{High}, A_{Medium}, and A_{Low}), each component has the following proportion limits: $0.40 \leq x_1 \leq 0.60$ for material 1, $0.25 \leq x_2 \leq 0.35$ for material 2, and $0.15 \leq x_3 \leq 0.35$ for material 3. These limits are used in the design process such that all the resulting formulations remain within the technically and practically acceptable range. In Full, The D-Optimal design points based on the point exchange algorithm in scenario one with $n = 12$ will be represented in the design area, which will illustrate the optimal point positions at each total mixture amount (see Figure 4).

(a)



(b)



(c)

Figure 4. The D-Optimal design region for $n = 16$, with the parameter values set at (a) $\eta = 1$, (b) $\eta = 5$, and (c) $\eta = 10$.

Figure 4 shows that the optimal points at $\eta = 1, \eta = 5,$ and $\eta = 10$ when $n = 12$ are the edge points located in the design area with a total mixture composition at a high level of 6 points, a medium level of 2 points, and a low level of 4 points.

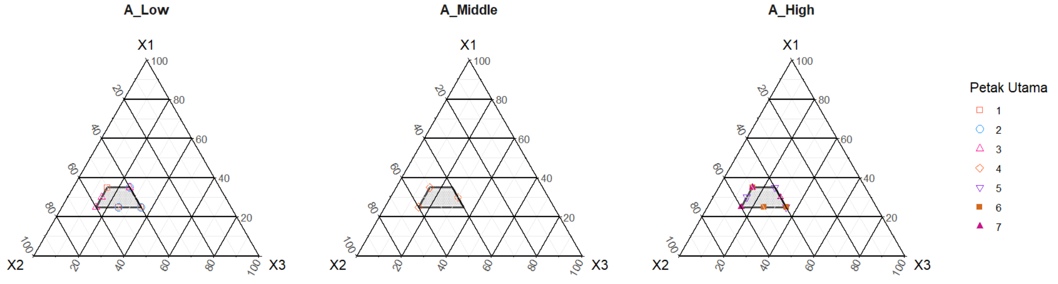
3.2.2. D-Optimal Design for Mixture Amount Experiment with $n = 21$

The experimental structure with $n = 21$ was analyzed using the same approach, with an arrangement of 7 main plots and 3 subplots. Optimal points based on the D-Optimal criterion of this structure provided additional information that when the experimental units were increased the determinant values when $\eta = 1, \eta = 5,$ and $\eta = 10$ also increased. The optimal design at $n = 21$ can be seen in Table (3) with determinants of 5.14E-17, 4.89E-19, 5.74E-20 respectively.

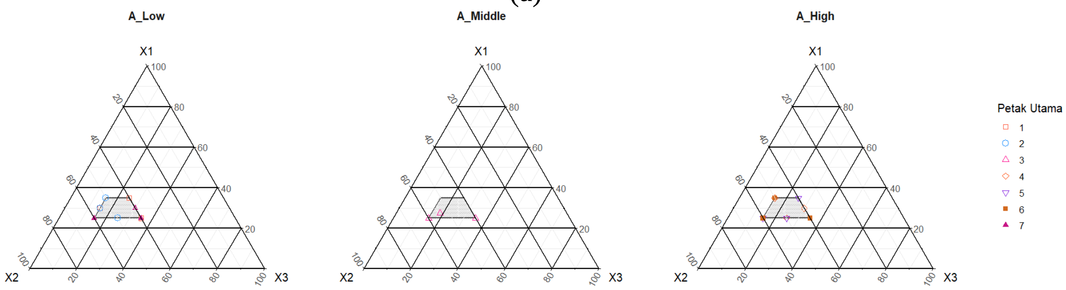
Table 3. D-Optimal results for MAE employing the algorithm with $n = 21$

Main-plot	Sub-Plot	$\eta = 1$				$\eta = 5$				$\eta = 10$			
		x_1	x_2	x_3	A	x_1	x_2	x_3	A	x_1	x_2	x_3	A
1	1	0.500	0.350	0.150	-1	0.400	0.350	0.250	-1	0.500	0.250	0.250	-1
1	2	0.400	0.250	0.350	-1	0.400	0.250	0.350	-1	0.550	0.300	0.150	-1
1	3	0.500	0.250	0.250	-1	0.550	0.300	0.150	-1	0.400	0.250	0.350	-1
2	1	0.400	0.350	0.250	-1	0.550	0.300	0.150	-1	0.400	0.350	0.250	-1
2	2	0.400	0.250	0.350	-1	0.500	0.350	0.150	-1	0.500	0.350	0.150	-1
2	3	0.500	0.250	0.250	-1	0.500	0.250	0.250	-1	0.400	0.250	0.350	-1
3	1	0.400	0.350	0.250	-1	0.538	0.275	0.187	0	0.600	0.250	0.150	1
3	2	0.550	0.300	0.150	-1	0.400	0.250	0.350	0	0.475	0.300	0.225	1
3	3	0.600	0.250	0.150	-1	0.600	0.250	0.150	0	0.400	0.350	0.250	1
4	1	0.500	0.350	0.150	0	0.500	0.350	0.150	1	0.400	0.250	0.350	0
4	2	0.400	0.300	0.300	0	0.500	0.250	0.250	1	0.500	0.350	0.150	0
4	3	0.600	0.250	0.150	0	0.400	0.300	0.300	1	0.500	0.250	0.250	0
5	1	0.400	0.250	0.350	1	0.500	0.250	0.250	1	0.500	0.250	0.250	1
5	2	0.400	0.350	0.250	1	0.400	0.350	0.250	1	0.400	0.300	0.300	1
5	3	0.550	0.300	0.150	1	0.600	0.250	0.150	1	0.600	0.250	0.150	1
6	1	0.400	0.250	0.350	1	0.600	0.250	0.150	1	0.500	0.350	0.150	1
6	2	0.500	0.350	0.150	1	0.400	0.250	0.350	1	0.400	0.250	0.350	1
6	3	0.500	0.250	0.250	1	0.500	0.350	0.150	1	0.400	0.350	0.250	1
7	1	0.600	0.250	0.150	1	0.400	0.300	0.300	-1	0.600	0.250	0.150	-1
7	2	0.400	0.300	0.300	1	0.400	0.250	0.350	-1	0.500	0.350	0.150	-1
7	3	0.500	0.350	0.150	1	0.600	0.250	0.150	-1	0.400	0.300	0.300	-1
Determinant		5.14E-17				4.89E-19				5.74E-20			

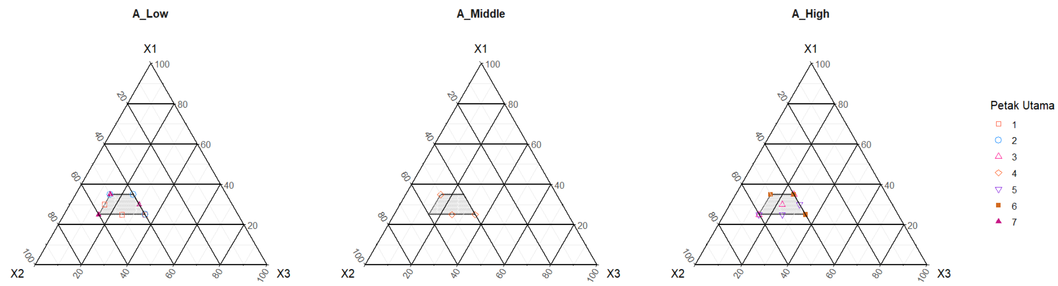
The interpretation of Table 3 aligns with that of Table 2, with the primary distinction being the number of experimental units (n). To facilitate the identification of the optimal point, Figure (5) illustrates this point within the design region's structure. The optimal point for the $n = 21$ structure generally corresponds to that of $n = 12$, specifically at the edge point of the optimal design for high, medium, or low levels.



(a)



(b)



(c)

Figure 5. The D-Optimal design region for $n = 21$, with the parameter values set at (a) $\eta = 1$, (b) $\eta = 5$, and (c) $\eta = 10$.

3.2.3. D-Optimal Design for Mixture Amount Experiment with $n = 32$

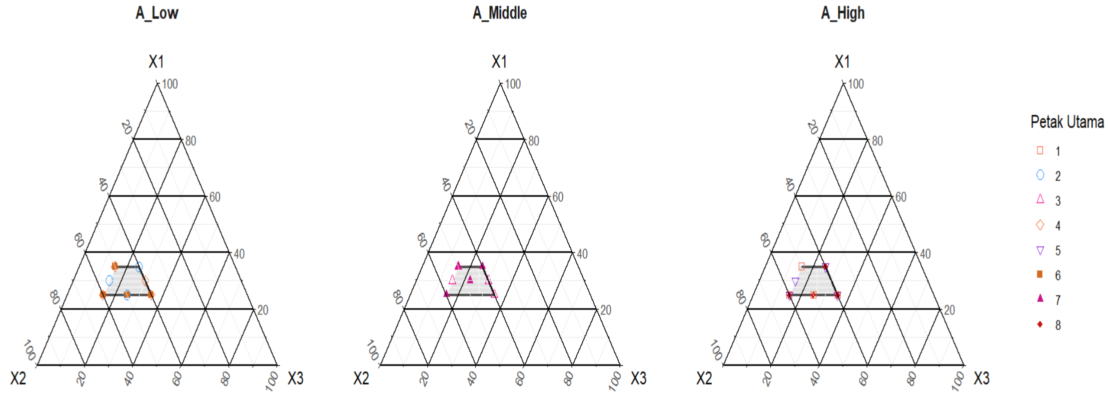
In the structure with the number of experimental units, $n = 32$, an arrangement of 8 main plots and four sub-plots is used. This analysis examines the effect of adding units on the efficiency of the information obtained. Using the same algorithm, the design resulting from this structure is expected to provide the maximum contribution to the quality of model estimation in the context of D-optimal design. Optimal points at different mixture amounts with $n = 32$ can be seen in Table 4.

Table 4. D-Optimal results for MAE employing the algorithm with $n = 32$

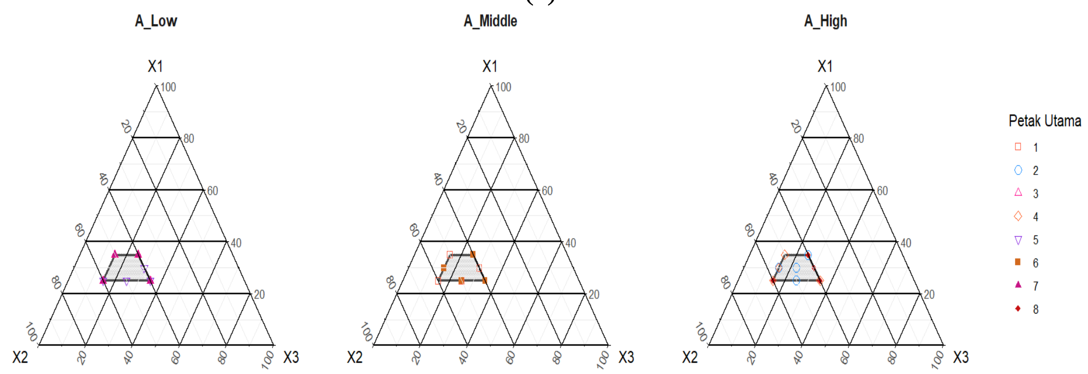
Main-plot	Sub-Plot	$\eta = 1$				$\eta = 5$				$\eta = 10$			
		x_1	x_2	x_3	A	x_1	x_2	x_3	A	x_1	x_2	x_3	A
1	1	0.600	0.250	0.150	1	0.500	0.350	0.150	0	0.600	0.250	0.150	-1
1	2	0.500	0.250	0.250	1	0.500	0.250	0.250	0	0.400	0.250	0.350	-1
1	3	0.400	0.250	0.350	1	0.400	0.300	0.300	0	0.500	0.350	0.150	-1
1	4	0.500	0.350	0.150	1	0.600	0.250	0.150	0	0.400	0.300	0.300	-1
2	1	0.550	0.300	0.150	-1	0.400	0.350	0.250	1	0.550	0.300	0.150	-1
2	2	0.400	0.350	0.250	-1	0.550	0.300	0.150	1	0.400	0.250	0.350	-1

2	3	0.400	0.350	0.250	-1	0.500	0.250	0.250	1	0.500	0.250	0.250	-1
2	4	0.500	0.250	0.250	-1	0.475	0.300	0.225	1	0.400	0.350	0.250	-1
3	1	0.400	0.250	0.350	0	0.500	0.350	0.150	-1	0.500	0.350	0.150	0
3	2	0.500	0.350	0.150	0	0.600	0.250	0.150	-1	0.600	0.250	0.150	0
3	3	0.550	0.300	0.150	0	0.400	0.250	0.350	-1	0.500	0.250	0.250	0
3	4	0.400	0.300	0.300	0	0.400	0.350	0.250	-1	0.400	0.300	0.300	0
4	1	0.600	0.250	0.150	-1	0.600	0.250	0.150	1	0.500	0.250	0.250	1
4	2	0.400	0.300	0.300	-1	0.400	0.250	0.350	1	0.500	0.350	0.150	1
4	3	0.400	0.250	0.350	-1	0.550	0.300	0.150	1	0.400	0.350	0.250	1
4	4	0.500	0.350	0.150	-1	0.500	0.350	0.150	1	0.600	0.250	0.150	1
5	1	0.550	0.300	0.150	1	0.500	0.250	0.250	-1	0.500	0.350	0.150	-1
5	2	0.600	0.250	0.150	1	0.400	0.250	0.350	-1	0.400	0.250	0.350	-1
5	3	0.400	0.350	0.250	1	0.600	0.250	0.150	-1	0.400	0.300	0.300	-1
5	4	0.400	0.250	0.350	1	0.400	0.300	0.300	-1	0.600	0.250	0.150	-1
6	1	0.500	0.250	0.250	-1	0.400	0.250	0.350	0	0.600	0.250	0.150	1
6	2	0.400	0.250	0.350	-1	0.550	0.300	0.150	0	0.550	0.300	0.150	1
6	3	0.500	0.350	0.150	-1	0.400	0.350	0.250	0	0.400	0.350	0.250	1
6	4	0.600	0.250	0.150	-1	0.500	0.250	0.250	0	0.400	0.250	0.350	1
7	1	0.600	0.250	0.150	0	0.500	0.350	0.150	-1	0.500	0.350	0.150	0
7	2	0.475	0.300	0.225	0	0.400	0.250	0.350	-1	0.400	0.350	0.250	0
7	3	0.500	0.350	0.150	0	0.400	0.350	0.250	-1	0.550	0.300	0.150	0
7	4	0.400	0.350	0.250	0	0.600	0.250	0.150	-1	0.600	0.250	0.150	0
8	1	0.600	0.250	0.150	1	0.400	0.250	0.350	1	0.400	0.250	0.350	1
8	2	0.400	0.250	0.350	1	0.400	0.350	0.250	1	0.400	0.250	0.350	1
8	3	0.400	0.350	0.250	1	0.400	0.300	0.300	1	0.500	0.250	0.250	1
8	4	0.500	0.250	0.250	1	0.600	0.250	0.150	1	0.500	0.350	0.150	1
Determinant		2.87E-15				3.22E-17				5.11E-18			

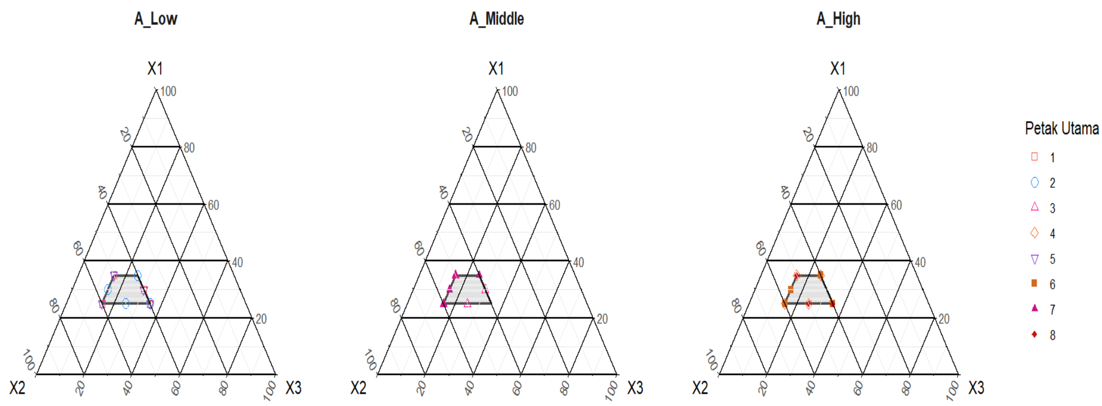
Table 4 presents the optimal points at various blend combinations and factor A levels for three parameter values of η $(1, 5$, and $10)$. The resulting combinations of blend proportions (x_1, x_2, x_3) and total blend amount factor show a rich variation in the design space, reflecting the flexibility of the design to parameter changes. The information matrix determinant values for each η value indicated that the highest information efficiency was achieved at η = 1, with a determinant value 2.87E-15. However, there is a sharp decline at η = 5 and η = 10, with determinant values of 3.22E-17 and 5.11E-18, respectively. This decrease indicates that the larger the value of η, the less effective the information can be obtained from the design. Overall, these results suggest that adding units to the experimental structure can improve the efficiency of model estimation, especially when the η value is at a lower level. A visual interpretation of the distribution of points in the design space can be seen in Figure (6).



(a)



(b)



(c)

Figure 6. The D-Optimal design region for $n = 32$, with the parameter values set at (a) $\eta = 1$, (b) $\eta = 5$, and (c) $\eta = 10$.

Figure 5 on the $n = 32$ structure shows that for values of $\eta = 1$ and $\eta = 5$, the optimal points are generally distributed at the edges of the design area, but there are also points in the centre. Meanwhile, at $\eta = 10$, the optimal points are only at the edges.

3.3 Practical and Applied Implications of Optimal Design

Based on the entire analysis series in this study, the mixture design involving the total mixture amount variable can be developed effectively through a modified split-plot approach with the D-optimal criterion. Compiling candidate design points using the cox-direction method allowed us to obtain a set of points representing the entire variation in material composition according to each case's limitations. Visualization of the design area shows that the optimal points tend to be on the sides or edges of the design area, particularly at high and low total mixture levels.

Furthermore, the results of the efficiency evaluation through the determinant value for various structures of the number of experimental units ($n = 12, 21$, and 32) and three assumptions of the variance ratio value η $(1, 5$, and $10)$ show that increasing the number of experimental units can improve the efficiency and stability of the design, as indicated by the increasing value of the D-Optimal determinant. This shows that the point-selection strategy using the point-exchange algorithm can consistently produce optimal formulations under various experimental conditions.

Thus, this discussion not only underscores methodological considerations but also demonstrates the applicability of the proposed design within a practical context, specifically in the food industry. In the formulation of food products, such as bread, beverages, or snacks, the total quantity of ingredients is frequently constrained by production capacity, nutritional value, or consumer preferences. Consequently, this design approach is particularly pertinent as it facilitates the exploration of ingredient combinations within defined total mixture constraints. By employing this experimental design, researchers and practitioners can ascertain the optimal ingredient proportions without the necessity of conducting numerous physical trials, thereby conserving time and reducing costs in the product development process. Furthermore, this approach offers a systematic framework to support data-driven decision-making in food product formulation strategies.

4. CONCLUTION

This study shows that a modified split-plot approach using the D-Optimal criteria can effectively develop an optimal mixture design involving the total mixture variable. Through point-exchange algorithms and variations in the experimental unit structure, this design can produce optimal formulation points spread across various combinations of the material proportions. The efficiency evaluation through the determinant value of the information matrix on three experimental structures ($n = 12, 21$, and 32) with variance ratios $\eta = 1, 5$, and 10 shows that increasing the number of experimental units improves the efficiency and stability of the design, especially when η is lower. The optimal points are generally located at the edges of the design region, particularly at the high and low total mixture levels. However, the points in the middle also appeared in the structures with more experiments. These results indicate that the point-selection strategy using the point-exchange algorithm consistently produced efficient and representative designs. Thus, this approach provides flexibility and a strong foundation for formulating optimal designs in subsequent stages of product development or mixture optimization that considers more than one level of the total composition.

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