ISSN 0971-0388

Aligarh Journal of Statistics
Vol. 39 (2019), 53-74

F-Squares Based Efficient Uniform Designs for Mixture Experiments in Three and Four Components

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[Received on September, 2019. Accepted on March 2020]

ABSTRACT

In this paper, two classes of uniform designs for mixture experiments based on F-squares are presented. The properties of these classes of designs using different mixture models are investigated. The two classes of designs are (i) mixture designs constructed using uniform designs based on cyclic F-squares for two designs with different runs and (ii) mixture designs constructed by projecting the uniform designs based on good lattice point method based on two designs with different runs. Design efficiencies of the most uniform designs of the two classes of designs are also computed and compared.

1. Introduction

In the general mixture experimental setup, the usual constraints on the component proportions are that they are non-negative and should sum to unity. As a result, the factor space reduces to a regular $(q - 1)$ dimensional simplex

$$S_{q-1} = \left\{x : (x_1, x_2, ..., x_q) \middle| \sum_{i=1}^{q} x_i = 1, x_i \geq 0, i = 1, 2, ..., q\right\} \qquad (1.1)$$

Scheffé (1958) was the first author to introduce models and designs for experiments with mixtures. These extraneous factors do not form any portion of the mixture but their different levels could significantly affect the blending properties of the ingredients. For example, the driving speed and automobile size may affect the blending behaviours of fuels being tested to compare the average mileage of the fuels individually as well as when blended together. Scheffé (1963) introduced the problem of mixture experiments involving process variables.

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In many situations, there may be additional constraints on some or all of the factors. For instance, the factors may lie within the lower (L_i) and upper (U_i) bounds

$$0 \le L_i \le x_i \le U_i \le 1, \qquad i = 1, 2, \ldots, n \tag{1.2}$$

In such cases, the experimental region is a part of the Simplex $S_{q\text{-}1}$. For exploring the restricted region, Mclean and Anderson (1966) have developed extreme vertices designs (EVD) which satisfy both the constraints (1.1) and (1.2). Saxena and Nigam (1977) gave a transformation that provides designs constructed through symmetric simplex designs. Cornell (2002) has given an excellent review on the problem of experiments with mixtures. Various model forms for mixture experiments are suggested in literature. The following are the models considered by us in this study:

Model I: Scheffé's (1958) quadratic model

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{1 \le i < j \le q} \beta_{ij} x_i x_j \tag{1.3}$$

Model II: Scheffé's (1958) special cubic model

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{1 \le i < j \le q} \beta_{ij} x_i x_j + \sum_{1 \le i < j < k \le q} \beta_{ijk} x_i x_j x_k \tag{1.4}$$

Model III: Scheffé's (1958) full cubic model

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{1 \le i < j \le q} \beta_{ij} x_i x_j + \sum_{1 \le i < j \le q} \beta_{ij} x_i x_j (x_i - x_j) + \sum_{1 \le i < j < k \le q} \beta_{ijk} x_i x_j x_k \tag{1.5}$$

Model IV: Darroch and Waller's (1985) additive quadratic model

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q} \beta_{ii} x_i (1 - x_i) \tag{1.6}$$

Model H_i: Becker's (1968) homogeneous models of degree one

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i<j} \beta_{ij} f(x_i, x_j) + \ldots + \sum_{i_1 < i_2 < \ldots < i_n} \beta_{i_1 i_2 \ldots i_n} f(x_i, x_j, \ldots, x_{i_n}) \tag{1.7}$$

where

$$f(x_{i_1}, x_{i_2}, \ldots, x_{i_n}) = \min(x_{i_1}, x_{i_2}, \ldots, x_{i_n}) \qquad \text{for } \textit{Model } H_1$$

$$= \frac{(x_{i_1} x_{i_2} \ldots x_{i_n})}{(x_{i_1} + x_{i_2} + \ldots + x_{i_n})^{n-1}} \qquad \text{for } \textit{Model } H_2$$

$$= \left(x_{i_1} x_{i_2} \ldots\ x_{i_n}\right)^{1/n} \qquad \text{for } Model\ H_3$$

and $2 \leq n \leq q$

In H_2 if any denominator is zero, the value of corresponding term is taken to be zero. *Models I, II* and *III* are the most commonly used models in mixture experiments. *Model IV* is additive in mixture components and is suitable for the design of industrial or agricultural products where mixture components have additive effects on the response function. The models introduced by Becker (1968) are homogeneous models of degree one and are applied in different scientific areas.

Aggarwal et al. (2009) have studied mixture designs in orthogonal blocks using F-squares. Husain and Sharma (2015) obtained optimal orthogonal designs in two blocks based on F-squares for mixture inverse model in four components. In this paper, we have obtained mixture designs in three and four components by projecting the two families of designs based on good lattice point method. The uniformity measure for these designs is also calculated and tabulated. The D-, A- and G-efficiencies of these designs are also compared. We have also constructed designs for the restricted exploration of mixtures, using the transformation given by Saxena and Nigam (1977).The method has been illustrated with the help of examples.

2. F-Squares

Laywine (1989) obtained F-squares by making substitutions based on numbers for latin squares. F-squares and orthogonal F-squares are a generalization of latin squares and orthogonal latin squares, respectively. Hedayat and Seiden (1970) gave the following definition:

***Definition* 1.1:** Let **A** = $[a_{ij}]$ be an $n \times n$ matrix and let $\mathbf{\Sigma} = (c_1, c_2, \ldots, c_m)$ be the ordered set of distinct elements of **A**. In addition, suppose that for each $k = 1, 2, \ldots, m$, 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 $\mathbf{\Sigma}$ with frequency vector $(\lambda_1, \lambda_2, \ldots, \lambda_m)$ and is denoted by $F(n; \lambda_1, \lambda_2, \ldots, \lambda_m)$.

Laywine (1989) studied F-squares by making substitutions on the symbols of latin squares. For example, consider the following latin square of order 4. By substituting the symbol $d = a$ in the latin square, F (4; 2, 1, 1) defined on $\mathbf{\Sigma} = (a, b, c)$ is obtained. Aggrawal et al. (2009) denoted this F-square as FSI(4), where 4 in the parenthesis denotes the number of components.

Latin Square of order 4				FSI(4) Square number 1				FSI(4) Square number 2				FSI(4) Square number 3			
a	*b*	*c*	*d*	*a*	*B*	*c*	*a*	*a*	*c*	*a*	*b*	*a*	*a*	*b*	*c*
b	*a*	*d*	*c*	*b*	*A*	*a*	*c*	*b*	*a*	*c*	*a*	*b*	*c*	*a*	*a*
c	*d*	*a*	*b*	*c*	*A*	*a*	*b*	*c*	*a*	*b*	*a*	*c*	*b*	*a*	*a*
d	*c*	*b*	*a*	*a*	*C*	*b*	*a*	*a*	*b*	*a*	*c*	*a*	*a*	*c*	*b*

FSI(4) generates two distinct F-squares, namely Square number 2 and Square number 3 via permutations of the last three columns. Aggrawal et al. (2009) identified F-squares by simply writing down the first row of the square and represented Square number 2 by writing its first row as *a c a b*. Aggrawal et al. (2009) gave the following definitions.

***Definition* 1.2:** An F-square with the first row and first column in natural order is called a standard F-square where by natural order we mean to imply that each element is followed by the same element (if it assumes an equal proportion) or the next element cyclically. For example, for four components if $d = a$, then $\boldsymbol{\Sigma} = (a, b, c)$, the order could either be a, a, b, c or a, b, c, a.

***Definition* 1.3:** Two F-squares are equivalent if one can be derived from the other by permutations of rows and/or permutations of columns and/or permutations of elements.

***Definition* 1.4:** Two F-squares are conjugates if the rows of one are the columns of the other.

3. Uniform Designs and Uniformity Measures

Uniformity is an important concept in uniform designs. Fang and Wang (1994) have described uniform designs (UD) in which the points are scattered uniformly over the experimental domain. This is based on cyclic F-squares. The UD generated by them have smaller discrepancies than those based on the good lattice point method.

The rationale for construction of good UD is based on the following Koksma-Hlawka inequality

$$|E\{h(x)\} - h| \leq D(\mathcal{P})V(h)$$

where $V(h)$ is a measure of variation of h and $D(\mathcal{P})$ is the discrepancy of $\mathcal{P}$ which is a measure of the uniformity of $\mathcal{P}$.

Warnock (1972) gave the following analytical expression for calculating L_2-discrepancy.

$$(D_2(P))^2 = 3^{-s} - \frac{2^{1-s}}{n}\sum_{k=1}^{n}\prod_{l=1}^{s}(1-x_{kl}^2) + \frac{1}{n^2}\sum_{k=1}^{n}\sum_{j=1}^{n}\prod_{i=1}^{s}(1-max(x_{ki}, x_{ji})) \quad (3.1)$$

where, $P = \{x_1, x_2, \ldots, x_n\}$ is a set of n points in $C^s = [0,1]^s$.

Hickernell (1998) gave three modified L_2-discrepancies: the symmetric L_2-discrepancy (SD_2), the centered L_2-discrepancy (CD_2) and modified L_2-discrepancy (MD_2). These uniformity measures are described in Fang et al. (2001).

Hickernell (1998) gave an analytical expression for the centered L_2-discrepancy

$$\left(CD_2(P)\right)^2 = \left(\frac{13}{12}\right)^s - \frac{2}{n}\sum_{k=1}^{n}\prod_{l=1}^{s}\left(1+\frac{1}{2}|x_{kl}-0.5|-\frac{1}{2}|x_{kl}-0.5|^2\right) + \frac{1}{n^2}\sum_{k=1}^{n}\sum_{j=1}^{n}\prod_{i=1}^{s}\left(1+\frac{1}{2}|x_{ki}-0.5|+\frac{1}{2}\left|x_{ji}-0.5\right| - \right.$$

$$12xki-xji \quad (3.2)$$

The centered L_2-discrepancy (CD_2) considers the uniformity of P not only over C^s= [0, 1]^s but also of all the projected uniformity of P over C^u where u is a non-empty subset of the set of coordinate indices $\boldsymbol{X} = \{1, 2, \ldots, q\}$.
In this paper, we have used the centered L_2-discrepancy (CD_2) as a measure of uniformity and the minimum value of $CD_2$is desirable for F-square based uniform designs.

4. Design Efficiencies Based on Optimality Criteria

Design optimality criteria are often used to evaluate the proposed experimental design for a particular experiment of interest. The following three are the most popular design optimality criteria available in the literature where $\boldsymbol{X}$ denotes the extended design matrix.

- *D*-criterion: maximize the determinant of $\boldsymbol{X'X}$
- *A*-criterion: minimize the trace of $(\boldsymbol{X'X})^{-1}$
- *G*-criterion: minimize the maximum of the prediction variance over a specified set of design points.

In order to compare the different designs efficiencies, we use the following most commonly used design optimality measures.

$$D\text{-}eff = 100\left(\frac{|\boldsymbol{X'X}|^{1/p}}{n}\right)$$

$$A\text{-}eff = 100\left(\frac{p}{n \times trace(\boldsymbol{X}'\boldsymbol{X})^{-1}}\right)$$

$$G\text{-}eff = 100\left(\frac{p}{n \times d}\right) \quad (4.1)$$

where,

n = number of design points in the design

p = number of parameters in the model

d = max $\{\nu = \boldsymbol{X}(\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{X}'\}$ over a specified set of design points (the row vectors) $\boldsymbol{x}$ in $\boldsymbol{X}$.

In this paper, the efficiencies given is (4.1) are generated using Matlab software and are denoted here simply by *D*, *A* and *G* for the sake of notational convenience. The maximum value of *D*, *A* and minimum value of *G* are desirable.

5. Projection Designs

Prescott (2000) and Box and Hau (2001) have discussed the construction of projection designs for the cases when the design variables are subject to linear constraints. A design satisfying linear constraints has been obtained by projecting unconstrained design onto the constrained space. If q operational factors $x = \{x_i\}$, where $i = 1, 2, \ldots, q$, are subject to m constraints so that

$$\boldsymbol{C}x = \boldsymbol{c} \quad (5.1)$$

where, $\boldsymbol{C}$ is an $m \times q$ matrix and $\boldsymbol{c}$ is an $m \times 1$ column vector. Suppose $\boldsymbol{x_o}$ is the chosen origin for the levels of the experimental design then $\boldsymbol{Cx_o} = \boldsymbol{c}$. Let the region of interest be the neighborhood $x_{jo} \pm r_j$ around $\boldsymbol{x_o}$ where r_j's are some positive numbers, then the coded variables $\xi_j = \frac{x_j - x_{jo}}{ar_j}$ satisfy the constraints

$$\boldsymbol{A}x = \boldsymbol{0} \quad (5.2)$$

where, ξ is a $q \times 1$ vector of coded variables ξ_i's, $\boldsymbol{A} = (a_{ij})$ is an $m \times q$ matrix of constraints such that $a_{ij} = r_j c_{ij}$ and $\boldsymbol{0}$ is an $m \times 1$ vector of 0's and a is the number to be determined.

Let the $n \times q$ matrix $\boldsymbol{D}_z$ be that of some unconstrained generating design, and $\boldsymbol{D}_\xi$ be that of the corresponding constrained design obtained through projection to satisfy (5.2) so that

$$\boldsymbol{D}_\xi = \boldsymbol{D}_z\boldsymbol{P} \quad (5.3)$$

where, $\boldsymbol{P}$ is the $q \times q$ idempotent projection matrix satisfying

$$\boldsymbol{P} = \boldsymbol{I} - \boldsymbol{A}^T(\boldsymbol{A}\boldsymbol{A}^T)^{-1}\boldsymbol{A} \quad (5.4)$$

Then, as required

$$\boldsymbol{D}_{\xi}\boldsymbol{A}^T = \boldsymbol{D}_z\boldsymbol{P}\,\boldsymbol{A}^T = \mathbf{0} \qquad (5.5)$$

and the levels of the design $\boldsymbol{D}_X$ may be obtained from

$$x_j = ar_j\xi_j + x_{jo} \qquad (5.6)$$

where, 'a' is the number such that all the entries of $a\boldsymbol{D}_{\xi}$ are between -1 and 1.

6. Unconstrained Mixture Experiments

Aggarwal and Singh (2008) suggested a method to construct latin square based design of n runs for mixture of q components. We describe the following method to construct mixture designs through cyclic F-squares. *UF*-type designs may be obtained by selecting s linearly independent columns of cyclic F-squares (Husain and Sharma (2016)).

Method

To construct F-squares based n run mixture design in q components for *UF*-type designs.

Step 1: Choose a *UF*-type design $UF(n;q^n)$ and any $s(\leq n)$ columns of an F square form a *UF*-type design $UF(n;q^s)$.

Step 2: For a given n, there are n! left cyclic F-squares. Among all these n! left cyclic F-squares of order n, find an F-square $F = fij$ that has the smallest discrepancy using the expression given in (3.2).

Step 3: Search $s = (q\text{-}1)$ out of n columns of the F-square to form a UF-type design UF$(n;q^s)$. In all, there are ${}^{n}C_{s}$ such UF-type designs.

Step 4: From these ${}^{n}C_{s}$UF-type designs UF$(n;q^s)$, choose a design UF$_n(n^s)$ such that I thas the smallest discrepancy (as given in (3.2)) among all UF$(n;q^s)$ designs generated in Step-3. This design is nearly uniform design. We now have UF$n(n^s)$ on C^s where C = [0,1].

Step 5: Let $U = (u_{ki})$,$u_{ki} = kh_i$ (mod n),i = 1, 2, …,s; k = 1, 2, …, n be the uniform design as obtained in Step-4. Calculate $C_{ki} = (u_{ki} - 0.5) / n$ and make the transformation given in Fang and Wang (1994, p.231).

$$x_{ki} = \prod_{j=1}^{i-1} C_{kj}^{\frac{1}{q-j}}\left(1 - C_{ki}^{\frac{1}{q-i}}\right), \qquad i = 1, 2, \ldots, q\text{-}1$$

$$x_{kq} = \prod_{j=1}^{q-1} C_{kj}^{\frac{1}{q-j}}, \qquad k = 1, 2, \ldots, n. \qquad (6.1)$$

then $x_k = (x_{k1}, x_{k2}, \ldots, x_{kq})$, $k = 1, 2, \ldots, n$; is a uniform design on S_{q-1}.

Note that, the number of possible h_i is given by the Euler function $\varphi(n)$ defined by Hua (1956) as follows:

$$\varphi(n) = n \prod_{p|n} \left(1 - \frac{1}{p}\right)$$

where, p runs over the prime divisors of n. For example,

$$\varphi(9) = 9\left(1 - \frac{1}{3}\right) = 6$$

Because 9 = 3 × 3, and the possible associated h_i are 1, 2, 4, 5, 7 and 8.

In this paper, we have presented two classes of uniform designs for mixture application. We have studied properties of these classes of designs for various models. The two classes of considered designs are (i) mixture designs constructed using uniform designs based on cyclic F-squares for two designs with different runs and (ii) mixture designs constructed by projecting the uniform designs based on good lattice point method based on two designs with different runs. Consider the case n = 4. We have taken 4 = 3 as it yields centered L_2-discrepancy (CD_2), the natural order being 1 2 3 3. This sequence is denoted with last-previous (LP) value. For the other sequence, we have taken 4 = 1 as it yields centered L_2-discrepancy (CD_2), the natural order being 1 2 3 1. We have denoted this sequence with last-first (LF) value. Hence the considered designs with different runs are denoted by LP and LF.

Using the method given above, we have first obtained the most uniform designs based on cyclic F-squares. These designs are denoted here by D_{UF}. Then using step 5, we have obtained mixture designs for three and four component mixtures. These are denoted here by D_F. We have also generated mixture designs through projection of uniform designs D_{UF} as described in Section 5. So we now have two classes of designs D_F and D_{UF} for both the sequences LP and LF, respectively. The uniformity measure for each of these classes of designs is calculated using (3.2). The discrepancies CD_2 for each of the classes of mixture designs in three and four components are given in Table 1 and Table 2, respectively. The most uniform six run designs for three and four component mixtures for each of the families D_F and D_{UF} are given in Table 3 and Table 4, respectively.

From Table 1, we observe that for three component mixtures, the design $D_{UF}(LF)$ is most uniform for run sizes n = 15 for the two classes D_F and D_{UF}. Moreover, D_{UF} (LP) is most uniform for all run sizes except n = 15. All designs are most uniform for n = 4 runs. From Table 2, we observe that for four component mixtures, the design $D_{UF}(LF)$ is most uniform for run sizes n = 4 and 7 and the design $D_{UF}(LP)$ is most uniform for all run sizes except n = 4 and 7. When n = 4, the designs $D_F(LP)$ and $D_F(LF)$ are most uniform. When n = 7, the designs $D_{UF}(LP)$ and $D_{UF}(LF)$ are most uniform. The designs $D_{UF}(LP)$ and $D_{UF}(LF)$ are obtained through projection of the designs $D_F(LP)$ and $D_F(LF)$. The designs $D_F(LP)$ and $D_{UF}(LF)$ are most uniform better than the designs $D_F(LF)$ and $D_{UF}(LP)$, respectively.

Table 1: Discrepancies of the most uniform three component mixture designs.

n	***D_F(LP)***	***D_F(LF)***	***D_{UF}(LP)***	***D_{UF}(LF)***
4	0.672528	0.691977	0.621585	0.626328
5	0.680799	0.703155	0.628167	0.632778
6	0.685391	0.705232	0.631856	0.636886
7	0.690831	0.703638	0.628626	0.631139
8	0.702989	0.711591	0.641498	0.644163
9	0.698299	0.708262	0.641358	0.644810
10	0.699276	0.710290	0.633579	0.636435
11	0.688570	0.709356	0.634094	0.636934
12	0.708748	0.716759	0.639239	0.641616
13	0.705680	0.714011	0.637435	0.640063
14	0.691894	0.698163	0.634056	0.635911
15	0.702877	0.702108	0.649156	0.648736
16	0.711894	0.716383	0.645628	0.647331
17	0.707727	0.708599	0.652365	0.652775
18	0.707581	0.708261	0.652794	0.653077

19	0.712794	0.717675	0.643690	0.645387
20	0.707428	0.707763	0.648146	0.653689

Table 2: Discrepancies for most uniform four component mixture designs.

n	$D_F(LP)$	$D_F(LF)$	$D_{UF}(LP)$	$D_{UF}(LF)$
4	0.861168	0.869273	0.827342	0.824991
5	0.865524	0.875347	0.825065	0.825174
6	0.865774	0.878883	0.825161	0.825317
7	0.866703	0.878016	0.825020	0.824307
8	0.874400	0.877889	0.825441	0.825482
9	0.877626	0.884636	0.825589	0.825948
10	0.875257	0.880175	0.825236	0.825302
11	0.874308	0.879774	0.825245	0.825308
12	0.882991	0.887693	0.825711	0.825810
13	0.876571	0.881749	0.825347	0.825432
14	0.873899	0.874391	0.825328	0.825329
15	0.884629	0.886514	0.826045	0.826092
16	0.886045	0.888752	0.826175	0.826248
17	0.885668	0.889178	0.826141	0.826252
18	0.874407	0.877318	0.825825	0.825917
19	0.876212	1.745720	0.825630	0.825706
20	0.872871	0.874093	0.825864	0.825909

Table 3: The most uniform six run designs for three component mixtures.

$D_F(LP)$			$D_F(LF)$		
0.7113	0.2165	0.0722	0.7113	0.2165	0.0722
0.2362	0.4455	0.3182	0.2362	0.4455	0.3182
0.5000	0.1250	0.3750	0.5000	0.1250	0.3750
0.3545	0.1614	0.4841	0.3545	0.5917	0.0538
0.1340	0.7939	0.0722	0.1340	0.7939	0.0722
0.1340	0.3608	0.5052	0.7113	0.1203	0.1684
$D_{UF}(LP)$			$D_{UF}(LF)$		
0.5074	0.2795	0.2131	0.5074	0.2795	0.2131
0.2886	0.3850	0.3264	0.2886	0.3850	0.3264
0.4101	0.2374	0.3525	0.4101	0.2374	0.3525
0.3431	0.2542	0.4028	0.3431	0.4523	0.2046
0.2415	0.5454	0.2131	0.2415	0.5454	0.2131
0. 2415	0.3460	0.4125	0. 5074	0.2352	0.2574

Table 4: The most uniform six run designs for four component mixtures.

$D_F(LP)$				$D_F(LF)$			
0.5632	0.2184	0.0546	0.1638	0.5632	0.2184	0.0546	0.1638
0.1645	0.2962	0.1348	0.4045	0.1645	0.2962	0.4944	0.0449
0.3700	0.0844	0.5001	0.0455	0.3700	0.0844	0.5001	0.0455
0.2531	0.1001	0.2695	0.3773	0.2531	0.5313	0.0898	0.1258
0.0914	0.6463	0.1967	0.0656	0.0914	0.6463	0.1967	0.0656
0.0914	0.2146	0.4048	0.2891	0.5632	0.1032	0.1946	0.1390
$D_{UF}(LP)$				$D_{UF}(LF)$			
0.2810	0.2469	0.2306	0.2415	0.2810	0.2469	0.2306	0.2415
0.2415	0.2546	0.2386	0.2653	0.2415	0.2546	0.2742	0.2297

0.2619	0.2336	0.2748	0.2297	0.2619	0.2336	0.2748	0.2297
0.2503	0.2351	0.2519	0.2626	0.2503	0.2779	0.2341	0.2377
0.2343	0.2893	0.2447	0.2317	0.2343	0.2893	0.2447	0.2317
0.2343	0.2465	0.2653	0.2539	0.2810	0.2355	0.2445	0.2390

We have fitted *Model I* to *Model IV* to the minimum point most uniform mixture designs in three and four components for each of the two classes. *Models* H_i; i = 1, 2, 3 to the minimum point most uniform mixture designs in three components and *Models* H_i; i = 2, 3 to the minimum point most uniform mixture designs in four components for each of the two classes *LP* and *LF*, respectively. The design efficiencies D, A and G are as given in Table 5 and Table 6, respectively.

From Table 5, we observe that for three component mixtures, the designs generated from D_F(*LP*) and D_F(*LF*) are in general more efficient than the designs generated from D_{UF}(*LP*) and D_{UF}(*LF*) for all *Model I* to *IV* and *Models* H_i; i = 1, 2, 3 as regards D-efficiency. As regards A-efficiency, for *Model II* to *IV*, the designs generated from D_F(*LP*) and D_F(*LF*) are more efficient than the designs generated from D_{UF}(*LP*) and D_{UF}(*LF*) and in other models, the designs generated from D_{UF}(*LP*) and D_{UF}(*LF*) are more efficient than the designs generated from D_F(*LP*) and D_F(*LF*). From Table 6, we observe that the designs generated from D_{UF}(*LP*) and D_{UF}(*LF*) are in general more efficient than the designs generated from D_F(*LP*) and D_F(*LF*). Designs based on D_{UF}(*LP*) is better in terms of efficiencies for *Model I*, *III* and H_2. Designs based on D_{UF}(*LF*) is better in terms of efficiencies for *Model II*. Designs based on D_F(*LP*) are better for *Model IV* in terms of D-efficiency.

Table 5: Efficiencies of the minimum point uniform mixture designs for three components.

Model	***p***	***$D_F(LP)$***			***$D_F(LF)$***			***$D_{UF}(LP)$***			***$D_{UF}(LF)$***		
		D-eff	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***
I	6	9.7375E-06	1.2782E-15	1.5E-12	4.42975E-06	1.4154E-16	1.5E-14	3.4636E-06	3.8078E-15	1.5E-13	2.552E-06	7.8113E-16	1.5E-12
II	7	1.8645E-07	1.7255E-16	1.75E-13	1.1053E-07	3.2596E-18	1.75E-13	8.4310E-08	1.3334E-16	1.75E-12	5.5437E-08	3.7433E-17	1.75E-13
III	10	2.4114E-10	4.3806E-17	2.5E-14	2.4021E-10	1.1599E-14	2.5E-14	9.7349E-11	2.7202E-17	2.5E-13	1.1219E-10	6.2470E-18	2.5E-13
IV	6	8.6172E-06	8.9648E-15	1.5E-13	1.2528E-05	8.6331E-16	1.5E-13	2.8269E-06	1.1255E-15	1.5E-13	4.5387E-06	1.1724E-14	1.5E-13
$H_1(r=3)$	7	4.3705E-07	1.4747E-16	1.675E-13	2.4378E-07	4.6357E-17	1.675E-13	2.9958E-07	7.4569E-16	1.675E-12	2.7274E-07	1.8211E-14	1.675E-14
$H_2(r=3)$	7	2.4565E-07	1.1828E-16	1.675E-12	1.2875E-07	8.4545E-17	1.675E-12	7.3837E-08	7.8328E-16	1.675E-11	5.1464E-08	1.0229E-16	1.675E-12
$H_3(r=3)$	7	6.1651E-07	1.2793E-15	1.675E-12	5.2911E-07	7.5107E-16	1.675E-13	1.1305E-07	2.6181E-15	1.675E-12	2.7114E-07	2.1387E-15	1.675E-12

Table 6: Efficiencies of the minimum point uniform mixture designs for four components.

Model	***p***	***$D_F(LP)$***			***$D_F(LF)$***			***$D_{UF}(LP)$***			***$D_{UF}(LF)$***		
		D-eff	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***
I	10	4.9095E-07	3.9135E-16	1.6667E-13	2.7296E-07	8.2451E-16	1.6667E-13	9.054E-07	2.2291E-16	1.6667E-13	1.0433E-08	7.2376E-16	1.6667E-13
II	14	1.7115E-12	7.1987E-16	3.5E-14	1.3150E-12	7.1987E-16	3.5E-14	1.8794E-10	4.1135E-16	2E-13	2.7359E-11	4.1135E-16	2E-14
III	20	4.9714E-14	1.0284E-15	5E-14	3.6525E-14	1.0284E-15	5E-14	9.1968E-13	5.8765E-16	2.8571E-13	1.7064E-13	2.7822E-19	3.3333E-14
IV	8	0.00016838	2.1635E-16	1.3333E-14	3.7762E-08	4.1135E-16	2E-13	2.3483E-06	3.0953E-16	1.3333E-13	2.3588E-05	2.3506E-16	1.1428E-05
$H_2(r=3)$	14	4.2878E-12	7.1987E-16	3.5E-13	3.2031E-10	7.1987E-16	3.5E-13	3.5250E-10	4.1135E-16	4.1135E-16	6.3163E-11	2.691E-15	2.3333E-13
$H_3(r=3)$	14	4.8407E-09	7.1987E-16	2.3333E-13	2.2733E-09	1.3990E-15	2.3333E-13	3.7487E-10	4.1135E-16	2.3333E-13	2.6706E-10	4.1135E-16	2E-11

7. Restricted Exploration of Mixtures

For restricted exploration of mixtures i.e. when (1.2) is satisfied, Saxena and Nigam (1977) have given a transformation which provides designs constructed through symmetric simplex designs. Aggarwal and Singh (2008) suggested the following steps to generate projections designs.

Step 1: Rank the components in order of their increasing ranges ($U_i - L_i$). x_1 has the smallest range and x_q has the largest range.

Step 2: Consider a mixture design Z satisfying (1.1). This can be selected from the four classes of designs obtained in Section 5.

Step 3: Compute B and B', the minimum and maximum proportions of any component Z_i in the design so that $0 \le B \le Z_i \le B' \le 1$ for all Z_i.

Step 4: Make the transformation as given by Saxena and Nigam (1977) i.e.,

$$x_{iu} = \lambda_i + \mu_i z_{iu}, \qquad i = 1, 2, \ldots, t; \quad u = 1, 2, \ldots, n$$

where

$$\lambda_i = \frac{L_i B' - U_i B}{B' - B} \qquad \textit{and} \qquad \mu_i = \frac{U_i - L_i}{B' - B}$$

and

$$x_{iu} = \frac{\left(1 - \sum_{h=1}^{t} x_{hu}\right)}{\left(1 - \sum_{h=1}^{t} z_{hu}\right)} z_{iu} \qquad i = t+1, \ldots, q; \quad u = 1, 2, \ldots, n$$

where $t \le (q\text{-}1)$ is the number of components constrained by (1.2).

When all the components are constrained by (1.2), then the levels of x_q may be obtained by $x_q = 1 - (x_1 + x_2 + \ldots + x_{q-1})$.

Step 5: While determining the value of x_q in Step-4, if any point x_q lies outside the range $L_q \le x_q \le U_q$, it may be adjusted by setting x_q equal to the violated bound and adjusting the level of x_{q-1} so that (1.2) is satisfied.

Step 6: The design points from Step-4 combined with different combinations of adjusted points result in a number of designs. The design that is most uniform and optimal with certain optimality criteria is taken as the best design.

The steps given above are illustrated with the help of examples for three and four component mixtures based on F-squares.

Example 1: Let us first consider a three component example taken from Snee and Marquardt (1974) with components ranked in order of their increasing ranges.

$$0.1 \le x_1 \le 0.6$$
$$0.1 \le x_2 \le 0.7$$

$$0.0 \leq x_3 \leq 0.7$$

Using Steps 1 to 4, we obtain designs with different run sizes for each of the two considered classes. The discrepancies of these designs are calculated for different run sizes and are given in Table 7. Table 8 presents the six run uniform designs based on designs given in Table 3.

Table 7: Discrepancies of most uniform three component designs for Example 1.

1	D_F(LP)	D_F(LF)	D_{UF}(LP)	D_{UF}(LF)
4	0.681488	0.672802	0.616094	0.616212
5	0.680474	0.660865	0.616276	0.615941
6	0.659181	0.660720	0.615957	0.616017
7	0.664419	0.666565	0.615895	0.615682
8	0.666437	0.658828	0.616133	0.615996
9	0.662670	0.654420	0.616009	0.615910
10	0.661016	0.663801	0.615732	0.615798
11	0.662063	0.672179	0.615694	0.615697
12	0.669371	0.666703	0.616244	0.616067
13	0.666600	0.664524	0.615912	0.615806
14	0.653625	0.652520	0.615572	0.615567
15	0.657551	0.652834	0.616093	0.615860
16	0.665891	0.663251	0.616098	0.616029
17	0.662207	0.655734	0.616225	0.615988
18	0.662159	0.655818	0.616187	0.615928
19	0.667399	0.665513	0.616057	0.616043
20	0.660461	0.656061	0.616198	0.616054

Table 8: The most uniform six run designs for three component mixtures.

$D_F(LP)$			$D_F(LF)$		
0.5409	0.2332	0.2259	0.5417	0.2393	0.2190
0.2242	0.4164	0.3594	0.2291	0.4202	0.3507
0.4000	0.1600	0.4400	0.4026	0.1671	0.4303
0.3030	0.1891	0.5079	0.3069	0.5356	0.1575
0.1560	0.6951	0.1489	0.1618	0.6951	0.1430
0.1560	0.3487	0.4953	0.5417	0.1634	0.2950
$D_{UF}(LP)$			$D_{UF}(LF)$		
0.3584	0.3213	0.3204	0.3585	0.3220	0.3195
0.3202	0.3434	0.3365	0.3208	0.3438	0.3354
0.5414	0.3124	0.3462	0.3417	0.3133	0.3450
0.3297	0.3159	0.3544	0.3301	0.3577	0.3121
0.3119	0.3770	0.3111	0.3127	0.3770	0.3104
0.3119	0.3352	0.3529	0. 3585	0.3128	0.3287

From Table 7, we observe that for Example 1 when n = 4, 6, 10 and 11, $D_{UF}(LP)$ is most uniform and $D_{UF}(LF)$ is most uniform for n = 5, 7, 8, 9, 12, 13, 14, 15, 16, 17, 18, 19 and 20. For n = 6, 7, 10 and 11, $D_F(LP)$ is most uniform and $D_F(LF)$ is most uniform for n = 4, 5, 8, 9, 12, 13, 14, 15, 16, 17, 18, 19 and 20.

We have fitted *Model I* to *Model IV* and *Models* H_i; i = 1, 2, 3 to the minimum point most uniform mixture designs in three components for each of the four classes. The design efficiencies D, A and G are given in Table 11.

Example 2: Let us now consider a four component example taken from Snee (1975) with components ranked in order of their increasing ranges.

$$0.07 \le x_1 \le 0.18$$
$$0.00 \le x_2 \le 0.15$$
$$0.00 \le x_3 \le 0.30$$
$$0.37 \le x_4 \le 0.70$$

Using Steps 1 to 4, we obtain designs with different run sizes for each of the four classes. The discrepancies of these designs are calculated for different run sizes and are given in Table 9. Table 10 presents the six run uniform designs based on designs given in Table 4.

Table 9: Discrepancies of most uniform four component designs for Example 2.

n	$D_F(LP)$	$D_F(LF)$	$D_{UF}(LP)$	$D_{UF}(LF)$
4	0.934520	0.905616	0.827773	0.826239
5	0.923403	0.912608	0.827227	0.826572
6	0.917171	0.918592	0.826819	0.826915
7	0.918128	0.912089	0.827080	0.826632
8	0.927806	0.922770	0.827196	0.827245
9	0.937198	0.929950	0.828361	0.827862
10	0.924913	0.920287	0.827630	0.827409
11	0.923215	0.918763	0.827208	0.826723
12	0.933938	0.929168	0.828241	0.827850
13	0.922307	0.919754	0.827224	0.827026
14	0.929262	0.924009	0.827877	0.827445
15	0.934432	0.931890	0.828199	0.827787
16	0.938489	0.934687	0.828649	0.828062
17	0.934953	0.930937	0.828237	0.829316
18	0.933185	0.930975	0.827793	0.827632
19	0.930816	0.932483	0.827756	0.827501
20	0.934301	0.930737	0.827856	0.827671

Table 10: The most uniform six run designs for four component mixtures.

$D_F(LP)$				$D_F(LF)$			
0.1788	0.0503	0.0075	0.7634	0.1643	0.0437	0.0066	0.7854
0.0956	0.0724	0.0531	0.7788	0.0923	0.0628	0.2233	0.6216
0.1385	0.0122	0.2609	0.5884	0.1294	0.0106	0.2261	0.6338
0.1141	0.0167	0.1297	0.7395	0.1083	0.1208	0.0240	0.7470
0.0804	0.1720	0.0883	0.6593	0.0791	0.1491	0.0766	0.6952
0.0804	0.0492	0.2066	0.6637	0.1643	0.0153	0.0756	0.7448

$D_{UF}(LP)$				$D_{UF}(LF)$			
0.2406	0.2236	0.2179	0.3179	0.2385	0.2224	0.2174	0.3216
0.2296	0.2265	0.2240	0.3199	0.2289	0.2250	0.2464	0.2997
0.2353	0.2186	0.2514	0.2947	0.2339	0.2180	0.2468	0.3014
0.2320	0.2192	0.2341	0.3147	0.2310	0.2327	0.2198	0.3165
0.2276	0.2397	0.2286	0.3041	0.2271	0.2365	0.2268	0.3096
0.2276	0.2235	0.2443	0.3047	0.2385	0.2186	0.2267	0.3162

From Table 9, we observe that for Example 2 when $n = 4$, the designs $D_F(LF)$ and $D_{UF}(LF)$ are most uniform. For $n = 6$, the designs $D_F(LP)$ and $D_{UF}(LP)$ are most uniform. We also observe that for four component mixture designs D_F and D_{UF} with this example are most uniform for all runs as compared to Aggarwal and Singh (2008).

We have fitted *Model I* to *Model IV* and *Models* H_i; $i = 2$ and 3 to the minimum point most uniform mixture designs in four components for each of the four classes. The design efficiencies D, A and G are as given in Table 12.

From Table 11, we observe that the designs generated from $D_F(LP)$ is better in terms of D- efficiency for *Model II*, *III*, H_2 and H_3 while $D_F(LF)$ is better for *Model I*, *IV* and H_1. The designs generated from $D_F(LP)$ is better in terms of A-efficiency for *Model III* and H_2 and $D_F(LF)$ is better for *Model I*, *II*, *IV*, H_1 and H_3. The designs generated from $D_F(LP)$ are better in terms of G-efficiency for *Model* H_2, $D_F(LF)$ is better for *Model II*, $D_{UF}(LP)$ is better for *Model III*, H_1 and H_3 while $D_{UF}(LF)$ is better for *Model I* and *IV*.

From Table 12, we observe that the designs generated from $D_F(LP)$ and $D_F(LF)$ are in general more efficient than the designs generated from $D_{UF}(LP)$ and $D_{UF}(LF)$. Designs based on $D_{UF}(LP)$ is better in terms of efficiencies for *Model III* and H_2. Designs based on $D_F(LP)$ is better in terms of efficiencies for *Model I*, *II*, *IV* and H_3. Designs based on $D_{UF}(LP)$ is better for *Model III* and H_2 as regards D-efficiency.

Table 11: Efficiencies of the minimum point uniform mixture designs for three components in Example 1.

Model	***p***	***$D_F(LP)$***			***$D_F(LF)$***			***$D_{UF}(LP)$***			***$D_{UF}(LF)$***		
		D-eff	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***
I	6	0.16350	0.00110	46.7720	0.1861	0.00323	49.7471	0.411E-3	0.43E-6	49.8629	0.324E-3	0.422E-6	53.1726
II	7	0.04740	0.162E-4	54.4781	0.0471	0.742E-4	58.0450	0.742E-4	0.362E-7	53.2255	0.766E-4	0.7694E-7	54.3951
III	10	0.00440	0.716E-5	71.4357	0.0043	0.702E-5	71.4357	0.519E-5	0.114E-7	78.7352	0.522E-5	0.110E-7	67.4554
IV	6	0.20561	0.00243	46.7057	0.2342	0.00784	49.7125	0.00032	0.346E-6	49.0637	0.361E-4	0.556E-6	53.9016
$H_1(r=3)$	7	0.41792	0.01543	58.1531	0.4830	0.04021	52.4329	0.00611	0.882E-4	58.1666	0.00712	0.255E-4	52.5045
$H_2(r=3)$	7	0.04593	0.521E-4	54.9390	0.0436	0.101E-4	54.0482	0.561E-4	0.414E-7	53.8039	0.702E-4	0.205E-7	53.9084
$H_3(r=3)$	7	0.06651	0.196E-4	54.2299	0.0622	0.439E-4	56.1987	0.892E-4	0.783E-7	260.8242	0.961E-4	0.921E-7	50.3221

Table 12: Efficiencies of the minimum point uniform mixture designs for four components in Example 2.

Model	***p***	***$D_F(LP)$***			***$D_F(LF)$***			***$D_{UF}(LP)$***			***$D_{UF}(LF)$***		
		D-eff	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***	***D-eff***	***A-eff***	***G-eff***
I	10	4.47176E-08	1.03244E-17	1.66667E-14	9.64023E-11	3.56379E-17	2.5E-13	7.6344E-09	1.38819E-15	1.6667E-12	5.58969E-11	3.34685E-16	2.5E-13
II	14	2.34133E-11	1.70428E-19	2.33333E-13	4.19051E-13	1.43056E-18	3.5E-14	2.23283E-11	1.27588E-17	2.3333E-13	5.7071E-13	1.55039E-17	3.5E-13
III	20	2.55634E-13	2.19775E-19	3.33333E-15	1.64889E-13	5.17507E-18	5.0E-14	5.19251E-13	5.9459E-18	3.3333E-13	2.26439E-14	3.49895E-20	5E-14
IV	8	1.1314E-05	6.24132E-16	1.33333E-13	1.00699E-08	4.02325E-16	2.0E-14	2.47232E-06	4.85838E-15	1.3333E-12	4.32156E-09	1.81587E-15	2E-13
$H_2(r=3)$	14	9.44261E-11	2.94449E-18	2.33333E-14	1.15283E-12	3.83852E-18	3.5E-13	1.70278E-10	9.13099E-16	2.3333E-13	1.55869E-12	4.89052E-17	3.5E-13
$H_3(r=3)$	14	9.68000E-10	2.03553E-16	2.33333E-14	1.35677E-11	7.30628E-16	3.5E-13	1.83134E-10	2.002E-16	2.3333E-15	1.00185E-11	6.37616E-15	3.5E-13

8. Conclusions

For three and four components, D-, A- and G-efficient uniform designs for mixture experiments based on latin squares for Scheffe's quadratic model (1958), Darroch and Waller's (1985) model and Becker's (1968) model were obtained by Aggarwal and Singh (2008). In this paper, we have found the centered L_2-discrepancy with three and four components for the considered classes of designs based on F-squares. The design $D_{UF}(LF)$ is most uniform for run sizes $n = 4$ and 7 in three components. When $n = 4$, all the designs are most uniform for three components. The designs $D_{UF}(LP)$ and $D_{UF}(LF)$ are obtained through projection of the designs $D_F(LP)$ and $D_F(LF)$ based on four components. The designs $D_F(LP)$ and $D_{UF}(LF)$ are most uniform better than the designs $D_F(LF)$ and $D_{UF}(LP)$ for four components.

In this paper, we have computed the D-, A- and G- efficiencies of three and four component mixture experiments based on F-squares for Scheffe's (1958) quadratic model, Darroch and Waller's (1985) model and Becker's (1968) model. For three components, the designs generated from $D_F(LP)$ is better in terms of D-efficiency for *Model II*, *III*, H_2 and H_3 and $D_F(LF)$ is better in terms of D-efficiency for *Model I*, *IV* and H_1. The designs generated from $D_F(LP)$ is better in terms of A- efficiency for *Model III* and H_2 and $D_F(LF)$ is better for *Model I*, *II*, *IV*, H_1 and H_3. The designs generated from $D_F(LP)$ is better in terms of G-efficiency for *Model* H_2, $D_F(LF)$ is better for *Model II*, $D_{UF}(LP)$ is better for *Model III*, H_1 and H_3 and $D_{UF}(LF)$ is better for *Model I* and *IV*. For four components, the designs generated from $D_F(LP)$ and $D_F(LF)$ are in general more efficient than the designs generated from $D_{UF}(LP)$ and $D_{UF}(LF)$. Designs based on $D_{UF}(LP)$ is better in terms of D- efficiency for *Model III* and H_2. Designs based on $D_F(LP)$ is better in terms of D- efficiency for *Model I*, *II*, *IV* and H_3. Designs based on $D_{UF}(LP)$ is better for *Model III* and H_2 as regards D-efficiency.

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