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OPTIMAL AND NEARLY OPTIMAL ORTHOGONALLY BLOCKED DESIGNS FOR AN ADDITIVE QUADRATIC MIXTURE MODEL IN THREE COMPONENTS.

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ABSTRACT

Orthogonal block designs for Scheffé's quadratic model in three and four components were given 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 (1985) quadratic mixture model in three and four components. Prescott (1998) suggested nearly optimal orthogonally blocked designs for a quadratic mixture model in q components. Husain and Parveen (2016) obtained F- square based four component D-, A-, and E- optimal orthogonal block designs for an additive quadratic mixture model. In this paper, we have obtained Latin square based D-, A- and E-optimal and nearly optimal orthogonal designs in three components for the model presented by Husain and Parveen (2016).

Key Words - Mixture Experiments; Process variables; Orthogonality; Additive quadratic mixture model; D-optimality; A-optimality; E-optimality.

AMS 2010 subject classifications: 62K05, 62K10

1. INTRODUCTION

In mixture experiments, the measured response is assumed to depend on the proportions of the ingredients and not on the total amount of the mixture. Scheffé (1958) introduced models and designs for experiments with mixtures. In many practical situations, extraneous factors known as process variables are present. Scheffé (1963) discussed the problem of mixture experiments involving process variables. These variables do not form any physical portion of the mixture but their levels may affect the response(s) of interest. It becomes necessary to use blocking to deal with mixture experiments involving process variables. Orthogonal blocking facilitates estimation of the parameters of the mixture components independently of the estimation of the parameters of the process variables. In such type of experiments, the proportion of a mixture of q ($\geq$2) components may be expressed as a q-vector $x = (x_1, x_2, \ldots, x_q)$ in the (q-1) dimensional simplex S_{q-1}.

$$S_{q-1} = \{(x_1, x_2, \ldots, x_q) : \sum x_i = 1, x_i \geq 0 \ (i=1, 2, \ldots, q)\} \qquad (1.1)$$

Nigam (1970, 1976) obtained conditions for the orthogonal blocking of blends for Scheffé's quadratic model and constructed designs satisfying those conditions. John (1984) gave simple conditions for orthogonal blocking of blends for the Scheffé's quadratic model and presented designs based on Latin squares. Czitrom (1988, 1989, 1992) 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) studied mixture designs for five mixture components. Chan and Sandhu (1999) obtained A- and E-optimal orthogonal block designs for three component mixture experiments using the class of designs proposed by John (1984). Aggarwal *et al*. (2002) obtained D-, A- and E-optimal orthogonal block designs for Becker's (1968) model in three and four components. Singh (2003) obtained optimal orthogonal designs in two blocks for Darroch and Waller's (1985) quadratic mixture model in three and four components.

Optimal designs are obtained by selecting suitable pairs of Latin squares known as mates. Barring the centroid points, these designs consist of binary blends of mixture components and hence are not pure mixtures in the real sense. Practical implication may require at least minimum proportion of each ingredient to be physically present in the mixture. Prescott (1998) presented an interesting idea of nearly optimal block designs to meet this requirement. Prescott (1998) obtained three and four component nearly D-optimal orthogonal blocked designs without affecting the orthogonality of the designs. Nearly optimal designs are obtained by reparametrisation of each component. This method of shrinking the optimal design towards the centroid yields an alternative nearly optimal design comprising of pure mixtures.

In this paper, we have considered the class of design proposed by John (1984) to obtain D-, A- and E-optimal and nearly optimal orthogonal block designs for the mixture model presented by Husain and Parveen (2016).

2. BLOCKING CONDITIONS

Scheffé (1958) proposed the following quadratic model $y(x, z)$.

$$y(x, z) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i \leq j \leq q} \beta_{ij} x_i x_j + \varepsilon_u \qquad i, j = 1, 2, \ldots\ldots, q \quad i \neq j \qquad (2.1)$$

Husain and Parveen (2016) considered the following additive quadratic mixture model given in (2.2)

$$\eta\,(x, z) = \sum_{i=1}^{q}\beta_i x_i + \sum_{i=1}^{q}\sum_{j=1}^{q}\beta_{ij} x_i (x_i - x_j) + e_u \qquad i, j = 1, 2,..., q \quad i < j \tag{2.2}$$

The above model is additive in the mixture blends and is specifically useful in situations when the product of the components with the inter differences between various components affects the response of interest. These models are beneficial in the formulation of new drugs where the interactions between various drugs is to be studied. In particular, for three components model $\eta\,(x, z)$ reduces to

$$\eta\,(x, z) = \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{12} x_1 (x_1 - x_2) + \beta_{13} x_1 (x_1 - x_3) + \beta_{23} x_2 (x_2 - x_3) \tag{2.3}$$

Nigam's (1970, 1976) orthogonal blocking conditions were limited by some unnecessary restrictions which were removed by John (1983, 1984) who used Box and Hunter's (1957) orthogonality conditions and presented blocking conditions for Scheffé's quadratic model. Two or more blocks of blends are orthogonal if the least squares estimate of the blending coefficients of the fitted model are uncorrelated to the least squares estimate of the coefficients of terms involving the process variables. Husain and Parveen (2016) obtained the following orthogonality conditions for the model $\eta\,(x, z)$.

$$\sum_k x_{ik} = u_i \qquad \text{For each block;} \quad i = 1, 2, \ldots, q$$

$$\sum_k x_{ik} (x_{ik} - x_{jk}) = u_{ij} \qquad \text{For each block;} \quad i, j = 1, 2, \ldots, q, \quad i < j \tag{2.4}$$

where x_{ik} is the value of x_i for the k^{th} blend in a block and the u's are constants. The summations are extending over all the blends in a given block.

The following are the orthogonality conditions for three component mixtures.

3. REPARAMETRISATION OF THE COORDINATE SYSTEM

Prescott (1998) suggested raparametrisation of the coordinates in order to modify the optimal designs so that some or all of the runs used in the experiment include a minimum proportion of each mixture ingredient. For $q = 3$, the reparametrisation presented by Prescott (1998) for a point P (a, b, c) with $a \geq b \geq c$ takes the form of one shrinkage. The coordinates of O are (1/3, 1/3, 1/3) and the coordinates of Q are (f, 1-f, 0) which are obtained by extending the line OP to the edge of the simplex.

If $\dfrac{PO}{QP} = \dfrac{1-s}{s}$ i.e., P is situated at a proportion s along the line QO, then by simple geometry

$$a = (1-s)f + \frac{s}{3}$$

$$b = (1-s)(1-f) + \frac{s}{3}$$

$$c = \frac{s}{3} \tag{3.1}$$

Now the coordinates of point P can be shown in terms of *f* and *s*, where *f* identifies the point Q on the edge of the simplex and *s* is a shrinkage parameter which moves Q towards the centroid O.

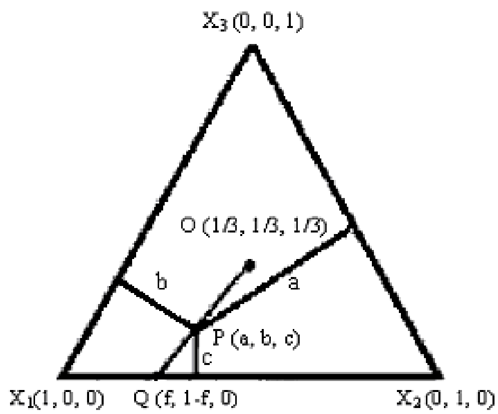

Fig. 3.1 Reparametrisation of P from (a, b, c) with $a \geq b \geq c$ to (f, s)

4. THREE COMPONENT MIXTURES

For three component mixtures seven distinct runs are required to estimate all the parameters in (2.3). A design with two blocks is required for a single process variable at two levels. If the process variable is represented by Z, then we may set Z = -1 in one block and Z = +1 in the other block. John (1984) proposed the class of design given in (4.1) for the Scheffé's quadratic model.

$$B_1 = \begin{bmatrix} a & b & c \\ b & c & a \\ c & a & b \\ 1/3 & 1/3 & 1/3 \end{bmatrix} \qquad B_2 = \begin{bmatrix} a & c & b \\ b & a & c \\ c & b & a \\ 1/3 & 1/3 & 1/3 \end{bmatrix} \tag{4.1}$$

The design consists of seven distinct runs arranged in two blocks B_1 and B_2. Here a, b and c are numbers between 0 and 1 and their sum is unity. These restrictions imply that (a, b, c) must lie on or inside a triangular simplex S_2. These two blocks are based on orthogonal Latin squares with an added observation at the centroid to remove singularity. Using the same class of design, Czitrom (1988) obtained D- optimal orthogonal block design and Chan and Sandhu (1999) obtained A- and E- optimal orthogonal block design for Scheffe's quadratic model in three components. Singh (2003) used this class of design to obtain D-, A- and E- optimal orthogonal block designs for Darroch and Waller's quadratic model. In this paper, we use this class of design to obtain D-, A- and E- optimal orthogonal block designs for the model (2.3).

For the model (2.3), the following blocking conditions are satisfied for the two blocks B_1 and B_2 given in (4.1).

$$u_1 = u_2 = u_3 = a + b + c + \frac{1}{3}$$

$$u_{12} = u_{13} = u_{23} = a^2 + b^2 + c^2 - ab - bc - ca \tag{4.2}$$

Hence the two blocks in (4.1) are orthogonal and we need to consider the matrix $\mathbf{X'X}$ only in order to derive the optimal designs. The matrix $\mathbf{X'X}$ for the additive quadratic model (2.3) is

$$\mathbf{X'X} = \begin{bmatrix} A & B & B & C & C & D \\ B & A & B & E & D & C \\ B & B & A & D & E & E \\ C & E & D & F & G & H \\ C & D & E & G & F & I \\ D & C & E & H & I & F \end{bmatrix} \tag{4.3}$$

where,

$$A = \frac{2}{9} + 2a^2 + 2b^2 + 2c^2$$

$$B = \frac{2}{9} + 2ab + 2ac + 2bc$$

$$C = a^2(a-b) + b^2(b-a) + a^2(a-c) + b^2(b-c) + c^2(c-a) + c^2(c-b)$$

$$D = ab(a-c) + ab(b-c) + a(a-b)c + b(b-a)c + bc(c-a) + ac(c-b)$$

$$E = a(a-b)b + ab(b-a) + a(a-c)c + b(b-c)c + ac(c-a) + bc(c-b)$$

$$F = a^2(a-b)^2 + b^2(b-a)^2 + a^2(a-c)^2 + b^2(b-c)^2 + c^2(c-a)^2 + c^2(c-b)^2$$

$$G = 2a^2(a-b)(a-c) + 2b^2(b-a)(b-c) + 2c^2(c-a)(c-b)$$
$$H = ab(b-a)(a-c) + a(a-b)b(b-c) + a(a-b)c(c-a) + b(b-c)c(-a+c) + b(b-a)c(c-b)$$
$$a(a-c)c(c-b)$$
$$I = 2ab(a-c)(b-c) + 2b(b-a)c(c-a) + 2a(a-b)c(c-b) \tag{4.4}$$

4.1. THREE COMPONENT OPTIMAL DESIGNS

In order to obtain D-, A- and E-optimality for model (2.3), we need to find the values of a, b and c that maximize $|\mathbf{X'X}|$, minimize T= trace $(\mathbf{X'X})^{-1}$ and maximize the minimum of the eigenvalues of $\mathbf{X'X}$, respectively. The expressions for $|\mathbf{X'X}|$ and T= trace $(\mathbf{X'X})^{-1}$ are given in (4.5) and (4.6), respectively. However, the expressions for the eigenvalues are very lengthy and hence not presented here.

$$|\mathbf{X'X}| = 48(a-b)^4(a-c)^4(b-c)^4(a^2 - ab + b^2 - ac - bc + c^2)^2 \tag{4.5}$$

$T = T_1/T_2$

$T_1=$

$$(a^8(8+81(b-c)^2) - 2a^7(10+81(b-c)^2)(b+c) + 8c^6(4+c^2) - 4bc^5(24+5c^2) + b^8(8+81c^2) - 2b^7c$$
$$(10+81c^2) - 8b^5c(12+10c^2+81c^4) - 2b^3c^3(139+40c^2+81c^4) + b^2c^4(219+146c^2+81c^4) + b^4c^2$$
$$(219-76c^2+405c^4) + b^6(32+146c^2+405c^4) + a^6(32+405b^4-162b^3c+146c^2+405c^4+b^2(146-486c^2)$$
$$-2bc(94+81c^2)) - 2a^5(b+c)(48+324b^4-405b^3c+40c^2+324c^4+2b^2(20+81c^2)-bc(22+405c^2))$$
$$-2a^3(b+c)(81b^6+b^4(40-243c^2)-2bc(59+58c^2)+4b^3c(-29+81c^2)+b^2(139+294c^2-243c^4)+c^2$$
$$(139+40c^2+81c^4)) + 2a(-81b^8c-2c^5(24+5c^2)+3b^5(2+3c^2)(-8+9c^2)+b^7(-10+81c^2)-b^6c(94+81c^2)$$
$$+bc^4(21-94c^2-81c^4)+b^3c^2(-21+76c^2-81c^4)+3b^2c^3(-7-6c^2+27c^4)+b^4c(21+76c^2+81c^4))$$
$$+a^4(405b^6+162b^5c+2b^3c(76-81c^2)-2b^4(38+243c^2)+3b^2(73+96c^2-162c^4)+2bc(21+76c^2+81c^4)$$
$$+c^2(219-76c^2+405c^4)) + a^2(81b^8+162b^7c+b^6(146-486c^2)+18b^5c(-2+27c^2)+3b^4(73+96c^2-162c^4)$$
$$+18b^2c^2(7+16c^2-27c^4)+6bc^3(-7-6c^2+27c^4)+c^4(219+146c^2+81c^4)+b^3(-42c-356c^3+486c^5)))$$

$$T_2 = (36(a-b)^2(a-c)^2(b-c)^2(a^2+b^2-bc+c^2-a(b+c))^2 \tag{4.6}$$

We observe that $|\mathbf{X'X}| = 0$ when $a = b$ or $b = c$ or $c = a$. Moreover $|\mathbf{X'X}|$, T and eigenvalues are symmetric functions of a, b and c. Hence, we obtain the same results for $a = 0$, $b = 0$ and $c = 0$. Here we consider the case $c = 0$. So we need to find the values of a and b that maximize $|\mathbf{X'X}|$, minimize T and maximize the minimum of the eigenvalues λ_i (i= 1, 2, ..., 6). Also since a+b=1, on substituting b=1 - a we obtain $|\mathbf{X'X}|$, T and the eigenvalues λ_i (i= 1, 2, ..., 6) as functions of a alone. We have obtained different values of $|\mathbf{X'X}|$, T and λ_1 for $a \in [0, 1]$. Their graphs are shown in Figure 4.1. The design matrix $\mathbf{X'X}$ for $c = 0$ is as given in (4.3), where now

$$A = \frac{2}{9} + 2a^2 + 2b^2$$

$$B = \frac{2}{9} + 2ab$$

$$C = a^3 + a^2(a-b) + b^3 + b^2(b-a)$$

$$D = a^2b + ab^2$$

$$E = a(a-b)b + ab(b-a)$$

$$F = a^4 + a^2(a-b)^2 + b^4 + b^2(b-a)^2$$

$$G = 2a^3(a-b) + 2b^3(b-a)$$

$$H = a(a-b)b^2 + a^2b(b-a)$$

$$I = 2a^2b^2 \tag{4.7}$$

We have obtained the expressions for $|\mathbf{X'X}|$, T and λ_i (i= 1, 2,..., 6). The expressions for the eigenvalues are very lengthy and hence not discussed here. The expressions for $|\mathbf{X'X}|$ and T are given in (4.8) and (4.9), respectively.

$$|\mathbf{X'X}| = 48a^4(a-b)^4b^4(a^2-ab+b^2)^2 \tag{4.8}$$

$$T = \frac{1}{36}\left(81 + \frac{8}{a^2} + \frac{32(1+a^2)}{a^2(a-b)^2} + \frac{32}{a^3(a-b)} + \frac{8(4+a^2)}{a^2b^2} + \frac{4(8+3a^2)}{a^3b} + \frac{27(1-9a^4+10ab+9a^3b)}{(a^2-ab+b^2)^2} + \frac{9(10+9a(3a+2b))}{a^2-ab+b^2}\right) \tag{4.9}$$

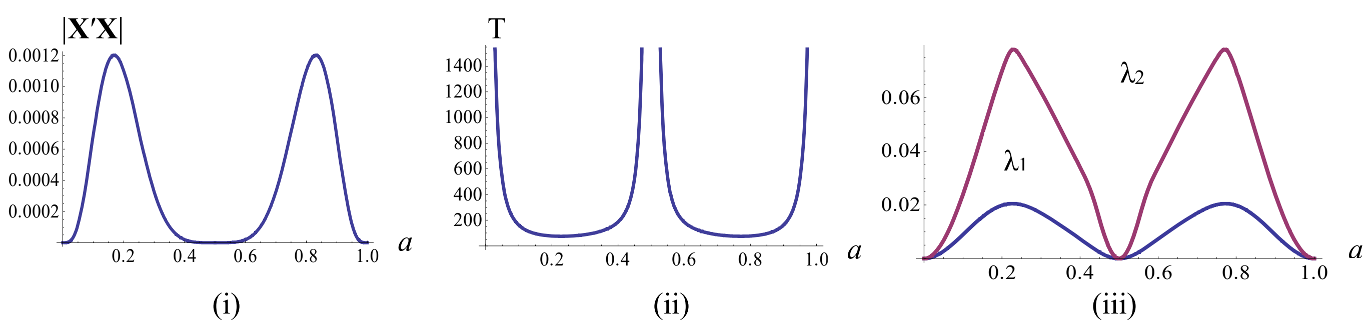

Figure.4.1 Graphs of $|\mathbf{X'X}|$, T and the eigenvalues λ_1 and λ_2 against a.

We observe both numerically and graphically that

1. $|\mathbf{X'X}| = 0$ when $a = 0$, 0.5 and 1.
2. The curve of $|\mathbf{X'X}|$ is an m- shaped curve. Its maximum (= 0.00120092) is attained when $a = 0.168497, 0.831503$.
3. T attains its minimum (= 74.7588) when $a = 0.228141, 0.771859$
4. We observe that $\lambda_2 > \lambda_1$ for $a \in [0, 1]$. Therefore $\lambda_0 = \lambda_1$, where λ_1 is an m- shaped curve with $\lambda_0 = 0$ when $a = 0$, 0.5 and 1. Thus λ_0 attains its absolute maximum (=0.0204984) when $a = 0.2273$ and $a = 0.7727$.

The D-, A- and E-optimalities obtained on all the boundary points $a = 0$, $b = 0$ and $c = 0$ are the same. Table 1 presents the numerical values of the design parameters for the three component mixtures for Scheffé's (1958) quadratic mixture model, Darroch and Waller's (1985) quadratic model and the additive quadratic mixture model (2.2).

Table 1: The numerical values of the design parameters for three component mixtures.

Optimality criteria	Scheffe's quadratic model			Darroch and Waller's quadratic model			Additive quadratic mixture model		
	a	b	c	a	b	c	a	b	c
D-optimality	0.16850	0.83150	0	0.16850	0.83150	0	0.1685	0.83150	0
A-optimality	0.18333	0.81667	0	0.2522	0.7478	0	0.228141	0.771859	0
E-optimality	0.15457	0.84543	0	0.2794	0.7206	0	0.2273	0.7727	0

5. THREE COMPONENT NEARLY OPTIMAL ORTHOGONALLY BLOCKED DESIGNS

In this section, we obtain nearly optimal designs based on Latin squares for the additive quadratic mixture model (2.3). The design shown in (4.1) was proposed by John (1984) and used by Czitrom (1988) for D- optimality and Chan and Sandhu (1999) for A- and E-optimality. We now shrink John's (1984) design towards the centroid by using the transformation (3.1) as suggested by Prescott (1998).

For the model (2.2), the same form of $|\mathbf{X'X}|$ as the one given in (4.3) is obtained. The general form of $|\mathbf{X'X}|$ is maximized at the point $b = a$, $1 - a$ for which $|\mathbf{X'X}|$ transforms to the following:

$$|\mathbf{X'X}|_0 = 48(1-a)^4 a^4 (-1+2a)^4 ((1-a)^2 - (1-a)a + a^2)^2 \qquad (5.1)$$

This takes its maximum value of 0.00120092 at $a = 0.168497, 0.831503$. With the reparametrisation of the coordinates (a, b, c) for the points in this design, the form of the general determinant in terms of f and s is a simple reduction in scale towards the centroid by a factor s and is given in (5.2).

$$|\mathbf{X'X}| = 48(1-f)^4 f^4 (-1+2f)^4 ((1-f)^2 - (1-f)f + f^2)^2 (1-s)^{16} \qquad (5.2)$$

$|\mathbf{X'X}|$ is a strictly decreasing function of s as $s \to 1$ and for any fixed value of s, $|\mathbf{X'X}|$ is maximised for $f = 0.168497, 0.831503$. Thus a nearly optimal design is obtained by shrinking the optimal design towards the centroid. The D- criterion is D = $|\mathbf{X'X}|^{1/p}$, where p is the number of parameters. Efficiency of the nearly optimal design, in terms of D- criterion is

$$\text{D-Efficiency} = |\mathbf{X'X}|^{1/p} / |\mathbf{X'X}|_0^{1/p} \times 100 \text{ percent} \qquad (5.3)$$

This reduces to a simple form $(1 - s)^{16/7} \times 100$, which is shown below for different values of s. Table 2 presents the efficiency of the nearly D-optimal design for the additive quadratic mixture model (2.3). Note that the D-efficiency of three component mixture design obtained for the additive model (2.3) is the same as that obtained by Prescott (1988) for Scheffé's quadratic model.

Table 2: Efficiency of the nearly D-optimal design for the Additive quadratic mixture model (2.3)

s	D-Efficiency
0.05	88.9
0.10	78.6
0.15	69.0
0.20	60.0
0.25	52.8

Table 3 presents the nearly D-optimal orthogonal block design with $f = 0.1685$ and $s = 0.05$ for the additive quadratic mixture model (2.3).

Table 3: Nearly D- optimal orthogonal block design with $f = 0.1685$ and $s = 0.05$ for the Additive quadratic mixture model (2.3).

B_1			B_2		
0.1767	0.8065	0.0167	0.1767	0.0167	0.8065
0.8065	0.0167	0.1767	0.8065	0.1767	0.0167
0.0167	0.1767	0.8065	0.0167	0.8065	0.1767
0.3333	0.3333	0.3333	0.3333	0.3333	0.3333

Every blend used in this shrunken nearly optimal design uses a proportion of all the three ingredients and so are true mixtures.

Equation (4.5) represents the general form of T = trace $(\mathbf{X'X})^{-1}$ for the design shown in (4.1). Minimum occurs at a = 0.228141, $b = 1 - a$ and c = 0 for which the form of T= trace $(\mathbf{X'X})^{-1}$ is

$$T=\frac{1}{36}\left[\begin{array}{l}81+\frac{8}{a^2}+\frac{32}{a^3(-1+2a)}+\frac{32(1+a^2)}{a^2(-1+2a)^2}+\frac{8(4+a^2)}{(1-a)^2a^2}+\frac{4(8+3a^2)}{(1-a)a^3}+\frac{27(1+10(1-a)a+9(1-a)a^3-9a^4)}{((1-a)^2-(1-a)+a^2)^2}\\+\frac{9(10+9a(2(1-a)+3a))}{(1-a)^2-(1-a)a+a^2}\end{array}\right] \quad (5.4)$$

With the reparametrisation (3.1) of the of the coordinates (a, b, c) for the points in this design, the general form of the T = trace $(\mathbf{X'X})^{-1}$ in terms of f and s is

$$T=\frac{1}{54}\left[\begin{array}{l}81+\frac{8(7+9(-1+f)f(7+3(-1+f)f(8+11(-1+f)f)))}{(1-2f)^2(-1+f)^2f^2(1+3(-1+f)f)^2(-1+s)^4}+\frac{2(2+9(-1+f)f)}{f^2(1-3f+2f^2)^2(-1+s)^3}+\\\frac{8(1+3(-1+f)f)^2}{f^2(1-3f+2f^2)^2(-1+s)^2}\end{array}\right] \quad (5.5)$$

We get T as a function of f alone by putting different values of s. We obtain the min T at s = 0 and f = 0.228141. Chan and Guan (2001) gave the following formula for obtaining efficiency of the A- Optimal designs.

A-efficiency = T_0/ Min (T) ×100. (5.6)

where T_0 is the minimum T obtained by substituting optimal f in original T. From Table 4 we observe that with a little loss in A-efficiency we obtain a true mixture which contains some proportions of all the ingredients.

Table 4: Efficiency of the Nearly A-optimal design against the shrinkage parameter s for the Additive quadratic mixture model (2.1).

s	Opt f	Min(T)	T_0	A-efficiency
0	0.228141, 0.771859	74.7588	74.7588	100
0.05	0.227918, 0.772082	91.1149	74.7589	82.04
0.1	0.227713, 0.772287	112.372	74.7593	66.53
0.2	0.227361, 0.772639	178.009	74.7603	41.99

Table 5: Nearly A-optimal orthogonal block design with f = 0.227918 and s = 0.05 for the Additive quadratic mixture model (2.3)

B_1			B_2		
0.233188	0.750144	0.016667	0.233188	0.016667	0.750144
0.750144	0.016667	0.233188	0.750144	0.233188	0.016667
0.016667	0.233188	0.750144	0.016667	0.750144	0.233188
0.33333	0.33333	0.33333	0.33333	0.33333	0.33333

The efficiency of E-optimal design is obtained by the following formula.

E-efficiency = Abs {Max (λ_0)}/ Abs {Max (λ_0)}$_0$× 100 (5.7)

We have λ_0 = min (λ_1, λ_2), λ_0 attains its absolute maximum (= 0.0204984) at c = 0, b = 1-a, a, where a = 0.2272.. By employing transformation (3.1) we get the eigenvalues in terms of f and s. The expressions for the eigenvalues are very lengthy and hence not discussed here.

Table 6: Efficiency of the nearly E-optimal design against the shrinkage parameter s for the Additive quadratic mixture model (2.3)

s	Opt f	Abs Max (λ_0)	Abs{Max (λ_0)}$_0$	E- efficiency
0	0.22728, 0.772715	0.0204984	0.0204984	100
0.05	0.22763, 0.772368	0.0166607	0.0204983	81.27
0.1	0.22797, 0.77203	0.0133896	0.0204980	65.32
0.2	0.22866, 0.771344	0.0083165	0.0204968	40.57

Table 7: Nearly E-optimal orthogonal block design with f = 0.22763 and s = 0.05 for the Additive quadratic mixture model (2.3)

B_1			B_2		
0.23291612	0.75041722	0.01666667	0.23291612	0.01666667	0.75041712
0.75041722	0.01666667	0.23291612	0.75041722	0.23291612	0.01666667
0.01666667	0.23291612	0.75041722	0.01666667	0.75041722	0.23291612
0.33333	0.33333	0.33333	0.33333	0.33333	0.33333

We see from tables 6 and 7, that when f = 0.22763 and s = 0.05, then with a little loss in E-efficiency, we get true mixtures which contain some proportion of all the ingredients.

5.1. DESIGNS USING TWO PAIRS OF SQUARE FOR q = 3

Table 7: Orthogonal block design with two squares for q = 3

Run	x_1	x_2	x_3	Run	x_1	x_2	x_3
1	a	b	c	8	a	c	b
2	b	c	a	9	b	a	c
3	c	a	b	10	c	b	a
4	a'	c'	b'	11	a'	b'	c'
5	b'	a'	c'	12	b'	c'	a'
6	c'	b'	a'	13	c'	a'	b'
7	1/3	1/3	1/3	14	1/3	1/3	1/3

Prescott (1998) suggested that by adding extra Latin squares to each block we may obtain more flexible designs while maintaining the orthogonality. For the design shown in Table 7, the values in the second square need not be the same as those in the first square for the orthogonality conditions to be satisfied. Now, we consider the case when they are same.

5.1.1 Design formed by shrinking both the pairs of Latin squares

Consider the case when both the pairs of Latin squares in Table 7 have same values i.e. $a' = a$, $b' = b$, $c' = c$ and as a result we obtain a symmetric design. We shrink both pairs of Latin squares towards the centroid of the design. By reparametrisation of the coordinate system as done in section (5), nearly optimal designs are constructed. The form of the general determinant obtained by shrinking both pairs of Latin squares is

$$|\mathbf{X}'\mathbf{X}| = 1536(a-b)^4(a-c)^4(b-c)^4(a^2-ab+b^2-ac-bc+c^2)^2 \tag{5.8}$$

This form of $|\mathbf{X}'\mathbf{X}|$ is 36 times the corresponding determinant obtained for the one square design given in (4.1). D-optimal designs provides maximum of $|\mathbf{X}'\mathbf{X}|$ for the additive model (2.3) on the boundary of the simplex at $a = f = 0.831503$, $b = 1\text{-}f = 0.168497$ and $c = 0$. These designs have the same efficiencies relative to the D- optimal design as the one square design shown in (4.1).On shrinking both the pairs of Latin squares towards the centroid, we obtain the minimum value of T (44.4981) for $s = 0$ at $a = f = 0.212427$, $b = 1\text{-}f = 0.787573$, $c = 0$. The efficiencies of the nearly A-optimal design by shrinking both the Latin squares are given in Table 9.

Table 9: Properties of the nearly A-optimal design with shrinkage parameter s applied to design 5.2.1

s	Opt f	Min T	T_0	A-efficiency
0	0.212427, 0.787573	44.4981	44.4981	100
0.05	0.212167, 0.787833	54.1133	44.982	82.2
0.1	0.211926, 0.788074	66.6112	44.4985	66.80
0.15	0.211705, 0.788295	83.0982	44.4989	53.5
0.2	0.211504, 0.788496	105.211	44.4994	42.29

We have obtained nearly E-optimality (= 0.0354362) at $a = f = 0.206354$, $b = 1\text{-}f = 0.793655$, $c = 0$. Again by reparametrisation, we get the general form of the minimum eigenvalue i.e., λ_1 in terms of f and s. Table 10 provides the maximum of the minimum eigenvalues for some specific values of s and the respective efficiencies of the nearly E-optimal designs.

Table 10: Properties of the nearly E-optimal design with shrinkage parameter s applied to design 5.2.1

s	Opt f	Abs Max{ λ_0}	Abs{Max{ λ_0}}$_0$	E-efficiency
0	0.206354, 0.793655	0.0354362	0.0354362	100
0.05	0.206931, 0.793069	0.0288434	0.0354357	81.3
0.1	0.207465, 0.792529	0.0232069	0.0354341	65.49
0.15	0.207964, 0.792036	0.0184345	0.0354318	52.02
0.2	0.208434, 0.791565	0.0144364	0.0354289	40.74

5.1.2. Design formed by shrinking one pair of Latin squares

Prescott (1998) constructed nearly D-optimal design for Scheffe's quadratic model by shrinking only one Latin square in each block of design as shown in Table 7. We use it to construct nearly D-, A- and E-optimal designs for the additive quadratic mixture model (2.3). When only one Latin square is shrunk towards the centroid of the design, other Latin squares are left on the edge of the simplex. As a result, the design consists of 13 distinct blends. It contains some binary blends and some three ingredient blends covering the simplex region more uniformly.

The determinant of $\mathbf{X'X}$ for the design (5.1.2) is very lengthy and hence not discussed here. Table 11 shows the optimum f, $D = |\mathbf{X'X}|^{1/7}$, $D_0 = |\mathbf{X'X}|_0{}^{1/7}$, and the efficiency of the nearly D-optimum designs for selected values of the shrinkage parameter s.

Table 11: Properties of the nearly D-optimal design with shrinkage parameter s applied to design 5.1.2.

s	Opt f	$\lvert\mathbf{X'X}\rvert$	$D = \lvert\mathbf{X'X}\rvert^{1/7}$	$D_0 = \lvert\mathbf{X'X}\rvert_0{}^{1/7}$	D-efficiency
0	0.168497, 0.831503	0.0384296	0.627783	0.627783	100
0.05	0.168173, 0.831825	0.0264574	0.595182	0.627782	94.80
0.1	0.167142, 0.832855	0.0192842	0.568891	0.627767	90.62
0.15	0.165323, 0.834677	0.0149066	0.548345	0.627698	87.35
0.2	0.162654, 0.837344	0.0121845	0.053277	0.627494	84.90

We have obtained nearly A-optimal design by shrinking only one Latin square towards the centroid. On employing the transformation (3.1), we get a very lengthy expression for the general form of T in terms of f and s. T is minimised for $f = 0.212427, 0.787573$ at $s = 0$. Table 12 provides the properties of nearly A-optimal design against the shrinkage parameter s.

Table 12: Properties of the nearly A-optimal design with shrinkage parameter s applied to design 5.1.2.

s	Opt f	Min T	T_0	A-efficiency
0	0.212427, 0.787573	44.4981	44.4981	100
0.05	0.212647, 0.787352	48.5342	44.4982	91.68
0.10	0.213579, 0.786421	52.0448	44.5002	85.50
0.15	0.215118, 0.784882	54.8983	44.5093	81.07
0.20	0.217048, 0.782952	57.0927	44.5311	77.97

For the design 5.1.2, we have obtained nearly E-optimal design against the shrinkage parameter s for the additive quadratic mixture model (2.3). Table 13 provides the properties of the nearly E-optimal designs with shrinkage parameter s.

Table 13: Properties of the nearly E-optimal design with shrinkage parameter *s* applied to design 5.1.2.

s	Opt *f*	Abs Max{λ_0}	Abs{Max{λ_0}}$_0$	D-efficiency
0	0.206354, 0.793646	0.0354362	0.0354362	100
0.05	0.207177, 0.792828	0.0323175	0.0354351	91.20
0.10	0.209033, 0.790973	0.0299707	0.0354241	84.60
0.15	0.211743, 0.788256	0.0282405	0.0353876	79.80
0.20	0.214945, 0.785057	0.0269693	0.035314	76.36

6. CONCLUSIONS

In this paper, we have obtained the D-, A- and E-optimal and nearly optimal orthogonally blocked designs for Husain and Parveen's (2016) additive quadratic mixture model in three components for Latin square based designs. Three component D-, A- and E-optimal designs are obtained at a = 0.168497, 0.228141 and 0.22728, respectively. We observe from Table 1 that the D-optimality obtained for the model (2.3) is at the same points as obtained by Czitrom (1988) for Scheffé's quadratic model and Singh (2003) for Darroch and Waller's quadratic mixture model.

Practically, we need designs in which at least the minimum proportion of each ingredient is available and the optimal designs consist of only binary blends with the exception of the centroid. Following Prescott (1998), we have shrunk the optimal designs towards the centroid in order to obtain pure mixtures. Nearly D-, A- and E-optimal designs are obtained at $a = f =$ 0.168497, 0.228141 and 0.22728. Further by shrinking only one Latin square in each block towards the centroid, as in Design 5.1.2, higher efficiency as compared to Design 5.1.1 is achieved. D-, A- and E-efficiencies for s = 0.05 are 88.9%, 82.2% and 81.3%, respectively for the Design 5.1.1 while for the Design 5.1.2 the corresponding values are 94.80%, 91.68% and 91.20%, respectively. Note that the D-efficiency for the single Latin square based designs presented in section 5 is the same as that obtained for the design 5.1.1. The design 5.1.2 is also more efficient as compared to the design presented in Section 5 for John's (1984) single Latin square based design given in (4.1).

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