Statistica Sinica **8**(1998), 979-990

A-OPTIMAL DESIGNS FOR AN ADDITIVE QUADRATIC MIXTURE MODEL

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Abstract: Quadratic models are widely used in the analysis of experiments involving mixtures. This paper gives A-optimal designs for an additive quadratic mixture model for $q \geq 3$ mixture components. It is proved that in these A-optimal designs, vertices of the simplex S^{q-1} are support points, and other support points shift gradually from barycentres of depth 1 to barycentres of depth 3 as q increases. A-optimal designs with minimal support are also discussed.

Key words and phrases: Additive model, A-optimal design, experiments with mixtures.

1. Introduction

Suppose that in an experiment with a mixture of $q \geq 2$ ingredients, the response depends on the relative proportions $x_1,\ldots,x_q$ of the ingredients. Let $\mathbf{x}' = (x_1, \ldots, x_q)$, so that $\mathbf{x}$ belongs to the $(q-1)$-dimensional simplex $S^{q-1} = \{(x_1, \ldots, x_q)' : x_1 + \cdots + x_q = 1, x_i \geq 0, 1 \leq i \leq q\}$. Let the observed response be expressed as $y = \eta(\mathbf{x}) + \varepsilon(\mathbf{x})$, where $\eta(\mathbf{x})$ is the expected response and $\varepsilon(\mathbf{x})$ is the error at $\mathbf{x}$. We assume that for independent observations, the errors $\varepsilon(\mathbf{x})$ are statistically independent and have mean zero and the same variance. The following quadratic mixture model for $\eta(\mathbf{x})$ was first studied by Darroch and Waller (1985) for the case $q = 3$:

$$\eta_{DW2}(\mathbf{x}) = \sum_{1\leq i\leq q} \alpha_i x_i + \sum_{1\leq i\leq q} \alpha_{ii} x_i(1 - x_i). \tag{1.1}$$

Another commonly used mixture model is the following full quadratic model due to Scheffé (1958):

$$\eta_{q,2}(\mathbf{x}) = \sum_{1\leq i\leq q} \beta_i x_i + \sum_{1\leq i<j\leq q} \beta_{ij} x_i x_j. \tag{1.2}$$

When $q = 2, 3$, models $\eta_{DW2}(\mathbf{x})$ and $\eta_{q,2}(\mathbf{x})$ are equivalent, but when $q = 2$ the coefficients α_{11} and α_{22} in $\eta_{DW2}(\mathbf{x})$ are not uniquely determined. When $q \geq 4$,

$\eta_{DW2}(\mathbf{x})$ is a special case of $\eta_{q,2}(\mathbf{x})$ expressed by (1.2) in which the coefficients β_{ij} are governed by a system of linear constraints.

Model $\eta_{DW2}(\mathbf{x})$ expressed by (1.1) is additive (Hastie and Tibshirani (1990), Sections 1.1 and 4.3) in the mixture components, in the sense that it is a sum of separate functions of $x_1, \ldots, x_q$. When $x_1, \ldots, x_q$ vary but the sums $x_1 + \cdots + x_s$ and $x_{s+1} + \cdots + x_q (1 < s < q)$ are kept fixed, the total effect on $\eta_{DW2}(\mathbf{x})$ is the sum of the effects of varying $x_1, \ldots, x_s$ and $x_{s+1}, \ldots, x_q$ separately. This additivity property of $\eta_{DW2}(\mathbf{x})$ can be used to study additivity effects of mixture components on the response, while Scheffé's full quadratic model $\eta_{q,2}(\mathbf{x})$ is not appropriate for this study because it contains all 2-factor interaction terms $x_i x_j$.

The objective of this paper is to obtain A-optimal designs analytically for model $\eta_{DW2}(\mathbf{x})$ expressed by (1.1). Various results on optimal designs are available for other mixture models (cf. Kiefer (1961), Lim (1990), Mikaeili (1989, 1993), Uranisi (1964), He and Guan (1990), Chan (1988, 1992), Xue and Guan (1993), and so on). Chan (1995) provides a comprehensive review on optimal designs of mixture models.

2. Designs and Barycentres on S^{q-1}

The model $\eta_{DW2}(\mathbf{x})$ can be expressed as $\eta_{DW2}(\mathbf{x}) = \boldsymbol{\theta}'\mathbf{f}(\mathbf{x})$, where $\boldsymbol{\theta}$ and $\mathbf{f}(\mathbf{x})$ are column vectors of length $2q$ defined by $\boldsymbol{\theta} = (\alpha_1, \ldots, \alpha_q, \alpha_{11}, \ldots, \alpha_{qq})'$ and $\mathbf{f}(\mathbf{x}) = (x_1, \ldots, x_q, x_1(1-x_1), \ldots, x_q(1-x_q))'$.

Given N support points $\mathbf{x}_1, \ldots, \mathbf{x}_N$ in the design space, the design matrix is defined as $(\mathbf{x}_1 \ldots, \mathbf{x}_N)'$, and the model matrix (Pukelsheim (1993), Section 1.25) or extended design matrix (Atkinson and Donev (1996), Section 5.2) is defined as $(\mathbf{f}(\mathbf{x}_1), \ldots, \mathbf{f}(\mathbf{x}_N))'$. We shall denote by $\boldsymbol{\xi}$ the design which assigns a weight ξ_j to the point $\mathbf{x}_j (j = 1, \ldots, N)$. A design $\boldsymbol{\xi}$ with non-singular moment matrix (Pukelsheim (1993), Section 1.24) $M(\boldsymbol{\xi}) = \sum_{j=1}^{N} \xi_j \mathbf{f}(\mathbf{x}_j)\mathbf{f}'(\mathbf{x}_j)$ is said to be A-optimal if it minimizes $\mathrm{tr}M^{-1}(\boldsymbol{\xi})$.

A point $\mathbf{x} \in S^{q-1}$ is called a barycentre of depth j $(0 \le j \le q-1)$ if $j+1$ of its q coordinates are equal to $1/(j+1)$ and the remaining ones are equal to zero (Galil and Kiefer (1977)). The collection of all barycentres of depth j is denoted by J_j. We define $J = \bigcup_{j=0}^{q-1} J_j$. A design which assigns a weight r_{j+1} to each point in $J_j (0 \le j \le q-1)$ is called a symmetric weighted centroid design (cf. Scheffé (1963)). For the rest of this paper, we shall reserve the symbols $r_1, \ldots, r_q$ to denote such weights assigned to each point in $J_0, \ldots, J_{q-1}$, respectively. In this paper, for convenience we shall use $C(q,j)$ to denote the binomial coefficient $q!/[j!(q-j)!]$. By the definition of $r_j (j = 1, \ldots, q)$, it is obvious that we require $C(q,1)r_1 + \cdots + C(q,j)r_j + \cdots + C(q,q)r_q = 1$.

In the next section, we shall see that only barycentres are possible support points for A-optimal designs for model $\eta_{DW2}(\mathbf{x})$.

3. A-optimal Designs for $\eta_{DW2}(\mathbf{x})$

According to an equivalence theorem of Kiefer (1974, 1975), a design $\boldsymbol{\xi}$ is A-optimal if and only if $\mathbf{f}'(\mathbf{x})M^{-2}(\boldsymbol{\xi})\mathbf{f}(\mathbf{x}) \leq \operatorname{tr} M^{-1}(\boldsymbol{\xi})$ for all $\mathbf{x}$ in the design space, and all points in the support of an A-optimal design must achieve the equality.

In what follows we shall write

$$d_2(\mathbf{x},\boldsymbol{\xi}) = \mathbf{f}'(\mathbf{x})M^{-2}(\boldsymbol{\xi})\mathbf{f}(\mathbf{x}). \tag{3.1}$$

It is not difficult to mimic an argument of Atwood (1969), pp.1573–1574 to show that for model $\eta_{DW2}(\mathbf{x})$, $d_2(\mathbf{x},\boldsymbol{\xi})$ attains its maximum only at the barycentres of S^{q-1}. Hence only the barycentres are possible support points for A-optimal designs, and in order to prove that a design $\boldsymbol{\xi}$ is A-optimal on S^{q-1} it suffices to prove that

$$\operatorname{tr} M^{-1}(\boldsymbol{\xi}) - d_2(\mathbf{x},\boldsymbol{\xi}) \geq 0 \tag{3.2}$$

for all $\mathbf{x} \in J$.

For model $\eta_{DW2}(\mathbf{x})$, it is clear that a model matrix generated by all points in J_0 is $(\boldsymbol{I}_q, \mathbf{0}_q)$, where $\boldsymbol{I}_q$ and $\mathbf{0}_q$ are the $q \times q$ identity matrix and zero matrix, respectively. It is also straightforward to see that for any fixed integer $i = 1,\ldots,q-1$, a model matrix generated by all points in J_{i-1} is $(i^{-1}M_i, (i-1)i^{-2}M_i)$, where M_i is a $C(q,i) \times q$ matrix, such that the first i elements in the first row of M_i are 1 and the remaining elements in the first row are 0, and the remaining $C(q,i)-1$ rows of M_i are the different permutations of the first row according to lexicographical order. (For example, when $q = 4$ and $i = 2$, M_i is a 6×4 matrix, and its 1st, 2nd,…, 6th rows are (1,1,0,0), (1,0,1,0), (1,0,0,1), (0,1,1,0), (0,1,0,1), (0,0,1,1), respectively.)

In what follows, for any integer $i, j (1 < i < j \leq q)$, we shall denote by $\boldsymbol{\xi}_{1,i}$ the symmetric weighted centroid design in which $r_k = 0$ except for $k = 1$ and $k = i$, and denote by $\boldsymbol{\xi}_{1,i,j}$ the symmetric weighted centroid design in which $r_k = 0$ except for $k = 1, k = i$ and $k = j$.

The matrix $M(\boldsymbol{\xi}_{1,i})$ is given by

$$M(\boldsymbol{\xi}_{1,i}) = \begin{pmatrix} r_1\boldsymbol{I}_q + r_i i^{-2}M_i'M_i & (i-1)r_i i^{-3}M_i'M_i \\ (i-1)r_i i^{-3}M_i'M_i & (i-1)^2 r_i i^{-4}M_i'M_i \end{pmatrix}, \tag{3.3}$$

and by Morrison (1976), Section 2.11 we have

$$M^{-1}(\boldsymbol{\xi}_{1,i}) = \begin{pmatrix} r_1^{-1}\boldsymbol{I}_q & -i(i-1)^{-1}r_1^{-1}\boldsymbol{I}_q \\ -i(i-1)^{-1}r_1^{-1}\boldsymbol{I}_q & i^2(i-1)^{-2}(r_1^{-1}\boldsymbol{I}_q + i^2 r_i^{-1}(M_i'M_i)^{-1}) \end{pmatrix}. \tag{3.4}$$

It is straightforward to show that

$$M_i'M_i = C(q-2,i-1)\boldsymbol{I}_q + C(q-2,i-2)\boldsymbol{J}_q, \tag{3.5}$$

where $\boldsymbol{J}_q$ is the $q \times q$ matrix with all elements equal to 1, and that

$$(M_i'M_i)^{-1} = (\boldsymbol{I}_q - (i-1)i^{-1}(q-1)^{-1}\boldsymbol{J}_q)/C(q-2,i-1). \tag{3.6}$$

Consequently, we have

$$\operatorname{tr} M^{-1}(\boldsymbol{\xi}_{1,i}) = \frac{q(2i^2-2i+1)}{(i-1)^2 r_1} + \frac{i^3 q(qi-2i+1)}{(i-1)^2(q-1)C(q-2,i-1)r_i}. \tag{3.7}$$

By the method of Lagrange multipliers, it can be shown that under the constraint $C(q,1)r_1 + C(q,i)r_i = 1$, the only critical point of $\mathrm{tr}M^{-1}(\boldsymbol{\xi}_{1,i})$ is attained at

$$C(q,1)r_1 = 1/(1+\alpha(q,i)), C(q,i)r_i = \alpha(q,i)/(1+\alpha(q,i)), \tag{3.8}$$

where

$$\alpha(q,i) = i(qi-2i+1)^{1/2}((q-i)(2i^2-2i+1))^{-1/2}. \tag{3.9}$$

Since $\mathrm{tr}M^{-1}(\boldsymbol{\xi}_{1,i}) \geq 0$ and $\mathrm{tr}M^{-1}(\boldsymbol{\xi}_{1,i}) \to \infty$ as $r_1 \to 0+$ or $r_i \to 0+$, the above critical point must be an absolute minimum.

We have the following results for A-optimal designs for $\eta_{DW2}(\mathbf{x})$:

Theorem 1. *When $q = 3, 4$, the design $\boldsymbol{\xi}_{1,2}$ is A-optimal for $\eta_{DW2}(\mathbf{x})$ on the design space S^{q-1}, where r_1, r_2 are given by* (3.8) *and* (3.9) *($i = 2$).*

Theorem 2. *When $5 \leq q \leq 21$, the design $\boldsymbol{\xi}_{1,3}$ is A-optimal for $\eta_{DW2}(\mathbf{x})$ on the design space S^{q-1}, where r_1, r_3 are given by* (3.8) *and* (3.9) *($i = 3$).*

Theorem 3. *When $q \geq 26$, the design $\boldsymbol{\xi}_{1,4}$ is A-optimal for $\eta_{DW2}(\mathbf{x})$ on the design space S^{q-1}, where r_1, r_4 are given by* (3.8) *and* (3.9) *($i = 4$).*

Numerical values of r_1, r_2, r_3, r_4 in Theorems 1 - 3 are given in Table 1 in Section 5. It is verified numerically that when $q = 22, 23, 24, 25$, the design $\boldsymbol{\xi}_{1,3,4}$ is A-optimal for $\eta_{DW2}(\mathbf{x})$ on the design space S^{q-1}, where the numerical values of r_1, r_3, r_4, rounded off at the 4th decimal place, are also given in Table 1 in Section 5.

The proofs of Theorems 1 - 3 and the algebraic computations for $q = 22, 23, 24, 25$ will be given in the Appendix.

4. Asymmetric Weighted Centroid Design

Note that the inverse of the moment matrix of an A-optimal design for $\eta_{DW2}(\mathbf{x})$ is unique and is determined by (3.4). Out of all the points in J, only those in J_0 can generate the identity matrix $\boldsymbol{I}_q$ in (3.3) and (3.4). Therefore all the points in J_0 must be included in each A-optimal design, and the weights assigned to these points must be equal. So, an alternative A-optimal design will vary only by assigning different frequencies λ_j to each $\mathbf{x}_j \in J_{i-1}$. Thus the symmetric matrix $M_i'M_i$ in (3.3) and (3.4) determined by (3.5) has to fulfill the equation:

$$M_i'M_i = C(q-2, i-1)\boldsymbol{I}_q + C(q-2, i-2)\boldsymbol{J}_q = \sum_{\mathbf{x}_j \in J_{i-1}} \lambda_j \mathbf{x}_j \mathbf{x}_j'. \tag{4.1}$$

This results in a system of $q(q+1)/2$ linear equations in λ_j, where $0 \le \lambda_j \le C(q,i)$ and $\sum_{j=1}^{C(q,i)} \lambda_j = C(q,i)$. Clearly, the design with $\lambda_j = 1$ for all points in J_{i-1} is one solution. When $q \ge 7$, other solutions for λ_j can be obtained, because the total number of points in J_2 is $C(q,3) > 1 + q(q+1)/2$ (cf. Carathéodory's Theorem (Silvey (1980), p. 72)). This results in asymmetric designs. When $q = 6$, asymmetric designs can also be constructed.

As an illustration of construction of an asymmetric A-optimal design, consider the case $q = 6$. Theorem 2 implies that $i = 3$, and this leads to $M_i'M_i = 6\boldsymbol{I}_6 + 4\boldsymbol{J}_6$ in (4.1). Arrange all the 20 points in J_2 in such a way that $\mathbf{x}_1 = (1/3, 1/3, 1/3, 0, 0, 0)'$, and $\mathbf{x}_2, \ldots, \mathbf{x}_{20}$ are obtained by permutating the three $1/3$'s in $\mathbf{x}_1$ according to lexicographical order. Using *Mathematica* (Wolfram (1991)), (4.1) can be simplified, and a solution for the frequencies λ_j is found as follows:

$$\lambda_1 = \lambda_3 = \lambda_7 = \lambda_8 = \lambda_9 = \lambda_{11} = \lambda_{15} = \lambda_{16} = \lambda_{17} = \lambda_{19} = 2,$$
$$\lambda_2 = \lambda_4 = \lambda_5 = \lambda_6 = \lambda_{10} = \lambda_{12} = \lambda_{13} = \lambda_{14} = \lambda_{18} = \lambda_{20} = 0.$$

The weights of the corresponding A-optimal design derived using (3.8) and (3.9) are $(\sqrt{3}-1)/12$ for each of the six points in J_0, $(3-\sqrt{3})/20$ for each of the 10 points $\mathbf{x}_1, \mathbf{x}_3, \mathbf{x}_7, \mathbf{x}_8, \mathbf{x}_9, \mathbf{x}_{11}, \mathbf{x}_{15}, \mathbf{x}_{16}, \mathbf{x}_{17}, \mathbf{x}_{19}$ in J_2, and 0 for the remaining points in J_2. The total number of support points in this asymmetric weighted centroid design is 16, which is less than 26, the number of support points in the symmetric weighted centroid design $\boldsymbol{\xi}_{1,3}$.

When $q = 3, 4, 5$, equation (4.1) has the unique solution $\lambda_1 = \lambda_2 = \cdots = 1$. Hence the designs $\boldsymbol{\xi}_{1,2}$ and $\boldsymbol{\xi}_{1,3}$ are the unique A-optimal designs when $q = 3, 4$ and when $q = 5$, respectively.

5. Conclusion

In this paper we have obtained symmetric and asymmetric A-optimal designs for model $\eta_{DW2}(\mathbf{x})$ for $q \ge 3$. The numerical results for symmetric designs are summarized in Table 1. When $q=3$, J_3 does not exist, and r_4 in Table 1 has no meaning.

Table 1. A-optimal symmetric centroid designs for $\eta_{DW2}(\mathbf{x})$

q	$C(q,1)r_1$	$C(q,2)r_2$	$C(q,3)r_3$	$C(q,4)r_4$
3	0.3923	0.6077	0	–
4	0.4142	0.5858	0	0
5	0.3496	0	0.6504	0
⋮	⋮	⋮	⋮	⋮
21	0.4010	0	0.5990	0
22	0.3946	0	0.4687	0.1367
23	0.3881	0	0.3328	0.2791
24	0.3818	0	0.1974	0.4208
25	0.3769	0	0.0676	0.5565
26	0.3732	0	0	0.6268
⋮	⋮	⋮	⋮	⋮
$\to\infty$	0.3846	0	0	0.6154

As for D-optimal designs for model $\eta_{DW2}(\mathbf{x})$, the following results are proved by Zhang and Guan (1992):

$$
\begin{array}{llll}
q=3,4: & C(q,1)r_1=1/2, & C(q,2)r_2=1/2; & \\
q=5: & C(q,1)r_1=0.4984, & C(q,2)r_2=0.4506, & C(q,3)r_3=0.0510; \\
q=6: & C(q,1)r_1=0.4959, & C(q,2)r_2=0.2753, & C(q,3)r_3=0.2288; \\
q=7: & C(q,1)r_1=0.4977, & C(q,2)r_2=0.0877, & C(q,3)r_3=0.4146; \\
q\geq 8: & C(q,1)r_1=1/2, & C(q,3)r_3=1/2. &
\end{array}
$$

The comparison of A- and D-optimal designs for $\eta_{DW2}(\mathbf{x})$ shows that all points in J_0 are possible support points, but other possible support points shift gradually from J_1 to J_2 or J_3 as q increases. A similar behaviour can be observed for model $\eta_{q,2}(\mathbf{x})$ for A-optimal designs, but not for D-optimal designs. Kiefer (1961) proved that the weighted centroid design with $r_1=r_2=2/(q(q+1))$ and $r_3=\cdots=r_q=0$ is D-optimal for model $\eta_{q,2}(\mathbf{x})$ for all $q\geq 3$. Yu and Guan (1993) showed that an A-optimal design for $\eta_{q,2}(\mathbf{x})$ for $q\geq 4$ is the one with $r_1=(4q-3)^{1/2}/(q(4q-3)^{1/2}+2q(q-1)), r_2=4r_1/(4q-3)^{1/2}$ and $r_3=\cdots=r_q=0$, and for $q=3$ the numerical solution is $r_1=0.1418075, r_2=0.1872667$, and $r_3=0.0127745$. When $3\leq q\leq 11$, Yu and Guan's results and the numerical results given in Table 1 of Galil and Kiefer (1977) are in agreement.

In the mixture models discussed above, the expected response depends only on the relative amounts but not the actual amounts of the mixture components. It is difficult to interpret from these models the effect of each individual component on the expected response. For example, if different mixtures of nitrates and chlorides are to be compared as fertilizers, using these mixture models we cannot tell how much of the effect was from nitrates helping the plants to grow and how much was from chlorides damaging the plants. One possible remedy to overcome this weakness is to include the actual amounts of the mixture components in the models (cf. Piepel and Cornell (1985, 1987)).

Acknowledgement

The authors wish to express their sincere thanks to the referees and the Chair-Editor for their valuable suggestions in improving the presentation of this paper and for pointing out a number of typographical errors in the manuscript.

Appendix. Proofs of Theorems 1-4

If $i = 1,\ldots,q$ is a fixed integer and $\mathbf{x} \in S^{q-1}$, it follows from (3.1), (3.4), (3.6) that

$$d_2(\mathbf{x}, \boldsymbol{\xi}_{1,i}) = \sum_{k=1}^{q}\{ax_k^2 + 2bx_k^2(1-x_k) + 2cx_k(1-x_k) + dx_k^2(1-x_k)^2\} + e\Big\{\sum_{k=1}^{q} x_k(1-x_k)\Big\}^2, \tag{A.1}$$

where

$$a = \frac{1}{r_1^2} + \frac{i^2}{(i-1)^2 r_1^2},$$

$$b = \frac{-i}{(i-1)r_1^2} + \frac{-i^3}{(i-1)^3 r_1^2} + \frac{-i^5}{(i-1)^3 r_1 r_i C(q-2,i-1)},$$

$$c = \frac{i^4}{r_1 r_i (i-1)^2 (q-1) C(q-2,i-1)},$$

$$d = \frac{i^2}{(i-1)^2 r_1^2} + \Big(\frac{i^2}{(i-1)^2 r_1} + \frac{i^4}{(i-1)^2 r_i C(q-2,i-1)}\Big)^2,$$

$$e = \frac{-2i^5}{(i-1)^3 r_1 r_i (q-1) C(q-2,i-1)} + \frac{-2i^7(q-1) + i^6 q(i-1)}{(i-1)^3 r_i^2 (q-1)^2 (C(q-2,i-1))^2}.$$

Suppose that j is an integer, $1 \le j \le q$, and $\mathbf{x} \in J_{j-1}$. It follows from (A.1) that

$$d_2(\mathbf{x}, \boldsymbol{\xi}) = aj^{-1} + 2b(j-1)j^{-2} + 2c(j-1)j^{-1} + d(j-1)^2 j^{-3} + e(j-1)^2 j^{-2},$$

where a, b, c, d, e are as given above. Define

$$g_{1,i}(j) = j^3 \operatorname{tr} M^{-1}(\boldsymbol{\xi}_{1,i}) - aj^2 - 2b(j-1)j - 2c(j-1)j^2 - d(j-1)^2 - e(j-1)^2 j, \tag{A.2}$$

where $\mathrm{tr}M^{-1}(\boldsymbol{\xi}_{1,i})$ is given by (3.7), (3.8) and (3.9).

It is clear that (3.2) is equivalent to

$$g_{1,i}(j) \geq 0 (1 \leq j \leq q). \tag{A.3}$$

Proof of Theorem 1. Let $i = 2$. We need to prove (A.3) when $q = 3, 4$. From (3.9), we have

$$\alpha(q, 2) = 2(2q - 3)^{1/2}(5(q - 2))^{-1/2}. \tag{A.4}$$

From (3.7), (3.8) and (A.4), we find that $g_{1,2}(1) = g_{1,2}(2) = 0$ and $g_{1,2}(3) = Q_1(q) \times P_1(q)$, where

$$Q_1(q) = -2q^5(1 + \alpha(q, 2))/((2q - 3)(q - 2)),$$
$$P_1(q) = 5(2q - 3)(7q - 30)\alpha(q, 2) + (94q^2 - 481q + 490).$$

The algebraic calculations are lengthy and tedious, but can be carried out efficiently using softwares such as *Mathematica*. This also happens to the proofs of Theorems 2-3.

It is easy to verify that $P_1(q) < 0$ when $q = 3, 4$, and hence $g_{1,2}(3) > 0$ when $q = 3, 4$. As $g_{1,2}(j)$ defined by (A.2) is a cubic polynomial in j, and $g_{1,2}(0) = -d < 0, g_{1,2}(1) = g_{1,2}(2) = 0, g_{1,2}(3) > 0$, we have $g_{1,2}(j) > 0$ for all $j \geq 3$. Hence the design $\boldsymbol{\xi}_{1,2}$ with r_1 and r_2 defined by (3.8) and (3.9) ($i = 2$) is A-optimal when $q = 3, 4$, and only points in J_0, J_1 are possible support points.

When $q \geq 5$, we have $P_1(q) > 0$, $g_{1,2}(3) < 0$, and the design $\boldsymbol{\xi}_{1,2}$ is not A-optimal when $q \geq 5$.

Proof of Theorem 2. Let $i = 3$. We need to prove that (A.3) is satisfied when $5 \leq q \leq 21$. From (3.9) we have

$$\alpha(q, 3) = 3(3q - 5)^{1/2}(13(q - 3))^{-1/2}. \tag{A.5}$$

Using (3.7), (3.8) and (A.5) we find that $g_{1,3}(1) = g_{1,3}(3) = 0$, and

$$\begin{aligned} g_{1,3}(2) &= Q_2(q)(13\alpha(q, 3)P_2(q) + P_3(q)), \\ g_{1,3}(4) &= -3Q_2(q)(13\alpha(q, 3)P_4(q) + P_5(q)), \end{aligned} \tag{A.6}$$

where

$$Q_2(q) = q^2(1 + \alpha(q, 3))/(8(3q - 5)(q - 3)),$$
$$P_2(q) = 15q^2 - 112q + 147,$$
$$P_3(q) = 321q^2 - 1804q + 2037,$$
$$P_4(q) = 3q^2 - 140q + 231,$$
$$P_5(q) = 165q^2 - 2336q + 3201.$$

Since $P_2(q) > 0$ and $P_3(q) > 0$ when $q \geq 6$, $g_{1,3}(2) > 0$ when $q \geq 6$. When $q = 5$, from (A.6), (A.5), and the definitions of $P_2(q)$ and $P_3(q)$, we have $g_{1,3}(2) > 0$.

Let $P_6(q) = 13\alpha(q,3)P_4(q) + P_5(q)$. It is straightforward to prove that $P_4(q) < 0$ when $5 \leq q \leq 44$, $P_5(q) < 0$ when $5 \leq q \leq 12$, and $P_5(q) > 0$ when $q \geq 13$.

Hence when $5 \leq q \leq 12$ we have $P_6(q) < 0$ and $g_{1,3}(4) > 0$. When $13 \leq q \leq 21$ we have $19.945 > 3(13)^{1/2}(3 + 4/(13-3))^{1/2} \geq 3(13)^{1/2}(3 + 4/(q-3))^{1/2} = 13\alpha(q,3) \geq 3(13)^{1/2}(3 + 4/(21-3))^{1/2} > 19.41648$. Hence when $13 \leq q \leq 21$, we have $P_6(q) < 19.41648P_4(q) + P_5(q) = 223.149q^2 - 5054.31q + 7686.21 < 0$, since the zeros of the last quadratic polynomial are $q = 1.639\ldots$ and $q = 21.0003\ldots$ Thus $g_{1,3}(4) > 0$ when $13 \leq q \leq 21$. Therefore when $5 \leq q \leq 21$, we have $g_{1,3}(4) > 0$ and $g_{1,3}(2) > 0$. Since $g_{1,3}(j)$ is a cubic polynomial in j, and $g_{1,3}(0) = -d < 0, g_{1,3}(1) = 0, g_{1,3}(2) > 0, g_{1,3}(3) = 0, g_{1,3}(4) > 0$, we deduce that $g_{1,3}(j) > 0$ for all $j \geq 4$. Therefore the design $\boldsymbol{\xi}_{1,3}$ with r_1 and r_3 defined by (3.8) and (3.9) ($i = 3$) is A-optimal when $5 \leq q \leq 21$, and only points in J_0 and J_2 are possible support points.

When $22 \leq q \leq 44$, we have $P_6(q) > 19.41648P_4(q) + P_5(q) > 0$. When $q \geq 45$, both $P_4(q)$ and $P_5(q)$ are positive, and so is $P_6(q)$. Thus when $q \geq 22$ we have $g_{1,3}(4) < 0$, and the design $\boldsymbol{\xi}_{1,3}$ is not A-optimal when $q \geq 22$. When $q = 4$, from (A.6), (A.5), and the definitions of $P_2(q)$ and $P_3(q)$, we have $g_{1,3}(2) < 0$, and the design $\boldsymbol{\xi}_{1,3}$ is not A-optimal.

Proof of Theorem 3. Let $i = 4$. We need to prove that (A.3) is satisfied when $q \geq 26$. From (3.9) we have

$$\alpha(q,4) = (4/5)\Big((4q-7)/(q-4)\Big)^{1/2}. \tag{A.7}$$

From (3.7), (3.8), (A.2) and (A.7) we find that $g_{1,4}(1) = g_{1,4}(4) = 0$, and

$$\begin{aligned}
g_{1,4}(2) &= 2Q_3(q)(25\alpha(q,4)P_7(q) + P_8(q)),\\
g_{1,4}(3) &= Q_3(q)(25\alpha(q,4)P_9(q) + P_{10}(q)),\\
g_{1,4}(5) &= 2Q_3(q)(25\alpha(q,4)P_{11}(q) + P_{12}(q)),
\end{aligned}$$

where

$$\begin{aligned}
Q_3(q) &= 2q^2(1 + \alpha(q,4))/(81(4q-7)(q-4)),\\
P_7(q) &= 20q^2 - 247q + 380,\\
P_8(q) &= 1124q^2 - 7927q + 9980,\\
P_9(q) &= 4q^2 - 551q + 988,\\
P_{10}(q) &= 1348q^2 - 18215q + 24948,\\
P_{11}(q) &= 68q^2 + 665q - 1444,\\
P_{12}(q) &= 452q^2 + 22937q - 37924.
\end{aligned}$$

When $q \geq 26$, it is clear that $P_7(q) > 0, P_8(q) > 0, P_{11}(q) > 0, P_{12}(q) > 0$, and therefore $g_{1,4}(2) > 0, g_{1,4}(5) > 0$. Let $P_{13}(q) = 25\alpha(q,4)P_9(q) + P_{10}(q)$. When $q \geq 26$, we have $2.1 > (4+9/22)^{1/2} > ((4q-7)/(q-4))^{1/2} = (5/4)\alpha(q,4) > 2$, and when $4 < q \leq 25$ we have $((4q-7)/(q-4))^{1/2} \geq (4+9/21)^{1/2} > 2.1$. It is clear that $P_9(q) > 0$ when $q \geq 136$, and $P_9(q) < 0$ when $26 \leq q \leq 135$. Therefore when $q \geq 136$, we have $P_{13}(q) > 2\times 20P_9(q)+P_{10}(q) = 1508q^2-40255q+65468 > 0$. When $135 \geq q \geq 26$, we have $P_{13}(q) > 2.1\times 20P_9(q)+P_{10}(q) = 1516q^2-41357q+67444$, and the last inequality is reversed when $4 < q \leq 25$. The quadratic expression $1516q^2 - 41357q + 67444$ is positive when $q \geq 26$ and negative when $4 < q \leq 25$. Hence $P_{13}(q) > 0$ when $135 \geq q \geq 26$, and $P_{13}(q) < 0$ when $4 < q \leq 25$. Since $P_{13}(q) > 0$ when $q \geq 26$, we have $g_{1,4}(3) > 0$ when $q \geq 26$. Therefore when $q \geq 26$ we have $g_{1,4}(0) = -d < 0, g_{1,4}(1) = 0, g_{1,4}(2) > 0, g_{1,4}(3) > 0, g_{1,4}(4) = 0, g_{1,4}(5) > 0$. Since $g_{1,4}(j)$ is a cubic polynomial in j, we have $g_{1,4}(j) > 0$ for all $j \geq 5$. Hence the design $\boldsymbol{\xi}_{1,4}$ with r_1 and r_4 defined by (3.8) and (3.9) ($i = 4$) is A-optimal when $q \geq 26$, and only points in J_0 and J_3 are possible support points.

When $q \leq 25$, we have $P_{13}(q) < 0$ and $g_{1,4}(3) < 0$, and the design $\boldsymbol{\xi}_{1,4}$ is not A-optimal when $q \leq 25$.

Algebraic computations for $q = 22, 23, 24, 25$. We shall obtain the numerical values of r_1, r_3, r_4 which correspond to an A-optimal design. The model matrix and the moment matrix of $\boldsymbol{\xi}_{1,3,4}$ are given by

$$\begin{pmatrix} \boldsymbol{I}_q & \boldsymbol{0}_q \\ M_3/3 & 2M_3/9 \\ M_4/4 & 3M_4/16 \end{pmatrix}$$

and

$$M(\boldsymbol{\xi}_{1,3,4}) = \begin{pmatrix} r_1\boldsymbol{I}_q + \frac{1}{9}r_3M_3'M_3 + \frac{1}{16}r_4M_4'M_4 & \frac{2}{27}r_3M_3'M_3 + \frac{3}{64}r_4M_4'M_4 \\ \frac{2}{27}r_3M_3'M_3 + \frac{3}{64}r_4M_4'M_4 & \frac{4}{81}r_3M_3'M_3 + \frac{9}{256}r_4M_4'M_4 \end{pmatrix}, \tag{A.8}$$

respectively. Using (3.5), the matrix on the right hand side of (A.8) can be expressed in terms of $\boldsymbol{I}_q$ and $\boldsymbol{J}_q$. Calculation shows that

$$M^{-1}(\boldsymbol{\xi}_{1,3,4}) = \begin{pmatrix} A\boldsymbol{I}_q + B\boldsymbol{J}_q & C\boldsymbol{I}_q + D\boldsymbol{J}_q \\ C\boldsymbol{I}_q + D\boldsymbol{J}_q & E\boldsymbol{I}_q + F\boldsymbol{J}_q \end{pmatrix},$$

where A, B, C, D, E, F are algebraic functions of r_1, r_3, r_4.

An A-optimal design minimizes $\mathrm{tr}M^{-1}(\boldsymbol{\xi}_{1,3,4}) = q(A + B + E + F)$ under the constraints $C(q,1)r_1 + C(q,3)r_3 + C(q,4)r_4 = 1, r_i \geq 0 (i = 1,3,4)$. Using *Mathematica* (Wolfram (1991)) we found that the solutions for $q = 22, 23, 24, 25$, rounded off at the 4th decimal place, are as given in Table 1. By substituting values of r_1, r_3, r_4 in $M^{-1}(\boldsymbol{\xi}_{1,3,4})$, condition (3.2) is verified numerically up to the 14th decimal place.

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(Received September 1995; accepted June 1997)