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Optimal orthogonal designs in two blocks based on F-squares for Darroch and Waller's quadratic mixture model in four components

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Abstract

Orthogonal block designs for Scheffé's quadratic model in three and four components were considered by John (1984), Czitrom (1988, 1989, 1992), Draper et al. (1993), Chan and Sandhu (1999), and Ghosh and Liu (1999). Singh (2003) considered optimal orthogonal designs in two blocks for Darroch and Waller's quadratic mixture model in three and four components. Aggarwal et al. (2008) have studied mixture designs in orthogonal blocks using F-squares. In this paper, we have considered the case of equal volume fractions and have constructed optimal orthogonal designs in two blocks based on F-squares for Darroch and Waller's quadratic mixture model in four components. Conditions required for orthogonality are also given.

Key words: Mixture experiments; Process variables; Orthogonality; Darroch and Waller's Model; F-squares; D-optimality; A-optimality; E-optimality.

1 Introduction

In studies involving mixtures of ingredients, the response is a function of the proportions of the q components present in the mixture and is independent of the total amount of the mixture. The factor space is therefore a regular $(q-1)$ dimensional simplex S_{q-1}.

$$S_{q-1} = \left\{ x : (x_1, x_2, \ldots, x_q) \middle| \sum_{i=1}^{q} x_i = 1, x_i \geq 0 \right\} \tag{1.1}$$

Sometimes along with the mixture ingredients, an experiment may involve some variables known as process variables that are not physically linked to the mixture variables but affect the response nonetheless. For example, in cake baking formulations, oven temperature and the time of bake could serve as the two process variables.

Scheffé (1958) introduced models and designs for experiments with mixtures. Scheffé (1963) introduced the problem of mixture experiments involving process variables. Nigam (1970) obtained conditions for orthogonal blocking of blends for Scheffé's quadratic model and constructed designs satisfying these conditions. John (1984) simplified the conditions for orthogonal blocking of blends for Scheffé's quadratic model and constructed designs using latin squares. Czitrom (1988, 1989) and Draper et al. (1993) studied mixture designs for three and four components in orthogonal blocks for Scheffé's quadratic model. Prescott et al. (1993) considered these designs for five mixture components.

Lewis et al. (1994) gave general methods of constructing designs for $q \geq 3$ mixture components in two or more orthogonal blocks using latin squares with specific properties. Chan and Sandhu (1999) obtained A- and E-optimal orthogonal block designs for three component mixture experiments. Ghosh and Liu (1999) obtained A-optimal orthogonal block designs for Scheffé's quadratic model in four component mixtures. Aggarwal et al. (2002) obtained D-, A- and E-optimal orthogonal block designs for Becker's models in three and four components. Singh (2003) obtained optimal orthogonal designs in two blocks for Darroch and Waller's quadratic mixture model in three and four components. Optimal orthogonal block designs in two blocks for second degree K-model were studied by Aggarwal et al. (2004).

Aggarwal et al. (2008) studied orthogonal blocking of blends for Scheffé's quadratic model using F-squares in the case when some of the components assume equal volume fractions. They also presented a general method for obtaining mates that are required to construct orthogonal blocks using F-squares. Moreover, the D-, A- and E-optimalities of these designs for $q = 4$ are also given by Aggarwal et al. (2008).

In this paper, we give conditions for orthogonal blocking of blends for Darroch and Waller's quadratic model and construct D-, A- and E-optimal orthogonal block designs in four components for the classes of designs that satisfy the blocking conditions for Darroch and Waller's quadratic model. These classes of designs are based on F-squares and are considered in Aggarwal et al. (2008).

2 Blocking conditions

Darroch and Waller (1985) gave the following additive quadratic model for experiments with mixtures

$$E(y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q} \beta_{ii} x_i^2 \tag{2.1}$$

The model is additive in the mixture components, in the sense that it is a sum of separate functions of $x_1, x_2, \ldots, x_q$. When mixture components $x_1, x_2, \ldots, x_q$ vary but the sums $x_1 + x_2 + \ldots + x_s$ and $x_{s+1} + \ldots + x_q$ $(1 \leq s < q)$ are fixed, the total effect on the expected response is the sum of the effects of varying $x_1 + x_2 + \ldots + x_s$ and $x_{s+1} + \ldots + x_q$ separately. This model is suitable for the design of industrial products where mixture components have additive effects on the response function. An example where this model can be applied is discussed in Chan et al. (1998; p. 361) in which he has described the study and design of solder plate used in surface-mount technology in electronic manufacturing.

When m mixture blends (not necessarily all distinct) are arranged in two blocks B_1 and B_2 with m_1 and m_2 blends respectively and $m_1 + m_2 = m$, the model (2.1) with block effect γ is

$$Y_u = \sum_{i=1}^{q} \beta_i x_{iu} + \sum_{i=1}^{q} \beta_{ii} x_{iu}^2 + \gamma Z_u + e_u; \quad u = 1, 2, \ldots, m \tag{2.2}$$

Here $Z_u = -1$, for the blends in block B_1, and $Z_u = 1$, for the blends in block B_2 and e_u's are random errors which are independently distributed with mean 0 and same variance σ^2. The model (2.2) does not contain the product terms of x_i and Z_u, whereas in general the model with block effect may contain cross-product terms. Rewriting (2.2) in matrix notation, we have

$$E(y) = X\beta + Z\gamma \tag{2.3}$$

where X is the $m \times 2q$ matrix corresponding to the mixture part, β is the $2q \times 1$ column vector of unknown parameters, γ is the block effect parameter and Z is the $m \times 1$ column vector corresponding to the block variable Z. The condition $X'Z = 0$ ensures that the two blocks of mixture blends will be orthogonal in the sense that the block effects do not affect the estimates of the coefficients in the mixture model. The following conditions should be satisfied in order to achieve orthogonal blocking.

$$\sum_{u=1}^{m_w} x_{iu} = k_i, \sum_{u=1}^{m_w} x_{iu}^2 = k_{ii} \quad \forall\ w = 1, 2 \text{ and } i = 1, 2, 3, \ldots, q \qquad (2.4)$$

where k_i's and k_{ii}'s are constants.

We consider mixture experiments in which some of the components have equal volume fractions. Such a situation may arise when we are interested in studying the effect on the response involving two or more ingredients at the same level or there is some past data indicating that the difference between some of the volume fractions considered as distinct in the previous study is actually negligible. We construct designs using F-squares and achieve orthogonal blocking of blends by satisfying the conditions given in (2.4). The use of F-squares improves the precision of the least squares estimates of the model parameters. This also simplifies the calculations involved in obtaining optimal orthogonal designs which otherwise is rather difficult and time consuming. Equal volume fractions have previously been considered by John (1984) for the case $q = 5$. The specific design of Cornell (1990) involving six blocks each of $5 + n$ blends where n is the number of common runs added to make design nonsingular has been considered further by Prescott et al. (1993) with two pairs of components assuming the same volume fractions.

Draper et al. (1993) considered an industrial application in which four flours are mixed into doughs in various proportions and obtained a class of designs in which two equal volume fractions are zero. The class of design proposed by Draper et al. (1993) has also been considered by Ghosh and Liu (1999), Aggarwal et al. (2002), Singh (2003) and Aggarwal et al. (2004).

3 F-squares

F-squares have been considered by Finney (1945, 1946a, 1946b), Freeman (1966) and Addleman (1967). Hedayat and Seiden (1970) considered F-squares and orthogonal F-squares as a generalization of latin squares and orthogonal latin squares. Hedayat, Raghavarao and Seiden (1975) made further contributions to the theory of F-squares design. An F-square may be preferred to a latin square especially when the number of treatments is smaller than the order of the square and one would like to take advantage of the available experimental units in order to improve the precision of the estimates of at least some of the treatment effects.

Definition 3.1. Let $A = [a_{ij}]$ be an $n \times n$ matrix and let $\Sigma = (c_1, c_2, \ldots, c_m)$ be the ordered set of distinct elements of A. In addition, suppose that for each $k = 1, \ldots, m, c_k$ appears precisely λ_k times ($\lambda_k \geq 1$) in each row and in each column of A. Then, A will be called a frequency square or more concisely, an F-square of order n on Σ with frequency vector $(\lambda_1, \lambda_2, \ldots, \lambda_m)$ and is denoted by $\mathrm{F}(n; \lambda_1, \lambda_2, \ldots, \lambda_m)$. This notation is contracted by use of powers to denote successive equal values of λ's. Thus, $\mathrm{F}(n; \lambda^m)$ represents $\mathrm{F}(n; \lambda, \lambda, \ldots, \lambda)$ while $\mathrm{F}(n; \lambda_1^2, \lambda_3, \lambda_4^2, \lambda_6, \ldots, \lambda_m)$ represents $\mathrm{F}(n; \lambda_1, \lambda_1, \lambda_3, \lambda_4, \lambda_4, \ldots, \lambda_m)$. In particular, in an $\mathrm{F}(n; \lambda^m)$ square, m is determined uniquely by n and λ and hence such a square is represented simply by $\mathrm{F}(n; \lambda)$.

An $\mathrm{F}(n, 1)$ square is simply a Latin square of order n and thus exists for all n. Consequently, $\mathrm{F}(n; \lambda_1, \lambda_2, \ldots, \lambda_m)$ square on $\Sigma = (c_1, c_2, \ldots, c_m)$ exists if and only if $n = \lambda_1 + \lambda_2 + \ldots + \lambda_m$. It is quite natural to study F-squares by making substitutions on the symbols of Latin squares. This idea was exploited by Laywine (1989). For example, consider the following Latin square of side 4.

$$\begin{array}{cccc} a & b & c & d \\ b & a & d & c \\ c & d & a & b \\ d & c & b & a \end{array}$$

By substituting the symbol $d = a$ in the above Latin square, we obtain the following $\mathrm{F}(4; 2, 1, 1)$ defined on $\Sigma = (a, b, c)$. We denote this F-square as FSI(4).

FSI(4)

Square number 1

$$\begin{array}{cccc} a & b & c & a \\ b & a & a & c \\ c & a & a & b \\ a & c & b & a \end{array}$$

FSI(4) generates two distinct F-squares via permutation of the last three columns, viz;

Square number 2

$$\begin{array}{cccc} a & c & a & b \\ b & a & c & a \\ c & a & b & a \\ a & b & a & c \end{array}$$

Square number 3

$$\begin{array}{cccc} a & a & b & c \\ b & c & a & a \\ c & b & a & a \\ a & a & c & b \end{array}$$

We identify F-squares by simply writing down the first row. For example, we represent FSI(4) Square number 2 by writing the first row as $a\,c\,a\,b$.

We seek pairs of F-squares that are mates in order to estimate the effects of mixture components independent of block variables, i.e., to obtain orthogonally blocked mixture designs based on F-squares. Distinct F-squares of order q are mates if they have identical cross product sums. Aggarwal et al. (2008) have presented a general method for obtaining mates required for constructing orthogonal blocks using F-squares.

4 Orthogonally blocked four component mixture designs using F-squares

For four components, nine distinct runs are required to estimate all the parameters in (2.2). With a single block variable at two levels, $Z = -1$ and $Z = +1$, we take one block at $Z = -1$ and the other block at $Z = +1$. Aggarwal et al. (2008) suggested the class of designs given in (4.1), (4.2) and (4.3) that are based on F-squares with an added observation at centroid and a, b, c as numbers between 0 and 1 such that $2a + b + c = 1$. Here, without loss of generality, we have assumed that the two equal volume fractions assume the component proportion a. These designs have 18 runs in two blocks. Each block contains 9 runs representing the specific four component mixtures. Two quaternary blends are present in both the blocks viz; (c,a,a,b) and (b,a,a,c) for Design 1, (b,c,a,a) and (c,b,a,a) for Design 2 and (c,a,b,a) and (b,a,c,a) for Design 3. There is a single repeat within the two blocks for all the three designs, viz; (a,b,c,a) in B_1 and (a,c,b,a) in B_2 for Design 1; (a,a,b,c) in B_1 and (a,a,c,b) in B_2 for Design 2 and (a,c,a,b) in B$_1$ and (a,b,a,c) in B_2 for Design 3.

DESIGN 1:

$$B_1 = \begin{bmatrix} a & b & c & a \\ b & c & a & a \\ c & a & a & b \\ a & a & b & c \\ a & c & a & b \\ b & a & a & c \\ c & a & b & a \\ a & b & c & a \\ 1/4 & 1/4 & 1/4 & 1/4 \end{bmatrix} \quad \text{and} \quad B_2 = \begin{bmatrix} a & a & c & b \\ b & a & a & c \\ c & b & a & a \\ a & c & b & a \\ a & c & b & a \\ b & a & c & a \\ c & a & a & b \\ a & b & a & c \\ 1/4 & 1/4 & 1/4 & 1/4 \end{bmatrix} \tag{4.1}$$

DESIGN 2:

$$B_1 = \begin{bmatrix} a & b & c & a \\ b & c & a & a \\ c & a & a & b \\ a & a & b & c \\ a & a & b & c \\ b & a & c & a \\ c & b & a & a \\ a & c & a & b \\ 1/4 & 1/4 & 1/4 & 1/4 \end{bmatrix} \quad \text{and} \quad B_2 = \begin{bmatrix} a & a & c & b \\ b & a & a & c \\ c & b & a & a \\ a & c & b & a \\ a & b & a & c \\ b & c & a & a \\ c & a & b & a \\ a & a & c & b \\ 1/4 & 1/4 & 1/4 & 1/4 \end{bmatrix} \tag{4.2}$$

DESIGN 3:

$$B_1 = \begin{bmatrix} a & c & a & b \\ b & a & a & c \\ c & a & b & a \\ a & b & c & a \\ a & a & b & c \\ b & a & c & a \\ c & b & a & a \\ a & c & a & b \\ 1/4 & 1/4 & 1/4 & 1/4 \end{bmatrix} \quad \text{and} \quad B_2 = \begin{bmatrix} a & c & b & a \\ b & a & c & a \\ c & a & a & b \\ a & b & a & c \\ a & b & a & c \\ b & c & a & a \\ c & a & b & a \\ a & a & c & b \\ 1/4 & 1/4 & 1/4 & 1/4 \end{bmatrix} \tag{4.3}$$

This class of designs consists of 13 distinct quaternary blends. In this paper, we use this class of designs to obtain D-, A- and E-optimal orthogonal block designs for Darroch and Waller's quadratic model in four components. When an equal number of observations are made, the two blocks in all the three designs satisfy the orthogonality conditions in (2.4) and we have orthogonal design in two blocks. Moreover, since the blocks in Design 1, Design 2 and Design 3 are orthogonal, it is unnecessary to consider the block variable Z while optimizing the mixture design and we need to concentrate on matrix $X'X$ only, where X is the extended design matrix for model (2.1). The form of $X'X$ for Design 1, Design 2 and Design 3 are as given in (4.4), (4.5) and (4.6) respectively.

$$X'X = \left[\begin{array}{cccc|cccc} A & C & C & B & D & F & F & E \\ C & A & B & C & F & D & E & F \\ C & B & A & C & F & E & D & F \\ B & C & C & A & E & F & F & D \\ \hline D & F & F & E & G & I & I & H \\ F & D & E & F & I & G & H & I \\ F & E & D & F & I & H & G & I \\ E & F & F & D & H & I & I & G \end{array}\right] \tag{4.4}$$

$$X'X = \left[\begin{array}{cccc|cccc} A & B & C & C & D & E & F & F \\ B & A & C & C & E & D & F & F \\ C & C & A & B & F & F & D & E \\ C & C & B & A & F & F & E & D \\ \hline D & E & F & F & G & H & I & I \\ E & D & F & F & H & G & I & I \\ F & F & D & E & I & I & G & H \\ F & F & E & D & I & I & H & G \end{array}\right] \tag{4.5}$$

$$X'X = \left[\begin{array}{cccc|cccc} A & C & B & C & D & F & E & F \\ C & A & C & B & F & D & F & E \\ B & C & A & C & E & F & D & F \\ C & B & C & A & F & E & F & D \\ \hline D & F & E & F & G & I & H & I \\ F & D & F & E & I & G & I & H \\ E & F & D & F & H & I & G & I \\ F & E & F & D & I & H & I & G \end{array}\right] \tag{4.6}$$

where,

$$\begin{aligned} A &= 8a^2+4b^2+4c^2+1/8, \qquad B = 4a^2+4ab+4ac+4bc+1/8, \\ C &= 2a^2+6ab+6ac+2bc+1/8, \qquad D = 8a^3+4b^3+4c^3+1/32, \\ E &= 4a^3+2a^2b+2ab^2+2a^2c+2bc^2+2b^2c+2ac^2+1/32, \\ F &= 2a^3+3a^2b+3ab^2+3a^2c+b^2c+3ac^2+bc^2+1/32, \\ G &= 8a^4+4b^4+4c^4+1/128, \quad H = 4a^4+4a^2b^2+4a^2c^2+4b^2c^2+1/128, \\ I &= 2a^4+6a^2b^2+6a^2c^2+2b^2c^2+1/128 \end{aligned} \tag{4.7}$$

5 Optimal designs

In order to obtain D-, A- and E-optimal designs, we obtain the expressions for $|X'X|$, $T = \text{Trace}(X'X)^{-1}$ and the eigen values λ_i $(i = 1, 2, \ldots, 8)$ of $X'X$ which are as given in (5.1), (5.2) and (5.3), respectively.

$$|X'X| = 288(a-b)^6(a-c)^6(b-c)^6(-2a+8a^2-b+4b^2-c+4c^2)^2 \tag{5.1}$$

$$\begin{aligned} T &= \frac{1}{12}\Bigg(24+\frac{11}{(a-b)^2}+\frac{11(1+(a+b)^2)}{(a-b)^2(a-c)^2}+\frac{14+2(a+b)(3a+11b)}{(a-b)^3(a-c)} \\ &\quad +\frac{14(1+(a+b)^2)}{(a-b)^2(b-c)^2}-\frac{14(1+2a(a+b))}{(a-b)^3(b-c)} \end{aligned}$$

$$+\frac{51(1+2b+4a(1+4a+8b)+2c+16(2a+b)c)}{(2(1-4a)a+b-4b^2+c-4c^2)^2}$$
$$+\left.\frac{6(17+16a+8b+8c)}{2a(-1+4a)+b(-1+4b)+c(-1+4c)}\right) \quad (5.2)$$

and the eigen values λ_i $(i=1,2,\ldots,8)$ are

$$\begin{aligned}
\lambda_1 &= \frac{1}{64}\Bigg(17+128(4a^4+b^4+2bc+c^2+c^4+4a(b+c)+4a^2(1+b^2+c^2)+b^2(1+2c^2))\\
&\quad -\sqrt{\begin{matrix}(-512(2(1-4a)a+b-4b^2+c-4c^2)^2+(-17-128(4a^4\\ +b^4+2bc+c^2+c^4+4a(b+c)+4a^2(1+b^2+c^2)+b^2(1+2c^2)))^2)\end{matrix}}\Bigg)\\
\lambda_2 &= \frac{1}{64}\Bigg(17+128(4a^4+b^4+2bc+c^2+c^4+4a(b+c)+4a^2(1+b^2+c^2)+b^2(1+2c^2))\\
&\quad +\sqrt{\begin{matrix}(-512(2(1-4a)a+b-4b^2+c-4c^2)^2+(-17-128(4a^4\\ +b^4+2bc+c^2+c^4+4a(b+c)+4a^2(1+b^2+c^2)+b^2(1+2c^2)))^2)\end{matrix}}\Bigg)\\
\lambda_3 &= 2\Bigg(2a^4+b^2+b^4+c^2+c^4-2a(b+c)-2a^2(-1+b^2+c^2)\\
&\quad -\sqrt{\begin{matrix}(-4(a-b)^2(a-c)^2(b-c)^2+(2a^4+b^2+b^4\\ +c^2+c^4-2a(b+c)-2a^2(-1+b^2+c^2))^2)\end{matrix}}\Bigg)\\
\lambda_4 &= 2\Bigg(2a^4+b^2+b^4+c^2+c^4-2a(b+c)-2a^2(-1+b^2+c^2)\\
&\quad +\sqrt{\begin{matrix}(-4(a-b)^2(a-c)^2(b-c)^2+(2a^4+b^2+b^4\\ +c^2+c^4-2a(b+c)-2a^2(-1+b^2+c^2))^2)\end{matrix}}\Bigg)\\
\lambda_6 &= 2\Bigg(a^4+b^4-bc+c^2+c^4-a(b+c)-b^2(-1+c^2)-a^2(-1+b^2+c^2)\\
&\quad -\sqrt{\begin{matrix}(-3(a-b)^2(a-c)^2(b-c)^2+(a^4+b^4-bc+c^2+c^4-a(b+c)\\ -b^2(-1+c^2)-a^2(-1+b^2+c^2))^2)\end{matrix}}\Bigg)\\
\lambda_7 &= 2\Bigg(a^4+b^4-bc+c^2+c^4-a(b+c)-b^2(-1+c^2)-a^2(-1+b^2+c^2)\\
&\quad +\sqrt{\begin{matrix}(-3(a-b)^2(a-c)^2(b-c)^2+(a^4+b^4-bc+c^2+c^4-a(b+c)\\ -b^2(-1+c^2)-a^2(-1+b^2+c^2))^2)\end{matrix}}\Bigg)\\
\lambda_5 &= \lambda_6, \quad \lambda_8=\lambda_7
\end{aligned} \quad (5.3)$$

We consider the following particular cases for all the three designs viz; Design 1, Design 2 and Design 3:

Case 1: $\boldsymbol{a=0}$
Case 2: $\boldsymbol{b=0}$
Case 3: $\boldsymbol{c=0}$

5.1 Case 1: $a = 0$

The form of $X'X$ for Design 1, Design 2 and Design 3 in this case is as given in (4.4), (4.5) and (4.6) respectively, with

$$\begin{aligned}
A &= 4b^2+4c^2+1/8, \quad B=4bc+1/8, \quad C=2bc+1/8,\\
D &= 4b^3+4c^3+1/32, \quad E=2b^2c+2bc^2+1/32, \quad F=b^2c+bc^2+1/32,\\
G &= 4b^4+4c^4+1/128, \quad H=4b^2c^2+1/128, \quad I=2b^2c^2+1/128 \qquad (5.4)
\end{aligned}$$

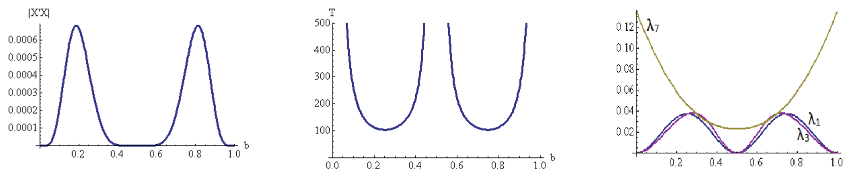
For all the three designs viz, Design 1, Design 2 and Design 3, we obtain the following results.

$$\begin{aligned}
|X'X| &= 288b^6c^6(b-c)^6(-b+4b^2-c+4c^2)^2 \qquad (5.5)\\
T &= \frac{1}{12}(24+\frac{11}{b^2}+\frac{14(1+b^2)}{b^2(b-c)^2}+\frac{14}{b^3(b-c)}+\frac{11(1+b^2)}{b^2c^2}+\frac{2(7+11b^2)}{b^3c}\\
&\quad +\frac{102+48b+48c}{b(-1+4b)+c(-1+4c)}+\frac{51(1+2c+b(2+16c))}{(b(-1+4b)+c(-1+4c))^2}) \qquad (5.6)
\end{aligned}$$

and the eigen values λ_i $(i=1,2,\ldots,8)$ as

$$\begin{aligned}
\lambda_1 &= 2(b^2+b^4+c^2+c^4-\sqrt{-4b^2(b-c)^2c^2+(b^2+b^4+c^2+c^4)^2})\\
\lambda_2 &= 2(b^2+b^4+c^2+c^4+\sqrt{-4b^2(b-c)^2c^2+(b^2+b^4+c^2+c^4)^2})\\
\lambda_4 &= 2(b^4-bc+c^2+c^4-b^2(-1+c^2)\\
&\quad -\sqrt{-3b^2(b-c)^2c^2+(b^4-bc+c^2+c^4-b^2(-1+c^2))^2}\\
\lambda_5 &= 2(b^4-bc+c^2+c^4-b^2(-1+c^2)\\
&\quad +\sqrt{-3b^2(b-c)^2c^2+(b^4-bc+c^2+c^4-b^2(-1+c^2))^2})\\
\lambda_7 &= \frac{17}{64}+2b^4+4bc+2c^2+2c^4+2b^2(1+2c^2)\\
&\quad -\frac{1}{64}\sqrt{\begin{array}{c}-512(-b+4b^2-c+4c^2)^2\\ +(-17-128(b^4+2bc+c^2+c^4+b^2(1+2c^2)))^2\end{array}}\\
\lambda_8 &= \frac{17}{64}+2b^4+4bc+2c^2+2c^4+2b^2(1+2c^2)\\
&\quad +\frac{1}{64}\sqrt{\begin{array}{c}-512(-b+4b^2-c+4c^2)^2\\ +(-17-128(b^4+2bc+c^2+c^4+b^2(1+2c^2)))^2\end{array}} \qquad (5.7)\\
\lambda_3 &= \lambda_4, \lambda_6=\lambda_5
\end{aligned}$$

Since the model (2.1) is symmetrical in x_1, x_2, x_3 and x_4, $|X'X|$, T and the eigen values λ_i $(i=1,2,\ldots,8)$ are symmetric functions of b and c. In order to find D-, A- and E-optimal designs, we need to find the values of b and c that maximize $|X'X|$, minimize T and maximize the minimum of the eigen values λ_i $(i=1,2,\ldots,8)$, respectively. If $\lambda_0 = \min(\lambda_i,\ i=1,2,\ldots,8)$, then from (5.7), we have $\lambda_0=\min(\lambda_1,\lambda_3,\lambda_7)$. Also, since $b+c=1$, on substituting $c=1-b$, we obtain $|X'X|$, T and λ_i $(i=1,2,\ldots,8)$ as functions of b alone. We have obtained different values of $|X'X|$, T, λ_1, λ_3, and λ_7 for $b \in [0, 1]$. Their graphs are shown in Figure 1.



(i) (ii) (iii)

Figure 1. Graphs of $|X'X|$, T and the eigen values λ_1, λ_3 and λ_7 against b for the particular case $a = 0$.

We observe both numerically and graphically that

1. $|X'X| = 0$ when $b = 0$, $\frac{1}{2}$ or 1.

2. The curve of $|X'X|$ is an m-shaped curve. Its maximum ($=$ 0.000680936) is attained when $b = 0.18667, 0.81333$.

3. T attains its minimum ($= 103.362$) when $b = 0.251687, 0.748313$.

4. $\lambda_0 = \min(\lambda_1, \lambda_3)$ attains its absolute maximum ($= 0.037507587$) at
   $b = 0.258570$, 0.74143.

We may note here that the form of $X'X$ for Design 3 as well as the form of $|X'X|$ for all the three designs viz, Design 1, Design 2 and Design 3 coincide with that obtained earlier by Singh (2003) for the class of designs that were earlier considered by Ghosh and Liu (1999). Hence the same D-, A-, and E-optimal values for Design 1, Design 2 and Design 3 are obtained as those obtained by Singh (2003). However, there are some practical differences between our designs and the design proposed by Ghosh and Liu (1999) that might make one design more suitable for use than the other. Ghosh and Liu's (1999) design is characterized by the presence of four binary blends in both the blocks and four distinct binary blends in each block. Our designs are characterized by the presence of two binary blends in both the blocks, a single repeat within the two blocks and four distinct blends in each block. The quaternary blend $(1/4, 1/4, 1/4, 1/4)$ is common to all the blocks irrespective of the design.

5.2 Case 2: $b = 0$ or Case 3: $c = 0$

Since Design 1, Design 2 and Design 3 are symmetric in b and c, we get similar results for the Case 2: $b = 0$ and Case 3: $c = 0$. We therefore consider the case $c = 0$.

Case 3: $c = 0$

The form of $X'X$ for Design 1, Design 2 and Design 3 in this case is as given in (4.4), (4.5) and (4.6) respectively, with the following modifications.

$$\begin{aligned} A &= 8a^2+4b^2+1/8, \quad B = 4a^2+4ab+1/8, \quad C = 2a^2+6ab+1/8, \\ D &= 8a^3+4b^3+1/32, \quad E = 4a^3+2a^2b+2ab^2+1/32, \\ F &= 2a^3+3a^2b+3ab^2+1/32, \quad G = 8a^4+4b^4+1/128, \\ H &= 4a^4+4a^2b^2+1/128, \quad I = 2a^4+6a^2b^2+1/128 \end{aligned} \tag{5.8}$$

For all the three designs viz, Design 1, Design 2 and Design 3, we obtain the following results.

$$|X'X| = 288a^6(a-b)^6b^6(-2a+8a^2-b+4b^2)^2 \tag{5.9}$$

$$\begin{aligned} T &= \frac{1}{12}\Big(24+\frac{11}{a^2}+\frac{11(1+a^2)}{a^2(a-b)^2}+\frac{2(7+3a^2)}{a^3(a-b)}+\frac{14(1+a^2)}{a^2b^2} \\ &+ \frac{14(1+2a^2)}{a^3b}+\frac{6(17+16a+8b)}{2a(-1+4a)+b(-1+4b)} \\ &+ \frac{51(1+2b+4a(1+4a+8b))}{(2a(-1+4a)+b(-1+4b))^2}\Big) \end{aligned} \tag{5.10}$$

and the eigen values λ_i $(i = 1, 2, \ldots, 8)$ as

$$\begin{aligned} \lambda_1 &= 8a^4+8ab+2b^2+2b^4+8a^2(1+b^2)+\frac{17}{64} \\ &-\frac{1}{64}\sqrt{\begin{array}{c}(-512(-2a+8a^2-b+8b^2)^2 \\ +(-17-512a^4-512ab-128b^2-128b^4-512a^2-512a^2b^2)^2)\end{array}} \\ \lambda_2 &= 8a^4+8ab+2b^2+2b^4+8a^2(1+b^2)+\frac{17}{64} \\ &+\frac{1}{64}\sqrt{\begin{array}{c}(-512(-2a+8a^2-b+8b^2)^2 \\ +(-17-512a^4-512ab-128b^2-128b^4-512a^2-512a^2b^2)^2)\end{array}} \\ \lambda_3 &= 4a^4-4ab+2b^2-2b^4+4a^2-4a^2b^2 \\ &-2\sqrt{-4a^2b^2(a-b)^2+(2a^4-2ab+b^2+b^4+2a^2-2a^2b^2)^2} \\ \lambda_4 &= 4a^4-4ab+2b^2-2b^4+4a^2-4a^2b^2 \\ &+2\sqrt{-4a^2b^2(a-b)^2+(2a^4-2ab+b^2+b^4+2a^2-2a^2b^2)^2} \\ \lambda_6 &= 2a^4-2ab+2b^2+2b^4+2a^2-2a^2b^2 \\ &-2\sqrt{-3a^2b^2(a-b)^2+(a^4-ab+b^2+b^4+a^2-a^2b^2)^2} \\ \lambda_7 &= 2a^4-2ab+2b^2+2b^4+2a^2-2a^2b^2 \\ &+2\sqrt{-3a^2b^2(a-b)^2+(a^4-ab+b^2+b^4+a^2-a^2b^2)^2} \quad (5.11) \\ \lambda_5 &= \lambda_6, \lambda_8 = \lambda_7 \end{aligned}$$

Again, to obtain D-, A- and E-optimal designs, we find the values of a and b that maximize $|X'X|$, minimize T and maximize the minimum of the eigen values λ_i $(i = 1, 2, \ldots, 8)$, respectively. If $\lambda_0 = \min(\lambda_i, i = 1, 2, \ldots, 8)$, then from (5.11), we have $\lambda_0 = \min(\lambda_1, \lambda_3, \lambda_5)$. Since $2a + b = 1$, on substituting $a = 0.5 - b/2$, we obtain $|X'X|$, T and λ_i $(i = 1, 2, \ldots, 8)$ as functions of b alone. We obtain different values of $|X'X|$, T, λ_1, λ_3 and λ_5 for $b \in [0, 1]$. Their graphs are shown in Figure 2 and clearly, these are not symmetrical in b.

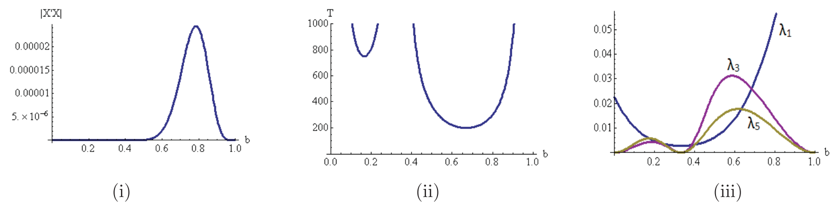


Figure 2. Graphs of $|X'X|$, T and the eigen values λ_1, λ_3 and λ_5 against b for the particular case $c = 0$.

We observe both numerically and graphically that

1. $|X'X| = 0$ when $0 \le b \le \frac{1}{2}$ or when $b = 1$.
2. The curve of $|X'X|$ is an inverted V-shaped curve. Its maximum $(= 0.0000243944)$ is attained when $b = 0.784893$.
3. T attains its minimum $(= 198.997)$ when $b = 0.669838$.
4. λ_0 attains its absolute maximum $(= 0.017699)$ at $b = 0.633436114$.

The following table depicts the values of parameters a, b and c for the Darroch and Waller's quadratic mixture model in four components for the particular cases $a = 0$ and $c = 0$.

Table 1. Numerical values of the design parameters for four component mixtures based on F-squares

Optimality Criteria	Particular case	
	$a = 0, c = 1 - b$	$c = 0, a = 0.5 - b/2$
	b	b
D $-$ optimality	$0.18667, 0.81333$	0.784893
A $-$ optimality	$0.251687, 0.748313$	0.669838
E $-$ optimality	$0.258570, 0.74143$	0.633436114

6 Conclusions

In this paper, we have constructed optimal orthogonal designs in two blocks based on F-squares for Darroch and Waller's quadratic mixture model in four components. From the results in Section 5.1 and Section 5.2, we find that for the case $a = 0$, Design 1, Design 2 and Design 3 are D-, A- and E-optimal when $b = 1 - c, c$ where $c = 0.18667, 0.251687, 0.258570$, respectively. Since for the case $a = 0$, the functions $|X'X|$, T, λ_1, λ_3, and λ_7 are symmetrical in b and c and hence optimality of the designs considered is maintained by the interchange of b and c. The same does not hold for the two equivalent cases $b = 0$ or $c = 0$. For the case $c = 0$, Design 1, Design 2 and Design 3 are D-, A- and E-optimal when $b = 0.784893, 0.669838$ and 0.633436114, respectively and $a = 0.5 - b/2$. Since Design 1, Design 2 and Design 3 are symmetrical in b and c, the results obtained for the case $b = 0$ are the same as that obtained for the case $c = 0$.

We observe that for the particular case $a = 0$, the D-, A- and E-optimal values for the designs Design 1, Design 2 and Design 3 coincide with the values obtained by Singh (2003) for the class of designs proposed by Ghosh and Liu (1999). However as discussed in Section 5.1, there are some practical differences between our designs and the design proposed by Ghosh and Liu (1999) that might make one design more suitable for use than the other.

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