International Journal of Sciences: Basic and Applied Research (IJSBAR)

ISSN 2307-4531
(Print & Online)

http://gssrr.org/index.php?journal=JournalOfBasicAndApplied

D-Optimal Design for Mixture Amount Experiment Involving Split-Plot Design

A. Muthiah Nur Angriany[a], Anik Djuraidah[b], Utami Dyah Syafitri[c*]

[a,b,c]IPB University, Jl. Raya Dramaga, Babakan, Dramaga District, Bogor City, West Java, Indonesia

[c]Email: utamids@apps.ipb.ac.id

Abstract

A mixture amount experiment (MAE) is a design that depends on the proportions of the ingredients and the total amounts. The classical MAE contains the classical mixture experiment on each total amount. Consequently, complete randomization is challenging to implement in MAE, so a split-plot design approach was proposed. In the MAE, the whole plot factor is the total amount of mixtures, while the subplot factor is the composition of the ingredients. Another problem in the MAE is if the number of ingredients and total amounts increase, the number of runs increases. The split-plot design with an optimal design approach was proposed. The study aimed to develop a point-exchange algorithm with a split-plot design approach. The case study used is a mixed design consisting of three ingredients and two total amounts of mixtures. The results obtained are that the algorithm compiled in this study produces optimal design points, namely the edge points in the design region.

Keywords: D-optimal; mixture amount experiment; split-plot design.

1. Introduction

Experimental design is a series of tests to observe and identify changes in the response output caused by changes in the input variables of a process [1]. Experimental designs are widely used in the industrial sector, which determines formulations in producing a product. A design that can be used for the formulation is a mixture experiment. The mixture experiment is a design that involves two or more components blended with the same or different proportions [2].

---

* Corresponding author.

The response of a mixture experiment depends only on the proportion of ingredients and not on the total amount of components. This condition is usually met in practice by keeping the entire components in all blends fixed. In some cases, the optimal compositions differ among different total amounts. The design that involves the total amount of the mixture is called the Mixture amount Experiment (MAE). The response is assumed to depend on the proportion of ingredients and the total amount of the mixture [3]. Reference [2] discuss producing complete and fractional designs in constrained and unrestricted for MAE. This study uses a model that explains the effect of amount on the blending properties of components by considering the regression coefficient of the usual mixture models proposed by [4]. In addition, Reference [5] developed a new model with a smaller number of parameters in the mixture amount experiment based on the A-Optimal and D-optimal criteria for estimating the model parameters.

In the MAE, the composition of the mixture is carried out on each total amount of the mixture. This situation has a consequence that complete randomization is hard to implement. The approach that can use is the split-plot design, which is a design that arises when structural randomization cannot be carried out [6]. In the Mixture amount experiment, the whole plot factor is the total amount of mixtures, while the subplot factors have the compositions of the mixture experiments. The use of split-plot modification in mixture experiments has been studied by [7], who researched the mixture process variable split-plot (MPVSP). The MPSVP design performed limited randomization between the mixture components and process variables.

The total amount of mixture variable can cause the experimental unit to increase, resulting in increased production costs. An alternative solution to handle this situation is an optimal design. Optimal design is part of an experimental design that is more flexible in building a design according to actual conditions. Creating an optimal design is based on a specific optimality criterion [8]. In this study, the optimal design criterion to be used is the D-optimality criterion, which is a criterion that emphasizes the quality of parameter estimates by maximizing the determinants of the information matrix [9]. An algorithm is needed to build the optimal design [10]. One of the algorithms is a point-exchange algorithm. The point-exchange algorithm is an algorithm that aims to improve the starting design by removing or adding points to the starting design. Reference [11] used a point exchange algorithm in a split-plot design. They applied it to the case of protein extraction experiments with the number and size of main plots using the D-optimality criterion. In this research, the point-exchange algorithm was proposed to obtain the D-Optimal design using a split-plot approach. To evaluate the designs, the optimal design was compared to the design from the coordinate-exchange algorithm, which already been implemented in the commercial software.

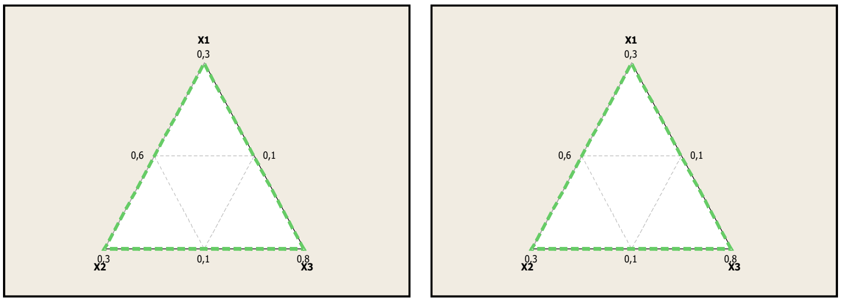
2. Methodology

2.1 A Practical Example

The case study consisted of three mixture components (x_1, x_2, x_3) and two total amounts: 50 grams and 150 grams. In addition, the case has a constraint function, namely the upper limit for each ingredient (Table 1). Figure 1 shows the constraints on each composition in the design region for each total amount of mixture. The design region is a triangular region but a part of a whole simplex.

Table 1: Constraints of each ingredient

ingredient	Constrains
1st	$x_1 \geq 0.1$
2nd	$x_2 \geq 0.1$
3rd	$x_3 \geq 0.6$



(a) (b)

Figure 1: The design region of the mixture amount experiment for (a) 50 gram (b) 150 grams

2.2 Analysis Procedure

To construct a D-optimal design, a point-exchange algorithm was developed. The guideline of the point-exchange algorithm for MAE with split-plot approach was outlined below:

1. Determine the assumption of the model mixture amount experiment. This paper used the mixture model, which was proposed by Piepel and Cornell (1987). The model involves the mixture and total amount variable model, and it is written as:

$$y_i = \beta_1^0 x_{1i} + \beta_2^0 x_{2i} + \beta_3^0 x_{3i} + \beta_4^0 x_{1i} x_{2i} + \beta_5^0 x_{1i} x_{3i} + \beta_6^0 x_{2i} x_{3i} + \{\beta_1^1 x_{1i} + \beta_2^1 x_{2i} + \beta_3^1 x_{3i} + \beta_4^1 x_{1i} x_{2i} + \beta_5^1 x_{1i} x_{3i} + \beta_6^1 x_{2i} x_{3i}\} A + \varepsilon \quad (1)$$

The model involves six parameters of the mixture experiments and the interaction between them and the total amount of mixture. In total, there are 12 model parameters

2. Determine the model of the mixture amount experiment using a split-plot design. The model which written in matrix notation is shown below [12]:

$$\boldsymbol{y} = \boldsymbol{X\beta} + \boldsymbol{Z\gamma} + \boldsymbol{\varepsilon} \quad (2)$$

where $\boldsymbol{X}$ represents model matrix $n \times p$ containing all whole plots and subplots. $\boldsymbol{\beta}$ represents model parameter vector $p \times 1$. The matrix $\boldsymbol{Z}$ represents model matrix $n \times b$ of matrix zeroes and ones in the entire plot. The vector $\boldsymbol{\gamma}$ represents random effects of the b whole plot, and the vector $\boldsymbol{\varepsilon}$ represents random errors. The assumptions of the model are:

$$\gamma \sim N(\mathbf{0}_b, \sigma_\gamma^2 \boldsymbol{I}_b)$$

$$\varepsilon \sim N(\mathbf{0}_n, \sigma_\varepsilon^2 \boldsymbol{I}_n)$$

$$cov(\gamma, \varepsilon) = \mathbf{0}_{nxb}$$

Under these assumptions, the covariance matrix of the observations, $cov\ (Y)$, can be written as:

$$\boldsymbol{V} = \sigma_\varepsilon^2 \boldsymbol{I}_n + \sigma_\gamma^2 \boldsymbol{Z}\boldsymbol{Z}` = \sigma_\varepsilon^2 (I_n + \eta \boldsymbol{Z}\boldsymbol{Z}') \qquad (3)$$

where $\eta = \frac{\sigma_\gamma^2}{\sigma_\varepsilon^2}$ is a measure of the extent to which the same whole plot's observations are correlated and is referred to as variance component ratio. The maximum likelihood estimator of the unknown model parameter $\boldsymbol{\beta}$ in the Equations (2) is the generalized least squares estimator [13] are:

$$\widehat{\boldsymbol{\beta}} = (\boldsymbol{X}'\boldsymbol{V}^{-1}\boldsymbol{X})^{-1}\boldsymbol{X}'\boldsymbol{V}^{-1}\boldsymbol{y} \qquad (4)$$

with covariance matrix

$$cov(\boldsymbol{\beta}) = (\boldsymbol{X}'\boldsymbol{V}^{-1}\boldsymbol{X})^{-1} = \sigma_\varepsilon^2 \{\, \boldsymbol{X}'(\boldsymbol{I}_n + \eta \boldsymbol{Z}\boldsymbol{Z}')^{-1}\boldsymbol{X}\}^{-1} \qquad (5)$$

The information matrix on the unknown model parameter vector $\boldsymbol{\beta}$ is given by

$$M = \boldsymbol{X}'\widehat{\boldsymbol{V}}^{-1}\boldsymbol{X} = \sigma_\varepsilon^{-2}\, \boldsymbol{X}'(\boldsymbol{I}_n + \eta \boldsymbol{Z}\boldsymbol{Z}')^{-1}\boldsymbol{X} \qquad (6)$$

3. Determine the value of η. Model in the Equations (2) use variance ratio of whole plot and subplot. In this case, the variance ratio was set to $\sigma_\gamma^2 = 1{,}5{,}10$ and $\sigma_\varepsilon^2 = 1$

4. Develop a point exchange Algorithm adapted from Goss and Vandebroek (2003). Steps to construct design based on the algorithm are:

a) Define a list of the candidate set following the cox-direction formula. Let the first component as x_i and other components as x_j, so changes for every component can be calculated [14]:

$$\tilde{x}_i = x_i + \delta \text{ dan } \tilde{x}_j = x_j - \frac{x_j \delta}{1 - x_i} \qquad (7)$$

for $i \neq j$ dan $i = 1,2, \ldots, p$, p is the parameter model number.

b) Determine the number of Whole Plot (b) and subplot (k_i). As the number of parameters was 12, whereas the number of runs was set to 16.

c) Define a starting design. The starting design was composed of design points which selected randomly from the candidate set.

d) Calculate the determinant of the information matrix and prediction variance from the starting design

e) Calculate the prediction variance of the candidate set.

f) Exchange points on the subplots of the starting design with the points of the candidate set. The smallest prediction variance of the starting design was replaced by the largest prediction variance of the candidate set.

g) Step (f) is repeated until it converges or there is no more significant change in the value of the determinant

h) Exchange the levels of the whole-plot factor settings.

i) Repeat step (h) until no more changes are possible

5. Repeat Step (4) 5000 times

6. Save the design that produces the largest value of the determinant of the information matrix

7. Compare the results of the Point exchange Algorithm with the candidate-set-free coordinate-exchange algorithms described in Jones and Goos [6] in the software package JMP

8. Evaluating the design results

This study uses an evaluation of the D-efficiency design. The formula to calculate D-efficiency are [15]:

$$Deff = \left(\frac{|\mathbf{M}_1|}{|\mathbf{M}_2|}\right)^{\frac{1}{p}} \qquad (8)$$

where p is the number of parameters in the model, $\mathbf{M}_1$ is the first design information matrix, and $\mathbf{M}_2$ is the second design information matrix

3. Result and Discussion

Nineteen candidate points were generated using cox-direction based on equation (7) shown in Tabel 2. The candidate points in the case can be described geometrically in the design region as Figure 2. The candidate set consists of 19 points in each total amount of mixture (50 grams, 150 grams) so that the whole candidate point was 19×2=38. The candidate set involves the corner points, the edges points, and the axial points.

Table 3 shows the results of the D-Optimal design on the mixture amount experiment based on the point-exchange algorithm.

The design point value (x_1, x_2, x_3) indicates the number of mixture proportions for each ingredient, and A indicates the total amount of mixture. The value of the determinant obtained at $\eta = 1$ was 5.90E-07, $\eta = 5$ was 2.55E-08, and $\eta = 10$ was 6.99E-09, respectively.

Table 2: Candidate set of mixture

No.	Proportion		
	x_1	x_2	x_3
1	0.20	0.20	0.60
2	0.10	0.25	0.65
3	0.20	0.15	0.65
4	0.25	0.15	0.60
5	0.15	0.10	0.75
6	0.25	0.10	0.65
7	0.23	0.13	0.63
8	0.10	0.15	0.75
9	0.10	0.20	0.70
10	0.17	0.17	0.67
11	0.30	0.10	0.60
12	0.15	0.25	0.60
13	0.15	0.15	0.70
14	0.10	0.30	0.60
15	0.13	0.23	0.63
16	0.10	0.10	0.80
17	0.15	0.20	0.65
18	0.20	0.10	0.70
19	0.13	0.13	0.73

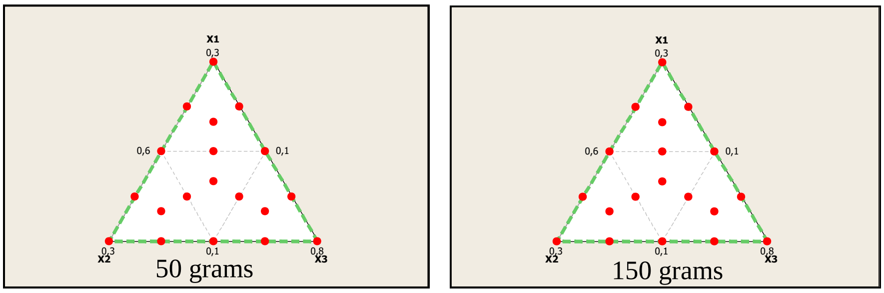
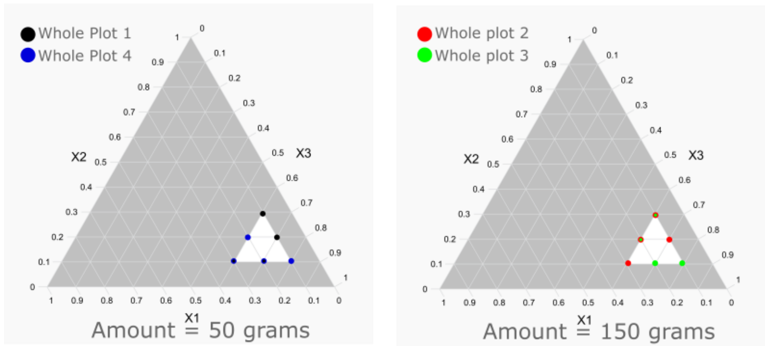


Figure 2: Candidate Set in each total amount of mixture

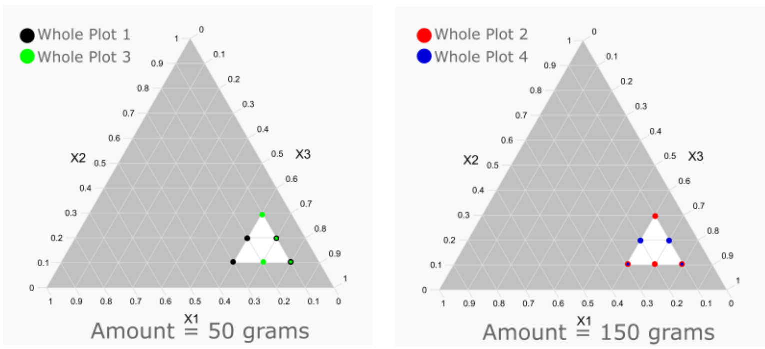
Table 3: D-Optimal design for MAE based on the point exchange algorithm

Whole Plot	subplot	$\eta = 1$				$\eta = 5$				$\eta = 10$			
		x_1	x_2	x_3	A	x_1	x_2	x_3	A	x_1	x_2	x_3	A
1	1	0.30	0.10	0.60	50	0.10	0.20	0.70	50	0.30	0.10	0.60	50
	1	0.10	0.30	0.60	50	0.20	0.20	0.60	50	0.10	0.30	0.60	50
	1	0.10	0.20	0.70	50	0.10	0.10	0.80	50	0.10	0.10	0.80	50
	1	0.20	0.10	0.70	50	0.30	0.10	0.60	50	0.20	0.10	0.70	50
2	2	0.30	0.10	0.60	150	0.20	0.10	0.70	150	0.10	0.30	0.60	150
	2	0.20	0.20	0.60	150	0.10	0.30	0.60	150	0.30	0.10	0.60	150
	2	0.10	0.20	0.70	150	0.10	0.10	0.80	150	0.10	0.20	0.70	150
	2	0.10	0.30	0.60	150	0.30	0.10	0.60	150	0.20	0.10	0.70	150
3	3	0.10	0.30	0.60	150	0.10	0.30	0.60	50	0.10	0.10	0.80	50
	3	0.10	0.10	0.80	150	0.10	0.10	0.80	50	0.20	0.20	0.60	50
	3	0.20	0.20	0.60	150	0.20	0.10	0.70	50	0.10	0.20	0.70	50
	3	0.20	0.10	0.70	150	0.10	0.20	0.70	50	0.10	0.30	0.60	50
4	4	0.20	0.10	0.70	50	0.20	0.20	0.60	150	0.10	0.30	0.60	150
	4	0.10	0.10	0.80	50	0.10	0.10	0.80	150	0.10	0.10	0.80	150
	4	0.30	0.10	0.60	50	0.30	0.10	0.60	150	0.30	0.10	0.60	150
	4	0.20	0.20	0.60	50	0.10	0.20	0.70	150	0.20	0.20	0.60	150
D-Optimal		5.90E-07				2.55E-08				6.99E-09			

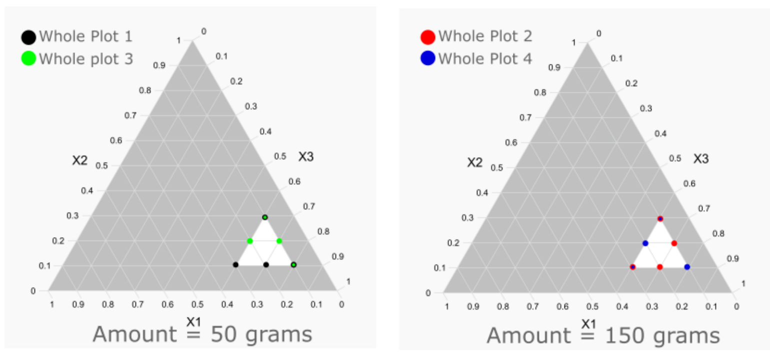
Figure 3 shows the D-optimal design region for MAE based on the point-exchange algorithm. The results of the D-Optimal design at $\eta = 1$ $\eta = 5$ dan $\eta = 10$ have the same structure. If two whole plots are joined, then the design is similar to a {3,2} simplex-lattice design in which involves the corner points and the middle of edges points. Furthermore, the nice things of the design is each whole plot consists of three or four design points that already known the optimal design points in the literature.



(a)



(b)



(c)

Figure 3: The D-Optimal design region based on the point exchange algorithm with a value of

(a) $\eta = 1$, (b) $\eta = 5$, dan (c) $\eta = 10$

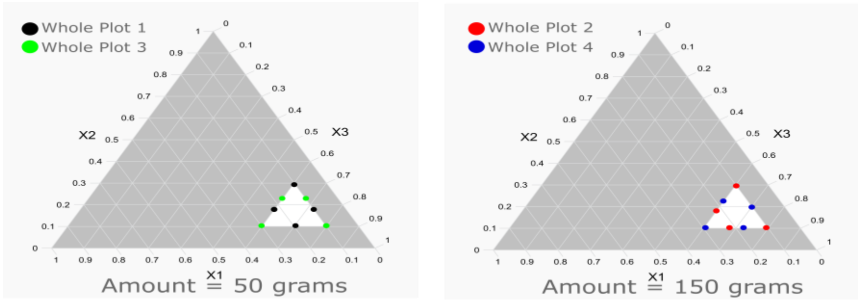
Table 4 shows the designs of the D-Optimal for MAE by the coordinate-exchange algorithm with values $\eta = 1$, $\eta = 5, \eta = 10$, represented in Figure 4. The D-optimal design points resulting from the coordinate exchange algorithm have the same structure as the point-exchange algorithm in Table 2 for $\eta = 1$, but not for $\eta = 5$ and $\eta = 10$. For $\eta = 1$, the designs based on the coordinate-exchange algorithm have the points slightly off the middle of the edges rather than at the edge centroids themselves. This is because the candidate set of the point-exchange algorithm only involved the corner points, the edges points, and the axial points. For $\eta = 5$ and $\eta = 10$, the design points resulted from the coordinate-exchange algorithm were different from the ones from the point-exchange algorithm. The designs consist of four different design points. To evaluate the two designs, D-efficiencies was used. Table 5 shows the D-efficiencies of the D-optimal MA designs in various η. Based on Table 5, the value of D-efficiency < 1 for $\eta = 1, \eta = 5, \eta = 10$ so that the results of the coordinate-exchange algorithm based on the software package JMP on the mixture amount experiment are slightly more efficient when compared to the point-exchange algorithm. The D-optimal for MAE of the coordinate-exchange algorithm was ± 3% better than the design of the proposed point-exchange algorithm in terms of D-efficiency. Hence, the proposed point-exchange algorithm is an alternative algorithm for finding the D-optimal for MAE. In conclusion, the coordinate-exchange algorithm resulted the different design from the point-exchange algorithm for $\eta = 5$, and $\eta = 10$, however the two designs are optimal.

Table 4: D-optimal design based on the coordinate-exchange algorithms described in the software package JMP

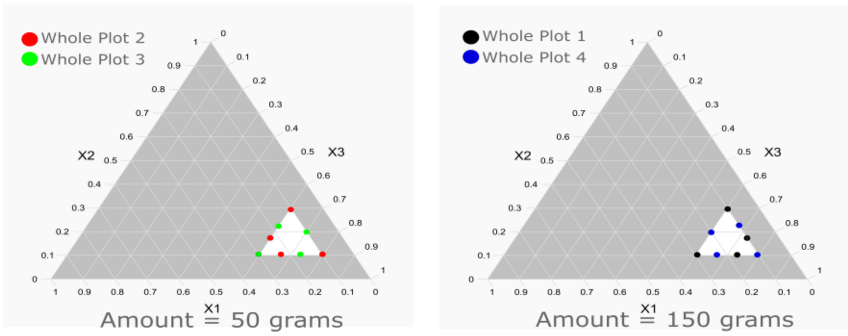
Whole Plot	Sub Plot	$\eta = 1$				$\eta = 5$				$\eta = 10$			
		x_1	x_2	x_3	A	x_1	x_2	x_3	A	x_1	x_2	x_3	A
1	1	0.10	0.18	0.72	50	0.30	0.10	0.60	150	0.22	0.18	0.60	150
	1	0.22	0.18	0.60	50	0.10	0.30	0.60	150	0.10	0.30	0.60	150
	1	0.20	0.10	0.70	50	0.10	0.18	0.72	150	0.20	0.10	0.70	150
	1	0.10	0.30	0.60	50	0.18	0.10	0.72	150	0.10	0.18	0.72	150
2	2	0.22	0.18	0.60	150	0.10	0.30	0.60	50	0.10	0.10	0.80	50
	2	0.22	0.10	0.68	150	0.10	0.10	0.80	50	0.22	0.18	0.60	50
	2	0.10	0.10	0.80	150	0.22	0.18	0.60	50	0.10	0.30	0.60	50
	2	0.10	0.30	0.60	150	0.22	0.10	0.68	50	0.22	0.10	0.68	50
3	3	0.30	0.10	0.60	50	0.30	0.10	0.60	50	0.10	0.10	0.80	150
	3	0.10	0.10	0.80	50	0.10	0.20	0.70	50	0.18	0.22	0.60	150
	3	0.10	0.22	0.68	50	0.18	0.10	0.72	50	0.10	0.22	0.68	150
	3	0.18	0.22	0.60	50	0.18	0.22	0.60	50	0.30	0.10	0.60	150
4	4	0.18	0.10	0.72	150	0.22	0.10	0.68	150	0.18	0.10	0.72	50
	4	0.10	0.20	0.70	150	0.10	0.10	0.80	150	0.10	0.20	0.70	50
	4	0.30	0.10	0.60	150	0.10	0.22	0.68	150	0.17	0.23	0.60	50
	4	0.18	0.22	0.60	150	0.20	0.20	0.60	150	0.30	0.10	0.60	50
D-Optimal		7.64E-07				3.78E-08				9.72E-09			

Table 5: D-Efficiency Calculation Results

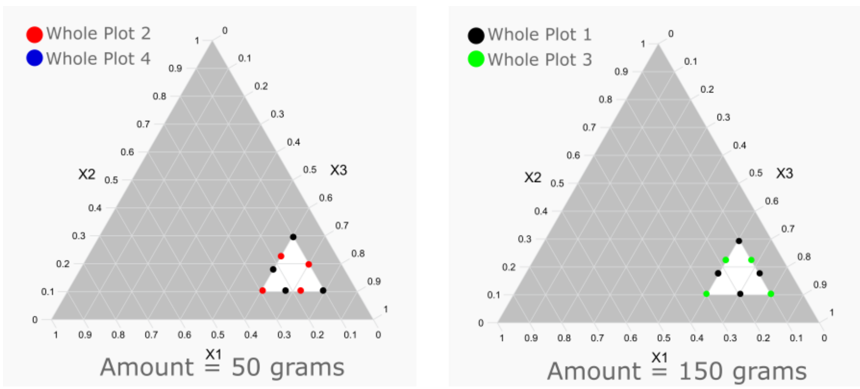
variance ratio of whole plot and subplot	$\eta = 1$	$\eta = 5$	$\eta = 10$
Point Exchange algorithms (M_1)	3.17E-17	3.85E-19	7.10E-20
Coordinate exchange algorithms (M_2)	4.80E-17	6.83E-19	9.25E-20
D-efficiency	0.966	0.953	0.978



(a)



(b)



(c)

Figure 4: The D-optimal design region based on the *coordinate exchange* algorithm with a value of

(a) $\eta = 1$, (b) $\eta = 5$, dan (c) $\eta = 10$

4. Conclusions

The point-exchange algorithm developed was successful in finding the D-optimal for MAE with a split-plot approach. Although the resulted designs are different, but the design from the point-exchange algorithm was as efficient as the one of the coordinate -exchange algorithm already implemented in commercial software.

Acknowledgments

The authors are incredibly grateful to the Ministry of Research, Technology, and Higher Education of the Republic of Indonesia for financial support. Then, the authors also thank our partnership company for funding and collaboration in this research.

References

[1] D. C. Montgomery, Design and analysis of experiments. John wiley & sons, 2001.

[2] G. F. Piepel and J. A. Cornell, “Designs for mixture-amount experiments,” J. Qual. Technol., vol. 19, no. 1, pp. 11–28, 1987.

[3] G. F. Piepel, “Models and designs for generalizations of mixture experiments where the response depends on the total amount,” PhD Thesis, University of Florida, 1985.

[4] H. Scheffé, “Experiments with mixtures,” J. R. Stat. Soc. Ser. B Methodol., vol. 20, no. 2, pp. 344–360, 1958.

[5] M. Pal and N. K. Mandal, “Optimum designs for estimation of parameters in a quadratic mixture-amount model,” Commun. Stat.-Theory Methods, vol. 41, no. 4, pp. 665–673, 2012.

[6] B. Jones and P. Goos, “A candidate-set-free algorithm for generating D-optimal split-plot designs,” J. R. Stat. Soc. Ser. C Appl. Stat., vol. 56, no. 3, pp. 347–364, 2007.

[7] S. M. Kowalski, J. A. Cornell, and G. G. Vining, “Split-plot designs and estimation methods for mixture experiments with process variables,” Technometrics, vol. 44, no. 1, pp. 72–79, 2002.

[8] P. Goos, The optimal design of blocked and split-plot experiments, vol. 164. Springer Science & Business Media, 2012.

[9] W. F. Smith, Experimental design for formulation. SIAM, 2005.

[10] F. Triefenbach, “Design of experiments: the D-optimal approach and its implementation as a computer algorithm,” Thesis Bachelor Degree Inf. Commun. Technol. Umea Univ. Swed., 2008.

[11] P. Goos and M. Vanderbroek, "D-optimal split-plot designs with given numbers and sizes of whole plots," Technometrics, vol. 45, no. 3, pp. 235–245, 2003.

[12] P. Goos and A. N. Donev, "Tailor-made split-plot designs for mixture and process variables," J. Qual. Technol., vol. 39, no. 4, pp. 326–339, 2007.

[13] G. G. Njoroge, J. A. Simbauni, and J. A. Koske, "An optimal split plot design for performing a mixture process experiment," Sci. J. Appl. Math. Stat., vol. 5, no. 1, p. 15, 2017.

[14] P. Goos and B. Jones, Optimal design of experiments: a case study approach. John Wiley & Sons, 2011.

[15] A. Atkinson, A. Donev, and R. Tobias, Optimum experimental designs, with SAS, vol. 34. Oxford University Press, 2007.