Axis-Slope-Rotatable Designs for Experiments with Mixtures

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ABSTACT

A new design concept, called axis-slope-rotatability, is presented for the design of experiments with mixtures. This is an analogue of the Box-Hunter(1957) rotatability for second order response surface designs. By choice of design, it is possible to make the variance of the estimated slopes along the component axes constant for all axial points equidistant from the center point of the factor space. This property is called axis-slope-rotatability for mixture experiments. When the Scheffe's second degree polynomial is used, it is shown that some symmetry conditions are sufficient for axis-slope-rotatability. Several designs having this property are illustrated.

KEY WORDS : Slope, Rotatability, Slope-rotatability, Axis-slope-rotatability, Mixture experiments

1. INTRODUCTION

Unlike the usual response suface problems where the concomitant variables represent quantitative amounts, in the mixture problems the commponents represent proportions of a mixture. In a q component mixture, if x_i is the proportion of the ith component ($i=1, 2, \cdots, q$), then

$$x_i \geq 0 \ (1 \leq i \leq q), \ \sum_{i=1}^{q} x_i = 1. \tag{1.1}$$

By virtue of the above restriction, the factor space containing the q components is a $(q-1)$-dimensional regular simplex.

A literature survey by Cornell (1979) for experiments with mixtures reveals that a great deal of attention has been recently focused on the design aspects. They are Murty and Das(1968), Thompson and Myers(1968), Saxena and Nigam(1973), Snee and

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Marquardt(1974, 1976), Snee(1975a), Galil and Kiefer(1977a, 1977b) and others. Also measuring the slope of the response surface along the component axes has been of interest to some researchers such as Cornell and Ott(1975) and Snee(1975b). In the context of such development in the experiments of mixtures, it is the purpose of this paper to present a new design concept which is an anlogue of the slope-rotatability property suggested by Hader and Park (1978). The slope rotatability property is, in turn, an analogue of the rotatabilty property originally suggested by Box and Hunter(1957).

The essence of Box and Hunter's rotatability is that by imposing certain restrictions on the moment matrix of the design it is possible to have equally reliable(same variance) estimates of the true response for all combinations of the independent variables equidistant from the design center. Similarly when the object is estimation of the derivative with respect to each independent variable, it is possible to find designs for which the derivative estimates are equally reliable at all combinations of the variables equidistant from the design center. such design property is called slope-rotatability by Hader and Park(1978).

When studying the shape of the surface in the mixture experiments, often it would be desirable to have the predicted response possess constant variance at points which are equidistant from the center of the design(Cornell and Khury(1979)). We also feel that it would be desirable to make the estimated slope along the component axis equally reliable at points which are equidistant from the center of the design. Along the axis of an individual component, say the ith component, the slope of the sunface reflects how quickly the response is changing relative to the proportion x_i of the ith component and the corresponding proportions $x_j=(1-x_i)/(q-1)$ of the other $q-1$ components. At a specific point on the axis, the slope of the response surface provides a measure of proximity of the location of the maximum (or minimum) of the surface. Hence, it is necessary to obtain the estimates of the slopes along the component axes which may be of great help in an attempt to uncover the characteristics of the mixture response surface under study.

Certain restrictions on the moment matrix of the design are suggested so that the variances of the estimated slopes along the component axes be equal at the axial points equidistant from the center point of the $(q-1)$-dimensional simplex. If we use the restrictions to construct a design, then the estimates of the slopes will be equally reliable for all axial points equidistant from the center point. We shall call this property as "axis-slope-rotability."

2. SUFFICIENT CONDITIONS FOR AXIS-SLOPE-ROTATABILITY

The general form of the second degree Scheffe's polynomial in q components is

$$\eta=\sum_{1\le i\le q}\beta_i x_i+\sum\sum_{1\le i<j\le q}\beta_{ij}x_i x_j. \tag{2.1}$$

An expression for the slope of the response surface with respect to the ith component can be obtained by first writing (2.1) as a function only of the proportion x_i. On the axis of the ith component $x_j=(1-x_i)/(q-1)$ for all $j\neq i$, and substituting this expression for x_j into (2.1), the expected response is

$$\eta=\beta_i x_i+\sum_{\substack{j=1\\ j\neq i}}^{q}\beta_j\frac{(1-x_i)}{q-1}+\sum_{l=1}^{i-1}\beta_{li}\frac{(1-x_i)x_i}{q-1}$$

$$+\sum_{j=i+1}^{q}\beta_{ij}x_i\frac{(1-x_i)}{q-1}+\sum\sum_{\substack{j<k\\ j,k\neq i}}^{q}\beta_{jk}\frac{(1-x_i)^2}{(q-1)^2}.$$

Then at proportion x_i on the axis of the ith component, the slope of the expected response with respect to the ith component is

$$\frac{\partial\eta}{\partial x_i}=\gamma_{0i}+\gamma_{1i}x_i$$

where

$$\gamma_{0i}=\frac{1}{q-1}[(q-1)\ \beta_i-\sum_{\substack{j=1\\ j\neq i}}^{q}\beta_j+\sum_{l=1}^{i-1}\beta_{li}$$

$$+\sum_{j=i+1}^{q}\beta_{ij}-2\sum\sum_{\substack{j<k\\ j,k\neq i}}^{q}\frac{\beta_{jk}}{q-1}] \tag{2.2}$$

and

$$\gamma_{1i}=\frac{2}{q-1}\left[\sum\sum_{\substack{j<k\\ j,k\neq i}}^{q}\frac{\beta_{jk}}{q-1}-\sum_{l=1}^{i-1}\beta_{li}-\sum_{j=i+1}^{q}\beta_{ij}\right]. \tag{2.3}$$

If we substitute b_i and b_{ij}, the least squares estimates of β_i and β_{ij}, into (2.2) and (2.3), we obtain $\hat{\gamma}_{0i}$ and $\hat{\gamma}_{1i}$, the estimates of γ_{01} and γ_{1i}, and the following estimate of the slope

$$\frac{\partial\hat{y}}{\partial x_i}=\hat{\gamma}_{0i}+\hat{\gamma}_{1i}x_i.$$

Hence the variance of the estimated slope is,

$$\mathrm{Var}\left(\frac{\partial \hat{y}}{\partial x_i}\right)=\mathrm{Var}(\hat{\gamma}_{0i})+2\mathrm{Cov}(\hat{\gamma}_{0i}, \hat{\gamma}_{1i})x_i+\mathrm{Var}(\hat{\gamma}_{1i})x_i^2. \tag{2.4}$$

Theorem: Suppose the Scheffe's second degree polynomial (2. 1) is adequate to represent the response relationship in a mixture experiment and the responses are distributed independently with equal variance σ^2. Then the following symmetry restrictions on the design moments are sufficient conditions for axis-slope-rotatability.

$$\begin{aligned}
&\sum_u x_{iu}^2 &&=\text{Constant}\\
&\sum_u x_{iu}x_{ju} &&=\text{Constant}\\
&\sum_u x_{iu}^2 x_{ju} &&=\text{Constant}\\
&\sum_u x_{iu}x_{ju}x_{ku} &&=\text{Constant}\\
&\sum_u x_{iu}^2 x_{ju}^2 &&=\text{Constant}\\
&\sum_u x_{iu}^2 x_{ju}x_{ku} &&=\text{Constant}\\
&\sum_u x_{iu}x_{ju}x_{ku}x_{lu} &&=\text{Constant}
\end{aligned} \tag{2.5}$$

for all i, j, k and $l(i \neq j \neq k \neq l)$ ranging over the columns of the design matrix, the summation on u being over all the design points.

3. EXAMPLES OF AXIS-SLOPE-ROTATABLE DESIGNS

In this section we will illustrate some axis-slope-rotatable designs.

CASE 1: The symmetric simplex designs suggested by Murty and Das(1968) are axis-slope-rotatable, since the symmetry conditions of Murty and Das satisfy the restrictions (2. 5) on the design moments.

CASE 2 : The simplex lattice designs and the simplex centroid designs proposed by Scheffe(1958, 1963) satisfy the restrictions in (2. 5). Hence, these designs are axis-slope-rotatable.

CASE 3 : This case shows an axis-slope-rotatable design which is neither a simplex lattice design nor a simplex centroid design. Consider Figure 1 which contains the following 13 points.

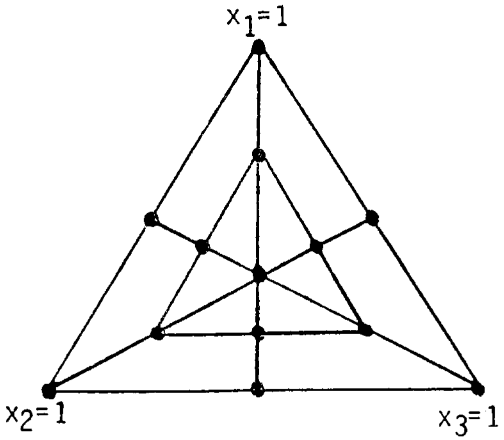
(1, 0, 0), (0, 1, 0), (0, 0, 1)

$$\left(\frac{1}{2}, \frac{1}{2}, 0\right), \left(\frac{1}{2}, 0, \frac{1}{2}\right), \left(0, \frac{1}{2}, \frac{1}{2}\right)$$

$$\left(\frac{1}{3}, \frac{1}{3}, \frac{1}{3}\right)$$

$$\left(\frac{2}{3}, \frac{1}{6}, \frac{1}{6}\right)\left(\frac{1}{6}, \frac{2}{3}, \frac{1}{6}\right)\left(\frac{1}{6}, \frac{1}{6}, \frac{2}{3}\right)$$

$$\left(\frac{1}{6}, \frac{5}{12}, \frac{5}{12}\right)\left(\frac{5}{12}, \frac{1}{6}, \frac{5}{12}\right)\left(\frac{5}{12}, \frac{5}{12}, \frac{1}{6}\right)$$



Figuer 1. **An axis-slope-rotatable design**

The values of moments are

$$\sum_{u=1}^{13} x_{iu}^2 = 2.4861 \quad i=1, 2, 3$$

$$\sum_{u=1}^{13} x_{iu}x_{ju} = 0.9236 \quad i \neq j=1, 2, 3$$

$$\sum_{u=1}^{13} x_{iu}^2 x_{ju} = 0.3721 \quad i \neq j=1, 2, 3$$

$$\sum_{u=1}^{13} x_{1u}x_{2u}x_{3u} = 0.1794$$

$$\sum_{u=1}^{13} x_{iu}^2 x_{ju}^2 = 0.1401 \quad i \neq j=1, 2, 3$$

$$\sum_{u=1}^{13} x_{iu}^2 x_{ju}x_{ku} = 0.0598 \quad i \neq j \neq k=1, 2, 3$$

We can see that the design satisfies the conditions (2. 5). Hence, it is an axis-slope-rotatable design. This design is obviously neither a simplex lattice design nor a simplex centroid design.

CASE 4 : For the three component mixture experiment, if θ is the angle between the line parallel to the line $x_1=0$ and the line which connects a point in the simplex and the centroid, we can express the coordinates of the point in terms of θ and ρ, where ρ is the distance from the centroid (Figure 2). That is

$$x_1=\frac{1}{3}+\frac{2}{\sqrt{6}}\rho\sin\theta$$

$$x_2=\frac{1}{3}-\frac{1}{\sqrt{2}}\rho\cos\theta-\frac{1}{\sqrt{6}}\rho\sin\theta$$

$$x_3=\frac{1}{3}+\frac{1}{\sqrt{2}}\rho\cos\theta-\frac{1}{\sqrt{6}}\rho\sin\theta$$

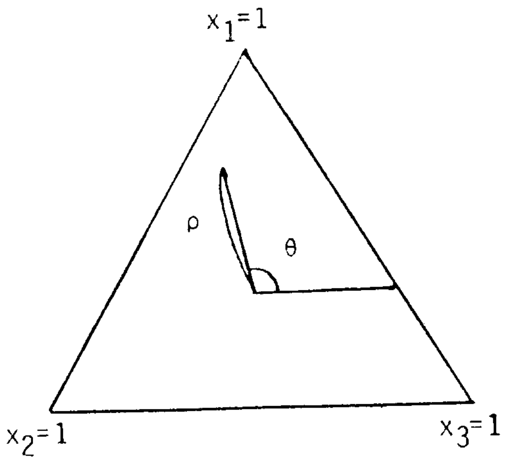


Figure 2.

Hence, if we arrange n points $(n\geq 5)$ equally spaced on a circle centered at the centroid with radius ρ, coordinates of each point are

$$x_{1k}=\frac{1}{3}+\frac{2}{\sqrt{6}}\rho\sin\left(\theta+\frac{2k\pi}{n}\right)$$

$$x_{2k}=\frac{1}{3}-\frac{1}{\sqrt{2}}\rho\cos\left(\theta+\frac{2k\pi}{n}\right)-\frac{1}{\sqrt{6}}\rho\sin\left(\theta+\frac{2k\pi}{n}\right)$$

$$x_{3k}=\frac{1}{3}+\frac{1}{\sqrt{2}}\rho\cos\left(\theta+\frac{2k\pi}{n}\right)-\frac{1}{\sqrt{6}}\rho\sin\left(\theta+\frac{2k\pi}{n}\right) \quad (3.1)$$

$k=0,\ 1, 2,\ \cdots,\ n-1$

Using the equalities,

$$\sum_{k=0}^{n-1} \sin\left(\frac{2k\pi}{n}\right)=\sum_{k=0}^{n-1} \cos\left(\frac{2k\pi}{n}\right)=0 \qquad n\geq 2$$

$$\sum_{k=0}^{n-1} \sin\left(\frac{4k\pi}{n}\right)=\sum_{k=0}^{n-1} \cos\left(\frac{4k\pi}{n}\right)=0 \qquad n\geq 3$$

$$\sum_{k=0}^{n-1} \sin\left(\frac{6k\pi}{n}\right)=\sum_{k=0}^{n-1} \cos\left(\frac{6k\pi}{n}\right)=0 \qquad n\geq 4$$

$$\sum_{k=0}^{n-1} \sin\left(\frac{8k\pi}{n}\right)=\sum_{k=0}^{n-1} \cos\left(\frac{8k\pi}{n}\right)=0 \qquad n\geq 5$$

it can be shown that

$$\begin{aligned}
&\sum_{k=0}^{n-1} x_{ik}^2 &&=\frac{n}{9}+\frac{n}{3}\rho^2 && i=1,2,3\\
&\sum_{k=0}^{n-1} x_{ik}x_{jk} &&=\frac{n}{9}-\frac{n}{6}\rho^2 && i\neq j=1,2,3\\
&\sum_{k=0}^{n-1} x_{1k}x_{2k}x_{3k} &&=\frac{n}{27}-\frac{n}{6}\rho^2 &&\\
&\sum_{k=0}^{n-1} x_{ik}^2x_{jk} &&=\frac{n}{27} && i\neq j=1,2,3\\
&\sum_{k=0}^{n-1} x_{ik}^2x_{jk}^2 &&=\frac{n}{81}+\frac{n}{12}\rho^4 && i\neq j=1,2,3\\
&\sum_{k=0}^{n-1} x_{ik}^2x_{jk}x_{lk} &&=\frac{n}{81}-\frac{n}{18}\rho^2 && i\neq j\neq l=1,2,3
\end{aligned} \tag{3.2}$$

Therefore, a design with n points ($n\geq 5$) equally spaced on a circle centered at the centroid is axis-slope-rotatable.

Consider a design which consists of equally spaced n_1 points on a circle centered at the centroid and equally spaced n_2 points on another circle, etc ($n_i\geq 5$), then we can see from (3.2) that such designs satisfy the conditions (2.5) and are also axis-slope-rotatable designs. For example, consider the following with 12 points.

i) Five points are obtained by substituting $n=5$, $\theta=0$, $\rho=\frac{1}{\sqrt{6}}$ into (3.1)
(0.3333, 0.0447, 0.6220), (0.6504, 0.0856, 0.2640) (0.5293, 0.4689, 0.0018), (0.0163, 0.4026, 0.5810) (0.1374, 0.6648, 0.1978).

ii) Three points are obtained by substituting $n=3$, $\theta=\frac{\pi}{2}$, $\rho=\sqrt{6}/4$ into (3.1)
(0.8333, 0.0833, 0.0833), (0.0833, 0.8333, 0.0833) (0.0833, 0.0833, 0.8333)

iii) Three points are obtained by substituting $n=3$, $\theta=\frac{\pi}{6}$, $\rho=\sqrt{6}/12$ into (3.1)
(0.1666, 0.4167, 0.4167), (0.4167, 0.1666, 0.4167) (0.4167, 0.4167, 0.1666)

iv) Centroid.

(0.3333, 0.3333, 0.3333)

These 12 points are displayed as in Figure 3.

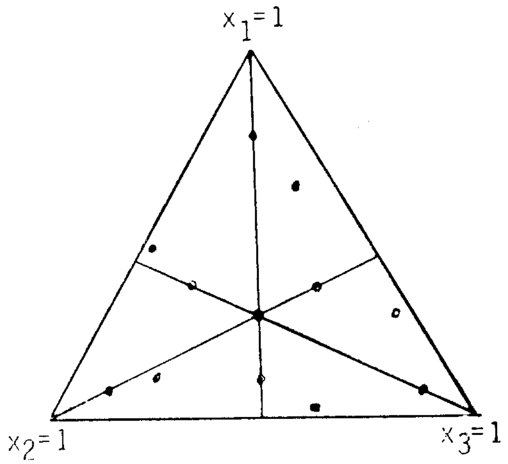


Figure 3.

4. SUMMARY AND CONCLUDING REMARKS

In this paper, a new design concept, called axis-slope-rotatability has been presented. This is an analogue of the Box-Hunter(1957) rotatability and the Hader-Park(1978) slope-rotatability for second order response surface designs. It requires that the variance of the estimated slopes along the component axes be constant at the axial points equidistant from the center point of the factor space.

When the Scheffe's seconid degree polynomial is fitted for mixture experiments, sufficient conditons for axis-slope-rotatability are presented and proved. Also it is shown that all the symmetric simplex designs of Murty and Das(1968), including the simplex lattice designs and the simplex centroid designs of Scheffe(1958, 1963), are axis-slope-rotatable designs. A few examples of axis-slope-rotatable designs are illustrated in section 3.

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