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Mixture Designs Generated Using Orthogonal Arrays From Mutually Orthogonal Latin Squares

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Abstract

This paper proposes an algorithm for constructing mixture designs based on orthogonal arrays of index unity containing the smallest number of runs for a given number of levels and a given strength using mutually orthogonal Latin squares. The algorithm allows the generation of cost effective and efficient mixture designs for Scheffé's canonical polynomials.

Key words: Mixture experiments; Mutually orthogonal Latin squares; Restricted region; G-efficiency.

1. Introduction

In experiments with mixtures, the response is a function only of the proportions of the q components present in the mixture and not of the total amount of the mixture. If x_i is the proportion of ith component, i = 1, 2, …, q, then

$$0 \leq x_i \leq 1, \quad \sum_{i=1}^{q} x_i = 1 \tag{1}$$

These restrictions force the factor space of the q components to take form of a $(q-1)$ dimensional simplex. The general purpose of mixture experimentation is to estimate the properties of an entire multicomponent system from only a limited number of observations. These observations are taken at preselected combinations of the components to determine which of the combinations in some sense optimize the response.

In many practical situations, one can encounter certain additional constraints that are placed on some or all component proportions besides (1). These are of the form

$$0 \leq L_i \leq x_i \leq U_i \leq 1; \; i = 1, 2, \ldots, q \tag{2}$$

where, L_i and U_i denote the lower bound and upper bound for the component proportion x_i; i = 1, 2, …, q. These supplementary restrictions limit the experimentation to some sub-region of the simplex, thereby altering the shape of the experimental region from a simplex to an irregularly shaped convex polyhedron inside the simplex. In such situations, directing the design and modelling only to the sub-region can help in lowering the experimentation cost and time and increasing the precision of model estimates.

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Mixture designs have a variety of applications in several industries. Amongst many others, Cafaggi *et al.* (2003) illustrated the application of a constrained mixture design to a pharmaceutical formulation. Mirabedini *et al.* (2012) discussed the application of mixture designs for the formulation of thermoplastic road markings. Schrevens and Cornell (1993) analysed the mixture designs for plant nutrition research. Buruk *et al.* (2016) reviewed the recent applications of mixture designs in the food industry.

Scheffé (1958, 1963) was the first to develop simplex lattice and simplex centroid designs for fitting the canonical polynomial models:

Linear model: $Y=\sum_{i=1}^{q}\beta_i x_i+\varepsilon$ (3)

Quadratic model: $Y=\sum_{i=1}^{q}\beta_i x_i+\sum\sum_{i<j}^{q}\beta_{ij}x_i x_j+\varepsilon$ (4)

Special cubic model: $Y=\sum_{i=1}^{q}\beta_i x_i+\sum\sum_{i<j}^{q}\beta_{ij}x_i x_j+\sum\sum\sum_{i<j<k}^{q}\beta_{ijk}x_i x_j x_k+\varepsilon$ (5)

McLean and Anderson (1966) developed extreme vertices designs (EVD) which satisfy both the constraints (1) and (2). A partial solution to the restricted exploration problem is the work of Thompson and Myers (1968) who considered an ellipsoidal region centred about a point of maximum interest. Snee and Marquardt (1974) obtained subsets of the extreme vertices which provide precise estimates of the parameters of a linear model. Snee (1975) used the computer to develop designs in constrained mixture spaces for the quadratic model. Saxena and Nigam (1977) explored the restricted mixture region using symmetric simplex design. Murthy and Murty (1983) discussed a method of construction of mixture designs for the exploration of the restricted region using factorials.

Much of the work on Latin squares has been done by various authors, for example, Bose (1938), Mann (1942) Parker (1959 a, b), Bose, Shrikhande and Parker (1960), Menon (1961) and Wallis (1984), who gave the methods of construction of mutually orthogonal Latin squares in various ways.

In this paper, we present an algorithm for constructing orthogonal arrays based mixture designs. The orthogonal arrays used in the proposed algorithm are constructed using a complete set of mutually orthogonal Latin squares. These orthogonal arrays have index unity and contain the smallest number of runs for a given number of levels and a given strength. This algorithm, therefore, leads to designs with small number of distinct runs.

We have examined and compared the designs constructed through this algorithm with the existing designs based on G-efficiency. The manageable number of design points help in reducing the cost and time in statistical experiments.

2. Orthogonal Arrays Based on Mutually Orthogonal Latin Squares

Hypercubes of strength *'d'* were defined by Rao (1946). Later, Rao (1947) extended the definition of hypercubes of strength d to cover a wider class of arrays called orthogonal arrays. An $N \times k$ array $\boldsymbol{A}$ with entries from S is said to be an orthogonal array $OA(N, k, s, t)$ with s levels, strength t and index λ (for some t in the range $0 \le t \le k$) if every $N \times t$ sub-array of $\boldsymbol{A}$ contains each t-tuple based on S exactly λ times as a row. If $\lambda = 1$, then such arrays are referred to as orthogonal arrays of index unity. (Bush 1952). Orthogonal arrays can be constructed using mutually orthogonal Latin squares.

A Latin square arrangement is an arrangement of s symbols in s rows and s columns, such that every symbol occurs once in each row and each column. When two Latin squares of same order are superimposed on one another, in the resultant array if every ordered pair of symbols occurs exactly once, then the two Latin squares are said to be orthogonal. A collection of ω Latin squares of order s, every pair of which is orthogonal, is called a set of mutually orthogonal Latin squares, and is denoted by $MOLS(s, \omega)$. Such a collection constitutes a complete set of mutually orthogonal Latin squares when $\omega = s - 1$.

2.1. Design criteria

If $\boldsymbol{X}$ denotes the $N \times k$ design matrix, then a useful criterion for evaluating the design is the minimum-maximum variance criterion. This refers to minimizing the maximum variance of prediction over the experimental region, where the prediction variance at the point $\boldsymbol{x}$ ($1 \times k$ row vector) is given by $\sigma^2 \upsilon$ and $\upsilon = \boldsymbol{x}(\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{x}'$. Computation of the maximum variance provides a criterion of how close is a design to optimality. The G-efficiency or Global efficiency of the design is given by,

$$G\text{-}efficiency\ (percent\ of\ optimum) = 100p/Nd$$

where, p is the number of model parameters, N is the number of design points and d is the maximum value of υ over the experimental region.

Wheeler (1972) stated as a practical rule of thumb that any design with a *G-efficiency* $\geq 50\%$ could be called good for practical purposes.

3. OABMD Algorithm

Let s be a prime or a power of a prime, then there are $(s-1)$ mutually orthogonal Latin squares of order s. Superimpose these $(s-1)$ mutually orthogonal Latin squares on one another. Label the rows and columns of this array with 0, 1, …, $s-1$. Prefix labels of the rows and post fix the labels of the columns to the entries of the superimposed mutually orthogonal Latin squares. The elements of the resultant give an $OA(s^2, s+1, s, 2)$ with the maximum number of factors. We now present Orthogonal Array Based Mixture Design (OABMD) algorithm for constructing q component mixture designs using mutually orthogonal Latin squares.

Step 1: Construct an orthogonal array $OA(s^2, s+1, s, 2)$ using the set of $(s-1)$ mutually orthogonal Latin squares, with $q = (s+1)$ factors. Denote this matrix by $\boldsymbol{A}$.

Step 2: Create a matrix $\boldsymbol{M}$ of order $q \times q$ which is symmetrical but not orthogonal, having all elements as integers with sum of elements in each row and each column being zero. The choice of $\boldsymbol{M}$ is arbitrary and is useful in getting more vertices of the experimental region in the design as mentioned by Murthy and Murty (1983).

Step 3: Identify the minimum value in each column of $\boldsymbol{A} \times \boldsymbol{M}$ and subtract it from all the entries of that corresponding column to create a new matrix $\boldsymbol{T}$.

Step 4: Obtain the row totals for matrix $\boldsymbol{T}$. Divide the entries of each row of $\boldsymbol{T}$ by its corresponding row total to obtain a new matrix $\boldsymbol{Z}$. The resultant matrix is a mixture design satisfying

$$0 \leq z_i \leq 1 \text{ and } \sum_{i=1}^{q} z_i = 1,$$

z_i being the proportion for ith component.

Further, if the mixture experiment has to be performed in the restricted region, where each component is bounded by lower or upper bounds, or both, then proceed as follows:

Step 5: Rank the components in order of their increasing ranges $R_i = (U_i - L_i)$ such that X_1 has the smallest range and X_q has the largest range, assuming range to be inversely proportional to the importance (in terms of cost, effectiveness, *etc.*) of the components in the experiment.

Step 6: Using the transformation given by Saxena and Nigam (1977), $X_i = L_i + (R_i \times z_i)$, compute the entries for the first $(q-1)$ components of the design matrix $\boldsymbol{X}$. The levels of X_q are obtained by $X_q = 1 - \sum_{i=1}^{q-1} X_i$.

Step 7: In case X_q lies beyond the specified bounds, generate candidate design points. There may be multiple candidate points corresponding to a given design point. The candidate points are generated by adjusting the level of one of the components by a quantity equal to the difference between the substituted upper or lower bound and the computed value for X_q. Additional points are produced only from those components whose adjusted levels remain within the limits of the components.

We have illustrated the OABMD algorithm for generating designs for three, four and five components. These designs have been found to be efficient designs.

4. Mixture Designs For Three, Four And Five Components

4.1. Three component example

Consider the three-component mixture experiment, where all the components satisfy (1). Construct an orthogonal array $OA(4, 3, 2, 2)$ with three factors. Denote it by $\boldsymbol{A}$.

$$\boldsymbol{A^T} = \begin{pmatrix} 0 & 0 & 1 & 1 \\ 0 & 1 & 1 & 0 \\ 0 & 1 & 0 & 1 \end{pmatrix}$$

Multiply it with a symmetric and non-orthogonal matrix, $\boldsymbol{M}$,

$$\boldsymbol{M} = \begin{pmatrix} -2 & 1 & 1 \\ 1 & -2 & 1 \\ 1 & 1 & -2 \end{pmatrix}$$

having row sums and column sums as zero, to obtain $\boldsymbol{T}$,

$$\boldsymbol{T} = \begin{pmatrix} 1 & 1 & 1 \\ 3 & 0 & 0 \\ 0 & 0 & 3 \\ 0 & 3 & 0 \end{pmatrix}$$

Using step 4 of the OABMD algorithm, we obtain the design matrix $\boldsymbol{Z}$ for unrestricted region as follows:

$$\boldsymbol{Z}=\begin{pmatrix} 1/3 & 1/3 & 1/3 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix} \tag{6}$$

We observe that $0 \leq z_i \leq 1$ and sum of the elements for each row is unity. This design has a G-efficiency of 81.8% for fitting (3).

Next, using step 6 of the OABMD algorithm, we compute the design matrix for the constrained region, as discussed below.

Example 1: Consider the following three component mixture experiment, discussed by Murthy and Murty (1983), in order of increasing ranges:

$$0.2 \leq X_1 \leq 0.3$$
$$0.3 \leq X_2 \leq 0.5$$
$$\text{and} \quad 0.2 \leq X_3 \leq 0.5$$

The first $(q-1)$ columns of the design matrix $\boldsymbol{X}$ are constructed using $X_i = L_i + (R_i \times z_i)$, where z_i is the proportion of the ith component of $\boldsymbol{Z}$ in (6). The levels of X_q are obtained by $X_q = 1 - \sum_{i=1}^{q-1} X_i$. The four design points of the resulting design matrix $\boldsymbol{X}$ are given in (7).

$$\boldsymbol{X}=\begin{pmatrix} 0.233 & 0.367 & 0.4 \\ 0.3 & 0.3 & 0.4 \\ 0.2 & 0.3 & 0.5 \\ 0.2 & 0.5 & 0.3 \end{pmatrix} \tag{7}$$

This design has a G-efficiency of 81.8% for fitting (3).

Other choices of $\boldsymbol{M}$ and resultant unconstrained and constrained design matrix are listed in Table 1.

Table 1: Unconstrained and constrained mixture designs corresponding to different choices of *M*

Choice of $\boldsymbol{M}$	Resultant $\boldsymbol{Z}$	Resultant $\boldsymbol{X}$
$\boldsymbol{M_1}=\begin{pmatrix} 1 & -1 & 0 \\ -1 & 2 & -1 \\ 0 & -1 & 1 \end{pmatrix}$	$\boldsymbol{Z_1}=\begin{pmatrix} 1/4 & 1/2 & 1/4 \\ 0 & 3/4 & 1/4 \\ 1/4 & 3/4 & 0 \\ 1/2 & 0 & 1/2 \end{pmatrix}$	$\boldsymbol{X_1}=\begin{pmatrix} 0.225 & 0.4 & 0.375 \\ 0.2 & 0.45 & 0.35 \\ 0.225 & 0.45 & 0.325 \\ 0.25 & 0.3 & 0.45 \end{pmatrix}$
$\boldsymbol{M_2}=\begin{pmatrix} -1 & 1 & 0 \\ 1 & -2 & 1 \\ 0 & 1 & -1 \end{pmatrix}$	$\boldsymbol{Z_2}=\begin{pmatrix} 1/3 & 1/3 & 1/3 \\ 2/3 & 0 & 1/3 \\ 1/3 & 0 & 2/3 \\ 0 & 1 & 0 \end{pmatrix}$	$\boldsymbol{X_2}=\begin{pmatrix} 0.233 & 0.367 & 0.4 \\ 0.267 & 0.3 & 0.433 \\ 0.233 & 0.3 & 0.467 \\ 0.2 & 0.5 & 0.3 \end{pmatrix}$

Choice of $\boldsymbol{M}$	Resultant $\boldsymbol{Z}$	Resultant $\boldsymbol{X}$
$\boldsymbol{M}_3=\begin{pmatrix} 1 & -1 & 0 \\ -1 & 0 & 1 \\ 0 & 1 & -1 \end{pmatrix}$	$\boldsymbol{Z}_3=\begin{pmatrix} 1/3 & 1/3 & 1/3 \\ 0 & 2/3 & 1/3 \\ 1/3 & 0 & 2/3 \\ 2/3 & 1/3 & 0 \end{pmatrix}$	$\boldsymbol{X}_3=\begin{pmatrix} 0.233 & 0.367 & 0.4 \\ 0.2 & 0.433 & 0.367 \\ 0.233 & 0.3 & 0.467 \\ 0.267 & 0.367 & 0.367 \end{pmatrix}$
$\boldsymbol{M}_4=\begin{pmatrix} -1 & 1 & 0 \\ 1 & 0 & -1 \\ 0 & -1 & 1 \end{pmatrix}$	$\boldsymbol{Z}_4=\begin{pmatrix} 1/3 & 1/3 & 1/3 \\ 2/3 & 0 & 1/3 \\ 1/3 & 2/3 & 0 \\ 0 & 1/3 & 2/3 \end{pmatrix}$	$\boldsymbol{X}_4=\begin{pmatrix} 0.233 & 0.367 & 0.4 \\ 0.267 & 0.3 & 0.433 \\ 0.233 & 0.433 & 0.333 \\ 0.2 & 0.367 & 0.433 \end{pmatrix}$
$\boldsymbol{M}_5=\begin{pmatrix} 2 & -1 & -1 \\ -1 & 2 & -1 \\ -1 & -1 & 2 \end{pmatrix}$	$\boldsymbol{Z}_5=\begin{pmatrix} 1/3 & 1/3 & 1/3 \\ 0 & 1/2 & 1/2 \\ 1/2 & 1/2 & 0 \\ 1/2 & 0 & 1/2 \end{pmatrix}$	$\boldsymbol{X}_5=\begin{pmatrix} 0.233 & 0.367 & 0.4 \\ 0.2 & 0.4 & 0.4 \\ 0.25 & 0.4 & 0.35 \\ 0.25 & 0.3 & 0.45 \end{pmatrix}$

The unconstrained and constrained design matrices obtained using different choices of $\boldsymbol{M}$ listed in the table above also yield a G-efficiency of 81.8% for fitting (3).

Example 2: Consider the following three component mixture experiment, discussed by Snee and Marquardt (1974) and by Saxena and Nigam (1977), in order of increasing ranges:

$$0.1 \le X_1 \le 0.6$$
$$0.1 \le X_2 \le 0.7$$
$$\text{and} \quad 0 \le X_3 \le 0.7$$

Using $\boldsymbol{M}$,

$$\boldsymbol{M}=\begin{pmatrix} -2 & 1 & 1 \\ 1 & -2 & 1 \\ 1 & 1 & -2 \end{pmatrix}$$

the four design points of the resulting design matrix $\boldsymbol{X}$ are given in (8).

$$\boldsymbol{X}=\begin{pmatrix} 0.267 & 0.3 & 0.433 \\ 0.6 & 0.1 & 0.3 \\ 0.1 & 0.1 & 0.8 \\ 0.1 & 0.7 & 0.2 \end{pmatrix} \tag{8}$$

We observe that the limits for the third component of the third design point of (8) lies outside the specified bounds, so we shall adjust the run (0.1, 0.1, 0.8), using step 7 of the OABMD algorithm, to create two candidate sub-points (0.1, 0.2, 0.7) and (0.2, 0.1, 0.7).

Case (a): Four-point design with candidate point (0.1, 0.2, 0.7)

The design matrix in (8) is modified to incorporate the candidate sub-point (0.1, 0.2, 0.7) to yield the following four design runs.

$$\boldsymbol{X_a} = \begin{pmatrix} 0.267 & 0.3 & 0.433 \\ 0.6 & 0.1 & 0.3 \\ 0.1 & 0.2 & 0.7 \\ 0.1 & 0.7 & 0.2 \end{pmatrix}$$

The G-efficiency for the above design matrix for fitting Scheffé's linear model given in (3) is 79.19%.

Case (b): Four-point design with candidate point (0.2, 0.1, 0.7)

The design matrix in (8) is adjusted to include the candidate sub-point (0.2, 0.1, 0.7) to give the following four design runs.

$$\boldsymbol{X_b} = \begin{pmatrix} 0.267 & 0.3 & 0.433 \\ 0.6 & 0.1 & 0.3 \\ 0.2 & 0.1 & 0.7 \\ 0.1 & 0.7 & 0.2 \end{pmatrix}$$

The G-efficiency for the above design matrix for fitting Scheffé's linear model given in (3) is 78.65%.

4.2. Four component example

To construct a mixture design in four components satisfying (1), construct an orthogonal array with four factors, say, $\boldsymbol{A} = OA(9, 4, 3, 2)$ and a symmetric and non-orthogonal matrix, $\boldsymbol{M}$ as

$$\boldsymbol{A^T} = \begin{pmatrix} 0 & 0 & 0 & 1 & 1 & 1 & 2 & 2 & 2 \\ 0 & 1 & 2 & 1 & 2 & 0 & 2 & 0 & 1 \\ 0 & 1 & 2 & 2 & 0 & 1 & 1 & 2 & 0 \\ 0 & 1 & 2 & 0 & 1 & 2 & 0 & 1 & 2 \end{pmatrix} \text{ and } \boldsymbol{M} = \begin{pmatrix} -3 & 1 & 1 & 1 \\ 1 & -3 & 1 & 1 \\ 1 & 1 & -3 & 1 \\ 1 & 1 & 1 & -3 \end{pmatrix}$$

The unconstrained mixture design $\boldsymbol{Z}$, is

$$\boldsymbol{Z} = \begin{pmatrix} 0.2 & 0.27 & 0.27 & 0.26 \\ 0.4 & 0.2 & 0.2 & 0.2 \\ 0.6 & 0.13 & 0.13 & 0.14 \\ 0.2 & 0.27 & 0 & 0.53 \\ 0.2 & 0 & 0.53 & 0.27 \\ 0.2 & 0.53 & 0.27 & 0 \\ 0 & 0.07 & 0.33 & 0.6 \\ 0 & 0.6 & 0.07 & 0.33 \\ 0 & 0.33 & 0.6 & 0.07 \end{pmatrix} \tag{9}$$

Consider the constrained four component mixture experiment as discussed by McLean and Anderson (1966). In manufacturing one particular type of flare the chemical constituents are magnesium (x_1), sodium nitrate (x_2), strontium nitrate (x_3), and binder (x_4). Engineering experience has indicated that the following constraints (in order of increasing ranges) on a proportion by weight basis should be utilized:

$$0.03 \leq X_1 \leq 0.08$$
$$0.40 \leq X_2 \leq 0.60$$
$$0.10 \leq X_3 \leq 0.50$$
$$\text{and } 0.10 \leq X_4 \leq 0.50$$

Using the OABMD algorithm, the constrained mixture design, ***X***, is

$$\boldsymbol{X} = \begin{pmatrix} 0.04 & 0.453 & 0.207 & 0.3 \\ 0.05 & 0.44 & 0.18 & 0.33 \\ 0.06 & 0.427 & 0.153 & 0.36 \\ 0.04 & 0.453 & 0.1 & 0.407 \\ 0.04 & 0.4 & 0.313 & 0.247 \\ 0.04 & 0.506 & 0.207 & 0.247 \\ 0.03 & 0.413 & 0.233 & 0.324 \\ 0.03 & 0.52 & 0.127 & 0.323 \\ 0.03 & 0.467 & 0.34 & 0.163 \end{pmatrix} \tag{10}$$

The design matrix ***Z*** in (9) as well as matrix ***X*** in (10) have a G-efficiency of 72.72% for fitting (3). The design matrix achieved using OABMD algorithm proves to be more economical in terms of number of design points and exploration of the constrained region than the XVERT algorithm.

If limitation of resources demands a reduction in the number of design points, we may use the method of normalization, as discussed by McLean and Anderson (1966). The operative idea is to compute a normalized distance d_{ij} between points of the design and randomly omit points that are less than a certain minimum distance from other design points.

$$d_{ij} = \left(\sum_{r=1}^{q} \left(\frac{x_{ir}\text{-}x_{jr}}{b_r\text{-}a_r} \right)^2 \right)^{\frac{1}{2}}$$

The design matrices, $\boldsymbol{Z_{1*}}$ and $\boldsymbol{X_{1*}}$, corresponding to the unrestricted and restricted region, consisting of just four design points, obtained using the above-mentioned technique are:

$$\boldsymbol{Z}_{1*} = \begin{pmatrix} 0.6 & 0.13 & 0.13 & 0.13 \\ 0.2 & 0.27 & 0 & 0.53 \\ 0 & 0.07 & 0.33 & 0.6 \\ 0 & 0.6 & 0.07 & 0.33 \end{pmatrix} \tag{11}$$

and

$$\boldsymbol{X}_{1*} = \begin{pmatrix} 0.06 & 0.427 & 0.153 & 0.36 \\ 0.04 & 0.453 & 0.1 & 0.407 \\ 0.04 & 0.4 & 0.313 & 0.247 \\ 0.03 & 0.52 & 0.127 & 0.323 \end{pmatrix} \tag{12}$$

The design matrix $\boldsymbol{Z}_{1^*}$ in (11) as well as matrix $\boldsymbol{X}_{1^*}$ in (12) have a G-efficiency of 100% for fitting (3). To allow estimation of error variance, we may add another design point, (0, 0.33, 0.6, 0.07) to the design matrix $\boldsymbol{Z}_{1^*}$in (11), then the resultant design consisting of five runs has a G-efficiency of 84.89% for fitting (3). Similarly, adding one point of the restricted region, say, (0.03, 0.467, 0.34, 0.163) to the design matrix $\boldsymbol{X}_{1^*}$ in (12) yields a G-efficiency of 80% for fitting (3).

In practical situations, fitting linear model is not always suitable. A higher model may provide a better fit to the given design. Consider the design matrices, ***Z*** and ***X***, given in (6) and (7). Further, in addition to these points, we may add the boundary points, centroids or the extreme vertices to facilitate the computation of G-efficiency for fitting higher order models as stated in (4) and (5). Addition of three centroid points to the unconstrained design matrix ***Z*** of (6) and corresponding design points in ***X*** of (7) yield two 7-point designs, both of which have a G-efficiency of 85.7% for fitting Scheffé quadratic model specified in (4). Similarly, adding one more point to the 7-point design of ***Z*** and ***X*** gives an 8-point design, both of which have a G-efficiency of 87.5% for fitting Scheffé cubic model mentioned in (5). Likewise, adding boundary points to the unconstrained and constrained design matrices, for any choice of ***M***, as stated in Table 1, yield a G-efficiency of 85.7% and 87.5% for fitting (4) and (5) respectively.

Similarly, for the four-component example, 11-point designs obtained by adding two boundary points, (0, 1, 0, 0) and (0, 0, 1, 0) to (9) and adding the points (0.03, 0.4, 0.1, 0.47) and (0.08, 0.6, 0.1, 0.22) to (10) yield a G-efficiency of 90.90% for fitting (4). This value of G-efficiency is computed using $\boldsymbol{X}_{\boldsymbol{E}}$ in place of ***X***, where $\boldsymbol{X}_{\boldsymbol{E}}$ given below is the extended design matrix for model (4).

$$\boldsymbol{X}_{\boldsymbol{E}} = \begin{pmatrix} 0.04 & 0.453 & 0.207 & 0.3 & 0.018 & 0.008 & 0.012 & 0.093 & 0.136 & 0.062 \\ 0.05 & 0.44 & 0.18 & 0.33 & 0.022 & 0.009 & 0.016 & 0.079 & 0.145 & 0.059 \\ 0.06 & 0.427 & 0.153 & 0.36 & 0.025 & 0.009 & 0.021 & 0.065 & 0.153 & 0.055 \\ 0.04 & 0.453 & 0.1 & 0.407 & 0.018 & 0.004 & 0.016 & 0.045 & 0.184 & 0.041 \\ 0.04 & 0.4 & 0.313 & 0.247 & 0.016 & 0.012 & 0.009 & 0.125 & 0.098 & 0.077 \\ 0.04 & 0.506 & 0.207 & 0.247 & 0.020 & 0.008 & 0.009 & 0.104 & 0.125 & 0.051 \\ 0.03 & 0.413 & 0.233 & 0.324 & 0.012 & 0.007 & 0.009 & 0.096 & 0.134 & 0.075 \\ 0.03 & 0.52 & 0.127 & 0.323 & 0.015 & 0.003 & 0.009 & 0.066 & 0.168 & 0.041 \\ 0.03 & 0.467 & 0.34 & 0.163 & 0.014 & 0.010 & 0.005 & 0.158 & 0.076 & 0.055 \\ 0.03 & 0.4 & 0.1 & 0.47 & 0.012 & 0.003 & 0.014 & 0.04 & 0.188 & 0.047 \\ 0.08 & 0.6 & 0.1 & 0.22 & 0.048 & 0.008 & 0.017 & 0.06 & 0.132 & 0.022 \end{pmatrix}$$

The G-efficiency values of ***Z*** and ***X*** for fitting (3) and (4) for different choices of ***M*** are listed in Table 2.

Table 2: G-efficiency of unconstrained and constrained mixture designs corresponding to different choices of *M*

Choice of $\boldsymbol{M}$	G-efficiency for fitting (3)		G-efficiency for fitting (3)		G-efficiency for fitting (4)	
	9-point design		5-point design		11-point design	
	$\boldsymbol{Z}$	$\boldsymbol{X}$	$\boldsymbol{Z}$	$\boldsymbol{X}$	$\boldsymbol{Z}$	$\boldsymbol{X}$
$\boldsymbol{M_1}=\begin{pmatrix} 1 & -1 & 0 & 0 \\ -1 & 2 & -1 & 0 \\ 0 & -1 & 2 & -1 \\ 0 & 0 & -1 & 1 \end{pmatrix}$	72.72%	72.72%	80%	80%	90.90%	90.90%
$\boldsymbol{M_2} = -\boldsymbol{M_1}$	72.72%	72.72%	80%	80%	90.90%	90.90%
$\boldsymbol{M_3}=\begin{pmatrix} 1 & -1 & 0 & 0 \\ -1 & 0 & 1 & 0 \\ 0 & 1 & 0 & -1 \\ 0 & 0 & -1 & 1 \end{pmatrix}$	72.72%	72.72%	84.89%	80%	90.90%	90.90%
$\boldsymbol{M_4} = -\boldsymbol{M_3}$	72.72%	72.72%	84.89%	80%	90.90%	90.90%
$\boldsymbol{M_5} = -\boldsymbol{M}$	72.72%	72.72%	84.89%	80%	90.90%	90.90%

4.3. Five component example

We may extend the application of our OABMD algorithm to five component constraints. To construct a mixture design, $\boldsymbol{Z}$, in five components satisfying (1), construct an orthogonal array with five factors, $\boldsymbol{A} = OA(16, 5, 4, 2)$.

$$\boldsymbol{A}^T = \begin{pmatrix} 0 & 0 & 0 & 0 & 1 & 1 & 1 & 1 & 2 & 2 & 2 & 2 & 3 & 3 & 3 & 3 \\ 0 & 1 & 2 & 3 & 1 & 0 & 3 & 2 & 2 & 3 & 0 & 1 & 3 & 2 & 1 & 0 \\ 0 & 1 & 2 & 3 & 2 & 3 & 0 & 1 & 3 & 2 & 1 & 0 & 1 & 0 & 3 & 2 \\ 0 & 1 & 2 & 3 & 3 & 2 & 1 & 0 & 1 & 0 & 3 & 2 & 2 & 3 & 0 & 1 \\ 0 & 1 & 2 & 3 & 0 & 1 & 2 & 3 & 0 & 1 & 2 & 3 & 0 & 1 & 2 & 3 \end{pmatrix}$$

and a symmetric and non-orthogonal matrix, $\boldsymbol{M}$ as

$$\boldsymbol{M} = \begin{pmatrix} -4 & 1 & 1 & 1 & 1 \\ 1 & -4 & 1 & 1 & 1 \\ 1 & 1 & -4 & 1 & 1 \\ 1 & 1 & 1 & -4 & 1 \\ 1 & 1 & 1 & 1 & -4 \end{pmatrix}$$

to give the unconstrained mixture design $\boldsymbol{Z}$,

$$\boldsymbol{Z} = \begin{pmatrix} 0.158 & 0.210 & 0.210 & 0.211 & 0.211 \\ 0.264 & 0.184 & 0.184 & 0.184 & 0.184 \\ 0.368 & 0.158 & 0.158 & 0.158 & 0.158 \\ 0.474 & 0.132 & 0.132 & 0.131 & 0.131 \\ 0.211 & 0.263 & 0.131 & 0 & 0.395 \\ 0.211 & 0.395 & 0 & 0.131 & 0.263 \\ 0.211 & 0 & 0.395 & 0.263 & 0.131 \\ 0.211 & 0.131 & 0.263 & 0.395 & 0 \\ 0.105 & 0.158 & 0.026 & 0.289 & 0.422 \\ 0.105 & 0.026 & 0.158 & 0.422 & 0.289 \\ 0.105 & 0.422 & 0.289 & 0.026 & 0.158 \\ 0.105 & 0.289 & 0.422 & 0.158 & 0.026 \\ 0 & 0.053 & 0.316 & 0.184 & 0.447 \\ 0 & 0.184 & 0.447 & 0.053 & 0.316 \\ 0 & 0.316 & 0.053 & 0.447 & 0.184 \\ 0 & 0.447 & 0.184 & 0.316 & 0.053 \end{pmatrix} \tag{13}$$

Using step 6 of the OABMD algorithm, we compute the design matrix of the constrained region. Consider the five-component example, discussed by Snee and Marquardt (1974). The gasoline blending model for a five-component system, namely, Butane (X_1), Alkylate (X_2), Lt. St. Run (X_3), Reformate (X_4) and Cat Cracked (X_5), with the following component ranges:

$$\begin{aligned} 0.00 \leq X_1 \leq 0.10 \\ 0.00 \leq X_2 \leq 0.10 \\ 0.05 \leq X_3 \leq 0.15 \\ 0.20 \leq X_4 \leq 0.40 \\ \text{and } 0.40 \leq X_5 \leq 0.60 \end{aligned}$$

Using the OABMD algorithm, the design matrix, $\boldsymbol{X}$ is as follows:

$$\boldsymbol{X} = \begin{pmatrix} 0.0158 & 0.0211 & 0.0710 & 0.2421 & 0.6500 \\ 0.0263 & 0.0184 & 0.0684 & 0.2369 & 0.6500 \\ 0.0368 & 0.0158 & 0.0658 & 0.2316 & 0.6500 \\ 0.0473 & 0.0132 & 0.0632 & 0.2263 & 0.6500 \\ 0.0211 & 0.0263 & 0.0632 & 0.2000 & 0.6894 \\ 0.0211 & 0.0394 & 0.0500 & 0.2263 & 0.6632 \\ 0.0211 & 0.0000 & 0.0895 & 0.2526 & 0.6368 \\ 0.0211 & 0.0132 & 0.0763 & 0.2789 & 0.6105 \\ 0.0105 & 0.0158 & 0.0526 & 0.2579 & 0.6632 \\ 0.0105 & 0.0026 & 0.0658 & 0.2842 & 0.6369 \\ 0.0105 & 0.0421 & 0.0789 & 0.2053 & 0.6632 \\ 0.0105 & 0.0289 & 0.0921 & 0.2316 & 0.6369 \\ 0.0000 & 0.0053 & 0.0816 & 0.2368 & 0.6763 \\ 0.0000 & 0.0184 & 0.0948 & 0.2105 & 0.6763 \\ 0.0000 & 0.0316 & 0.0552 & 0.2895 & 0.6237 \\ 0.0000 & 0.0447 & 0.0684 & 0.2632 & 0.6237 \end{pmatrix} \tag{14}$$

The design matrix $\boldsymbol{X}$ in (14) has many points, particularly of the fifth component, which lie beyond the specified limits of the component. Using step 7 of the OABMD algorithm, we adjust the matrix $\boldsymbol{X}$ in (14) to obtain the design matrix $\boldsymbol{X}^*$, given in (15), which has all the design points within the permissible limits of the components involved in the five-component example.

$$\boldsymbol{X}^* = \begin{pmatrix} 0.0158 & 0.0211 & 0.0710 & 0.2921 & 0.6000 \\ 0.0263 & 0.0184 & 0.0684 & 0.2869 & 0.6000 \\ 0.0368 & 0.0158 & 0.0658 & 0.2816 & 0.6000 \\ 0.0473 & 0.0132 & 0.0632 & 0.2763 & 0.6000 \\ 0.0211 & 0.0263 & 0.0632 & 0.2894 & 0.6000 \\ 0.0211 & 0.0394 & 0.0500 & 0.2895 & 0.6000 \\ 0.0211 & 0.0000 & 0.0895 & 0.2894 & 0.6000 \\ 0.0211 & 0.0132 & 0.0763 & 0.2894 & 0.6000 \\ 0.0105 & 0.0158 & 0.0526 & 0.3211 & 0.6000 \\ 0.0105 & 0.0026 & 0.0658 & 0.3211 & 0.6000 \\ 0.0105 & 0.0421 & 0.0789 & 0.2685 & 0.6000 \\ 0.0105 & 0.0289 & 0.0921 & 0.2685 & 0.6000 \\ 0.0000 & 0.0053 & 0.0816 & 0.3131 & 0.6000 \\ 0.0000 & 0.0184 & 0.0948 & 0.2868 & 0.6000 \\ 0.0000 & 0.0316 & 0.0552 & 0.3132 & 0.6000 \\ 0.0000 & 0.0447 & 0.0684 & 0.2869 & 0.6000 \end{pmatrix} \tag{15}$$

The design matrix $\boldsymbol{X}^*$ in (15), computed using the OABMD algorithm, has a G-efficiency of 58.10% for fitting (3). The design matrix $\boldsymbol{X}^*$ in (15) is space filling and allows for the greater exploration of the interior of the restricted region in contrast to the only extreme vertices generated by the XVERT algorithm.

Other choices of $\boldsymbol{M}$ with corresponding G-efficiency values of $\boldsymbol{Z}$ and $\boldsymbol{X}$ for fitting (3) and (4) are listed in Table 3.

Table 3: G-efficiency of unconstrained and constrained mixture designs corresponding to different choices of *M*

Choice of $\boldsymbol{M}$	G-efficiency for fitting (3)		G-efficiency for fitting (4)	
	$\boldsymbol{Z}$	$\boldsymbol{X}^*$	$\boldsymbol{Z}$	$\boldsymbol{X}^*$
$\boldsymbol{M}_1 = \begin{pmatrix} 1 & -1 & 0 & 0 & 0 \\ -1 & 2 & -1 & 0 & 0 \\ 0 & -1 & 2 & -1 & 0 \\ 0 & 0 & -1 & 2 & -1 \\ 0 & 0 & 0 & -1 & 1 \end{pmatrix}$	73.96%	88.57%	93.75%	92.20%
$\boldsymbol{M}_2 = -\boldsymbol{M}_1$	73.96%	74.55%	93.75%	39.70%*
$\boldsymbol{M}_3 = \begin{pmatrix} 1 & -1 & 0 & 0 & 0 \\ -1 & 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & -1 & 0 \\ 0 & 0 & -1 & 0 & 1 \\ 0 & 0 & 0 & 1 & -1 \end{pmatrix}$	73.96%	76.34%	93.75%	87.93%
$\boldsymbol{M}_4 = -\boldsymbol{M}_3$	73.96%	72.88%	93.75%	49.98%**

Choice of $\boldsymbol{M}$	G-efficiency for fitting (3)		G-efficiency for fitting (4)	
	$\boldsymbol{Z}$	$\boldsymbol{X}^*$	$\boldsymbol{Z}$	$\boldsymbol{X}^*$
$\boldsymbol{M_5} = -\boldsymbol{M}$	73.96%	70.42%	93.75%	94.26%
$\boldsymbol{M_6} = \begin{pmatrix} 0 & 1 & -1 & -1 & 1 \\ 1 & 0 & 1 & -1 & -1 \\ -1 & 1 & 0 & 1 & -1 \\ -1 & -1 & 1 & 0 & 1 \\ 1 & -1 & -1 & 1 & 0 \end{pmatrix}$	73.96%	81.76%	93.75%	81.39%
$\boldsymbol{M_7} = -\boldsymbol{M_6}$	73.96%	80.64%	93.75%	86.95%

*indicates non G-efficient designs as per the thumb rule stated by Wheeler (1972) and should not be used for fitting the quadratic model

**the value is almost equal to 50% and hence the design can be used for practical purposes, as suggested by Wheeler (1972)

5. Conclusion

The orthogonal arrays with index unity have been considered in our proposed OABMD algorithm. The designs, hence, constructed have the smallest number of runs for a given number of levels, thereby allowing higher cost efficiency. Furthermore, the flexibility in choice of matrix $\boldsymbol{M}$ allows for enhanced variety of design points. The manageable number of distinct design points help in reducing the cost and time in statistical experiments.

When the region of interest is pre-defined, the proposed OABMD algorithm can be customized to explore the restricted space. The constructed designs have a sufficiently high G-efficiency that make them suitable for practical purposes.

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