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R-optimal Mixture Designs for Special Cubic Model

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Abstract

R-optimality has been proposed in the literature as an alternative to the widely used D-optimality criterion, particularly when the goal is to construct rectangular confidence regions. In this study, R-optimal designs are investigated for the special cubic model in mixture experiments. The optimality of the proposed designs is verified using the equivalence theorem.

Key words: R-optimality; D-optimality; Special cubic model; Equivalence theorem; Mixture experiment.

AMS Subject Classifications: 62K05

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1. Introduction

The mixture experiments have a lot of applications in different industrial fields like the food industry, biological engineering, pharmaceutical industry, and building materials since many industrial products can be viewed as mixtures of several mixture components [*ref.* Moldes *et al.* (2007), Zaitri *et al.* (2014), and Liu *et al.* (2016), *etc.*]. Mixture experiments are a special type of design of experiments where the response of interest is only a function of the proportion of the ingredients present in the mixture.

Let x_i represents the proportion of the i^{th} component present in a q component mixture then

$$0 \leq x_i \leq 1, i = 1, 2, \ldots, q, \text{ and } \sum_{i=1}^{q} x_i = 1.$$

Due to the above restriction, the experimental region becomes a $(q-1)$-dimensional simplex given by

$$\mathrm{T} = \left\{ \boldsymbol{x} = (x_1, x_2, \ldots, x_q)' \in R^q \mid \sum_{i=1}^{q} x_i = 1, 0 \leq x_i \leq 1, i = 1, 2, \ldots, q \right\}. \tag{1}$$

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Consider a regression model of the form

$$\eta(\boldsymbol{x}) = \boldsymbol{f}'(\boldsymbol{x})\boldsymbol{\beta} \tag{2}$$

where $\eta(\boldsymbol{x})$ denotes the expected response, $\boldsymbol{x}$ is the vector of ingredient proportions, and $\boldsymbol{f}(\boldsymbol{x})$ is the model expansion of $\boldsymbol{x}$, and $\boldsymbol{\beta}$ is the vector of unknown parameters. Here we assume that the errors are independently and identically distributed with mean 0 and constant variance σ^2.

Different model forms have been proposed in the literature to analyze mixture data *e.g.*, Scheffè's canonical polynomial models, Becker's mixture models, Darroch and Waller's additive quadratic and cubic mixture models, log contrast models, *etc.* Among these models, Scheffè's quadratic polynomial models are the most widely used polynomial models to analyze mixture data. However, when the true underlying response surface exhibits considerable complexity or curvature, a cubic model typically yields a substantially better fit compared to a quadratic model. Cubic models are capable of capturing nonlinear blending effects and synergistic/antagonistic interactions among three components, phenomena that are commonly observed in complex mixtures such as those found in food, pharmaceutical, and polymer systems [*ref.* Cornell (2002)]. Further, the special cubic models (SCMs) reduce model complexity while retaining the ability to capture key higher-order interactions, thereby offering greater efficiency and a lower risk of overfitting compared to full cubic models.

The construction of optimal design based on a certain optimality criterion aims to make the predicted response nearer to the average response over a certain region of interest. In the last few years, many authors have contributed to the domain of optimal design for mixture experiments [*ref.* Aggrawal *et al.* (2011), Pal and Mandal (2012), Mandal and Pal (2017), Panda (2024b)]. The pioneering work on the construction of optimal design for cubic mixture models was due to Kiefer (1961). He obtained D-optimal designs for the full cubic model, cubic model without a 3-way effect, and special cubic model (SCM) for the three-component mixture. Uranisi (1964) proved that a $\{q, 3\}$ simplex centroid design that assigns equal weight to each support point is D-optimal for a special cubic mixture model. Farrell *et al.* (1967) derived the D-optimal designs for the general cubic polynomial model with two and three mixture components respectively. Lim (1990) obtained the D-optimal designs for the same model, when $4 \leq q \leq 10$. Mikaeili (1989) obtained the D-optimal designs for the cubic model without a 3-way effect. Mikaeili (1993) investigated the D-optimal designs for the full cubic model on the set T. Panda and Sahoo (2022) obtained saturated A-optimal designs for full cubic model, cubic model without a 3-way effect, and special cubic model in three mixture components. Zhu and Hao (2024) investigated the A-optimal designs for the special cubic mixture model. Recently, Panda (2024a) found saturated A-optimal designs for the cubic model without a 3-way effect.

The D-optimal design is one of the most widely used classical optimal design criteria. Its construction is relatively straightforward when the number of unknown parameters, denoted by p, is small (*e.g.*, 2 or 3). However, as the number of parameters increases, the computation of D-optimal designs becomes increasingly complex. To address this limitation, Dette (1997) introduced the concept of R-optimal design, which aims to minimize the volume of the p-dimensional rectangular confidence region based on a Bonferroni t-interval. Subsequently, Panda (2021) derived saturated R-optimal designs for all three forms of the cubic model, when $q = 3$. Hao *et al.* (2021) extended the investigation of R-optimal designs to the second-order Scheffé model for q number of mixture components. Building on this line of research, the present study derives the R-optimal design for the SCM when the mixture consists of q components.

The article is structured as follows. Section 2 discusses simplex centroid design. Section 3 explains the R-optimal design and the corresponding equivalence theorem. In Section 4, we obtain the R-optimal designs for the special cubic model where the mixture comprises q number of mixture ingredients. Section 5 obtains R-efficiencies of the corresponding A- and D-optimal designs. The article ends with some discussion in Section 6.

2. Simplex-centroid design

The simplex centroid design is a mixture design that consists of $2^q - 1$ number of support points, *i.e.*,

C_1^q number of pure components
C_2^q number of binary mixtures
C_3^q number of ternary mixtures
...
C_q^q number of q-nary mixtures.

Our focus is on the SCM with a simplex-centroid design consisting of pure components, binary mixtures, and ternary mixtures. This design may be called a $\{q, 3\}$ simplex centroid design. For example, the $\{3, 3\}$ simplex-centroid design consists of three pure components $(1, 0, 0), (0, 1, 0), (0, 0, 1)$; three binary blend points $(1/2, 1/2, 0), (1/2, 0, 1/2), (0, 1/2, 1/2)$; and one ternary mixture $(1/3, 1/3, 1/3)$.

To derive the R-optimal designs for a SCM, the search is restricted to the $\{q, 3\}$ simplex-centroid design. Notably, this design provides the precise set of mixture proportions necessary for the estimation of all model terms, including linear, binary (second-order), and ternary (third-order) interactions. Moreover, the number of support points in the $\{q, 3\}$ simplex-centroid design precisely matches the number of model terms, ensuring identifiability. The design points of this said design are evenly spread over the mixture space T. This balance ensures good coverage and reduces model bias.

3. R-optimal design and equivalence theorem

Let us consider a continuous design of the following form:

$$\xi = \left\{ \begin{array}{ccc} \boldsymbol{x}_1 & \ldots & \boldsymbol{x}_s \\ w_1 & \ldots & w_s \end{array} \right\}$$

where $\boldsymbol{x}_i \in \mathrm{T}, 0 < w_i < 1$ for $i = 1, 2, \ldots s$, and $\sum_{i=1}^{s} w_i = 1. w_i$ is the weight assigned to each of the points $\boldsymbol{x}_i$. For any continuous design $\xi \in \Delta$, the non-singular information matrix is defined as

$$\boldsymbol{M}(\xi) = \sum_{i=1}^{s} w_i \boldsymbol{f}(\boldsymbol{x}_i) \boldsymbol{f}'(\boldsymbol{x}_i) \tag{3}$$

where $\boldsymbol{f}(\boldsymbol{x})$ is of the form given in Equation (2). Let Δ denotes the collection of all continuous designs.

Definition 1: A design $\xi^* \in \Delta$ with a non-singular information matrix $\boldsymbol{M}\left(\xi^*\right)$ is said to be an R-optimal design for the model Equation (2) if it minimizes following function

$$\phi(\xi) = \prod_{i=1}^{p} \boldsymbol{e}_i' \boldsymbol{M}^{-1}(\xi) \boldsymbol{e}_i \tag{4}$$

over all the designs $\xi \in \Delta$, where $\boldsymbol{e_i}$ denotes the i$^{\text{th}}$ unit vector in R^p. Here p is the number of parameters associated with the model Equation (2). The R-optimal criterion corresponds to minimizing the product of the diagonal elements of the inverse information matrix, thereby reducing the joint confidence region associated with Bonferroni t-interval. The necessary and sufficient condition of the R-optimality can be verified using the corresponding equivalence theorem proposed by Dette (1997) which is as follows:

Theorem 1: Let us define the quadratic form

$$\Psi(\boldsymbol{x}, \xi) = \boldsymbol{f}'(\boldsymbol{x}) \boldsymbol{M}^{-1}(\xi) \left(\sum_{i=1}^{p} \frac{\boldsymbol{e}_i \boldsymbol{e}_i'}{\boldsymbol{e}_i' \boldsymbol{M}^{-1}(\xi) \boldsymbol{e}_i} \right) \boldsymbol{M}^{-1}(\xi) \boldsymbol{f}'(\boldsymbol{x}). \tag{5}$$

A design $\xi^* \in \Delta$ is R-optimal if and only if

$$\operatorname{Sup}_{\boldsymbol{x} \in \mathbf{T}} \Psi\left(\boldsymbol{x}, \xi^*\right) = p$$

with equality holds at the support points of ξ^*.

4. R-optimal designs for special cubic model

In this section, we derive R-optimal designs for the SCM with mixture experiment. The special cubic model for the mixture experiment can be expressed as

$$\begin{aligned} \eta(\boldsymbol{x}) &= \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j} \beta_{ij} x_i x_j + \sum_{i<j<k} \beta_{ijk} x_i x_j x_k \\ &= \boldsymbol{f}'(\boldsymbol{x}) \boldsymbol{\beta} \end{aligned} \tag{6}$$

where $\boldsymbol{f}(\boldsymbol{x})$ and $\boldsymbol{\beta}$ are column vectors of dimension $\frac{q\left(q^2+5\right)}{6} \times 1$ and are defined as

$$\boldsymbol{f}(\boldsymbol{x}) = \left(x_1, x_2, \ldots, x_q, x_1 x_2, x_1 x_3, \ldots, x_{q-1} x_q, x_1 x_2 x_3, x_1 x_2 x_4, \ldots, x_{q-2} x_{q-1} x_q\right)';$$
$$\boldsymbol{\beta} = \left(\beta_1, \beta_2, \ldots, \beta_q, \beta_{12}, \beta_{13}, \ldots, \beta_{(q-1)q}, \beta_{123}, \beta_{124}, \ldots, \beta_{(q-1)(q-2)q}\right)'.$$

Theorem 2: Under $\{q, 3\}$ simplex centroid designs, the allocations corresponding to R-optimality criterion for model Equation (6) are attached weights α_1 to the vertices, weights α_2 to the binary mixtures, weights α_3 to the ternary mixtures where weights α_1, α_2, and α_3

satisfy the following system of equations:

$$\left.\begin{aligned}
&\frac{1}{6\alpha_1\left(2\alpha_1+\alpha_2\right)\left(27\alpha_1\alpha_2+16\alpha_1\alpha_3+\alpha_2\alpha_3\right)}\Bigg(q-81\alpha_1\alpha_2\left(4\alpha_1+\alpha_2+q\alpha_2\right)\\
&\quad\times\left(192\alpha_1^2+2(32+q(q+21))\alpha_1\alpha_2+(5+q^2)\alpha_2^2\right)\alpha_3\\
&\quad+6\alpha_1(2\alpha_1+\alpha_2)(27\alpha_1\alpha_2+16\alpha_1\alpha_3+\alpha_2\alpha_3)\lambda\Bigg)=0\\[2ex]
&\frac{1}{6}q(q-1)\left(-\frac{2\alpha_1\left(81\alpha_1\alpha_2+16(q+1)\alpha_1\alpha_3+(8q-13)\right)}{\alpha_2(2\alpha_1+\alpha_2)(27\alpha_1\alpha_2+16\alpha_1\alpha_3+\alpha_2\alpha_3)}+\lambda\right)=0\\[2ex]
&\frac{1}{6}q(q-1)(q-2)\left(-\frac{27\alpha_1\alpha_2}{\alpha_3(27\alpha_1\alpha_2+16\alpha_1\alpha_3+\alpha_2\alpha_3)}+\lambda\right)=0\\[2ex]
&-1+\frac{1}{6}q\left(6\alpha_1+(q-1)\left(3\alpha_2+(q-2)\alpha_3\right)\right)=0
\end{aligned}\right\}\qquad(7)$$

Proof: A comprehensive proof of this theorem is presented in Panda (2025) and can be obtained from the author upon request. □

4.1. Establishment of equivalence theorem

In this section, we demonstrate that the R-optimal allocations derived in Theorem 2 satisfy the necessary and sufficient conditions of the equivalence theorem for R-optimal design. The verification of the equivalence theorem (Theorem 1), however, presents two primary challenges:

1. The complexity of the system of equations in (7) makes the derivation of a general closed-form solution for the optimal weights analytically challenging.
2. Establishing the equivalence theorem for a general value of q is difficult, primarily because of the complex structure of the matrix $\boldsymbol{M}^{-1}(\xi)$.

Although the explicit form of $\boldsymbol{M}^{-1}(\xi)$ is not included in this paper, its structure related mathematical details are discussed in Panda (2025). These materials are available from the author upon request. To address part (i), the optimal weights are computed numerically. The numerical value of optimal weights α_1, α_2, and α_3 for the design ξ^*, along with the maximum value of $\Psi(\boldsymbol{x},\xi)$ for various values of q in the range $3 \leq q \leq 18$, are displayed in Table 1.

Table 1: R-optimal allocations for the special cubic model and corresponding values of $\max_{\boldsymbol{x}\in\mathbf{T}} \Psi(\boldsymbol{x},\xi)$

(1)	(2)	(3)	(4)	(5)
q	α_1	α_2	α_3	$\max_{\boldsymbol{x}\in\mathbf{T}} \Psi(\boldsymbol{x},\xi)$
3	0.1796	0.1217	0.0960	7
4	0.0995	0.0675	0.0492	14
5	0.0611	0.0413	0.0281	25
6	0.0406	0.0271	0.0175	41
7	0.0286	0.0189	0.0115	63
8	0.0210	0.0137	0.0080	92
9	0.0160	0.0103	0.0058	129
10	0.0126	0.0080	0.0043	175
11	0.0101	0.0063	0.0033	231
12	0.0083	0.0051	0.0026	298
13	0.0070	0.0043	0.0020	377
14	0.0058	0.0035	0.0017	469
15	0.0049	0.0029	0.0014	575
16	0.0044	0.0026	0.0011	696
17	0.0037	0.0022	0.0009	833
18	0.0033	0.0019	0.0008	987

Subsequently, to handle the part (ii), we use appropriate MATLAB code by following the steps of Algorithm 1 and demonstrate numerically that $\mathrm{Max}_{\boldsymbol{x}\in\mathrm{T}} \Psi(\boldsymbol{x},\xi^*) = p$. We also examine that equality holds only at the support points of the design ξ^*. This confirms that the design ξ^* is the R -optimal design in the simplex region T.

Algorithm 1: Algorithm to demonstrate Equivalence theorem

Input:

Step 1: Set the value of q and the values of $\alpha_1, \alpha_2, \alpha_3$ using columns (2), (3), and (4) of Table 1 respectively.

Step 2: Input the column vector

$$\boldsymbol{f}(\boldsymbol{x}) = (x_1, x_2, \ldots, x_q, x_1x_2, x_1x_3, \ldots, x_{q-1}x_q, x_1x_2x_3, x_1x_2x_4, \ldots, x_{q-2}x_{q-1}x_q)'$$

Computation:

Step 3: Obtain the matrix $\boldsymbol{M}^{-1}(\xi^*)$ [*ref.* Panda (2025)].

Step 4: Find the functional form $\Psi(\boldsymbol{x},\xi)$

where

$$\Psi(\boldsymbol{x},\xi) = \boldsymbol{f}'(\boldsymbol{x})\boldsymbol{M}^{-1}(\xi^*)\left(\sum_{i=1}^{p} \frac{\boldsymbol{e_i}\boldsymbol{e_i}'}{\boldsymbol{e_i}'\boldsymbol{M}^{-1}(\xi^*)\,\boldsymbol{e_i}}\right)\boldsymbol{M}^{-1}(\xi^*)\,\boldsymbol{f}(\boldsymbol{x})$$

Step 5: Evaluate $\Psi(\boldsymbol{x}, \xi)$ at the support points of design ξ^*.

Step 6: Find $\text{Max}_{\boldsymbol{x} \in T}\, \Psi\left(\boldsymbol{x}, \xi^*\right)$.

5. Efficiency of A- and D-optimal designs

The R-efficiency denoted by $\Delta_R(\xi)$ of a design ξ relative to an R-optimal design $\xi^{(0)}$ is given by

$$\Delta_R(\xi) = \left(\frac{\phi\left(\xi^{(0)}\right)}{\phi(\xi)}\right)^{1/p}.$$

The value of $\Delta_R(\xi)$ closer to 1 indicates that the design has a high R-efficiency value. The R-efficiency values of D-optimal design [*ref.* Uranisi (1964)] and A-optimal design [*ref.* Zhu and Hao (2024)] of the SCM for different values of $q\,(3 \leq q \leq 18)$ have been computed and shown in Table 2. Further, the R-efficiency values of the corresponding D-optimal and A-optimal designs against various values of q are displayed in Figure 1.

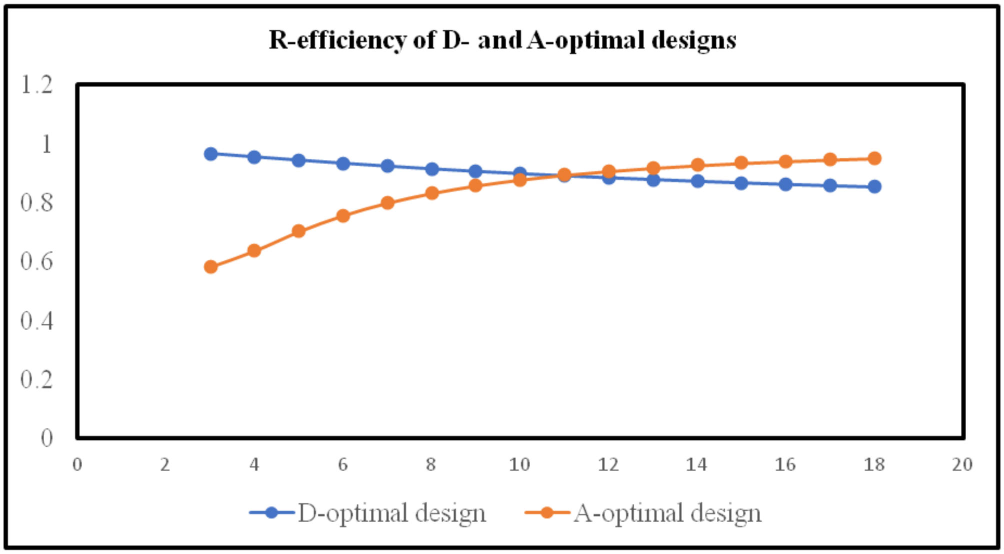

Figure 1: R-efficiency of D- and A-optimal designs for the special cubic model

Next, we consider a three-component example to establish the fact that the R-optimal design can improve the ability of parameter estimation over other designs.

Example 1: **The R-efficiency with three components example**

Cornell (*pg.* 57, 2002) cited a three-component mixture experiment that aims at the effect of the proportion of three artificial sweeteners such as glycerine, saccharin, and an enhancer on the intensity of sweetness. The amount of each of the sweeteners was fixed to 250 ml. To analyze the problem, we use the special cubic model. The R-optimal design for the said model is

$$\xi_R = \begin{pmatrix} \boldsymbol{x} \leftrightarrow (1,0,0) & \boldsymbol{x} \leftrightarrow (1/2,1/2,0) & \boldsymbol{x} \leftrightarrow (1/3,1/3,1/3) \\ 0.1796 & 0.1217 & 0.9996 \end{pmatrix}$$

where $\boldsymbol{x} \leftrightarrow (1,0,0)$ refers to the design point $(1,0,0)$ and its permutations $(0,1,0)$, and $(0,0,1)$; $\boldsymbol{x} \leftrightarrow (1/2,1/2,0)$ refers to the design point $(1/2,1/2,0)$ and its permutations $(1/2,0,1/2)$, and $(0,1/2,1/2)$; $x \leftrightarrow (1/3,1/3,1/3)$ means the design point itself. Here value of $\phi(\xi_R) = 1.06234 \times 10^{13}$. Let us consider the 10 support points that have been discussed in the same problem. Allocate a weight of $\frac{1}{10}$ to each of the support points. Let us denote the design by ξ_1.

$$\xi_1 = \begin{pmatrix} \boldsymbol{x} \leftrightarrow (1,0,0) & \boldsymbol{x} \leftrightarrow (1/2,1/2,0) & \boldsymbol{x} \leftrightarrow (1/3,1/3,1/3) & \boldsymbol{x} \leftrightarrow (2/3,1/6,1/6) \\ \frac{1}{10} & \frac{1}{10} & \frac{1}{10} & \frac{1}{10} \end{pmatrix}.$$

For the design ξ_1, the value of $\phi(\xi_1) = 1.1151 \times 10^{14}$. Consider another design ξ_2 as discussed in Cornell (2002) *i.e.*,

$$\xi_2 = \begin{pmatrix} \boldsymbol{x} \leftrightarrow (1,0,0) & \boldsymbol{x} \leftrightarrow (b,1-b,0) & \boldsymbol{x} \leftrightarrow (1/3,1/3,1/3) \\ \frac{1}{10} & \frac{1}{10} & \frac{1}{10} \end{pmatrix}$$

where $b = \frac{1-5^{-1/2}}{2}$. For this design, $\phi(\xi_2) = 9.88373 \times 10^{13}$. Similarly consider the following $\{3,3\}$ simplex-lattice design denoted by ξ_3 where equal weight has been allocated to each of the support points.

$$\xi_3 = \begin{pmatrix} \boldsymbol{x} \leftrightarrow (1,0,0) & \boldsymbol{x} \leftrightarrow (1/3,2/3,0) & \boldsymbol{x} \leftrightarrow (1/3,1/3,1/3) \\ \frac{1}{10} & \frac{1}{10} & \frac{1}{10} \end{pmatrix}$$

The R-efficiencies of the designs ξ_1, ξ_2, and ξ_3 are calculated as $0.7147, 0.7271$, and 0.7793 respectively. Thus, it can be depicted that ξ_R can improve the parameter estimation.

Table 2: R-efficiencies of various optimal designs for the special cubic mixture model

q	D-optimal design	A-optimal design
3	0.9661	0.5816
4	0.9549	0.6364
5	0.9441	0.7022
6	0.9336	0.7569
7	0.9238	0.7996
8	0.9148	0.8325
9	0.9064	0.8581
10	0.8986	0.8783
11	0.8915	0.8944
12	0.8849	0.9074
13	0.8788	0.9181
14	0.8730	0.9270
15	0.8677	0.9345
16	0.8627	0.9408
17	0.8581	0.9462
18	0.8537	0.9508

6. Discussion

The construction of R-optimal designs involves more challenges in comparison to the D-optimal design as the weights associated with the support points in the case of the former are different. Also, the weights vary as the value of q changes. This article finds R-optimal designs for the special cubic model with a mixture experiment based on q mixture components, where $3 \leq q \leq 18$. For $q = 3$, we also observe that the derived R-optimal design for the special cubic model is similar to the result derived by Panda(2021). From Example 1 as well as Table 2, we can see that the R-optimal designs have higher efficiency as compared to other designs. Based on Figure 1, it can be depicted that the R-efficiency value of the D-optimal design decreases with q whereas it increases in the case of the A-optimal design. The performance of the D-optimal design is better than the A-optimal design with q, when $3 \leq q \leq 10$ whereas the performance of the latter is better with q for $11 \leq q \leq 18$. Still, the performance of the R-optimal design is consistently better with different values of q.

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Conflict of interest

The authors do not have any financial or non-financial conflict of interest to declare for the research work included in this article.

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