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Measures Of Slope Rotatability For Mixture Experiment Designs

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Abstract

The concept of slope rotatability introduced by Hader and Park(1978) is available when we are interested in the difference of the responses. Since there can be constraints on the factor levels in mixture experiments, there arises a need for adaptation of the concept of slope rotatability and the measure to assess it. In this article, measures of slope rotatability in mixture experiments are proposed to quantify the amount of slope rotatability for a given design. Measures for a restricted region design as well as for an unrestricted region design are presented. Then, the designs having different optimalities are compared with respect to these measures by some examples.

Keywords : A-Optimality, D-Optimality, Extreme Vertices Design, Mixture Experiment, Slope Rotatability

1. Introduction

In response surface models, we assume that the dependent variable is adequately approximated by a low order polynomial in k independent variables. When we concentrate on the second order polynomial model, the algebraic form is as below.

$$\eta(\mathbf{x}) = \beta_0 + \sum_{i=1}^{k}\beta_i x_i + \sum_{i=1}^{k}\beta_{ii}x_i^2 + \sum_{i=1}^{k}\sum_{j>i}^{k}\beta_{ij}x_i x_j \tag{1}$$

It can be rewritten as $\eta(\mathbf{x}) = \mathbf{x_s}'\beta$ in matrix form, where $\mathbf{x}' = (x_1, x_2, \cdots, x_k)$, $\mathbf{x_s}' = (1, x_1, x_2, \cdots, x_k, x_1^2, \cdots, x_k^2, x_1x_2, \cdots, x_{k-1}x_k)$ and β is the corresponding $m \times 1$ column vector of regression coefficients, where $m = (k+1)(k+2)/2$.

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A mixture experiment is a special type of a response surface experiment in which the factors are the ingredients or components of a mixture and the response is a function of the proportions of each ingredient. In a q component mixture, if x_i is the proportion of the ith component, the restrictions on the components are $x_i \geq 0,\ i=1,2,\cdots,q,\ \sum_{i=1}^{q} x_i = 1$. So the factor space containing the q components is a $(q-1)$-dimensional regular simplex. Suppose an experimenter is interested in the difference of the responses in a mixture experiment. Then a design is wanted to have the same variances of the estimated slopes for all components at the same distance from the baseline where one component has the value of 0. Since this property is not easy to get, we consider measures to assess the amount of slope rotatability in mixture designs.

This article consists of five sections. In section 2, concepts of various rotatabilities are introduced. Measures of slope rotatability in mixture experiments and their applications are presented in section 3 and 4, respectively. Concluding remarks are in section 5.

2. Various Rotatabilities

2.1 Rotatability

The coefficients in (1) are to be estimated from N observations on the response variables. The observations are $y_i(\boldsymbol{x}) = \eta(\boldsymbol{x}) + \epsilon_i,\ i = 1,\cdots,N$, where ϵ_i's are assumed to be uncorrelated and have zero means and constant variances, σ^2. Then the β is estimated by the least squares method as $\boldsymbol{b} = (\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{X}'\boldsymbol{y}$, where $\boldsymbol{X}$ is an $N \times m$ matrix of the m-element $\boldsymbol{x_s}'$ vectors, taken at the design points, and $\boldsymbol{y}$ is an $N \times 1$ vector of observed responses. The predicted response value is given by $\hat{y}(\boldsymbol{x}) = \boldsymbol{x_s}'\boldsymbol{b}$ and the variance of the predicted response for a given point is $Var(\hat{y}(\boldsymbol{x})) = \boldsymbol{x_s}'(\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{x_s}\sigma^2$. So $Var(\hat{y}(\boldsymbol{x}))$ depends on a particular point $\boldsymbol{x}$ through $\boldsymbol{x_s}$ and depends on the design matrix $\boldsymbol{X}$ via $(\boldsymbol{X}'\boldsymbol{X})^{-1}$.

A design matrix $\boldsymbol{X}$ is said to be rotatable if the $Var(\hat{y}(\boldsymbol{x}))$ is only a function of $\rho = (x_1^2 + x_2^2 + \cdots + x_k^2)^{1/2}$, which is the distance of the point $\boldsymbol{x}$ from the center of the design. Since rotatability was introduced by Box and Hunter(1957), the concept of it has been an important design criterion. When the exact rotatable design can not be obtained, it is important to know how rotatable it is. Khuri(1988) and Park et al.(1993) introduced a measure quantifying the amount of rotatability in a given response surface design.

2.2 Slope rotatability

If the difference of the responses, not the response itself, is of our interest, we must introduce a somewhat different concept. Our concern is estimating the first derivative of $\eta(x)$ with respect to each independent variable. Then, for the second order model the first derivative of $\hat{y}(x)$ with respect to each variable x_i, $\frac{\partial \hat{y}(x)}{\partial x_i} = b_i + 2b_{ii}x_i + \sum_{j=1, j\neq i}^{k} b_{ij}x_j$, is wanted to have a property analogous to rotatability.

Hader and Park(1978) proposed that if a design satisfies the following conditions [C1] and [C2], estimates of the slopes over axial directions will be equally reliable for all points x equidistant from the design origin.

[C1] For each $i = 1,2,\cdots,k$, the variances of $\partial \hat{y}(x)/\partial x_i$ are equal for all x that are equidistant from the design origin, that is, $Var\left(\partial \hat{y}(x)/\partial x_i\right)$ depends on x only through $\rho = (x_1^2 + x_2^2 + \cdots + x_k^2)^{1/2}$.

[C2] The variances of $\partial \hat{y}(x)/\partial x_i$, $i = 1,2,\cdots,k$, are equal, that is, $Var\left(\frac{\partial \hat{y}(x)}{\partial x_1}\right) = Var\left(\frac{\partial \hat{y}(x)}{\partial x_2}\right) = \cdots = Var\left(\frac{\partial \hat{y}(x)}{\partial x_k}\right)$ for any point x.

They referred to this property as slope rotatability. Park and Kim(1992) suggested a measure of slope rotatability that enables us to assess the degree of slope rotatability for a given design. Recently Park et al.(2007), Jang(2005) and Kim et al.(2004) treated the slope rotatability in various respects.

2.3 Axis slope rotatability in mixture experiments

The concept of slope rotatability can be introduced in mixture experiments. The general form of the second-degree Scheffé polynomial in q components is $\eta(x) = \sum_{i=1}^{q}\beta_i x_i + \sum_{i=1}^{q}\sum_{j>i}^{q}\beta_{ij}x_i x_j$. On the axis of the ith component, the other components have the same value of $(1-x_i)/(q-1)$ and substituting this expression for x_j for all $j \neq i$ leads to

$$\eta(x) = \beta_i x_i + \sum_{j\neq i}^{q}\beta_j \frac{(1-x_i)}{q-1} + \sum_{l=1}^{i-1}\beta_{li}\frac{(1-x_i)x_i}{q-1} + \sum_{j=i+1}^{q}\beta_{ij}\frac{x_i(1-x_i)}{q-1} + \sum_{\substack{j=1 \\ j\neq i}}^{q}\sum_{\substack{k>j \\ k\neq i}}^{q}\beta_{jk}\frac{(1-x_i)^2}{(q-1)^2}$$

Then at the proportion x_i on the axis of the ith component, the slope of the expected response with respect to the ith component is $\frac{d\eta(x)}{dx_i} = \gamma_{0i} + \gamma_{1i}x_i$, where

$$\gamma_{0i} = \frac{1}{q-1}\left((q-1)\beta_i - \sum_{j\neq i}^{q}\beta_j + \sum_{l=1}^{i-1}\beta_{li} + \sum_{j=i+1}^{q}\beta_{ij} - \frac{2}{(q-1)}\sum_{\substack{j=1\\ j\neq i}}^{q}\sum_{\substack{k>j\\ k\neq i}}^{q}\beta_{jk},\right)$$

$$\gamma_{1i} = \frac{2}{q-1}\left(\frac{1}{(q-1)}\sum_{\substack{j=1\\ j\neq i}}^{q}\sum_{\substack{k>j\\ k\neq i}}^{q}\beta_{jk} - \sum_{l=1}^{i-1}\beta_{li} - \sum_{j=i+1}^{q}\beta_{ij}\right).$$

The estimates of γ_{0i} and γ_{1i} can be obtained by substituting the least squares estimates of β_i and β_{ij}. Hence the variance of the estimated slope is $Var\left(\frac{d\hat{y}(\boldsymbol{x})}{dx_i}\right) = Var(\widehat{\gamma_{0i}}) + 2Cov(\widehat{\gamma_{0i}}, \widehat{\gamma_{1i}})x_i + Var(\widehat{\gamma_{1i}})x_i^2$.

3. Measures of Slope Rotatability in Mixture Experiments

When a given mixture experiment design is not axis slope rotatable, we want a measure to assess the degree of slope rotatability. Focusing on the estimated slope of the response surface along the x_i-direction, it can be rewritten as $\frac{\partial\, \hat{y}(\boldsymbol{x})}{\partial\, x_i} = \boldsymbol{S}_i'(x_i)\boldsymbol{b}$, $i = 1, 2, \cdots q$, where $\boldsymbol{S}_i'(x_i)$ is a $1\times p$ vector of coefficients with which to multiply the elements of a $p\times 1$ vector $\boldsymbol{b} = (\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{X}'\boldsymbol{y}$. The subscript i associates $\boldsymbol{S}_i$ with the ith component.

For example, if $q = 3$ and the second-degree equation $\hat{y}(\boldsymbol{x}) = \boldsymbol{x_s}'\boldsymbol{b}$ contains $\boldsymbol{b} = (b_1, b_2, b_3, b_{12}, b_{13}, b_{23})$, then, for components 1, 2 and 3, respectively

$$\begin{aligned} \boldsymbol{S}_1'(x_1) &= \left(1, -\frac{1}{2}, -\frac{1}{2}, \frac{1-2x_1}{2}, \frac{1-2x_1}{2}, \frac{x_1-1}{2}\right) \\ \boldsymbol{S}_2'(x_2) &= \left(-\frac{1}{2}, 1, -\frac{1}{2}, \frac{1-2x_2}{2}, \frac{x_2-1}{2}, \frac{1-2x_2}{2}\right) \\ \boldsymbol{S}_3'(x_3) &= \left(-\frac{1}{2}, -\frac{1}{2}, 1, \frac{x_3-1}{2}, \frac{1-2x_3}{2}, \frac{1-2x_3}{2}\right) \end{aligned} \qquad (2)$$

The variance of the estimated slope along the x_i-direction at $x_i = t$, denoted by $Var_i(t)$, is $Var_i(t) = \boldsymbol{S}_i'(t)(\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{S}_i(t)\sigma^2$ and the average of $Var_i(t)$ for all components, denoted by $\overline{Var}(t)$, is $\overline{Var}(t) = \frac{1}{q}\sum_{i=1}^{q}\boldsymbol{S}_i'(t)(\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{S}_i(t)\sigma^2$.

To measure the discrepancy of variance in the ith component at $x_i = t$, let us introduce the following quantity.

$$D_i(t) = \frac{1}{\sigma^4}\left(Var_i(t) - \overline{Var}(t)\right)^2 = \left(\boldsymbol{S}_i'(t)(\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{S}_i(t) - \frac{1}{q}\sum_{j=1}^{q}\boldsymbol{S}_j'(t)(\boldsymbol{X}'\boldsymbol{X})^{-1}\boldsymbol{S}_j(t)\right)^2.$$

Then summing up on i and dividing by the number of components leads to the quantity, $D(t)$, which can be expressed as $D(t)=\frac{1}{q}S'(t)(I_q-\frac{J_q}{q})S(t)$, where $S'(t)=\left(S_1'(t)(X'X)^{-1}S_1(t),\cdots,S_q'(t)(X'X)^{-1}S_q(t)\right)$, I_q is a $q\times q$ unit matrix, and J_q is a $q\times q$ matrix whose elements are all 1.

When we are interested in all the range of components, that is, in case of the unrestricted region of interest, we must get the average of $D(t)$ for $0\le t\le 1$. Then the proposed measure of slope rotatability is $H_c(X)=\frac{1}{1+\int_0^1 D(t)dt}$. It ranges between 0 and 1 and equals 1 if and only if the design given by X is axis slope rotatable. It is close to 0 when the design is far from being axis slope rotatable. By the adjacency of this measure to 1, we can measure the axis slope rotatability for a given design matrix in an unrestricted region.

But, in many cases, the region of interest is restricted, and then the proposed measure of slope rotatability given above may not be practical. So we need to give a somewhat different measure with a narrower integration interval. Suppose that the bounds for each component are given by $a_i\le x_i\le b_i$, $i=1,2,\cdots,q$ and we can get $a_{\min}=\min\{a_1,a_2,\cdots,a_q\}$ and $b_{\max}=\max\{b_1,b_2,\cdots,b_q\}$. Then the proposed measure of slope rotatability may be modified to

$$H_r(X)=\frac{1}{1+\int_{a_{\min}}^{b_{\max}} D(t)dt/(b_{\max}-a_{\min})}. \tag{3}$$

$H_r(X)$ has the value of 1 for an axis slope rotatable design and the probable range of $H_r(X)$ is also between 0 and 1.

4. Applications of the Proposed Measures

4.1 Case of axis slope rotatable designs

It was stated that all axis slope rotatable designs have only one value of the measure, 1. To show this, consider the simplex lattice design with 3 components which has the following model matrix X_l.

$$X_l=\begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 2/3 & 1/6 & 1/6 & 1/9 & 1/9 & 1/36 \\ 1/6 & 2/3 & 1/6 & 1/9 & 1/36 & 1/9 \\ 1/6 & 1/6 & 2/3 & 1/36 & 1/9 & 1/9 \\ 0 & 1/2 & 1/2 & 0 & 0 & 1/4 \\ 1/2 & 0 & 1/2 & 0 & 1/4 & 0 \\ 1/2 & 1/2 & 0 & 1/4 & 0 & 0 \\ 1/3 & 1/3 & 1/3 & 1/9 & 1/9 & 1/9 \end{pmatrix}$$

This case has three components involved and $S_1'(x_1)$, $S_2'(x_2)$ and $S_3'(x_3)$ are shown in (2). By easy calculation, $S'(t)$ can be showed as $S'(t) = (11.75 - 42.72t + 44.45t^2, 11.75 - 42.72t + 44.45t^2, 11.75 - 42.72t + 44.45t^2)$. For the same t, the discrepancy does not occur, because the variance of the estimated slope along the x_i-direction has the same form. That is to say, $Var_i(t)$ has the same value as $\overline{Var}(t)$ for a given t. So $D_i(t)$'s and $D(t)$ of $\boldsymbol{X}_l$ are given by 0 and hence $H_c(\boldsymbol{X}_l)$ is 1, as is our expectation.

For illustration the design points of $\boldsymbol{X}_l$ is given in Figure 1. The filled circle is one design point.

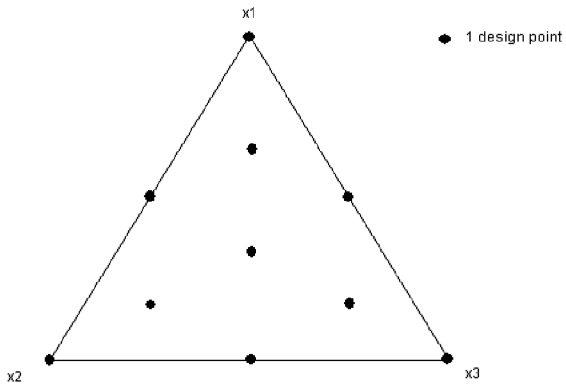

Figure 1. Design points of a simplex lattice design when $q = 3$

The representative of the axis slope rotatable designs is the symmetric simplex design by Murty and Das (1968). The simplex lattice design and the simplex centroid design are obviously particular cases of the symmetric simplex design, so they have the same value of measure $H_c(\cdot)$, 1.

4.2 Case of non slope rotatable designs with unrestricted regions

Among design optimality criteria, D-optimality is the best known and the most used one. A D-optimal design is one in which $\det(\boldsymbol{X}'\boldsymbol{X}/N^p)$ is maximized, where p is the number of parameters in the model and N is the number of data points. The second most popular one is A-optimality which makes the sum of the variances of the estimated coefficients minimized. Using the OPTEX procedure in SAS the 10 point D-optimal design $\boldsymbol{X}_d$ having $\boldsymbol{X}_l$ as candidate points can be obtained as below:

$$X_d = \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 1/2 & 1/2 & 0 & 0 & 1/4 \\ 1/2 & 0 & 1/2 & 0 & 1/4 & 0 \\ 1/2 & 1/2 & 0 & 1/4 & 0 & 0 \\ 1/2 & 1/2 & 0 & 1/4 & 0 & 0 \end{pmatrix}$$

$S'(t)$ of X_d is given as

$S'(t) = (10.75 - 37t + 37t^2, 10.75 - 37t + 37t^2, 10.75 - 41t + 43t^2)$. Then, $D(t)$ of X_d and the value of the measure can be obtained as

$D(t) = 3.55t^2 - 10.67t^3 + 8t^4$ and $H_c(X_d) = 0.894$.

The 10 point A-optimal design X_a having X_l as candidate points can be obtained as below.

$$X_a = \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 1/2 & 1/2 & 0 & 0 & 1/4 \\ 0 & 1/2 & 1/2 & 0 & 0 & 1/4 \\ 1/2 & 0 & 1/2 & 0 & 1/4 & 0 \\ 1/2 & 0 & 1/2 & 0 & 1/4 & 0 \\ 1/2 & 1/2 & 0 & 1/4 & 0 & 0 \\ 1/2 & 1/2 & 0 & 1/4 & 0 & 0 \end{pmatrix}$$

$D(t)$ of X_a is given as $D(t) = 0.03 - 0.58t + 3.97t^2 - 11.67t^3 + 12.54t^4$ and $H_c(X_a) = 0.607$. The replication of the vertex point of x_3 ends in breaking the slope rotatability. Comparing these two alphabetic optimal designs, though neither of them is slope rotatable, X_d is more slope rotatable than X_a in the measure of $H_c(\cdot)$.

4.3 Case of non slope rotatable designs with restricted regions

We now consider the case where there are both lower and upper bounds on the component proportions. In this case, the feasible mixture region is no longer a simplex. To illustrate, use the example in Myers and Montgomery(1995). Suppose that we wish to formulate a shampoo in terms of proportions of three components x_1 = lauryl sulfate, x_2 = cocamide, and x_3 = lauramide with bounds $0.4 \le x_1 \le 0.6$, $0.14 \le x_2 \le 0.2$ and $0.26 \le x_3 \le 0.4$. Here, $a_{\min}$ = 0.14 and $b_{\max}$ = 0.6.

The extreme vertices of the constrained region are formed by the combinations of the upper and lower bound constraints. The design matrix formed by this strategy, containing 13 data points, is given as below:

$$X_{re}=\begin{pmatrix} .60 & .140 & .260 & .08400 & .15600 & .036400 \\ .54 & .200 & .260 & .10800 & .14040 & .052000 \\ .40 & .200 & .400 & .08000 & .16000 & .080000 \\ .46 & .140 & .400 & .06440 & .18400 & .056000 \\ .57 & .170 & .260 & .09690 & .14820 & .044200 \\ .53 & .140 & .330 & .07420 & .17490 & .046200 \\ .47 & .200 & .330 & .09400 & .15510 & .066000 \\ .43 & .170 & .400 & .07310 & .17200 & .068000 \\ .55 & .155 & .295 & .08525 & .16225 & .045725 \\ .52 & .185 & .295 & .09620 & .15340 & .054575 \\ .45 & .185 & .365 & .08325 & .16425 & .067525 \\ .48 & .155 & .365 & .07440 & .17520 & .056575 \\ .50 & .170 & .330 & .08500 & .16500 & .056100 \end{pmatrix}$$

This design matrix contains the four vertices of the region, the four edge centers, the overall centroid, and the four axial runs that lie midway between the centroid, so there are 13 design points in all. The region of interest and the design points are in Figure 2, and the measure of slope rotatability has the value, $H_r(X_{re}) = 1.699 \times 10^{-10}$. The smallest parallelogram containing the design points is the region of interest in the shampoo foam experiment. The filled circle is one design point.

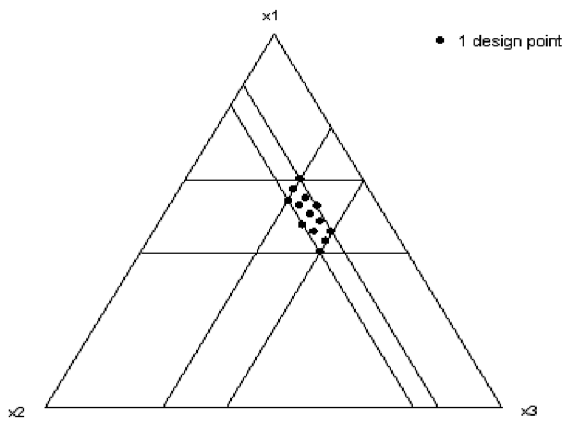

Figure 2. Design points of the a 13 point extreme vertices design

Now, we will construct a D-optimal design for this problem, assuming that the experimenter is planning to fit a quadratic model. We will specify the design points of the extreme vertices design mentioned previously as our candidate design points. We get the D-optimal design X_{ro}, and the region of interest and the design points are in Figure 3. The measure of slope rotatability has the value, $H_r(X_{ro}) = 6.41662 \times 10^{-11}$.

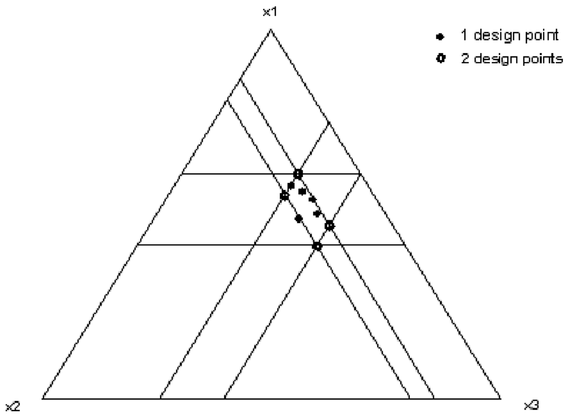

Figure 3. Design points of a 13 point D-optimal design

We also consider the distance-based design with four replicates for the shampoo form experiments, say, $\boldsymbol{X}_{rb}$, also. The measure of slope rotatability in (3) can be applied and has the value, $H_r(\boldsymbol{X}_{rb}) = 6.00406 \times 10^{-11}$. When the three designs are compared in terms of the proposed measure of slope rotatability, $\boldsymbol{X}_{re}$ is better than the others which have almost same values.

4.4 Further comments

When we think the integration is rather time-consuming and that only the design points are important to consider, we can use $H_m(\cdot)$, a discrete version of $H_r(\cdot)$.

$$H_m(\boldsymbol{X}) = \frac{1}{1+\sum_{i=1}^{p} D(d_i)/p} = \frac{1}{1+\sum_{i=1}^{p} \boldsymbol{S}'(d_i)(I_q - \frac{J_q}{q})\boldsymbol{S}(d_i)/pq},$$

where $\{d_1, \cdots, d_p\}$ are different design points and p is the number of different design points.

Since, for a slope rotatable design, $D(d_i)$ is zero for all d_i, this measure $H_m(\cdot)$ also ranges between 0 and 1. When a given design is slope rotatable, as all the $\boldsymbol{S}'(d_i)(I_q - \frac{J_q}{q})\boldsymbol{S}(d_i)$ $(i = 1, 2, \cdots, p)$ are zeros, the measure given above has the value of 1.

Though $H_r(\cdot)$ is more reasonable than $H_c(\cdot)$ when a region of interest is restricted, a more reasonable but a little complicated integration interval can be found. Since the upper and lower boundaries of each component are not always attainable, the boundary of one component can be adapted by the others. In (3)

$a_{\min}$ and $b_{\max}$ can be replaced by $a^*_{\min}$ and $b^*_{\max}$, where $a^*_{\min} = \max\{a_{\min}, a_{com}\}$, $b^*_{\max} = \min\{b_{\max}, b_{com}\}$, $a_{com} = \min\{(1-\sum_{j\neq i}^{q} b_j),\ i=1,\cdots,q\}$ and $b_{com} = \max\{(1-\sum_{j\neq i}^{q} a_j),\ i=1,\cdots,q\}$. Then the new measure denoted by $H_n(\cdot)$ can be obtained as below:

$$H_n(\boldsymbol{X}) = \frac{1}{1+\int_{a^*_{\min}}^{b^*_{\max}} D(t)dt/(b^*_{\max}-a^*_{\min})}.$$

5. Concluding remarks

Four similar but different measures of slope rotatability in mixture experiments are proposed in this article. The simplest $H_c(\cdot)$ can be used only when a region of interest is not constrained. But giving 0's and 1's to all a_i's and b_i's respectively, it is a mere special case of $H_r(\cdot)$. So $H_r(\cdot)$ is a more general form of the measure. The integration interval of $H_r(\cdot)$ is so loose that a tighter interval is needed. The result of such need is $H_n(\cdot)$. But in most cases, $H_n(\cdot)$ equals to $H_r(\cdot)$.

All the above three measures are considering the average discrepancy of variance in a continuous manner. But a clumsy measure, $H_m(\cdot)$, treats it in a discrete manner. Though it has a simpler form, the realization by a computer package is more difficult than the others.

As a D-optimal design is not always A-optimal, one design having a larger value than another design in one measure does not always have a larger value in another measure. Obviously, the slope rotatable design gives the value 1 for all the given measures. Often, these measures have very small values in restricted region cases.

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