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BLOCK DESIGNS FOR MIXTURE EXPERIMENTS

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1. Introduction. Scheffé (1958), (1963) introduced Simplex–Lattice and Simplex–Centroid designs for experiments with mixtures. Recently, Murty (1966) and Murty and Das (1968) have evolved Symmetric-Simplex designs which are the generalized form of Scheffé's designs.

One of the basic requirements for any response surface design according to Box and Hunter (1957) is that it should lend itself to blocking. Mixture designs so far available in literature lack this desirable characteristic. Murty (1966) did make some efforts for blocking the Symmetric–Simplex designs and reached the conclusion which, however, is empirical, saying that the actual blocking is not possible and the only possibility is to replicate the designs. Our investigations into this problem indicate that though the orthogonal blocking ensuring estimation of the regression parameters independent of the block effects is not possible without transforming the mixture variables, yet the parameters can be estimated by adjusting the parameters for the block effects. In the present paper we have derived the conditions required for blocking for estimating the parameters of a quadratic model. We have also constructed designs which satisfy these blocking conditions and hence are amenable to blocking. In the last section we have also constructed orthogonal blocking arrangements through suitable transformations. The case of cubic model will be dealt with in a separate paper.

2. The quadratic model.

2.1. *The blocking conditions.* Let the quadratic model proposed by Scheffé (1958) with block effects be

$$Y_u = \sum_{1 \leq i \leq n} \beta_i x_{iu} + \sum_{1 \leq i < j \leq n} \beta_{ij} x_{iu} x_{ju} + \sum_{w=1}^{t} \beta_w z_{wu} \tag{2.1}$$

where Y_u represents the response at the uth experimental point; $(u = 1, 2, \cdots, N)$, β_i and β_{ij} are the regression coefficients, x_{iu}'s are the mixture components such that $0 \leqq x_{iu} \leqq 1$ and $\sum_{1 \leqq i \leqq n} x_{iu} = 1$ for each $u = 1, 2, \cdots, N$, β_w is the expected value of the response in the wth block, $w = 1, 2, \cdots, t$ and

$$z_{wu} = 1 \quad \text{for those experimental points which fall in the } w\text{th block}$$
$$= 0 \quad \text{for all other points.}$$

Let the design with t blocks and N experimental points satisfy the following symmetry conditions of Murty and Das (1968):

$$\begin{aligned} &\sum x_{iu}^2 = A, \quad \sum x_{iu} x_{ju} = B, \quad \sum x_{iu}^2 x_{ju} = C, \\ &\sum x_{iu} x_{ju} x_{ku} = D, \quad \sum x_{iu}^2 x_{ju}^2 = E, \quad \sum x_{iu}^2 x_{ju} x_{ku} = F, \\ &\sum x_{iu} x_{ju} x_{ku} x_{1u} = G \qquad \text{for all} \quad i \neq j \neq k \neq 1. \end{aligned} \tag{2.2}$$

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The summations range over N experimental points and A, B, etc., are constants. Then, following the procedure similar to that of Murty and Das (1968), it is easy to derive the following two normal equations by differentiating

$$\sum_u (Y_u - \sum_i \beta_i x_{iu} - \sum_{i<j} \beta_{ij} x_{iu} x_{ju} - \sum_w \beta_w z_{wu})^2 \tag{2.3}$$

w.r. to any β_λ and $\beta_{\lambda\mu}$ respectively:

$$\sum_u x_{\lambda u} y_u = Ab_\lambda + B\sum_{1 \leq i \leq n \neq \lambda} b_i + C\sum_{1 \leq i \leq n \neq \lambda} b_{\lambda i} + D\sum_{1 \leq i < j \leq n \neq \lambda} b_{ij} + \sum_w b_w (\sum_{u=1}^{m_w} x_{\lambda u}) \tag{2.4}$$

$$\sum_u x_{\lambda u} x_{\mu u} y_u = C(b_\lambda + b_\mu) + D\sum_{1 \leq i \leq n \neq \lambda, \mu} b_i + Eb_{\lambda\mu} + F(\sum b_{\lambda i} + \sum b_{\mu i}) + G\sum_{1 \leq i < j \leq n \neq \lambda, \mu} b_{ij} + \sum_w b_w (\sum_{u=1}^{m_w} x_{\lambda u} x_{\mu u}) \quad i \neq \lambda, \mu \tag{2.5}$$

where b's are the estimates of the parameters β's.

Now, differentiating (2.3) w.r. to β_w, we get

$$\sum_{u=1}^{m_w} y_u = (\sum_{u=1}^{m_w} x_{iu}) \sum_{1 \leq i \leq n} b_i + (\sum_{u=1}^{m_w} x_{iu} x_{ju}) \sum_{1 \leq i < j \leq n} b_{ij} + m_w b_w \tag{2.6}$$

where m_w is the number of points in the wth block.

It may be observed from the equations (2.4), (2.5) and (2.6) that orthogonal blocking can be achieved if

$$\sum_{u=1}^{m_w} x_{iu} = 0 \quad \text{and} \quad \sum_{u=1}^{m_w} x_{iu} x_{ju} = 0 \qquad \text{for all } w = 1, 2, \cdots, t \text{ and } i \neq j. \tag{2.7}$$

But, an experiment with mixture requires $x_{iu} \geqq 0$. Obviously, the conditions (2.7) are satisfied when all x_{iu}'s are zero which is meaningless. Thus, it becomes clear that "orthogonal" blocking is not possible without transforming the mixture variables. We, therefore, attempted "non-orthogonal" blocking by adjusting the parameters for the block effects.

To facilitate the solution of the normal equations, let us assume that

$$\sum_{u=1}^{m_w} x_{iu} = \text{constant} = k_1 \quad \text{and} \tag{2.8}$$

$$\sum_{u=1}^{m_w} x_{iu} x_{ju} = \text{constant} = k_2$$

$$\text{for all } w = 1, 2, \cdots, t \text{ and } i \neq j;\ i, j = 1, 2, \cdots, n.$$

We then get from (2.6) the following solution for b_w

$$b_w = (\sum_{u=1}^{m_w} y_u - k_1 \sum_i b_i - k_2 \sum_{i<j} b_{ij}) / m_w. \tag{2.9}$$

Summing (2.9) over all $w = 1, 2, \cdots, t$ and putting $\sum_w 1/m_w = k_3$, we get

$$\sum_{w=1}^{t} b_w = \sum_w \sum_{u=1}^{m_w} y_u / m_w - k_1 k_3 \sum b_i - k_2 k_3 \sum b_{ij}. \tag{2.10}$$

Substituting for $\sum b_w$ in (2.4) and (2.5), we get the following two equations adjusted for block effects:

$$\begin{aligned}(2.11)\quad &\textstyle\sum_u x_{\lambda u}y_u - k_1\sum_w\sum_{u=1}^{m_w} y_u/m_w \\ &= (A-k_1{}^2k_3)b_\lambda + (B-k_1{}^2k_3)\textstyle\sum_{i\neq\lambda} b_i \\ &\quad + (C-k_1k_2k_3)\textstyle\sum_{i\neq\lambda} b_{\lambda i} + (D-k_1k_2k_3)\sum_{i<j\neq\lambda} b_{ij}\end{aligned}$$

$$\begin{aligned}(2.12)\quad &\textstyle\sum_u x_{\lambda u}x_{\mu u}y_u - k_2\sum_w\sum_{u=1}^{m_w} y_u/m_w \\ &= (C-k_1k_2k_3)(b_\lambda+b_\mu) + (D-k_1k_2k_3)\textstyle\sum_{i\neq\lambda,\mu} b_i \\ &\quad + (E-k_2{}^2k_3)b_{\lambda\mu} + (F-k_2{}^2k_3)(\textstyle\sum b_{\lambda i}+\sum b_{\mu i}) \\ &\quad + (G-k_2{}^2k_3)\textstyle\sum_{i<j\neq\lambda,\mu} b_{ij}. \qquad i\neq\lambda,\mu\end{aligned}$$

The l.h.s. in (2.11) and (2.12) are the adjusted sums of products.

Let us put

$$A-k_1{}^2k_3 = A^*,\quad B-k_1{}^2k_3 = B^*,\quad C-k_1k_2k_3 = C^*,\quad D-k_1k_2k_3 = D^*,$$

$$E-k_2{}^2k_3 = E^*,\quad F-k_2{}^2k_3 = F^*,\quad G-k_2{}^2k_3 = G^*.$$

$$\begin{aligned}(2.13)\quad &\textstyle\sum_u x_{\lambda u}y_u - k_1\sum_w\sum_{u=1}^{m_w} y_u/m_w = (\sum_u x_{\lambda u}y_u)^* \\ &\textstyle\sum_u x_{\lambda u}x_{\mu u}y_u - k_2\sum_w\sum_{u=1}^{m_w} y_u/m_w = (\sum_u x_{\lambda u}x_{\mu u}y_u)^*.\end{aligned}$$

Then, the equations (2.11) and (2.12) can be written as

$$(2.14)\quad (\textstyle\sum_u x_{\lambda u}y_u)^* = A^*b_\lambda + B^*\sum_{1\neq\lambda} b_i + C^*\sum_{i\neq\lambda} b_{\lambda i} + D^*\sum_{i<j\neq\lambda} b_{ij}$$

$$\begin{aligned}(2.15)\quad (\textstyle\sum_u x_{\lambda u}x_{\mu u}y_u)^* &= C^*(b_\lambda+b_\mu) + D^*\textstyle\sum_{i\neq\lambda,\mu} b_i + E^*b_{\lambda\mu} + F^*(\sum b_{\lambda i}+\sum b_{\mu i}) \\ &\quad + G^*\textstyle\sum_{i<j\neq\lambda,\mu} b_{ij} \qquad i\neq\lambda,\mu.\end{aligned}$$

These equations are similar to those of Murty and Das (1968) with the exception that the products $\sum_u x_{\lambda u}y_u$ and $\sum_u x_{\lambda u}x_{\mu u}y_u$ are replaced by adjusted products $(\sum_u x_{\lambda u}y_u)^*$ and $(\sum_u x_{\lambda u}x_{\mu u}y_u)^*$ respectively and the constants A, B, etc., of Murty and Das (1968) are replaced by the constants A^*, B^*, etc. We, therefore, straightway obtain the following solutions of b_λ and $b_{\lambda\mu}$:

$$\begin{aligned}(2.16)\quad PQb_\lambda &= PQ_4(\textstyle\sum_u x_{\lambda u}y_u)^* - PQ_2\sum_i(\sum_u x_{\lambda u}x_{iu}y_u)^* - \Delta_1\sum_i(\sum_u x_{iu}y_u)^* \\ &\quad + \Delta_2\textstyle\sum_{i<j}(\sum_u x_{iu}x_{ju}y_u)^*\end{aligned}$$

and

$$\begin{aligned}&PQ(E^*-2F^*+G^*)b_{\lambda\mu} \\ &\quad = PQ(\textstyle\sum_u x_{\lambda u}x_{\mu u}y_u)^* - P[Q_4(C^*-D^*)-Q_3(F^*-G^*)] \\ (2.17)\quad &\qquad\qquad \cdot[(\textstyle\sum_u x_{\lambda u}y_u)^* + (\sum_u x_{\mu u}y_u)^*] \\ &\quad + P[Q_2(C^*-D^*)-Q_1(F^*-G^*)][\textstyle\sum_i(\sum_u x_{\lambda u}x_{iu}y_u)^* + \sum_i(\sum_u x_{\mu u}x_{iu}y_u)^*] \\ &\quad - [(D^*P_4-G^*P_3)Q - 2(C^*-D^*)\Delta_1 + 2(F^*-G^*)\Delta_3]\textstyle\sum_i(\sum_u x_{iu}y_u)^* \\ &\quad + [(D^*P_2-G^*P_1)Q - 2(C^*-D^*)\Delta_2 + 2(F^*-G^*)\Delta_4]\textstyle\sum_{i<j}(\sum_u x_{iu}x_{ju}y_u)^*.\end{aligned}$$

where $P_1 = A^*+(n-1)B^*$

$$P_2 = 2C^*+(n-2)D^*$$
$$P_3 = (n-1)C^*+\binom{n-1}{2}D^*$$
$$P_4 = E^*+2(n-2)F^*+\binom{n-2}{2}G^*$$
$$P = P_1P_4-P_2P_3$$
$$Q_1 = A^*-B^*$$
$$Q_2 = C^*-D^*$$
$$Q_3 = (n-2)(C^*-D^*)$$
$$Q_4 = E^*+(n-4)F^*-(n-3)G^*$$
$$Q = Q_1Q_4-Q_2Q_3$$
$$\Delta_1 = P_4[B^*Q_4-Q_2(C^*+(n-2)D^*)]-P_3[D^*Q_4-Q_2(2F^*+(n-3)G^*)]$$
$$\Delta_2 = P_2[B^*Q_4-Q_2(C^*+(n-2)D^*)]-P_1[D^*Q_4-Q_2(2F^*+(n-3)G^*)]$$
$$\Delta_3 = P_4[B^*Q_3-Q_1(C^*+(n-2)D^*)]-P_3[D^*Q_3-Q_1(2F^*+(n-3)G^*)] \text{ and}$$
$$\Delta_4 = P_2[B^*Q_3-Q_1(C^*+(n-2)D^*)]-P_1[D^*Q_3-Q_1(2F^*+(n-3)G^*)].$$

The solutions for any b_λ and $b_{\lambda\mu}$ follow from the above because of the symmetry of the design. The block estimate b_w can easily be obtained by substituting for $\sum b_i$ and $\sum b_{ij}$ in (2.9).

Thus, it has been possible to estimate the parameters of the quadratic model (2.1) fitted through designs satisfying the relations (2.2) and (2.8). The conditions (2.8), in addition to the conditions (2.2) which are to be satisfied even if there is no blocking, therefore, must be satisfied for non-orthogonal blocking.

The variances and covariances of different estimates of the quadratic model (2.1) are easily obtained by following the method given by Murty and Das (1968).

2.2. *Analysis of variance.* Let the design of N points be replicated r times. Suppose there are t blocks. Then, the break-up of the degrees of freedom in the analysis of variance for the quadratic model (2.1) will be as follows:

Source of variation	df
Block	$t-1$
Regression	$n+\binom{n}{2}-1$
Lack of fit	$N-[n+\binom{n}{2}+t]+1$
Error	$N(r-1)$
Total	$Nr-1.$

The S.S. for blocks is found in the usual way. Regression S.S. is given by $S_R = \sum_{1 \leq i \leq n} b_i(\sum_u x_{iu} y_u)+\sum_{1 \leq i<j \leq n} b_{ij}(\sum_u x_{iu} x_{ju} y_u)-\text{C.F.}$, C.F. being the S.S. due to general mean. The error S.S. is found from the r replicated observations for each N design points. The lack of fit S.S. is found by subtraction.

3. Designs in blocks. We now proceed to obtain designs which satisfy the conditions (2.2) and (2.8) and hence are amenable to blocking.

3.1. *Designs through suitable choice of mixture points on a simplex.* Consider an n-component simplex–Centroid design of Scheffé (1963) without the total mixture $(1/n\ 1/n\ 1/n \cdots 1/n)$. Then by joining the points representing binary mixtures with equal proportions a triangle can be formed on each of the $\binom{n}{3}$ faces of the simplex whose vertices are of the type $(\frac{1}{2}\ \frac{1}{2}\ 0\ 0 \cdots 0)$.

Points distant $1/p$ (p taking non-zero integral values) from the vertices of this triangle, measured on each face along the sides in a certain direction, will form a set of $\binom{n}{3}$ triangles with vertices of the type $(\frac{1}{2}(p-1)/2p\ 1/2p\ 0\ 0 \cdots 0)$. One more set of $\binom{n}{3}$ triangle with vertices of the type $(\frac{1}{2}\ 1/2p\ (p-1)/2p\ 0\ 0 \cdots 0)$ can be formed by points lying at the complementary distance $(p-1)/p$ measured on each face in the same direction from the respective vertices.

We now state the following theorem:

THEOREM. *The points of the vertices of the two sets of triangles form a mixture design in two blocks, each set of vertex-points constituting a compact block.*

PROOF. Let us consider the points of the first block. The point $(\frac{1}{2}(p-1)/2p\ 1/2p\ 0\ 0 \cdots 0)$ generates $\binom{n}{3}$ points in which non-zero values occur in the same cyclical order irrespective of the position of zeros. Each of these points further gives rise to 3 points. For instance, in the case of $(\frac{1}{2}\ 0\ (p-1)2p\ 1/2p\ 0\ 0 \cdots 0)$, we have the following 3 points:

$$(\tfrac{1}{2}\ 0\ (p-1)/2p\ 1/2p\ 0\ 0 \cdots 0),$$
$$(1/2p\ 0\ \tfrac{1}{2}\ (p-1)/2p\ 0\ 0 \cdots 0), \quad \text{and}$$
$$((p-1)/2p\ 0\ 1/2p\ \tfrac{1}{2}\ 0\ 0 \cdots 0).$$

Obviously, all the $3\binom{n}{3}$ points of the block satisfy the relations (2.2) and (2.8). Similar is the case for the points of the second block. It, therefore, follows that all the $6\binom{n}{3}$ points of the two blocks also satisfy the relations (2.2). Hence the theorem.

EXAMPLE. Let us consider a 4-component design for $p = 3$. Then the two blocks formed out of four faces of the simplex will be as given below:

Faces	Block 1				Block 2			
	$1/p = \frac{1}{3}$				$(p-1)/p = \frac{2}{3}$			
1	$\frac{1}{2}$	$\frac{2}{6}$	$\frac{1}{6}$	0	$\frac{1}{2}$	$\frac{1}{6}$	$\frac{2}{6}$	0
	$\frac{2}{6}$	$\frac{1}{6}$	$\frac{1}{2}$	0	$\frac{1}{6}$	$\frac{2}{6}$	$\frac{1}{2}$	0
	$\frac{1}{6}$	$\frac{1}{2}$	$\frac{2}{6}$	0	$\frac{2}{6}$	$\frac{1}{2}$	$\frac{1}{6}$	0
2	$\frac{1}{2}$	$\frac{2}{6}$	0	$\frac{1}{6}$	$\frac{1}{2}$	$\frac{1}{6}$	0	$\frac{2}{6}$
	$\frac{2}{6}$	$\frac{1}{6}$	0	$\frac{1}{2}$	$\frac{1}{6}$	$\frac{2}{6}$	0	$\frac{1}{2}$
	$\frac{1}{6}$	$\frac{1}{2}$	0	$\frac{2}{6}$	$\frac{2}{6}$	$\frac{1}{2}$	0	$\frac{1}{6}$
3	$\frac{1}{2}$	0	$\frac{2}{6}$	$\frac{1}{6}$	$\frac{1}{2}$	0	$\frac{1}{6}$	$\frac{2}{6}$
	$\frac{2}{6}$	0	$\frac{1}{6}$	$\frac{1}{2}$	$\frac{1}{6}$	0	$\frac{2}{6}$	$\frac{1}{2}$
	$\frac{1}{6}$	0	$\frac{1}{2}$	$\frac{2}{6}$	$\frac{2}{6}$	0	$\frac{1}{2}$	$\frac{1}{6}$
4	0	$\frac{1}{2}$	$\frac{2}{6}$	$\frac{1}{6}$	0	$\frac{1}{2}$	$\frac{1}{6}$	$\frac{2}{6}$
	0	$\frac{2}{6}$	$\frac{1}{6}$	$\frac{1}{2}$	0	$\frac{1}{6}$	$\frac{2}{6}$	$\frac{1}{2}$
	0	$\frac{1}{6}$	$\frac{1}{2}$	$\frac{2}{6}$	0	$\frac{2}{6}$	$\frac{1}{2}$	$\frac{1}{6}$

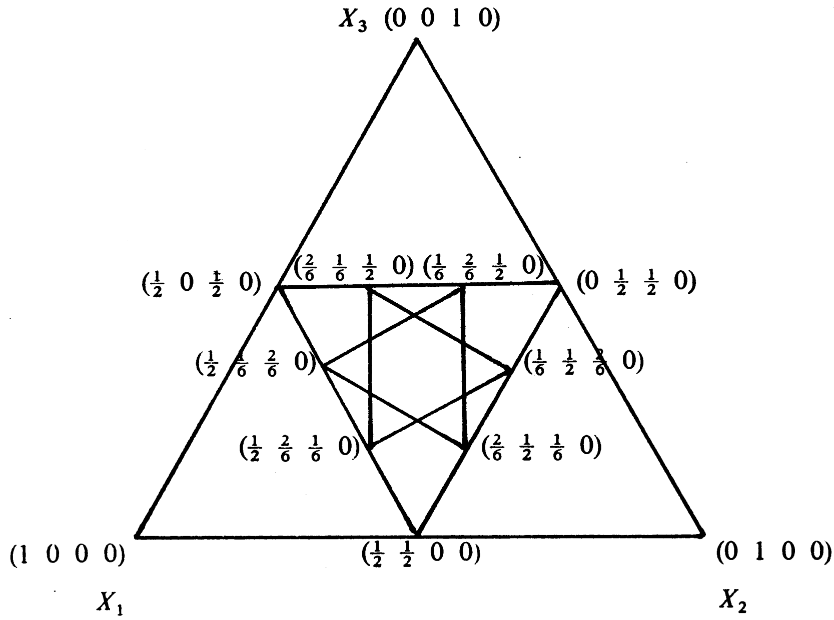

FIG. 1

The points of the first face for both the blocks are exhibited in Figure 1.

Thus, for an n-component mixture, the design will consist of $6\binom{n}{3}$ points, with $3\binom{n}{3}$ mixtures in each block. To these, suitable points at the interior and exterior can be added in common to each block to make a uniform exploration of the simplex and to provide replicated points for the estimation of a valid experimental error.

3.2. *Designs through mutually orthogonal Latin Squares* (MOLS). We first give the method of construction of mixture designs (without blocks) through MOLS.

3.2.1. *Construction of mixture designs through* MOLS. Suppose there is a set of $(s-1)$ MOLS of order s with symbols p_i, $1 \leqq i \leqq s$. Let the symbols p_i take the positive integral values (including zero). Then, if each element of the set of $(s-1)$ MOLS of order s is divided by $\sum_{1 \leqq i \leqq s} p_i$, then the set of $(s-1)$ MOLS forms a mixture design in s components.

It may be observed that, though the construction of mixture designs is quite simple, the method of estimation of regression parameters is somewhat cumbersome because some of the symmetry conditions (2.2) are not satisfied by these designs. We, therefore, propose the following restrictions on the choice of p_i, $1 \leqq i \leqq s$:

Case 1. All the p's are zero except two, say, p_i and p_j.

Case 2. All the p's are zero except three, say, p_i, p_j and p_k.

It can easily be verified that Case 1 satisfies all the conditions (2.2). Case 2 also satisfies all the conditions (2.2) when p_i, p_j and p_k are all different. But $\sum_u x_{iu}^2 x_{ju} x_{ku}$ has two values when $p_i = p_j \neq p_k$. The estimation procedure will not yet be difficult, for, it follows directly from the method of "Asymmetrical Mixture Experiments" given by Murty (1966).

3.2.2. *Designs in blocks through* MOLS. We first state a lemma due to Murty (1966):

LEMMA. *For odd s, the* $(s-1)$ MOLS *can be partitioned into two sets of* $(s-1)/2$ *Latin Squares each such that the* $s(s-1)/2$ *pairs of the s elements occur exactly once in any two columned submatrix of an array formed by* $s(s-1)/2$ *rows of any of the two sets.*

We now state the following theorem:

THEOREM. *If each of the elements of all the* $(s-1)$ MOLS *be divided by* $\sum_{1 \leqq i \leqq s} p_i$, *where* p_i, $1 \leqq i \leqq s$ *are selected according to one of the two cases of Section* 3.2.1, *then the two sets of* $(s-1)/2$ *Latin Squares each will form two different blocks of an s-component mixture design.*

PROOF. It follows from the lemma that $s(s-1)/2$ pairs of the s components occur exactly once in each block and this is true for all the columns. Thus, constancy of $\sum_u x_{iu} x_{ju}$ over each block is achieved. Further, $\sum_u x_{iu}$ is also the same for both the blocks because each p_i occurs $(s-1)/2$ times in each block. We thus see that both the conditions (2.8) for blocking are satisfied by the design. Hence the theorem.

EXAMPLE. For $s = 5$, we give below the two sets of two MOLS which with $p_i = 1$, $p_j = 2$, $p_k = 3$, $p_l = p_m = 0$ correspond to Case 2.

Set I					*Set* II				
0	1	3	0	2	0	0	2	1	3
1	2	0	0	3	1	0	3	2	0
2	3	0	1	0	2	1	0	3	0
3	0	1	2	0	3	2	0	0	1
0	0	2	3	1	0	3	1	0	2
0	3	0	2	1	0	2	1	3	0
1	0	0	3	2	1	3	2	0	0
2	0	1	0	3	2	0	3	0	1
3	1	2	0	0	3	0	0	1	2
0	2	3	1	0	0	1	0	2	3.

Then, by dividing each element by 6, we get a 5-component mixture design in two blocks and 20 mixtures.

3.3. *Designs through factorial experiments.* In this section we propose a method of obtaining orthogonal blocking arrangements by transforming the mixture variables.

3.3.1. *Preliminary.* In an n-component mixture experiment, if x_i is the proportion of the ith component, then

$$\sum_{1 \leqq i \leqq n} x_i = 1 \quad (3.1)$$

or

$$x_n = 1 - \sum_{1 \leqq i \leqq n-1} x_i. \quad (3.2)$$

Thus, there are only $(n-1)$ independent variables in the design. Murty (1966), instead of choosing the proportions of n components for satisfying (3.1), considered only $(n-1)$ components for choosing the proportions and then determined the proportion of the nth component such that (3.2) is satisfied. Utilising this, he constructed mixture designs through s^{n-1} factorial designs.

As regards the analysis, Murty (1966) showed that Scheffé's quadratic model in n mixture variables

$$Y_u = \sum_{1 \leqq i \leqq n} \beta_i x_{iu} + \sum_{1 \leqq i < j \leqq n} \beta_{ij} x_{iu} x_{ju} \quad (3.3)$$

reduces to the following general quadratic model in $(n-1)$ factorial variables by virtue of (3.2)

$$Y_u = B_0 + \sum_{1 \leqq i \leqq n-1} B_i x_{iu} + \sum_{1 \leqq i \leqq n-1} B_{ii} x_{iu}^2 + \sum_{1 \leqq i < j \leqq n-1} B_{ij} x_{iu} x_{ju} \quad (3.4)$$

where

$$B_0 = \beta_n$$

$$B_i = \beta_i - \beta_n + \beta_{in} \quad (3.5)$$

$$B_{ii} = -\beta_{in} \quad \text{and}$$

$$B_{ij} = \beta_{ij} - \beta_{in} - \beta_{jn}.$$

Conversely, if B's are known through the fitting of (3.4), Murty (1966) obtained the following

$$\beta_i = B_i + B_0 + B_{ii}$$

$$\beta_n = B_0 \quad (3.6)$$

$$\beta_{ij} = B_{ij} - B_{ii} - B_{jj} \quad \text{and}$$

$$\beta_{in} = -B_{ii}.$$

Murty (1966), thus established a correspondence between the model (3.3) in n mixture variables and the model (3.4) in $(n-1)$ factorial variables. He, therefore, suggested to fit the model in $(n-1)$ factorial variables (instead of fitting the model (3.3) in mixture variables) and then to transform this model into a model (3.3) for the mixture variables.

3.3.2. *Orthogonal arrangements in blocks.* With the s^{n-1} confounded factorial design, it is well known that the design ensures orthogonal blocking for the model (3.4) if the main effects and first order interactions are not confounded with block differences. Supposing that the s^{n-1} factorial design is an orthogonal arrangement in blocks, the B's of the model (3.4) can be estimated independent of the block effects. As the parameters β's of the model (3.3) are linear combinations in B's (by virtue of the relations (3.6)), the parameters β's can also be estimated independent of the block effects. Thus, an orthogonal arrangement in blocks for the $(n-1)$ factorial variables ensures orthogonal blocking of the design in n mixture variables.

It is known that the blocking can be orthogonal even if a fraction of the s^{n-1} factorial design be so taken that no four factor interaction or less is in the identity group and no first order interaction or less is confounded with the blocks. Similarly, the rotatable designs can also be used to achieve orthogonal blocking if the rotatable designs satisfy the blocking conditions as obtained by Box and Hunter (1957).

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