Mathematical Biosciences and Engineering

http://www.aimspress.com/journal/mbe

MBE, 19(5): 4737–4748.
DOI: 10.3934/mbe.2022221
Received: 26 December 2021
Revised: 29 January 2022
Accepted: 23 February 2022
Published: 10 March 2022

*Research article*

# D-optimal design of the additive mixture model with multi-response

**Zheng Gong**[1], **Xiaoyuan Zhu**[2] **and Chongqi Zhang**[1,*]

[1] School of Economics and Statistics, Guangzhou University, Guangzhou 510006, China

[2] School of Mathematics and Information Science, Guangzhou University, Guangzhou 510006, China

* **Correspondence:** Email: cqzhang@gzhu.edu.cn; Tel: +8613535128229.

**Abstract:** This paper proposes the D-optimal design for the additive mixture model with two-response, which is linear model with no interaction terms. The optimality was validated by using the general equivalence theorem, and the corresponding weights are found under which additive model satisfies D-optimality. In addition, relevant statistics and graphics are given to illustrate our results.

**Keywords:** mixture experiment; D-optimal design; additive model; multi-response

## 1. Introduction

Mixture experiment [1, 2] is a subject of great significance in engineering [3], pharmacy [4] and bioscience [5]. The response depends only on the proportions but not the total amount of the mixture. With further researches show the distinct progress of mixture experiment, the relevant research about algorithm [6–8], optimality [9, 10] and data analytics [11, 12] are well studied. Some general recommendations for the optimal design of general theory can be found in the monograph of Atkinson et al. [13], Cornell [14], Goos et al. [15] and Sinha et al. [16].

In various fields of research, experimental designs in multi-response situations are generally of interest and considered. Mixture experiment becomes more complicated due to the extension of multi-response. Fedorov [17] discussed the background of optimality of multi-response experiments, as well as its early research and influence. Draper and Hunter [18] details the design of experiment for parameter estimation in multi-response situations. Furthermore, a lot of multi-response problems were increasingly concerned about. For a detailed review about optimal design of mixture model with multi-response, see Imhof [19] and Rolz [20]. Nowadays, the relevant researches have wide coverage and practical application. The prime example is that Liu et al. [21] did the research about optimality for multi-response linear mixed models. Dette et al. [22] solved the application in thermal spraying by using multi-response method.

With the exception of multi-response, the method for finding appropriate mixture models is another major research area. There are also many mixture models based on various application conditions. A number of features of different mixture models have been introduced by Chan [23]. Among all mixture models, the Scheffé mixture model is the most commonly used and the easiest to be calculated. However, Scheffé mixture model can't function effectively when there is no interaction among all mixture components. For this reason, Darroch and Waller [24] proposed the additive mixture model, which is used to calculate the mixture model with no interaction.

During the last several decades, various optimal designs of additive model with single response were studied by Chan et al. [25, 26] and Zhang and Guan et al. [27, 28]. It is therefore worthy to extend the D-optimal design to additive model with multiple response, and investigate whether properties of D-optimal design in additive mixture model will change on account of multi-response. In order to better describe content. In Section 2, we first briefly review the basics of mixture experiments. Then we put forward the additive mixture model with multi-response. In Section 3, we obtain the principal results about the proof of D-optimality and equivalence theorem. Some concluding remarks are presented in the final section.

## 2. Model specification and preliminaries

The common mixture model involving $q$ ingredients $x_1, x_2, \ldots, x_q$ can be written as $Y(\mathrm{x}) = f^{\mathrm{T}}(\mathrm{x})\beta + \varepsilon(\mathrm{x})$, where $q \geqslant 2$ and $\mathrm{x} = (x_1, x_2, \ldots, x_q)^{\mathrm{T}}$ lies in a finite dimensional simplex

$$S^{q-1} = \{(x_1, x_2, \ldots, x_q) : \sum_{i=1}^{q} x_i = 1, 0 \leqslant x_i \leqslant 1, i = 1, 2, \ldots, q\}. \tag{2.1}$$

### *2.1. Additive mixture model*

The mixture experiment constraints have a substantial impact on the mixture model. For every square of $x_i$, it is a linear combination of $x_i$ and its cross-products with the other $(q-1)$ proportions. We usually write the square terms as follows:

$$x_i^2 = x_i(1 - \sum_{j=1, j\neq i}^{q} x_j) = x_i - x_i(1 - x_i).$$

In view of these considerations, we reform the second-order additive mixture model

$$E(Y) = \delta_0 + \sum_{i=1}^{q} \delta_i x_i + \sum_{i=1}^{q} \delta_{ii} x_i^2 = \sum_{i=1}^{q} (\delta_0 + \delta_i + \delta_{ii}) x_i + \sum_{i=1}^{q} (-\delta_{ii}) x_i (1 - x_i), \tag{2.2}$$

to

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q} \beta_{ii} x_i (1 - x_i). \tag{2.3}$$

On the basis of this theory, this paper consider the second-order additive mixture model with multi-response, which is given as

$$Y_j(\mathrm{x}) = f_j^{\mathrm{T}}(\mathrm{x})\beta_j + \varepsilon_j(\mathrm{x}), \tag{2.4}$$

$$\begin{cases} f_1^{\mathrm{T}}(x) = (x_1, x_2, ..., x_q) \\ f_2^{\mathrm{T}}(x) = (x_1, x_2, ..., x_q, x_1(1-x_1), x_2(1-x_2), ..., x_q(1-x_q)) \\ \beta_1 = (\beta_{11}, \beta_{12}, ..., \beta_{1q})^{\mathrm{T}} \\ \beta_2 = (\beta_{21}, \beta_{22}, ..., \beta_{2q}, \beta_{211}, \beta_{222}, ..., \beta_{2qq})^{\mathrm{T}} \\ E(\varepsilon_i) = 0 \\ \mathrm{Var}((\varepsilon_1, \varepsilon_2)^{\mathrm{T}}) = \Sigma = \begin{pmatrix} \sigma_1^2 & \rho\sigma_1\sigma_2 \\ \rho\sigma_1\sigma_2 & \sigma_2^2 \end{pmatrix} \end{cases}$$

### 2.2. *Experimental design*

Experimental design contains two parts: continuous design and exact design. Continuous space is more suitable for searching the optimal design and can iterate and approximate the best and optimal value. The domain of consideration provided by continuous space will not produce points that cannot be valued. Because discrete space has inherent limitations in iteration and approximation. We usually consider exact design under special conditions or restrictions. We generally only discussed continuous design.

The design problem for model (2.4) is to obtain an $n$-point design $\xi$ to estimate some function of the $k$-dimensional parameter vector $\beta$ with high efficiency, the design $\xi$ can be performed of the form

$$\xi = \begin{pmatrix} \tau_1 & \tau_2 & \dots & \tau_n \\ r_1 & r_2 & \dots & r_n \end{pmatrix},$$

where $\tau_i$ are support points in the interior of simplex region $S^{q-1}$, and the corresponding weights $r_i$ are nonnegative real numbers which sum to unity. For a given covariance matrix, the moment matrix is

$$M(\xi) = \int_{S^{q-1}} F(\tau)\Sigma^{-1}F^{\mathrm{T}}(\tau)d\xi(\tau),$$

where $F^{\mathrm{T}}$ is the block-diagonal matrix diag $(f_1^{\mathrm{T}}(\tau), f_2^{\mathrm{T}}(\tau))$, and D-optimal design aims to maximize $\det(M(\xi))$.

## 3. D-optimal design for the additive mixture model with multi-response

It is known that D-optimal designs for mixture model, including additive model, have support points in the barycenters of simplex region. But the main feature of additive model makes itself a little out of the ordinary. Apart from vertices, other support points of additive model vary according to the number of $q$, and they usually gather inward as $q$ increase. Based on generalized simplex-centroid design, we construct design $\xi_{1i}^*$:

There are two kinds of points in total: the $C_q^1$ permutations of the pure components, the $C_q^i$ permutations of the barycenters of deep $i$, which are of $S^{q-1}$ if $i$ of its q coordinates are equal to $\frac{1}{i}$ and others are zero. Geometric descriptions of the former and the later are separately vertices and $i^{th}$ barycenters of simplex.

That is, we consider the design $\xi_{1i}^*$ of following form:

$$\xi_{1i}^* = \begin{pmatrix} M_q^1 & M_q^i \\ r_1 & r_i \end{pmatrix},$$

where $M_q^1$ denotes any point from the pure components, $M_q^i$ denotes any point from the barycenters of deep $i$, which means $i$ of its $q$ coordinates are equal to $\frac{1}{i}$ and the remaining ones are equal to zero. We present the weight of vertices and the weight of $i^{th}$ barycenters separately by $r_1$ and $r_i$, they also satisfy $C_q^1 r_1 + C_q^i r_i = 1$.

For mixture model with multi-response, we have the equivalence theorem, presented by Kiefer [29], let:

$$\phi(\tau, \xi, \Sigma) = \mathrm{Tr}(\Sigma^{-1} F M^{-1} F^T), \tag{3.1}$$

and for any given design $\xi^*$ satisfying D-optimality, there is

$$\phi(\tau, \xi^*, \Sigma) \leqslant p, \tag{3.2}$$

for all points in simplex, equality in model (3.2) holds and only holds at $\tau \in \xi^*$, and the $p$ in model (2.4) is equal to $3q$.

### *3.1. D-optimal design for* $3 \leqslant q \leqslant 6$

**Theorem 1.** If $3 \leqslant q \leqslant 6$, then $\xi_{12}^*$ which assigns $r_1^*$ to pure component and $r_2^*$ to binary component is the D-optimal design for additive model (2.4), where

$$r_1^* = \frac{1}{q} - \frac{6q - 5 - \sqrt{(6q-5)^2 - 8(q-1)(3q-1)}}{2q(3q-1)},$$
$$r_2^* = \frac{6q - 5 - \sqrt{(6q-5)^2 - 8(q-1)(3q-1)}}{q(q-1)(3q-1)}.$$

where $r_1^*$ are the weights of pure component points and $r_2^*$ are the weights of points on edges, i.e., combinations of $(0.5, 0.5, 0)$.

**Proof.** D-optimal design typically maximizes $\det(M(\xi))$, it is necessary to identify the inverse of covariance matrix

$$\Sigma^{-1} = \frac{1}{(1-\rho^2)\sigma_1^2\sigma_2^2} \begin{pmatrix} \sigma_2^2 & -\rho\sigma_1\sigma_2 \\ -\rho\sigma_1\sigma_2 & \sigma_1^2 \end{pmatrix} = \begin{pmatrix} a & c \\ c & b \end{pmatrix}.$$

Straight forward calculation gives

$$\begin{aligned}
M(\xi) &= \sum_{i=1}^{q} r_1 F \Sigma^{-1} F^{\mathrm{T}} + \sum_{j=1}^{C_q^2} r_2 F \Sigma^{-1} F^{\mathrm{T}} \\
&= r_1 \begin{pmatrix} \begin{pmatrix} a & c \\ c & b \end{pmatrix} \otimes I_q & 0 \\ 0 & 0 \end{pmatrix} + \frac{q-1}{4} r_2 \begin{pmatrix} a & c & \frac{1}{2}c \\ c & b & \frac{1}{2}b \\ \frac{1}{2}c & \frac{1}{2}b & \frac{1}{4}b \end{pmatrix} \otimes I_q + \frac{r_2}{4} \begin{pmatrix} a & c & \frac{1}{2}c \\ c & b & \frac{1}{2}b \\ \frac{1}{2}c & \frac{1}{2}b & \frac{1}{4}b \end{pmatrix} \otimes U_q \\
&= \begin{pmatrix} \begin{pmatrix} a & c \\ c & b \end{pmatrix} \otimes ((r_1 + \frac{q-2}{4} r_2) I_q + \frac{r_2}{4} J_q) & \begin{pmatrix} c \\ b \end{pmatrix} \otimes (\frac{q-2}{8} r_2 I_q + \frac{r_2}{8} J_q) \\ \begin{pmatrix} c & b \end{pmatrix} \otimes (\frac{q-2}{8} r_2 I_q + \frac{r_2}{8} J_q) & b \otimes (\frac{q-2}{16} r_2 I_q + \frac{r_2}{16} J_q) \end{pmatrix} \\
&= \begin{pmatrix} A & B \\ C & D \end{pmatrix},
\end{aligned}$$

where $I_q$ is a $q$-dimensional identity matrix, $J_q$ is a $q{\times}q$ matrix with all elements equal to 1, $U_q = J_q - I_q$.

When $T = aI + bJ$, there is $T^{-1} = \frac{1}{a}I + \frac{b}{a(a+bq)}J$, we can get

$$D^{-1} = \frac{16}{br_2} \otimes (\frac{1}{q-2}I_q - \frac{1}{2(q-2)(q-1)}J_q)$$

$$BD^{-1}C = \frac{r_2}{4}\begin{pmatrix} \frac{c^2}{b} & c \\ c & b \end{pmatrix} \otimes ((q-2)I_q + J_q),$$

it follows that

$$A - BD^{-1}C = \begin{pmatrix} W & cr_1 I_q \\ cr_1 I_q & br_1 I_q \end{pmatrix},$$

where

$$W = (ar_1 + \frac{q-2}{4}r_2(a - \frac{c^2}{b}))I_q + \frac{r_2}{4}(a - \frac{c^2}{b})J_q,$$

then we can obtain

$$\begin{aligned} \det(M(\xi)) =& \det(A - BD^{-1}C)\det(D) \\ =& br_1^q \det\begin{pmatrix} W - \frac{c^2}{b}r_1 I_q & 0 \\ \frac{c^2}{b}I_q & I_q \end{pmatrix}\det(D) \\ =& \frac{br_1^q}{q}(a - \frac{c^2}{b})^q(r_1 + \frac{(q-2)r_2}{4})^{q-1}|b \otimes \frac{q-2}{16}r_2 I_q + \frac{r_2}{16}J_q| \\ =& \frac{br_1^q}{q}(a - \frac{c^2}{b})^q(r_1 + \frac{(q-2)r_2}{4})^{q-1}2(q-1)(q-2)^{q-1}(\frac{r_2}{16})^q. \end{aligned}$$

Clearly, $\det(M(\xi))$ is the function of $r_1$ and $r_2$. By the linear optimization method, we have

$$\Psi = sr_1^q(r_1 + \frac{(q-2)r_2}{4})^q r_2{}^q + \lambda(qr_1 + \frac{q(q-2)}{2}r_2 - 1),$$

where $s$ is a constant independent of our objective.

The Lagrange multiple method is applied to obtain:

$$\begin{cases} \frac{\partial\Psi}{\partial r_1} & = sr_2^q[qr_1^{q-1}(r_1 + \frac{(q-2)r_2}{4})^{q-1} + (q-1)r_1^q(r_1 + \frac{(q-2)r_2}{4})^{q-2}] + \lambda q \\ & = T_1 + \lambda q = 0 \\ \frac{\partial\Psi}{\partial r_2} & = sr_1^q[(q-1)(r_1 + \frac{(q-2)r_2}{4})^{q-2} + \frac{q-2}{4}r_2^q + (r_1 + \frac{(q-2)r_2}{4})^{q-1}]qr_2^{q-1} + \lambda\frac{q(q-1)}{2} \\ & = T_2 + \lambda\frac{q(q-1)}{2} = 0 \\ \frac{\partial\Psi}{\partial\lambda} & = qr_1 + \frac{q(q-1)}{2}r_2 - 1 = 0. \end{cases}$$

By calculating these equations, we get:

$$\begin{cases} r_1^* = \frac{1}{q} - \frac{6q-5-\sqrt{(6q-5)^2-8(q-1)(3q-1)}}{2q(3q-1)}, \\ r_2^* = \frac{6q-5-\sqrt{(6q-5)^2-8(q-1)(3q-1)}}{q(q-1)(3q-1)}. \end{cases}$$

At last, the D-optimality allocation $\xi_{12}^*$ can be found as:

$$\xi_{12}^* = \begin{pmatrix} M_q^1 & M_q^2 \\ \frac{1}{q} - \frac{6q-5-\sqrt{(6q-5)^2-8(q-1)(3q-1)}}{2q(3q-1)} & \frac{6q-5-\sqrt{(6q-5)^2-8(q-1)(3q-1)}}{q(q-1)(3q-1)} \end{pmatrix}.$$

For verifying equivalence theorem of design $\xi_{12}^*$, above all, we calculate the inverse of $M$

$$M^{-1} = \begin{pmatrix} L^{-1} & -\frac{c}{b}L^{-1} & 0 \\ -\frac{c}{b}L^{-1} & -\frac{1}{br_1}I_q + \frac{c^2}{b^2}L^{-1} & -\frac{2}{br_1}I_q \\ 0 & -\frac{2}{br_1}I & \frac{16}{br_2}(\frac{1}{q-2}I_q - \frac{1}{2(q-2)(q-1)J_q}) \end{pmatrix},$$

where $L^{-1} = \frac{1}{(r_1+\frac{q-2}{4}r_2)(a-\frac{c^2}{b})}(I_q - \frac{qr_2}{4}J_q)$. Then we can obtain

$$FM^{-1}F^{\mathrm{T}} = \begin{pmatrix} X_1L^{-1}X_1^{\mathrm{T}} & -\frac{c}{b}X_1L^{-1}X_1^{\mathrm{T}} \\ -\frac{c}{b}X_1L^{-1}X_1^{\mathrm{T}} & H \end{pmatrix},$$

where $X_1 = (x_1, x_2, ..., x_q)$, $X_2 = (x_1(1-x_1), x_2(1-x_2), ..., x_q(1-x_q))$, and

$$\begin{aligned} H = {} & \frac{1}{br_1}X_1X_1^{\mathrm{T}} + \frac{c^2}{b^2}X_1L^{-1}X_1^{\mathrm{T}} - \frac{2}{br_1}X_1X_2^{\mathrm{T}} + \frac{16}{br_2(q-2)}X_2X_2^{\mathrm{T}} \\ & - \frac{8}{br_2(q-2)(q-1)}X_2J_qX_2^{\mathrm{T}} + \frac{4}{br_1}X_2X_2^{\mathrm{T}}. \end{aligned}$$

At last, we have

$$\begin{aligned} \mathrm{Tr}(\Sigma^{-1}FM^{-1}F^{\mathrm{T}}) &= \mathrm{Tr}\left(\begin{pmatrix} a & c \\ c & b \end{pmatrix}\begin{pmatrix} X_1L^{-1}X_1^{\mathrm{T}} & -\frac{c}{b}X_1L^{-1}X_1^{\mathrm{T}} \\ -\frac{c}{b}X_1L^{-1}X_1^{\mathrm{T}} & H \end{pmatrix}\right) \\ &= \mathrm{Tr}\begin{pmatrix} aX_1L^{-1}X_1^{\mathrm{T}} - \frac{c^2}{b}X_1L^{-1}X_1^{\mathrm{T}} & -\frac{ac}{b}X_1L^{-1}X_1^{\mathrm{T}} + cH \\ 0 & -\frac{c^2}{b}X_1L^{-1}X_1^{\mathrm{T}} + bH \end{pmatrix} \\ &= \frac{1}{r_1 + \frac{q-2}{4}r_2}\left(\sum_{i=1}^{q} x_i^2 - \frac{q}{4}r_2\right) + \frac{1}{r_1}\sum_{i=1}^{q} x_i^2 - \frac{4}{r_1}\left(\sum_{i=1}^{q} x_i^2 - \sum_{i=1}^{q} x_i^3\right) \\ &\quad + \frac{16}{r_2(q-2)}\left(\sum_{i=1}^{q} x_i^2(1-x_i)^2\right) - \frac{8}{r_2(q-2)(q-1)}\left(1 - \sum_{i=1}^{q} x_i^2\right)^2 \\ &\quad + \frac{4}{r_1}\left(\sum_{i=1}^{q} x_i^2(1-x_i)^2\right). \end{aligned} \tag{3.3}$$

To verify D-optimality of the design $\xi_{12}^*$ in simplex-region. According to convex analysis and the theory of Atwood [1], the maximum value must lie in the boundary of the simplex $S^{q-1}$ and be one of barycenters. When $3 \leqslant q \leqslant 6$, the Table1 can be shown:

**Table 1.** Weights and values of variance function of support points.

| q | $r_1$ | $r_2$ | $\phi(M_q^1)$ | $\phi(M_q^2)$ | $\phi(M_q^3)$ | $C_q^1 r_1$ | $C_q^2 r_2$ | $\frac{\phi(M_q^3)}{3q}$ |
|---|---|---|---|---|---|---|---|---|
| 3 | 0.1959 | 0.1374 | 9 | 9 | 5.5009 | 0.5877 | 0.4123 | 0.6112 |
| 4 | 0.1460 | 0.0693 | 12 | 12 | 10.2627 | 0.5840 | 0.4160 | 0.8552 |
| 5 | 0.1165 | 0.0418 | 15 | 15 | 14.0425 | 0.5823 | 0.4177 | 0.9362 |
| 6 | 0.0969 | 0.0279 | 18 | 18 | 17.5775 | 0.5813 | 0.4187 | 0.9765 |

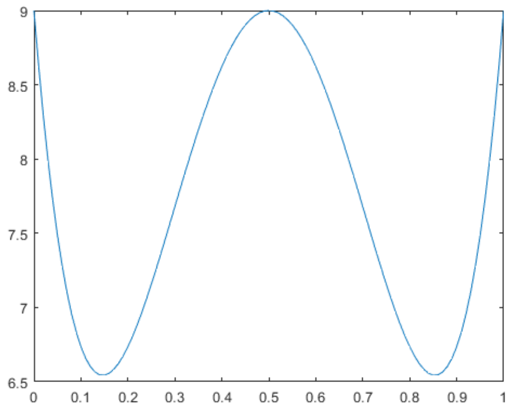


**Figure 1.** Sectional photograph.

By analyzing Table 1, at vertex and midpoint of edge, the value of $\mathrm{Tr}(\Sigma^{-1}FM^{-1}F^{\mathrm{T}})$ are $\phi(M_q^1)$, $\phi(M_q^2)$, which both equal to $3q$, and the value at other points are less than $3q$. That indicates the inequality in model (3.2) holds at all $x_i \in S^{q-1}$ except support points. Thus $\xi_{12}^*$ is in fact D-optimal design.

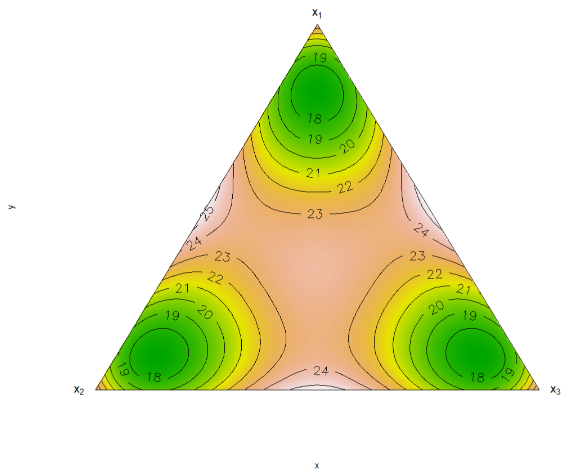


**Figure 2.** Contour map.

Typically, when $q = 3$, we plot the Figure 1, a section photograph in one edge of simplex region, and Figure 2, the contour map in simplex region.

Apparently the maximum value 9 must lie in the support points, and values of other points in $\xi_{12}^* \in S^2$ are less than 9.

Thus we have proved that the design $\xi_{12}^*$ is in fact D-optimal.

### 3.2. D-optimal design for $q \geqslant 16$

**Theorem 2.** If $q \geqslant 16$, then $\xi_{13}^*$ which assigns $r_1^*$ to pure component and $r_3^*$ to ternary component is the D-optimal design for additive model (2.4), where

$$r_1^* = \frac{1}{q} - \frac{5q - 4 - \sqrt{7q^2 - 16q + 10}}{2q(3q-1)},$$
$$r_3^* = \frac{15q - 12 - 3\sqrt{7q^2 - 16q + 10}}{q(q-1)(q-2)(3q-1)}$$

.

**Proof.** The process of proof is analogous, straight forward calculation gives

$$\begin{aligned} M(\xi) &= \sum_{i=1}^{q} r_1 F\Sigma^{-1}F^{\mathrm{T}} + \sum_{j=1}^{C_q^3} r_3 F\Sigma^{-1}F^{\mathrm{T}} \\ &= \begin{pmatrix} \begin{pmatrix} a & c \\ c & b \end{pmatrix} \otimes ((r_1 + \frac{(q-2)(q-3)}{18} r_3) I_q + \frac{q-2}{9} r_3 J_q) & \begin{pmatrix} c \\ b \end{pmatrix} \otimes (\frac{(q-2)(q-3)}{27} r_3 I_q + \frac{2(q-2)}{27} r_3 J_q) \\ \begin{pmatrix} c & b \end{pmatrix} \otimes (\frac{(q-2)(q-3)}{27} r_3 I_q + \frac{2(q-2)}{27} r_3 J_q) & b \otimes (\frac{2(q-2)(q-3)}{81} r_3 I_q + \frac{4(q-2)}{81} r_3 J_q) \end{pmatrix} \\ &= \begin{pmatrix} A & B \\ C & D \end{pmatrix}. \end{aligned}$$

After some algebraic manipulation, we can obtain

$$A - BD^{-1}C = \begin{pmatrix} W & cr_1 I_q \\ cr_1 I_q & br_1 I_q \end{pmatrix},$$

where

$$W = (ar_1 + \frac{(q-2)(q-3)}{18} r_3 (a - \frac{c^2}{b})) I_q + \frac{q-2}{9} r_3 (a - \frac{c^2}{b}) J_q.$$

Obeserving that

$$\begin{aligned} \det(M(\xi)) &= \det(A - BD^{-1}C)\det(D) \\ &= br_1^q \det \begin{pmatrix} W - \frac{c^2}{b} r_1 I_q & 0 \\ \frac{c^2}{b} I_q & I_q \end{pmatrix} \det(D) \\ &= \frac{3br_1^q}{q} (a - \frac{c^2}{b})^q (r_1 + \frac{(q-2)(q-3)}{4} r_3)^{q-1} (\frac{4}{81} r_3)^q (\frac{(q-1)(q-2)}{2})^q \end{aligned}$$

By the linear optimization method, we have the function:

$$\Psi = sr_1^q (r_1 + \frac{(q-2)(q-3)}{18} r_3)^q r_3{}^q + \lambda(qr_1 + \frac{q(q-1)(q-2)}{6} r_3 - 1).$$

According to the method mentioned above, there is

$$\begin{cases} \frac{\partial\Psi}{\partial r_1} & = sr_3^q[qr_1^{q-1}(r_1+\frac{(q-2)(q-3)}{18}r_3)^{q-1}+(q-1)r_1^q(r_1+\frac{(q-2)(q-3)}{18})^{q-2}r_3]+\lambda q \\ & = T_1+\lambda q = 0 \\ \frac{\partial\Psi}{\partial r_3} & = sr_1^q[(q-1)\frac{(q-2)(q-3)}{18}(r_1+\frac{(q-2)(q-3)}{18}r_3)^{q-2}+r_3^q \\ & +(r_1+\frac{(q-2)(q-3)}{18}(r_1+\frac{(q-2)(q-3)}{18}r_3)^{q-1}]qr_3^{q-1}+\lambda\frac{q(q-1)(q-2)}{6} \\ & = T_2+\lambda\frac{q(q-1)}{2} = 0 \\ \frac{\partial\Psi}{\partial\lambda} & = qr_1+\frac{q(q-1)(q-2)}{6}r_3-1=0. \end{cases}$$

By calculating these equations, we get:

$$\begin{cases} r_1^* = \frac{1}{q}-\frac{5q-4-\sqrt{7q^2-16q+10}}{2q(3q-1)}, \\ r_3^* = \frac{15q-12-3\sqrt{7q^2-16q+10}}{q(q-1)(q-2)(3q-1)}. \end{cases}$$

Then the D-optimality allocation $\xi_{13}^*$ can be written as:

$$\xi_{13}^* = \begin{pmatrix} M_q^1 & M_q^3 \\ \frac{1}{q}-\frac{5q-4-\sqrt{7q^2-16q+10}}{2q(3q-1)} & \frac{15q-12-3\sqrt{7q^2-16q+10}}{q(q-1)(q-2)(3q-1)} \end{pmatrix}.$$

The next step is to prove the equivalence theorem. After necessary calculations, there is

$$\begin{aligned} & \mathrm{Tr}(\Sigma^{-1}FM^{-1}F^{\mathrm{T}}) \\ & = \frac{1}{r_1+\frac{(q-2)(q-3)}{18}r_3}(\sum_{i=1}^q x_i^2-\frac{q(q-2)}{9}r_3)+\frac{1}{r_1}\sum_{i=1}^q x_i^2-\frac{3}{r_1}(\sum_{i=1}^q x_i^2-\sum_{i=1}^q x_i^3) \\ & +\frac{81}{2r_3(q-2)(q-3)}(\sum_{i=1}^q x_i^2(1-x_i)^2)-\frac{27}{r_3(q-1)(q-2)(q-3)}(1-\sum_{i=1}^q x_i^2)^2 \\ & +\frac{9}{4r_1}(\sum_{i=1}^q x_i^2(1-x_i)^2). \end{aligned}$$

Through some algebraic manipulation, we have the Table 2.

**Table 2.** Weights of support points and values of variance function.

| q | $r_1$ | $r_3$ | $\phi(M_q^1)$ | $\phi(M_q^2)$ | $\phi(M_q^3)$ | $C_q^1 r_1$ | $C_q^3 r_3$ | $\frac{\phi(M_q^2)}{3q}$ |
|---|---|---|---|---|---|---|---|---|
| 16 | 0.0381 | 6.9682e−04 | 48 | 47.8783 | 48 | 0.6098 | 0.3902 | 0.9975 |
| 17 | 0.0359 | 5.7405e−04 | 51 | 50.7327 | 51 | 0.6096 | 0.3904 | 0.9948 |
| 18 | 0.0339 | 4.7853e−04 | 54 | 53.5903 | 54 | 0.6095 | 0.3905 | 0.9924 |
| 19 | 0.0321 | 4.0308e−04 | 57 | 56.4506 | 57 | 0.6094 | 0.3906 | 0.9904 |
| 20 | 0.0305 | 3.4270e−04 | 60 | 59.3131 | 60 | 0.6093 | 0.3907 | 0.9886 |
| ⋮ | ⋮ | ⋮ | ⋮ | ⋮ | ⋮ | ⋮ | ⋮ | ⋮ |
| 100 | 0.0061 | 2.4246e−06 | 300 | 289.4101 | 3000 | 0.6079 | 0.3921 | 0.9647 |
| ⋮ | ⋮ | ⋮ | ⋮ | ⋮ | ⋮ | ⋮ | ⋮ | ⋮ |
| 200 | 0.0030 | 2.9863e−07 | 600 | 577.2789 | 600 | 0.6078 | 0.3922 | 0.9621 |

Clearly, when $q \geqslant 16$, $\phi(M_q^1)$ and $\phi(M_q^3)$ are equal to $3q$, $\frac{\phi(M_q^2)}{3q}$ is less than 1 and decrease as q increase, that indicates all $x_i \in S^{q-1}$ satisfying the inequality in model (3.2) and the equality in model (3.2) holds at all support points. We also notice that the $C_q^1 r_1$ and $C_q^3 r_3$ approach to 0.5, which means the layout of design are approximating to uniform distribution in simplex.

Thus we have proved that the design $\xi_{13}^*$ is in fact D-optimal.

## 4. Concluding remarks

Under the restriction of mixture experiments, this paper establishes the D-optimality for the additive model with multi-response. The corresponding equivalence theorems are presented and used to check optimality of designs in the illustrative examples.

The support points of additive model with multi-response still vary as $q$ increase, as additive model with single-response do. And we notice that the support points, which lie in the boundary of the simplex, gather inward slower as the number of response increase. Therefore, the further research about additive model should explore the relation and regularity between tendency of support points movement and changes of response.

## Acknowledgments

This work was supported by the National Nature Sciences Foundation of China (12071096).

## Conflict of interest

The authors declare there is no conflict of interest.

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