國立中山大學應用數學研究所

碩士論文

混合實驗在Scheffé模型之正合D-最適設計

Exact D-optimal designs for mixture experiments

in Scheffé's quadratic models

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摘要

關於多項式迴歸模型下之最適設計問題已經在很多文獻中被討論到。對於定義在[a,b]之多項式迴歸型，其正合D-最適設計的最小樣本數 Huang (1987)和 Gaffke (1987)都給出了相似的充分條件。在本文中我們則是對一混合實驗模型作探討。一混合實驗為一包含q個非負成分$\{x_1,\dots,x_q\}$，且$\sum_{i=1}^{q} x_i = 1$的$q-1$維之機率空間S^{q-1}上的實驗設計。Kiefer (1961)證明了在Scheffé 的二次混合實驗模型下之D-最適設計，而基於此一結果我們證明 2 維與 3 維在 Scheffé 的二次混合實驗模型下之正合D-最適設計，並對於 4 維至 9 維的模型給出一些數值的結果。

關鍵字：Scheffé 二次模型、D-最適設計、正合D-最適設計、訊息矩陣、正交多項式。

Exact D-optimal designs for mixture experiments in Scheffé's quadratic models

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Abstract

The exact D-optimal design problems for regression models has been investigated in many literatures. Huang (1987) and Gaffke (1987) provided a sufficient condition for the minimum sample size for an certain set of candidate designs to be exact D-optimal for polynomial regression models on a compact interval. In this work we consider a mixture experiment with q nonnegative components, where the proportions of components are subject to the simplex restriction $\sum_{i=1}^{q} x_i = 1, x_i \geq 0$. The exact D-optimal designs for mixture experiments for Scheffé's quadratic models are investigated. Based on results in Kiefer (1961) results about the exact D-optimal designs for mixture models with two or three ingredients are provided and numerical verifications for models with ingredients between four and nine are presented.

Keywords : Scheffé's quadratic models, D-optimal design, exact D-optimal design, information matrix, orthonormal polynomial

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1 Introduction

Consider a mixture experiment with q nonnegative components, where the proportions of components are subject to the simplex restriction $\sum_{i=1}^{q} x_i = 1, x_i \geq 0$. The q proportions can be expressed as a column vector $\mathbf{x} = (x_1, \ldots, x_q)'$ in S^{q-1} where

$$S^{q-1} = \{(x_1, \ldots, x_q)' \in [0,1]^q : x_1 + \cdots + x_q = 1, \quad x_i \geq 0 \quad i = 1, \ldots, q\}.$$

An observation $y(\mathbf{x})$ is obtained at $\mathbf{x} \in S^{q-1}$ with $E(y(\mathbf{x})) = \beta' f(\mathbf{x})$ and variance σ^2 independent of $\mathbf{x}$, where $f(\mathbf{x})$ is a known function and β is an unknown parameter vector. In Scheffé's quadratic model the expectation is expressed as

$$E(y(\mathbf{x})) = \beta_1 x_1 + \cdots + \beta_q x_q + \beta_{12} x_1 x_2 + \cdots + \beta_{(q-1)q} x_{q-1} x_q \tag{1}$$

with regression function $f(\mathbf{x}) = (x_1, \ldots, x_q, x_{12}, \ldots, x_{(q-1)q})'$. An exact design with sample size N is a probability measure on a design space which puts weight $p_i > 0$ at n distinct support points, $n \leq N$ such that $\sum_{i=1}^{n} p_i = 1$ and Np_i, $i = 1, \ldots, n$ are integers. An approximate design removes the integer restrictions on Np_i , $i = 1, \ldots, n$. Denote a probability measure ξ for a mixture experiment as follows

$$\xi = \begin{pmatrix} \mathbf{x}_1 & \cdots & \mathbf{x}_n \\ p_1 & \cdots & p_n \end{pmatrix} = \begin{pmatrix} \begin{pmatrix} x_{1,1} \\ \vdots \\ x_{1,q} \end{pmatrix} & \cdots & \begin{pmatrix} x_{n,1} \\ \vdots \\ x_{n,q} \end{pmatrix} \\ \\ p_1 & \cdots & p_n \end{pmatrix},$$

where $\mathbf{x}_1, \ldots, \mathbf{x}_n$ denote the finite supports with the corresponding weights $p_1, \ldots, p_n$. The information matrix is therefore defined by

$$M(\xi) = \sum_{i=1}^{n} \xi(\mathbf{x_i}) f(\mathbf{x_i}) f'(\mathbf{x_i}),$$

and the corresponding dispersion function is given by

$$d(\mathbf{x}, \xi) = f'(\mathbf{x}) M^{-1}(\xi) f(\mathbf{x}).$$

According to the Equivalence Theorem (Kiefer and Wolfowitz 1960) a design ξ^* is called an approximate D-optimal designs if ξ^* maximizes $\det(M(\xi))$ over all feasible designs on the design space. Kiefer (1961) showed that an approximate design for the Scheffé's quadratic model which assigns measure $2/q(q+1)$ to each point of the $(q-1, 2)$-lattice. If an exact design ξ_N^* maximizes $\det(M(\xi_N))$ over all feasible exact designs on the design space, then it is called an exact N-points D-optimal design.

For a polynomial model on a closed interval $[a, b]$, Salaevskii (1966) conjectured that an exact design which distributes the weights as even as possible on the support points of the approximate designs. The conjecture of Salaevskii had been studied by Constantine and Studden (1981), Gaffke and Krafft (1982), Gaffke (1987), Huang (1987) and had verified the Salaevskii conjecture holds for most of the cases. Chang and Chen (2004) discussed the exact D-optimal design problem for multivariate linear polynomial models on a simplex, parallelgram and quadratic polynomial models with or without intercept on the q-ball for some cases, and provided some numerical results.

In this work, we investigate the exact D-optimal design for mixture experiments in Scheffé's quadratic models based on results in Kiefer (1961) and provide some results for models with two and three ingredients and numerical verifications for models with ingredients between four and nine.

2 Preliminaries

Kiefer (1961) proved that an approximate D-optimal design ξ^* on a simplex is supported equally with weights $p = 2/q(q+1)$ on the $(q-1,2)$-lattice with $q(q+1)/2$ points, where $\mathbf{x}_i^*$, $i = 1,\ldots,q$ are the vertexes and $\mathbf{x}_{ij}^*$, $1 \le i < j \le q$ are the certres of the sides, i.e.

$$\xi^* = \left(\begin{array}{cccccccc} \begin{pmatrix} 1 \\ 0 \\ \vdots \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ 1 \\ \vdots \\ 0 \end{pmatrix} & \cdots & \begin{pmatrix} 0 \\ 0 \\ \vdots \\ 1 \end{pmatrix} & \begin{pmatrix} 1/2 \\ 1/2 \\ \vdots \\ 0 \end{pmatrix} & \begin{pmatrix} 1/2 \\ 0 \\ 1/2 \\ \vdots \end{pmatrix} & \cdots & \begin{pmatrix} 0 \\ \vdots \\ 1/2 \\ 1/2 \end{pmatrix} \\ p & p & \cdots & p & p & p & \cdots & p \end{array} \right).$$

For the $(q-1,2)$-lattice, Kiefer (1961) provided a $q(q+1)/2$ system of quadratic orthonormal polynomials with respect to the $q(q+1)/2$ support points in ξ^* such that each of which vanishes at all other support points expect at one point of the lattice. More explicitly the system consists of the functions $[2q(q+1)]^{\frac{1}{2}} x_i(x_i - \frac{1}{2})$, $1 \le i \le q$, and $[8q(q+1)]^{\frac{1}{2}} x_i x_j$, $1 \le i < j \le q$, these functions are very useful in expressing the corresponding dispersion function for design supports on the lattice points.

Now let $g(\mathbf{x})$ denote the vertor of the orthonormal polynomials mentioned above where

$$g(\mathbf{x}) = \begin{pmatrix} g_1(\mathbf{x}) \\ g_2(\mathbf{x}) \\ \vdots \\ g_q(\mathbf{x}) \\ g_{12}(\mathbf{x}) \\ \vdots \\ g_{(q-1)q}(\mathbf{x}) \end{pmatrix} = \begin{pmatrix} 2x_1(x_1 - \frac{1}{2}) \\ 2x_2(x_2 - \frac{1}{2}) \\ \vdots \\ 2x_q(x_q - \frac{1}{2}) \\ 4x_1x_2 \\ \vdots \\ 4x_{q-1}x_q \end{pmatrix}.$$

For designs supported on $S^* = \{\mathbf{x}_1^*, \ldots, \mathbf{x}_q^*, \mathbf{x}_{12}^*, \ldots, \mathbf{x}_{(q-1)q}^*\}$, we may transform the regression function $f(\mathbf{x})$ in the Scheffé's quadratic model into $g(\mathbf{x})$. More explicitly $g(\mathbf{x})$ may be expressed as

$$g(\mathbf{x}) = \mathbf{F}^{-1} f(\mathbf{x}), \quad \mathbf{x} \in S^{q-1}$$

where $\mathbf{F} = (f(\mathbf{x}_1^*), \ldots, f(\mathbf{x}_k^*), f(\mathbf{x}_{12}^*), \ldots, f(\mathbf{x}_{(q-1)q}^*)$.

Furthermore by the celebrated Kiefer-Wolfowitz Equivalence Theorem it can be shown

$$\sum_{i=1}^{q} g_i^2(\mathbf{x}) + \sum_{i\leq j} g_{ij}^2(\mathbf{x}) \leq 1, \ \ \forall \mathbf{x} \in S^{q-1}.$$

Similarly as in the Salaevskii conjecture, if the total number of trials is $N = kp + t$ where $k = q(q+1)/2$, $p, t \in \mathbb{N}$, $t < k$, there are $\left(\begin{array}{c} k \\ t \end{array} \right)$ different cnadidate exact designs $\xi_{N,T}^*$ with $|T| = t$, $T \subset \mathbb{T}$, originated from ξ^* and $\mathbb{T} = \{1, \ldots, k, 12, \ldots, (q-1)q\}$ is the index set. That is

$$\xi_{N,T}^* = \left\{ \begin{array}{cccccc} \mathbf{x}_1^* & \cdots & \mathbf{x}_q^* & \mathbf{x}_{12}^* & \cdots & \mathbf{x}_{(q-1)q}^* \\ n_1/N & \ldots & n_q/N & n_{q+1}/N & \ldots & n_k/N \end{array} \right\} \tag{2}$$

where $n_\ell = p + 1$ if $\ell \in T$ or $n_\ell = p$ if $\ell \notin T$. Then the information matrix of $\xi_{N,T}^*$ can be expressed as

$$\begin{aligned} M(\xi_{N,T}^*) &= \sum_{i\in\mathbb{T}} p_i f(x_i) f'(x_i) = \sum_{i\in\mathbb{T}} p_i F g(x_i) g'(x_i) F' \\ &= F \left(\begin{array}{ccc} p_1 & \cdots & 0 \\ \vdots & \ddots & \vdots \\ 0 & \cdots & p_{(q-1)q} \end{array} \right) F' \end{aligned}$$

and the inverse matrix of $M(\xi_{N,T}^*)$ is

$$M^{-1}(\xi_{N,T}^*) = (F^{-1})' \left(\begin{array}{ccc} p_1^{-1} & \cdots & 0 \\ \vdots & \ddots & \vdots \\ 0 & \cdots & p_{(q-1)q}^{-1} \end{array} \right) F^{-1}.$$

Hence, the dispersion function of $\xi_{N,T}^*$ is rewritten as

$$\begin{aligned} d(x, \xi_{N,T}^*) &= f'(x) M^{-1}(\xi_{N,T}^*) f(x) \\ &= g'(x) F' (F^{-1})' \left(\begin{array}{ccc} p_1^{-1} & \cdots & 0 \\ \vdots & \ddots & \vdots \\ 0 & \cdots & p_{(q-1)q}^{-1} \end{array} \right) F^{-1} F g(x) \\ &= \sum_{\ell\in\mathbb{T}} \frac{g_\ell^2(x)}{p_\ell}, \end{aligned}$$

where $p_\ell = (p+1)/N$ if $\ell \in T$ and $p_\ell = p/N$, $\ell \notin T$.

Gaffke and Krafft (1982) proved the exact D-optimality of the above candidate designs for quadratic regression on $[a, b]$, based on the geometric-arithmetic means inequality of the information matrix i.e.

$$\frac{\det(M(\xi_1))}{\det(M(\xi_2))} \leq \left[\frac{1}{k}\mathrm{Tr}(M^{-1}(\xi_2)M(\xi_1))\right]^k, \tag{3}$$

where ξ_1 and ξ_2 are two arbitrary designs defined on $[a, b]$. Note that

$$\mathrm{Tr}(M^{-1}(\xi_2)M(\xi_1)) = \frac{1}{N}\sum_{i=1}^{N} f'(\mathbf{x_i})M^{-1}(\xi_2)f(\mathbf{x_i}) = \frac{1}{N}\sum_{i=1}^{N} d(\mathbf{x_i}, \xi_2),$$

where $\mathbf{x_i}$, $i = 1, \ldots, N$ are the corresponding design points of ξ_1.

Gaffke (1987) and Huang (1987) independently verified Salaevskii's conjecture for the most of the cases of polynomial regression. In the following we provide some lemmas analogous to that in Huang (1987) which will be useful to prove the main results.

Lemma 1. *There exists $p_0 \in \mathbb{N}$ such that for all $p \geq p_0$*

$$\sum_{\ell \in \mathbb{T}} g_\ell^2(\mathbf{x}) \leq \frac{p_0}{p_0+1} + \frac{R^2(\mathbf{x})}{p_0+1} \leq \frac{p}{p+1} + \frac{R^2(\mathbf{x})}{p+1}, \tag{4}$$

where

$$R^2(\mathbf{x}) = \max\{g_\ell^2(\mathbf{x}), \ell \in \mathbb{T}\}, \quad \forall \mathbf{x} \in S^{q-1}. \tag{5}$$

Note that (4) can be rewritten in another form as

$$\frac{1 - R^2(\mathbf{x})}{1 - \sum_{\ell \in \mathbb{T}}^{k} g_\ell^2(\mathbf{x})} \leq p_0 + 1. \tag{6}$$

The proofs of Lemma 1 for $q = 2$ and 3 are deferred to Section 3.

Let

$$A_\ell = \{\mathbf{x} | R^2(\mathbf{x}) = g_\ell^2(\mathbf{x}),\ \mathbf{x} \in S^{q-1}\}, \quad \ell \in \mathbb{T}, \tag{7}$$

then we define the region A_T used in Lemma 2 where

$$A_T = \bigcup_{\ell \in T} A_\ell, \quad T \subset \mathbb{T}.$$

Lemma 2. *For all $p \geq p_0$, $p \in \mathbb{N}$, let $\xi^*_{N,T}$ be as defined in (2) with $T \subset \mathbb{T}$, $N = kp + t$, then*

$$\begin{aligned}
\frac{1}{N} d(\mathbf{x}, \xi^*_{N,T}) &\leq \frac{1}{p+1}, \ \forall \mathbf{x} \in A_T, \\
\frac{1}{N} d(\mathbf{x}, \xi^*_{N,T}) &\leq \frac{1}{p}, \qquad \forall \mathbf{x} \notin A_T.
\end{aligned}$$

Conjecture 1. *For $N = kp + t \geq kp_0$, where $1 \leq t \leq k - 1$, and p_0 is defined as in (4), each of the $\xi^*_{N,T}$ designs is exact D-optimal.*

The key steps of proving Conjecture 1 is based on the fact that if conditions in Lemmas 1 and 2 hold, then for any exact design with N trials supported on $\{\mathbf{x_1}, \ldots, \mathbf{x_N}\}$, there is $T \subset \mathbb{T}$ such that $n_1 \geq t(p + 1)$ where n_1 is the amount of trials in A_T, $1 \leq n_1 \leq N$, and for design $\xi^*_{N,T}$,

$$\begin{aligned}
\frac{1}{N} \sum_{i=1}^{N} d(\mathbf{x_i}, \xi^*_{N,T}) &\leq \frac{n_1}{p+1} + \frac{N - n_1}{p} = \frac{N(p+1) - n_1}{p(p+1)} \\
&\leq \frac{N(p+1) - t(p+1)}{p(p+1)} = k.
\end{aligned}$$

This implies that $\det(M(\xi)) \leq \det(M(\xi^*_{N,T}))$ by (3).

Hence, Lemma 2 and Conjecture 1 will hold for Scheffé's quadratic models if p_0 defined in Lemma 1 can be found.

In the following we find the exact D-optimal designs for Scheffé's quadratic models with two or three ingredients. We will show in two steps. First we will identify the maximum functions $R(\mathbf{x})$ in the design region and use it to partition S^{q-1} into several regions according to which $g_\ell(\mathbf{x})$ is the maximum functioin. Then we can find p_0 defined in Lemma 1 such that inequality (1) holds. Later, we find exact D-optimal designs for some sample sizes.

3 Two and three ingredients

In this section we would like to find the minimum sample size such that Conjecture 1 holds for two and three ingredients. Lemma 1 is the main tool to derive the results.

3.1 Two ingredients

We start from experiments with two ingredients that $q = 2$. Then the model is

$$E[y(\mathbf{x})] = \beta_1 x_1 + \beta_2 x_2 + \beta_{12} x_1 x_2, \quad \mathbf{x} \in S^1.$$

For this model, the approximate D-optimal design ξ^* is given by

$$\xi^* = \begin{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ 1 \end{pmatrix} & \begin{pmatrix} 1/2 \\ 1/2 \end{pmatrix} \\ 1/3 & 1/3 & 1/3 \end{pmatrix}$$

and the quadratic orthonormal polynomials with respect to ξ^* are $g_1^2(\mathbf{x})$, $g_2^2(\mathbf{x})$ and $g_{12}^2(\mathbf{x})$ as defined in Section 2.

For $q = 2$, we find $R^2(\mathbf{x})$ as defined in (5) and the regions A_ℓ as defined in (7) with respect to $g_1^2(\mathbf{x})$, $g_2^2(\mathbf{x})$ and $g_{12}^2(\mathbf{x})$. Then Theorem 1 holds for the special case with $q = 2$ as below.

Theorem 1. *The design $\xi^*_{N,T}$ as defined in (2) is an exact D-optimal design in Scheffé's quadratic models with two ingredients where $T \subset \mathbb{T} = \{1, 2, 12\}$ if $N \geq 3$.*

Proof. Firstly, we identify the sets A_ℓ, $\ell \in \mathbb{T}$ as defined in Section 2. Let $g_1^2(\mathbf{x}) \geq g_2^2(\mathbf{x})$ and $g_1^2(\mathbf{x}) \geq g_{12}^2(\mathbf{x})$ then we have $A_1 = \{\mathbf{x} | \frac{5}{6} \leq x_1 \leq 1, \mathbf{x} \in S^1\}$. Similarly, it can be found that $A_2 = \{\mathbf{x} | 0 \leq x_1 \leq \frac{1}{6}, \mathbf{x} \in S^1\}$ and $A_{12} = \{\mathbf{x} | \frac{1}{6} \leq x_1 \leq \frac{5}{6}, \mathbf{x} \in S^1\}$. Let

$L(\mathbf{x}) = (1 - R^2(\mathbf{x}))/(1 - \sum_{\ell \in \mathbb{T}} g_\ell^2(\mathbf{x}))$ and $L'(\mathbf{x})$ and $L''(\mathbf{x})$ denote the first and second order derivative of $L(\mathbf{x})$. Note that in A_1, $L(\mathbf{x}) = (1 - g_1^2(\mathbf{x}))/(1 - \sum_{\ell \in \mathbb{T}} g_\ell^2(\mathbf{x}))$. Since $x_2 = 1 - x_1$, then for $\mathbf{x} \in A_1$ we have

$$L(\mathbf{x}) = \frac{1 + x_1 + 4x_1^3}{6(1 - 2x_1)^2 x_1},$$

and

$$L'(\mathbf{x}) = \frac{1 - 6x_1 - 4x_1^2 - 8x_1^3}{6x_1^2(-1 + 2x_1)^3}.$$

It is easy to see that $L'(\mathbf{x}) < 0$ by $\mathbf{x} \in A_1$, then $L(\mathbf{x})$ is monotone decreasing in A_1 and the maximum of $L(\mathbf{x})$ is $28/15 \approx 1.8667$ on $x_1 = 5/6$. Since $L(\mathbf{x})$ is symmetric with $x_1 = x_2$, the maximum of $L(\mathbf{x})$ in A_2 is also $28/15$. Similarly for all $\mathbf{x} \in A_{12}$, $L(\mathbf{x}) = (1 - g_{12}^2(\mathbf{x}))/(1 - \sum_{\ell \in \mathbb{T}}^k g_\ell^2(\mathbf{x}))$, then

$$L(\mathbf{x}) = \frac{1 + x_1 - 4x_1^2}{6x_1 - 6x_1^2},$$

and

$$L''(\mathbf{x}) = \frac{-1 + 3x_1 - 3x_1^2}{3(-1 + x_1)^3 x_1^3}.$$

Also we find that $L''(\mathbf{x}) > 0$, $\forall \mathbf{x} \in A_{12}$, then $L(\mathbf{x})$ is convex in A_{12} and the maximum of $L(\mathbf{x})$ is $28/15$ on $x_1 = 1/6$ and $x_1 = 5/6$. From (6) we have $p_0 = 1$ and by Lemma 2, Conjecture 1 holds for Scheffé's quadratic models with two ingredients if the total trials $N \geq kp_0 = 3$. □

3.2 Three ingredients

Now we discuss the case with ingredients $q = 3$, the Scheffé's model turns to

$$E[y(\mathbf{x})] = \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3, \quad \mathbf{x} \in S^2.$$

Then the approximate D-optimal design is given by

$$\xi^* = \left(\begin{array}{cccccc} \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} & \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} & \begin{pmatrix} \frac{1}{2} \\ \frac{1}{2} \\ 0 \end{pmatrix} & \begin{pmatrix} \frac{1}{2} \\ 0 \\ \frac{1}{2} \end{pmatrix} & \begin{pmatrix} 0 \\ \frac{1}{2} \\ \frac{1}{2} \end{pmatrix} \\ 1/6 & 1/6 & 1/6 & 1/6 & 1/6 & 1/6 \end{array} \right),$$

and the corresponding orthonormal polynomials are $\{g_\ell(\mathbf{x}), \ell \in \mathbb{T}\}$ where $g_\ell(\mathbf{x})$, $\ell \in \mathbb{T}$ are defined as in Section 2 and $\mathbb{T} = \{1, 2, 3, 12, 13, 23\}$. Similarly, we must identify the regions of A_ℓ for every $g_\ell^2(\mathbf{x})$, $\ell \in \mathbb{T}$. In order to find each A_ℓ, $\ell \in \mathbb{T}$, we first define the following sets A_i^* and A_{ij}^*; for $i = 1, \ldots, k$,

$$A_i^* = \left\{\mathbf{x} | \mathbf{x} \in S^{q-1},\ g_i^2(\mathbf{x}) \geq g_j^2(\mathbf{x}), 1 \leq j \leq q, j \neq i\right\}$$

where $g_i(\mathbf{x})$ is the corresponding orthonormal polynomial with $g_i(\mathbf{x_i}^*) = 1$, and for $1 \leq i < j \leq k$,

$$A_{ij}^* = \{\mathbf{x} | \mathbf{x} \in S^{q-1},\ g_{ij}^2(\mathbf{x}) \geq g_{kl}^2(\mathbf{x}),\ g_{ij}^2(\mathbf{x}) \geq g_i^2(\mathbf{x}),$$
$$g_{ij}^2(\mathbf{x}) \geq g_j^2(\mathbf{x}),\ 1 \leq k < l \leq q,\ kl \neq ij\}$$

where $g_{ij}(\mathbf{x})$ is the corresponding orthonormal polynomial with $g_{ij}(\mathbf{x_{ij}}^*) = 1$.

For example, we have A_1^* and A_{12}^* being the sets that

$$A_1^* = \{\mathbf{x} | \mathbf{x} \in S^2,\ g_1^2(\mathbf{x}) \geq g_{12}^2(\mathbf{x}),\ g_1^2(\mathbf{x}) \geq g_{13}^2(\mathbf{x})\},$$
$$A_{12}^* = \{\mathbf{x} | \mathbf{x} \in S^2,\ g_{12}^2(\mathbf{x}) \geq g_1^2(\mathbf{x}),\ g_{12}^2(\mathbf{x}) \geq g_2^2(\mathbf{x}),$$
$$g_{12}^2(\mathbf{x}) \geq g_{13}^2(\mathbf{x}),\ g_{12}^2(\mathbf{x}) \geq g_{23}^2(\mathbf{x})\}.$$

It is obvious that

$$\bigcup_{\ell \in \mathbb{T}} A_\ell^* = S^{q-1}.$$

For $q = 3$, by comparing the functions $g_i^2(\mathbf{x})$ w.r.p $g_j^2(\mathbf{x})$ as well as with $g_{ij}^2(\mathbf{x})$, we have

$$A_i^* = \{\mathbf{x} | \mathbf{x} \in S^2,\ x_i \geq \frac{1}{2} + 2x_j,\ j \neq i, \},\quad i = 1, 2, 3,$$

as well as for $1 \leq i < j \leq 3$,

$$A^*_{ij} = \{\mathbf{x}|\mathbf{x} \in S^2,\ x_i \leq \frac{1}{2} + 2x_j,\ x_j \leq \frac{1}{2} + 2x_i,$$
$$x_i \geq x_k,\ x_j \geq x_k,\ k \neq i, j,\ k = 1, 2, 3\}.$$

In the following Lemma 3, it shows that $A_i = A^*_i$ and $A_{ij} = A^*_{ij}$.

Lemma 3. *For all* $\mathbf{x} \in A^*_\ell$*,* $g^2_\ell(\mathbf{x})$ *is the maximum among all* $g^2_i(\mathbf{x})$ *where* $\ell, i \in \mathbb{T}$*. That is* $A_\ell = A^*_\ell$*.*

Proof. Because in S^2, $g^2_2(\mathbf{x})$ and $g^2_3(\mathbf{x})$ are symmetric with $g^2_1(\mathbf{x})$ for $x_1 = x_2$ and $x_1 = x_3$, $g^2_{13}(\mathbf{x})$ and $g^2_{23}(\mathbf{x})$ are symmetric with $g^2_{12}(\mathbf{x})$ for $x_2 = x_3$ and $x_1 = x_3$, we only need to verify that $g_1(\mathbf{x})$ is the maximum in A_1 and $g_{12}(\mathbf{x})$ is the maximum in A_{12}. Firstly, form the definition of A^*_1, we have for all $\mathbf{x} \in A^*_1$, $g_1(\mathbf{x}) \geq g_{12}(\mathbf{x})$ and $g_1(\mathbf{x}) \geq g_{13}(\mathbf{x})$. Now we verify that $g^2_{12}(\mathbf{x}) \geq g^2_{23}(\mathbf{x})$, $\forall \mathbf{x} \in A^*_1$ which in turns implies $g_1(\mathbf{x}) \geq g_{23}(\mathbf{x})$. Note that $g^2_{12}(\mathbf{x})$ can be expressed as

$$\begin{aligned} g^2_{12}(\mathbf{x}) &= 16x_1^2x_2^2 = 16(1 - x_2 - x_3)^2x_2^2 \\ &= 16x_2^2x_3^2 + 16(1 + x_2^2 + x_3^2 - 2(x_2 + x_3) + 2x_2x_3)x_2^2. \end{aligned}$$

Since $x_2 + x_3 \leq \frac{1}{2}$, for all $\mathbf{x} \in A^*_1$, we have

$$g^2_{12}(\mathbf{x}) \geq 16x_2^2x_3^2 = g^2_{23}(\mathbf{x}).$$

Similarly, it can be shown that

$$\begin{aligned} g^2_{12}(\mathbf{x}) &= 16x_1^2x_2^2 \\ &\geq 16x_2^2(\frac{1}{2}(1 + 4x_2))^2 = 4x_2^2(1 + 4x_2)^2 \\ &\geq 4x_2^2(x_2 - \frac{1}{2})^2 = g^2_2(\mathbf{x}) \end{aligned}$$

then $g_1^2(\mathbf{x}) \geq g_{23}^2(\mathbf{x})$ and $g_1^2(\mathbf{x}) \geq g_2^2(\mathbf{x})$. Again, as $g_1^2(\mathbf{x}) \geq g_{13}^2(\mathbf{x})$, $\forall \mathbf{x} \in A_1^*$, it implies $g_1^2(\mathbf{x}) \geq g_{23}^2(\mathbf{x})$ and $g_1^2(\mathbf{x}) \geq g_3^2(\mathbf{x})$. Therefore $g_1^2(\mathbf{x})$ is the maximum function on A_1^* and $A_1^* \subset A_1$. Secondly, from definition of A_{12}^* is given above where

$$A_{12}^* = \{\mathbf{x}|\mathbf{x} \in S^2, x_1 \leq \frac{1}{2} + 2x_2, x_2 \leq \frac{1}{2} + 2x_1, x_1 \geq x_3, x_2 \geq x_3\}.$$

We now check $g_{12}^2(\mathbf{x}) \geq g_3^2(\mathbf{x})$ on A_{12}^* by expressing in the follow

$$g_{12}^2(\mathbf{x}) - g_3^2(\mathbf{x}) = (4x_1x_2 + 2x_3(x_3 - 1/2))(4x_1x_2 - 2x_3(x_3 - 1/2)).$$

Because for all $\mathbf{x} \in A_{12}^*$, $x_3 \leq 1/3$, then $(4x_1x_2 - 2x_3(x_3 - 1/2)) \geq 0$. In fact as $x_3 = 1 - x_1 - x_2$, the other part $4x_1x_2 + 2x_3(x_3 - 1/2)$ can be rewritten as $h(x_1, x_2)$ where

$$h(x_1, x_2) = 1 + 2x_1^2 - 3x_2 + 2x_2^2 + x_1(8x_2 - 3).$$

In order to find the extrme value of $h(x_1, x_2)$ in A_{12}^* we try to find the critical points. The critical points of $h(x_1, x_2)$ is $(\frac{1}{4}, \frac{1}{4}, \frac{1}{2})$ and is not in A_{12}^*. Hence the location of the minimum of $h(x_1, x_2)$ is on the boundary of A_{12}^* and the minimum value is $\frac{9}{32}$ at $(\frac{1}{8}, \frac{3}{4}, \frac{1}{8})$ or $(\frac{3}{4}, \frac{1}{8}, \frac{1}{8})$. Then $g_{12}^2(\mathbf{x}) - g_3^2(\mathbf{x}) \geq 0$, $\mathbf{x} \in A_{12}^*$ and $R(\mathbf{x}) = g_{12}^2(\mathbf{x})$ in A_{12}^*. Then we have $A_{12}^* \subset A_{12}$.

From the above discussions we have $A_\ell^* \subset A_\ell$, $\ell \in \mathbb{T}$. It is follows that $A_\ell^* = A_\ell$, $\ell \in \mathbb{T}$ since $\cup_{\ell \in \mathbb{T}} A_\ell^* = S^{q-1}$. □

From Lemma 3 there is a partition on the design region as in Figure 1 and this partition is $P = \{A_1, A_2, A_3, A_{12}, A_{13}, A_{23}\}$. In the following we prove that Lemma 1 is ture for $p_0 = 2$.

Lemma 4. *When $p \geq 2$ the inequality in (4) holds for $\mathbf{x} \in S^2$.*

Proof. Let

$$Q(\mathbf{x}) = \sum_{i=1}^{k} g_i^2(\mathbf{x}) - \frac{R^2(\mathbf{x})}{p_0 + 1},$$

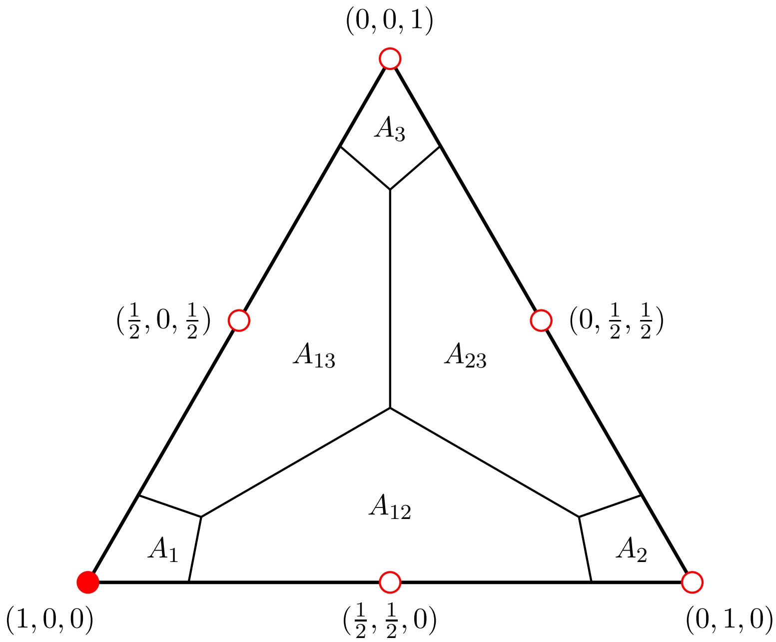

Figure 1: (2,2)-lattice

and we want to prove $Q(\mathbf{x}) \leq 2/3$. It is obvious that $Q(\mathbf{x})$ is symmetric with $x_1 = x_2$, $x_2 = x_3$, and $x_1 = x_3$. To find the maximums of $Q(\mathbf{x})$ for every $\mathbf{x} \in A_i$, is is sufficient to find the maximum of $Q(\mathbf{x})$ in A_1 and A_{12}. Then we only need to consider the regions in A_1 and A_{12}. Note that $\forall \mathbf{x} \in A_{12}$,

$$Q(\mathbf{x}) = \sum_{i=1}^{k} g_i^2(\mathbf{x}) - \frac{g_{12}^2(\mathbf{x})}{p_0 + 1}.$$

Let the parametric representation of $\mathbf{x}$ in A_{12} be $(t-s, t+s, 1-2t)$ where $1/3 \leq t \leq 1/2$ and $-a_t \leq s \leq a_t$, $a_t \in \mathbb{R}^+$. All parametric vectors for a fixed t are parallel to the vector from $(1,0,0)$ to $(0,1,0)$. Then $Q(\mathbf{x})$ can be expressed with a new function $q_{12}(s,t)$ as

$$q_{12}(s,t) = \frac{1}{3}(3 + 56s^4 - 36t + 258t^2 - 696t^3 + 632t^4 + 2s^2(51 - 228t + 232t^2)).$$

Because $q_{12}(s,t)$ is a 4^{th} degree polynomial with the highest coefficient to be positive for

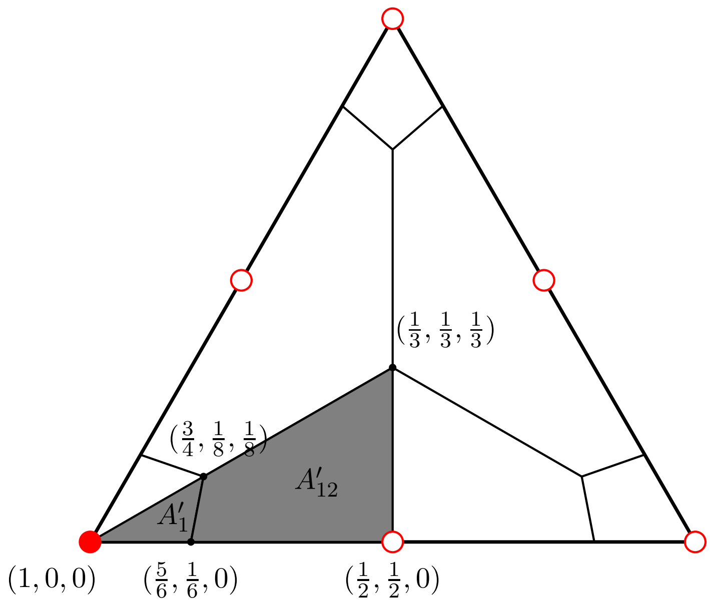

Figure 2: Subregion of A_1 and A_{12}

s and is symmetric with $s = 0$, then for a fixed t and a_t the maximum of $q_{12}(s, t)$ will be attained at $s = 0$, $s = a_t$ or $s = -a_t$. Hence, if we want to find the maximum of $Q(\mathbf{x})$ on A_{12}, it is just to find the maximum of $Q(\mathbf{x})$ on the boundary of A'_{12}, the half region of A_{12}, see Figure 2. It is obvious that the boundary of A'_{12} is composed of four segments and the boundary of A'_1 is composed of three segments. In the following we would like to find the maximum of $Q(\mathbf{x})$ on the boundary of A'_{12} and the segments are specified in the following. To this end let $p(s, t)$ or $p(t)$ denote the parametric vector of $\mathbf{x}$ on each segment.

(**i**) For segment from $(5/6, 1/6, 0)$ to $(1/6, 5/6, 0)$

It is apparent that the maximum of $Q(\mathbf{x})$ on this segment will occur at $(5/6, 1/6, 0)$, $(1/6, 5/6, 0)$ or $(1/2, 1/2, 0)$. Since $Q(\mathbf{x}) = 2/3$ at $(1/2, 1/2, 0)$ and $Q(\mathbf{x}) = 4/9$ at

$(5/6, 1/6, 0)$ and $(1/6, 5/6, 0)$, the maximum is $2/3$ in the segment.

(**ii**) For segment from $(1/3, 1/3, 1/3)$ to $(1/2, 1/2, 0)$

Let $s = 0$,

$$q_{12}(0,t) = 1 - 12t + 86t^2 - 232t^3 + \frac{632t^4}{3}, \quad \frac{1}{3} \leq t \leq \frac{1}{2},$$

then

$$q'_{12}(0,t) = \frac{4}{3}(-9 + 129t - 522t^2 + 632t^3).$$

By Bolzano Theorem, we are assured the interval where three roots of t for $q'_{12}(0,t) = 0$ locate, because $q'_{12}(1/2) = 16/3$ and $q'_{12}(1/3) = -64/81$, $q'_{12}(1/4) = 2/3$ and $q'_{12}(1/10) = -334/375$. Hence $q'_{12}(0,t)$ is positive in $1/3 \leq t \leq 1/2$ and the maximum will be attained at $t = 1/3$ or $1/2$. Moreover $q'_{12}(0, 1/3) = 137/243$ and $q'_{12}(0, 1/2) = 2/3$ then the maximum of $Q(\mathbf{x})$ in this segment is $2/3$.

(**iii**) For segment from $(1/3, 1/3, 1/3)$, to $(3/4, 1/8, 1/8)$

Let the parametric vector of $\mathbf{x}$ be $(1-t, t, t)$ where $1/8 \leq t \leq 1/3$, and let $p(t) = Q(\mathbf{x})$ then

$$p(t) = \frac{1}{3}(1 - 36t + 242t^2 - 632t^3 + 584t^4)$$

and

$$p'(t) = \frac{4}{3}(-9 + 121t - 474t^2 + 584t^3).$$

That is, there is only one real root 0.129911 for $p'(t) = 0$. Hence maximum can be found on $t = 1/8$, $t = 1/3$, or $t = 0.129911$. The maximum of $Q(\mathbf{x})$ is $127/243$ at $t = 1/3$.

(**iv**) For segment from $(3/4, 1/8, 1/8)$ to $(5/6, 1/6, 0)$

Let the parametric vector of x be $(5/6 - t/12, 1/6 - t/24, t/8)$ where $0 \leq t \leq 1$, and let $p(t) = Q(\mathbf{x})$ then

$$p(t) = \frac{1}{124416}(65536 - 45184t + 35376t^2 - 6832t^3 + 433t^4).$$

and

$$p''(t) = \frac{1}{10368}(5896 - 3416t + 433t^2).$$

Because $p''(t)$ is positive for $0 \le t \le 1$, $p(t)$ is convex and the maximum can be obtained with $t = 0$ or $t = 1$ and the maximum value is $128/243$ with $t = 0$, then the maximum of $Q(\mathbf{x})$ is $2/3$.

From the above (i)-(iv), we see that the maximum of $Q(\mathbf{x})$ on the boundary of A'_{12} is less than or equal to $2/3$, then the maximum of $Q(\mathbf{x})$ on A_{12} is $2/3$.

In A_1, we can obtain the maximum of $Q(\mathbf{x})$ with the same procedure. It can be found that $Q(\mathbf{x})$ is convex on each segment of the boundary of A_1 and the maximum is $2/3$ at $(1, 0, 0)$. Then $Q(\mathbf{x})$ is also less than or equal to $2/3$ in S^2 and the proof is complete. □

From Lemma 4 and Theorem 1, exact D-optimal design in Scheffé's quadratic models for $q = 3$ and $N \ge 12$ is stated explicity in Theorem 2.

Theorem 2. ξ_n^* *in* (2) *is an exact D-optimal design in Scheffé's quadratic models with three ingredients for sample size* $N \ge 12$.

4 Four or more ingredients

In Section 3, we have discussed cases with two and three ingredients. For four or more ingredients the same method as in Section 3 can be used, but it is more complicated to calculate the maximum on the subregion with respect to those orthonormal polynomials. In (4), we can see that the right side of the inequality is an increasing function of p for fixed $R^2(\mathbf{x})$. If a real solution $p \in \mathbf{R}$ can be found for the equality in (4) holds for each $\mathbf{x} \in S^{q-1}$ as expressed in (8), the minimum integer upper bound for these p, called p_0

can be obtained, then Conjecture 1 will hold in S^{q-1}. Here p_0 is the same as defined in Lemma 1.

$$\sum_{\ell \in \mathbb{T}} g_\ell^2(\mathbf{x}) = \frac{p}{p+1} + \frac{R^2(\mathbf{x})}{p+1} = 1 - \frac{1 - R^2(\mathbf{x})}{p+1}, \qquad \mathbf{x} \in S^{q-1}. \tag{8}$$

We try to compute the p_0 numerically by generating 100000 points uniformly on the design regions S^{q-1} and solve for p in (7) with each generated $\mathbf{x}$ in S^{q-1}, then find the corresponding p_0 through the maximum p resulted from the 100000 points. Table 1 lists the maximum p values for ingredients from 4 to 9.

Table 1: Maximum p for ingredients from 4 to 9

ingredients	3	4	5	6	7	8	9
p	1.16143	1.14157	1.10068	1.10919	1.12676	1.13648	1.10333

It is obvious that in this table p is always less than 7/6, where 7/6 is also the p value for (7) to hold when $\mathbf{x}$ is on the centroids of depth 3. Hence the integer supremum of p seems to be 2 for every ingredient. For this result we propose a conjecture below.

Conjecture 2. *For any finite sample size $N \geq kp_0$, $p_0 = 2$, $k = q(q+1)/2$, and q is the ingredient, there is at least one design as defined in* (2) *for mixture experiments in Scheffé's quadratic models to be exact D-optimal.*

5 Conclusion

In Section 3 we prove the exact D-optimality of candidate designs for mixture experiments with ingredients $q = 2$ and $q = 3$ for most of the sample size N. For experiments with the number of ingredients range from 4 to 9, we obtain the numerically computed

p_0, or the smallest sample size N for the candidate designs to be exact D-optimal. The proofs for the general cases will be investigated in the future.

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