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A Measure of Slope Rotatability for Mixture Experiments

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Abstract

A measure that quantifies the amount of slope rotatability for the second degree Scheffe polynomial model for mixture experiments is proposed and used to compare the several mixture designs which met the symmetric moments conditions in this article.

1. Introduction

The special nature of mixture experiments can be expressed in the following set of constraints: If x_i denote the proportion of the i_{th} component in the mixture, then

$$x_i \geq 0 \text{ for all } i, \text{ and } \sum_{i=1}^{q} x_i = 1. \tag{1.1}$$

For fitting a mixture response surface over the simplex factor space, much attention has been given to the use of the canonical polynomials suggested by Scheffe(1958). Let us assume the surface can be represented as a quadratic function in each of the q components. The second degree Scheffe polynomial in q components, $\underline{x}'=(x_1,x_2,\ldots,x_q)$, is

$$\eta(\underline{x}) = \sum_{i=1}^{q} \beta_i x_i + \sum_{j(>i)}^{q} \beta_{ij} x_i x_j. \tag{1.2}$$

The coefficients in the polynomial are to be estimated from observations on the response variable, $y_i(\underline{x}) = \eta(\underline{x}) + e_i$, where the observations are taken at n selected combinations of the $\underline{x}$ components. The e_i's are assumed to be uncorrelated random errors with zero means and constant variance σ^2. The β's are then estimated by the method of least squares, $\underline{b} = (X'X)^{-1}X'\underline{y}$, in which X is the n x m matrix of values of the m elements of $\underline{x}$'s taken at the design points and y is the n x 1 matrix of y observations with $m=q(q+1)/2$.

Box and Hunter(1957) suggested that, subject to a suitable scaling of the independent

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variables it would be desirable to have equally reliable estimates of the expected response for all points $\underline{x}$ equidistant from the design origin, that is, to have the variance of $y(\underline{x})$ be a function only of $\rho^2 = x_1^2 + x_2^2 + \ldots + x_q^2$. This requires $X'X$ to be invariant under rotation and the designs having this property were called rotatable designs.

If differences at points close together in the factor space are involved, the estimation of the local slopes(the rates of change) of the response surfaces becomes important. Thus, Hader and Park(1978) proposed an analog of the Box-Hunter rotatability criterion "Slope Rotatability", which requires that the variance of $\partial y(\underline{x})/\partial x_i$, the slope with respect to the axial direction of x_i, be constant at all combinations of the independent variables equidistant from the design center.

Recently, Khuri(1988) introduced a measure that quantifies the amount of rotatability in a given response surface design. The slope rotatability in experiments with mixtures has been discussed in Park and Kim(1988). However, no measure has been introduced yet that represents the degree of slope rotatability in experiments with mixtures. In this article, a measure that quantifies the amount of slope rotatability in a given response function with respect to the mixture components in mixture experiments is introduced.

2. A Measure of Slope Rotatability in Mixture Experiments

Suppose that estimation of slopes of $\eta(\underline{x})$ in the equation (1.2) at a point $\underline{x}$ is of interest. When the estimated slope of $y(\underline{x})$ with respect to x_i is

$$\partial \hat{y}(\underline{x})/\partial x_i = b_i + \sum_{\substack{k=1 \\ k\neq i}}^{q} b^*_{ik} x_k \tag{2.1}$$

where $b^*_{ik} = b_{ik}$ if $i < k$ and $b^*_{ik} = b_{ki}$ if $i > k$. The variance of this slope is written as

$$\begin{aligned} \mathrm{Var}(\partial \hat{y}(\underline{x})/\partial x_i) &= \mathrm{Var}(b_i) + \sum_{\substack{k=1 \\ k\neq i}}^{q} x_k^2 \mathrm{Var}(b_{ik}) + \sum_{\substack{k\neq l \\ k,l\neq i}}^{q}\sum^{q} x_k x_l \mathrm{Cov}(b^*_{ik}, b^*_{il}) + 2\sum_{\substack{k=1 \\ k\neq i}}^{q} x_k \mathrm{Cov}(b_i, b^*_{ik}) \\ &= v_i + \sum_{k=1}^{q} x_k^2 v_{ik} + \sum_{\substack{k\neq l \\ k,l\neq i}}^{q}\sum^{q} x_k x_l c_{ik,il} + 2\sum_{\substack{k=1 \\ k\neq i}}^{q} x_k c_{i,ik} \end{aligned} \tag{2.2}$$

where $v_i = \mathrm{Var}(b_i)$, $v_{ik} = \mathrm{Var}(b^*_{ik})$, $c_{i,ik} = \mathrm{Cov}(b_i, b^*_{ik})$, $c_{ik,il} = \mathrm{Cov}(b^*_{ik}, b^*_{il})$, $i\neq k\neq l$.

Consider the difference

$$d_{ij}(\underline{x}) = \mathrm{Var}(\partial \hat{y}(\underline{x})/\partial x_i) - \mathrm{Var}(\partial \hat{y}(\underline{x})/\partial x_j), \quad i\neq j. \tag{2.3}$$

To represent the quantity of slope rotatability, we define

$$Q_q(d) = \int_R \left\{ \sum_{i<j}^{q-1}\sum^{q} d_{ij}(\underline{x})^2 \right\} dx \tag{2.4}$$

where $R = \left\{ (x_1, x_2, \cdots, x_{q-1}) : x_1 \ge 0 \text{ and } \sum_{i=1}^{q-1} x_i \le 1 \ , \ x_q = 1 - \sum_{i=1}^{q-1} x_i \right\}$.

When the variance of the slopes with respect to each x_i is all the same, the above quantity $Q_q(d)$ has a value of zero. Conversely, "$Q_q(d)$ is zero" means all the differences d_{ij} are zero by definition of $Q_q(d)$ and hence the variances of the slopes are all the same. Furthermore, $Q_q(d)$ becomes larger as a design deviates from a slope rotatable design.

Now, $Q_q(d)$ needs to be expressed in an explicit form as a function of variances and covariances(v_i, v_{ik} and $c_{i,ik}$, $c_{ik,il}$). The following lemma appeared in Cornell(1981) will be used.

Lemma 1. Let $R = \left\{ (x_1, x_2, \cdots, x_{q-1}) : x_1 \ge 0 \text{ and } \sum_{i=1}^{q-1} x_i \le 1 \right\}$, where $x_q = 1 - \sum_{i=1}^{q-1} x_i$, then

$$\int_R x_1^{c_1} x_2^{c_2} \cdots x_{q-1}^{c_{q-1}} \; dx_1 dx_2 \cdots dx_{q-1} = \prod_{i=1}^{q-1} c_i! \int_0^1 y^{\left(\sum_{i=1}^{q-1} c_i + q - 2\right)} dy \Big/ \left(\sum_{i=1}^{q-1} c_i + q - 2\right)! \ . \tag{2.5}$$

Now it can be shown that $Q_q(d)$ is given by the following formula.

Theorem 1. The quantity of slope rotatability $Q_q(d)$ is

$$Q_q(d) = \sum_{i<j}\sum \{T_0(i,j) + T_1(i,j) + T_2(i,j) + T_4(i,j)\} \tag{2.6}$$

Proof. The proof of this equality is given in Appendix A.

Corollary 1. Let us use the notation such as [ii], [ijk], and [ijkl] to denote the pure second order moments, the mixed third order moments, and the mixed fourth order moments, respectively. Then for the mixture designs with symmetry conditions such as

[ii] = A for all i,	[ij] = B for all $i \neq j$,
[iij] = C for all $i \neq j$,	[ijk] = D for all $i \neq j \neq k$,
[iijj] = E for all $i \neq j$,	[iijk] = F for all $i \neq j \neq k$,
[ijkl] = G for all $i \neq j \neq k \neq l$,	

it can be shown that after some algebraic calculations v_{ij} , $c_{i,ij}$ and $c_{ij,ik}$ are constant for all $i \neq j \neq k$ and expressed as follows:

$$\begin{aligned} v &= [\ TU+2T\{(C-D)2-(A-B)(F-G)\}+U\{D(2C+(q-2)D) \\ &\quad -G(A+(q-1)B)\}-2R(C-D)+2S(F-G)]\ /\{TU(E-2F+G)\}, \\ c_1 &= R-T(C-D)/TU\ , \\ c_2 &= [\ T\{(C-D)2-(A-B)(F-G)\}+U\{D(2C+(q-2)D) \\ &\quad -G(A+(q-1)B)\}-2R(C-D)+2S(F-G)]\ /\{TU(E-2F+G)\}, \end{aligned} \tag{2.7}$$

where

$$R = \{2C+(q-2)D\}[\ B\{E+(q-4)F-(q-3)G\}-(C-D)\{C+(q-2)D\}] \\ -\{A+(q-1)B\}[\ D\{E+(q-2)F\}-C\{2F+(q-3)G\}]\ ,$$

$$S = \{2C+(q-2)D\}[\ (q-1)BC-A\{C+(q-2)B\}] \\ -A+(q-1)B[\ (q-2)D(C-D)-(A-B)\{2F+(q-3)G\}]\ ,$$

$$T = [\ \{A+(q-1)B\}\{2E+4(q-2)F+(q-2)(q-3)G\} \\ -(q-1)\{2C+(q-2)D\}^2]\ /\ 2,$$

$$U = [\ (A-B)\{2E+2(q-4)F-(q-2)(q-3)G\}-2(q-2)(C-D)^2]\ /\ 2.$$

Applying this Corollary to the formula (2.6) and after some algebraic calculations, the quantity of slope rotatability $Q_q(d)$ is obtained to be

$$Q_q(d) = q(q-1)[\ 5v^2+4\{(q+3)c_1+(q-2)c_2\}v+(q+2)(q+3)c_1^2 \\ +\ 2(q-2)(q+3)c_1c_2+(q-2)(q-1)c_2^2]\ /(q+3)!. \tag{2.8}$$

3. The Values of $Q_q(d)$ of Mixture Designs with Symmetry Conditions

Examples of experiments with mixtures can be found in various fields. Murty and Das(1968) suggested a symmetric simplex design, that is, a class of designs which satisfy the symmetry conditions and showed that the well known simplex-lattice and simplex-centroid design are particular cases of them. They define a symmetric simplex design as follows:

Definition. A symmetric simplex design for experiments with mixtures consists of some or all the groups G_d, d = 1,...,q, where every group G_d is obtained by permuting the different fractions over the q components in a d_{th} ordered mixture with d_1 components taking a proportion p_1, d_2 of them taking a proportion p_2, and so on, d_h of them taking a proportion p_h such that $d_1 + \dots + d_h = d$ and $d_1p_1 + \dots + d_hp_h = 1$.

Example 1. A simplex-lattice (q,m) design for q components consists of the ${}_{m+q-1}C_m$ points of the simplex representing all possible mixtures in which the proportion of each component has the m+1 equally spaced values 0, 1/m, 2/m, ... , 1. This design has a different number of groups G_d for every d = 1, ... , m. For example, there are three groups G_2 in a (q,6) simplex-lattice design with points of the type (1/6 5/6 0 ... 0), (1/3 2/3 0 ... 0) and (1/2 1/2 0 ... 0). Then, the values of the constants for a simplex-lattice (3,m) design are given as follows:

$$A = 1 + (m-1)(2m-1)/3m + (m-1)^2(m-2)/12m,$$
$$B = (m^2-1)/6m + (m-2)(m^2-1)/24m,$$
$$C = (m^2-1)/12m + (m-2)(m^2-1)(2m-1)/120m^2,$$
$$D = (m^2-1)(m^2-4)/120m^2,$$
$$E = (m^2-1)(m^2+1)/30m^3 + (m-2)(m^2-1)(2m^2-2m+3)/360m^3,$$
$$F = (m^2-1)(m^2-4)/360m^2,$$
$$G = 0.$$

By substituting these values in (2.6) and (2.7) we get the values of $Q_3(d)$ for the cases m = 2, 3, 4, 5 and 6 as in Table 3.1.

Table 3.1. Values of $Q_3(d)$ for (3,m) simplex-lattice design

m	2	3	4	5	6
$Q_3(d)$	18.067	6.989	3.325	1.646	0.817

Example 2. A simplex-centroid design with $2^q - 1$ points involves observations on mixtures consisting of all those combinations of the components where the proportion of each component present is equal. This design has all the q groups G_d, each of the nonzero fractions being equal to 1/d in every mixture. Then the constants in lemma 2.1 take the values as follows:

$$A = \sum_{k=1}^{q} {}_{q-1}C_{k-1} / k^2, \quad B = \sum_{k=2}^{q} {}_{q-2}C_{k-2} / k^2, \quad C = \sum_{k=2}^{q} {}_{q-2}C_{k-2} / k^2,$$
$$D = \sum_{k=3}^{q} {}_{q-3}C_{k-3} / k^3, \quad E = \sum_{k=2}^{q} {}_{q-2}C_{k-2} / k^4, \quad F = \sum_{k=3}^{q} {}_{q-3}C_{k-3} / k^4,$$
$$G = \sum_{k=4}^{q} {}_{q-4}C_{k-4} / k^4.$$

By substituting these values in (2.6) and (2.7) we get the values of $Q_q(d)$ for the cases q = 3, 4, and 5 as in Table 3.2.

Table 3.2. Values of $Q_q(d)$ for simplex-centroid design

q	3	4	5
$Q_q(d)$	12.503	2.392	0.317

Example 3. Consider the symmetric simplex design in q components with the three groups G_d; d = 1, 2, 3 where the components in each group G_d take the proportion 1/d. Then the values of the constants are given as follows:

$A = (2q^2 + 3q + 31) / 36$, $B = (4q + 1) / 36$, $C = (8q+11) / 216$, $D = 1 / 27$,
$E = (16q+49) / 1296$, $F = 1 / 81$, $G = 0$.

By substituting these values in (2.6) and (2.7) we get the values of $Q_q(d)$ for the cases q = 3, 4, and 5 as in Table 3.3.

Table 3.3. Values of $Q_q(d)$ for symmetric simplex design

q	3	4	5
$Q_q(d)$	12.503	2.606	0.438

Example 4. Consider the (3q+1)-point simplex screening design with q components described as follows:

name	number	composition
vertices	q	$x_i = 1$; $x_j = 0$ for all $j \neq i$
interior	q	$x_i = (q+1)/2q$; $x_j = 1/2q$ for all $j \neq i$
centroid	1	$x_i = 1/q$ for all i
end effects	q	$x_i = 0$; $x_j = 1/(q-1)$ for all $j \neq i$

Then, the values of the constants are given as follows:

$A = 5 / 4 + 3 / 4q + 1 / q^2 + 1 / (q-1)$,
$B = 3 / 4q + 1 / q^2 + (q-2) / (q-1)^2$,
$C = (q+4) / 8q^2 + 1 / q^3 + (q-2) / (q-1)^3$,
$D = 1 / 2q^2 + 1 / q^3 + (q-3) / (q-1)^3$,
$E = (2q+5) / 16q^3 + 1 / q^4 + (q-2) / (q-1)^4$,
$F = (q+5) / 16q^3 + 1 / q^4 + (q-3) / (q-1)^4$,
$G = 5 / 16q^3 + 1 / q^4 + (q-4) / (q-1)^4$.

By substituting these values in (2.6) and (2.7) we get the values of $Q_q(d)$ for the cases q = 3, 4, and 5 as in Table 3.4.

Table 3.4. Values of $Q_q(d)$ for simplex screening design

q	3	4	5
$Q_q(d)$	5.636	0.305	0.317

Now, we consider a design which is not a symmetric simplex design but has the symmetry conditions.

Example 5. Consider the three component mixture experiment such as the n design points are equally spaced on a circle centered at the centroid (1/3 1/3 1/3) with radius ρ. If θ denotes the angle between the x_1 axis and the line which connects a design point to the centroid, then the coordinates of each point are given by $(x_{1u}\ x_{2u}\ x_{3u})$, u = 0, 1, ... , n-1, where

$$x_{1u} = 1/3 + 6\rho\cos(\theta + 2u\pi/n)/3,$$
$$x_{2u} = 1/3 - 6\rho\cos(2\pi/3 - \theta - 2u\pi/n)/3,$$
$$x_{3u} = 1/3 + 6\rho\cos(\pi/3 - \theta - 2u\pi/n)/3.$$

Then, the values of the constants are given as follows:

$$A = n/9 + n\rho^2/3,\ B = n/9 - n\rho^2/6,\ C = n/27,$$
$$D = n/27 - n\rho^2/6,\quad E = n/81 - n\rho^2/18,$$
$$F = n/81 + n\rho^2/12,\quad G = 0.$$

By substituting these values in (2.6) and (2.7) we get the values of $Q_3(d)$ for the cases $\theta = 0$, $\rho^2 = 1/6$, n = 5, 10, 15, 20 and 25 as in Table 3.5.

Table 3.5. Values of $Q_3(d)$ for circular design

n	5	10	15	20	25
$Q_3(d)$	2.563	0.641	0.285	0.160	0.102

4. Concluding Remarks

In this paper, a quantity that represents the degree of slope rotatability in mixture experiments have been proposed and computed in oder to compare several mixture designs with symmetry conditions. We can found following facts:

(1) Increasing the number of design points seemes to be a cause to make a given mixture design more slope rotatable.

(2) Increasing the number of components also increase the number of design points and decrease the values of $Q_q(d)$.

(3) It can be evaluated for any set of points, with or without symmetries, and hence be applied to any kind of design and any number of independent variables.

But this quantity is not expressed as a percentage and does not have an upper bound, which is expected to be developed in later study.

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Appendix A

Proof of Theorem 1.

Applying the Lemma 1 to the formula (2.4) and after some tedious algebraic calculations the expression (2.6) can be obtained, where

$$T_0(i,j) = (v_i-v_j)^2/(q-1)! + 4(v_i-v_j)(c_{i,ij}-c_{j,ij})/q! + 8(c_{i,ij}^2-c_{i,ij}c_{j,ij}+c_{j,ij}^2)/(q+1)!$$
$$+ 16v_{ij}(c_{i,ij}+c_{j,ij})/(q+2)! + 40v_{ij}^2/(q+3)! ,$$

$$T_1(i,j) = \sum_{\substack{k=1 \\ k\neq i,j}}^{q} [4(v_i-v_j)(c_{i,jk}-c_{j,jk})/q! + \{2(v_i-v_j)(c_{ij,ik}-c_{ij,jk}+2v_{ik}-2v_{jk})$$
$$+ 8(c_{i,ij}-c_{j,ij})(c_{i,ik}-c_{j,jk}) + 8(c_{i,ik}-c_{j,jk})^2\}/(q+1)!$$
$$+ \{12(v_{ik}-v_{jk})(c_{i,ij}-c_{j,ij}+2c_{i,ik}-2c_{j,jk}) + 16(c_{i,ik}-c_{j,jk})(c_{ij,ik}-c_{ij,jk})$$
$$+ 8(2c_{i,ij}c_{ij,ik}-c_{i,ij}c_{ij,jk}-c_{j,ij}c_{ij,ik}+2c_{j,ij}c_{ij,ik})\}/(q+2)!$$
$$+ \{24(v_{ik}-v_{jk})^2+24(v_{ik}-v_{jk})(c_{ij,ik}-c_{ij,jk})+4v_{ij}(c_{ij,ik}+c_{ij,jk})$$
$$+ 16(c_{ij,ik}^2-c_{ij,ik}c_{ij,jk}+c_{ij,jk}^{2)}\}/(q+3)!] ,$$

$$T_2(i,j) = \sum_{\substack{k\neq l \\ k,l\neq i,j}}^{q}\sum^{q} [\{2(v_i-v_j)(c_{ik,il}-c_{jk,jl}-c_{ij,ik}+c_{ij,ik}) + 4(c_{i,ik}-c_{j,jk})(c_{i,il}-c_{j,jl})\}/(q+1)!$$
$$+ \{8(v_{ik}-v_{jk})(c_{i,il}-c_{j,jl}) + 8(c_{i,ik}-c_{j,jk})(c_{ij,il}-c_{ij,jl}-2c_{jk,jl}+2c_{ik,il})\}/(q+2)!$$
$$+ \{4(v_{ik}-v_{jk})(v_{il}-v_{jl}+2c_{ij,il}-2c_{ij,jl}+6c_{ik,il}-6c_{jk,jl}) - 4(c_{ij,il}c_{ij,jk}+c_{ij,ik}c_{ij,jl})$$
$$+ 16(c_{ij,ik}-c_{ij,jk})(c_{ik,il}-c_{jk,jl})\}/(q+3)!] ,$$

$$T_3(i,j) = \sum_{\substack{k \neq l \neq m \\ k,l,m \neq i,j}}^{q}\sum^{q}\sum^{q}\Big[\ 4(c_{i,ik}-c_{j,jk})(c_{il,im}+c_{jl,jm})/(q+2)! + 4\{(v_{ik}-v_{jk})(c_{il,im}-c_{jl,jm}) + 2(c_{ik,il}-c_{jk,jl})(c_{ik,im}-c_{jk,jm}) + (c_{ik,il}-c_{jk,jl})(c_{ij,im}-c_{ij,jm})\}/(q+3)!\Big],$$

$$T_4(i,j) = \sum_{\substack{k \neq l \neq m \neq n \\ k,l,m,n \neq i,j}}^{q}\sum^{q}\sum^{q}\sum^{q}(c_{ik,il}-c_{jk,jl})(c_{im,in}+c_{jm,jn})/(q+3)!\ .$$