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Optimal Slope Designs for Second Degree Kronecker Model Mixture Experiments

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Abstract: The aim of this paper is to investigate some optimal slope mixture designs in the second degree Kronecker model for mixture experiments. The study is restricted to weighted centroid designs, with the second degree Kronecker model. For the selected maximal parameter subsystem in the model, a method is devised for identifying the ingredients ratio that leads to an optimal response. The study also seeks to establish equivalence relations for the existence of optimal designs for the various optimality criteria. To achieve this for the feasible weighted centroid designs the information matrix of the designs is obtained. Derivations of D-, A- and E-optimal weighted centroid designs are then obtained from the information matrix. Basically this would be limited to classical optimality criteria. Results on a quadratic subspace of H-invariant symmetric matrices containing the information matrices involved in the design problem was used to obtain optimal designs for mixture experiments analytically. The discussion is based on Kronecker product algebra which clearly reflects the symmetries of the simplex experimental region.

Keywords: Slope Mixture designs Kronecker product, Optimal Designs, Weighted Centroid Designs, A-, D-, E-Optimality and H- invariant Symmetric Matrices

1. Introduction

The design of experiment involves selection of levels of one or more factors for optimizing one or more criteria. There are often many competing criteria that could be considered in selecting the design, and one is typically forced to make trade-offs between these objectives when choosing competing design. Several optimality criteria have been developed to address estimation or prediction through the use of variance characteristics. D- and A-optimality criteria provide a measure of the variance of the model coefficient through the moment matrix, $M = (X^{'}X)/\mathrm{N}$.

Many practical problems in research are associated with investigation of mixture ingredients $\left(t_1, t_2, ..., t_q\right)$ of m-factors with $\left(t_i \geq 0\right)$ and further restriction of $\sum t_i = 1$. The ingredients influence the response through ratios or proportions. In mixture experiment the factors $\left(t_1, t_2, ..., t_q, \ \ for\ q \geq 2\right)$, such that the mixture components t_i's satisfies the condition below;

$$0 \leq t_i \leq 1,\ i = 1, 2, , ..., q \tag{1}$$

The simplest mixture design is given by $q = 2$ resulting to straight line $(t_1 + t_2 = 1)$. The constraints in equation (1) yield a simplex experimental region. The $\{q, m\}$ simplex lattice designs and simplex-centroid designs were introduced by Scheffé (1958, 1963). Scheffé (1963) gave simplex centroid designs which consists of $2^q - 1$ points with q permutations of $(1,0,0,...0)$ (q pure blends), (qC_2) permutations of $\left(\frac{1}{2},\frac{1}{2},0,...,0\right)$ given by (qC_2) binary blends and the overall centroid $\left(\frac{1}{q},\frac{1}{q},...,\frac{1}{q}\right)$, the q-nary blend.

For weighted centroid, the weights $\alpha_1, \alpha_2,..., \alpha_n \geq 0$, with $\alpha_1+\alpha_2+...+\alpha_n = 1$, $\eta = \alpha_1\eta_1 + \alpha_2\eta_2 + \cdots + \alpha_n\eta_n$, is a weighted centroid design that constitutes a minimal complete class of designs for Kiefer ordering (Draper and Pukelsheim, (1998)). The definitive work by Cornel (1990) listed numerous examples of applications of mixture experiments and provides a thorough discussion for both theory and practice. Early seminal work done by Scheffe' (1958, 1963) suggested and analyzed canonical model forms when the regression function for the expected response is a polynomial of degree one, two or three for the expected response otherwise referred to as the S- models or S-polynomials. The main experimental domain is a probability simplex given by;

$$T_m = \left\{ t = (t_1,...t_m)' \in [0,1]^n : \sum_{i=1}^{m} t_i = 1 \right\} \tag{2}$$

under the experimental condition $t \in T_m$ where the response Y_t is taken to be the real valued random variable. In a polynomial regression model the expected value $E(Y_t)$ is a polynomial function of t.

In this paper, our concern is in the quadratic regression function represented by the homogeneous second degree Kronecker polynomial. The expected response assumes the form in equation (3), as given Draper and Pukelsheim (1998b, 1999).

$$E[Y_t] = \sum_{i=1}^{m}\sum_{j=1}^{m} t_i t_j \theta_{ij} = (t \otimes t)'\theta \tag{3}$$

The use of Kronecker representation forces each entry in the moment matrix to became;

$$M(\tau) = \int_{\tau} (t\otimes t)\,(t\otimes t)' d\tau \tag{1}$$

to be homogeneous of degree four.

According to Pukelsheim (2006), an information matrix ϕ on NND(s) is called $\mathcal{H}-$invariant if $\mathcal{H}$ is a subgroup of the general linear group GL(s) and all the transformation H$\in \mathcal{H}$ fulfil the equation below;

$$\phi(c) = \phi(HCH'), \forall\, C \in NND(s) \tag{2}$$

For invariance of matrix means, Pukelsheim (2006) proved that for $\mathcal{H}$ for which a subgroup of of GL(s), that for $p \in [-\infty: 0) \cup (0: \infty]$ the matrix mean ϕ_p is H-invariant iff $\mathcal{H}$ is a subgroup of the orthogonal matrices, i. e. $\mathcal{H} \in$ orth(s). On the converse he assumed $\mathcal{H}$ to be a subgroup of orthogonal matrices. But ϕ_p depends on C only through its eigenvalues, since the eigenvalues of C and HCH$'$ are identical and hence invariance.

It is important to note that invariance for the determinant criterion ϕ_0 holds relative to the group of unimodal transformations

$$\text{unim}(s) = \{H \in GL(s): \det H = \pm 1 \tag{3}.$$

For an arbitrary non-empty subset $\mathcal{H}$ of $s \times s$ matrices we define a symmetric of $s \times s$ C to be H-invariant iff;

$$C = HCH' \;\forall\, H \in \mathcal{H} \tag{4}$$

The set of all H-invariant symmetric $s \times s$ matrices are denoted by Sym (S, $\mathcal{H}$). Given a particular set $\mathcal{H}$ such that;

$$Sym(S,H) = \begin{cases} \{\Delta_z : z \in \Re^s & for \quad H = sign(s) \\ \{\alpha I_s + \beta I_s I_s' : \alpha, \beta, \in \Re) & for\, H = perm(s) \\ \{\;\alpha I_s : \alpha \in \Re & for\, H = orth(s) \end{cases} \tag{5}$$

H-invariant matrices are diagonal matrix if $\mathcal{H}$ is the sign change group sign(s). They are completely symmetric matrices if they have identical on diagonal entries and identical on off diagonal entries for permutation group perm(s). They are multiples of the identity matrix under the full orthogonal group orth(s).

In this paper we study the optimal slope mixture design using weighted centroid in the second order Kronecker mixtures model. Specifically, the study used the A-, D- and E-optimality criteria for maximal parameter subsystem of interest.

2. Design Problem

The main design problem for this paper is to obtain a design with maximum information for the maximal parameter subsystem $K'\theta$, subject to the side's conditions. The maximum is accomplished through the application of A-, D-, and E-optimality criteria of weighted centroid design which follows the Kiefer-Wolfowitz equivalence theorem.

We consider the second degree Kronecker model suggested by Draper and Pukelsheim (1998) given as;

$$E(Y_t) = f(t)'\theta = \sum_{i,j=1}^{m} \theta_{ii} t_i^2 + \sum_{i,j} (\theta_{ij} + \theta_{ji}) t_i t_j \tag{9}$$

where Y_t the observed response under the experimental conditions $t \in T$ is taken to be a scalar random variable and

$$\Theta = (\theta_{11}, \theta_{22}, ..., \theta_{mm})' \in R^{m^2} \tag{6}$$

is unknown parameter.

The moment matrix is given by

$$M(\tau)=\int_{T} f(t) f(t)' d\tau \qquad (7)$$

for the second-degree Kronecker-model has all entries homogeneous in degree four and reflects the statistical properties of a design τ. Kinyanjui (2007) and Ngigi (2009) showed that second degree mixture experiments for maximal parameter subsystem with $m \geq 2$ ingredients, unique D-and A-optimal weighted centroid designs for $K'\theta$ exist. In the same study E-optimal weighted centroid design mixture experiment with two ingredients only was derived. In this paper, we extend their work by deriving D-, A- and E-optimal weighted centroid designs for three ingredients.

The primary concern of the experimenter is to learn more about the subsystems of interest. This allows the designer to evaluate the performance of a design relative to the subsystems of interest only. The parameter system of the mixture experiments contains a lot of repeated terms making it rank deficient hence not all the parameters can be estimated efficiently. The parameter subsystem with $\binom{m+1}{2}$ parameters have been shown to have similar properties to those of the full parameter system. K is called a maximal coefficient matrix for M.

In this paper R-Gui (3.0.1) was used for analysis and in the computation of the A-, D- and E-optimality criteria.

3. Computation of the Coefficient, Moment and Information Matrices

For the full second-order model equation for three ingredient;

$$E(y) = \theta_{11}t_1^2 + \theta_{12}t_1t_2 + \theta_{13}t_1t_3 + \theta_{21}t_2t_1 + \theta_{22}t_2^2 + \theta_{23}t_2t_3 + \theta_{31}t_3t_1 + \theta_{32}t_3t_2 + \theta_{33}t_3^2 \qquad (8)$$

Our aim is to derive the coefficient (K), moment (M) and the information (C) matrices to obtain an optimal mixture.

3.1 Coefficient Matrix (K)

An experimenter may find it expensive and unnecessary to work with the full parameter system θ, and therefore may wish to study s out of the k $s \leq k$ components. This is achieved by studying the linear parameter subsystem of interest $K'\theta$ for some $k \times s$ matrix K. K is referred to as the coefficient matrix of the parameter sub-system K′θ.

Draper and Pukelshein (1998b) proposed a representation involving the Kronecker square $t \otimes t$, the $m^2 \times 1$ vector consisting of the squares and cross products of the components of t in lexicographic order.

Given the regression function (888888) we obtain the order as;

$$f(\mathrm{t}) = \theta_{11}t_1^2 + \theta_{12}t_1t_2 + \theta_{13}t_1t_3 + \theta_{21}t_2t_1 + \theta_{22}t_2^2 + \theta_{23}t_2t_3 + \theta_{33}t_3^2 \qquad (9)$$

The subsystem of interest is given as in equation (15);

$$K'\theta = \begin{pmatrix} \theta_{11} \\ \frac{\theta_{12}+\theta_{21}}{2} \\ \frac{\theta_{13}+\theta_{31}}{2} \\ \theta_{22} \\ \frac{\theta_{23}+\theta_{32}}{2} \\ \theta_{33} \end{pmatrix} \qquad (10)$$

3.2. Moment Matrix (M)

From the General Equivalence Theorem, if $M \in \mathcal{M}$ is a competing moment matrix that is feasible for $K'\theta$ with information matrix $C = C_K(M)$. Then M is $\phi -$ optimal for $K'\theta$ in $\mathcal{M}$ if and only if there exists an NND(s) matrix D, that solves the polarity equation

$$\phi(C)\phi^{-\propto}(D) = trace\ CD = 1$$

And also there exists a generalised inverse G of M, such that the matrix $N = GKCDCK'G'$ that satisfies the normality inequality

$$trace\ AN \leq 1\ for\ all\ A \in \mathcal{M}$$

But for optimality, equality is obtained in the normality inequality if M is inserted instead of A.

Using the Kronecker product, the three factors, $(t_1, t_2, t_3,)$=(1,0,0), (0,1,0) and (0,0,1), for the pure mixture blends are obtained as $(t \otimes t)(t \otimes t)'$; resulting to matrices for each of the design point. This procedure was repeated for the three design points namely, pure, binary and the centroid.

Step 1:- Using pure blends the design is given by combining the three Kronecker matrices to be the design η_1. Further the Moment Matrix $m(\eta_1)$ corresponding to this design (η_1) was obtained.

Step 2:- For the binary mixture blends (1/2, 1/2, 0), (1/2, 0, 1/2) and (0, 1/2, 1/2), we work out the Kronecker product matrices as follows, $(t_1 \otimes t_1), (t_2 \otimes t_2)\, and\, (t_3 \otimes t_3)$. Using the binary blends the design and by combining the three Kronecker matrices then design η_2 was constructed with further Moment Matrix corresponding to the design η_2 of binary blends $m(\eta_2)$.

Step 3:- Finally, we obtain the Kronecker product matrix for the centre point with the following coordinates $\left(\frac{1}{3}, \frac{1}{3}, \frac{1}{3}\right)$.

$\boldsymbol{M(\eta)}$ for the Weighted Centroid Design can be obtained using elementary designs $\boldsymbol{\eta_1}$, $\boldsymbol{\eta_2}$ and $\boldsymbol{\eta_3}$ are used to generate the weighted centroid design $\boldsymbol{\eta}$ with points $t_1 = \begin{pmatrix}1 & 0 & 0 & \frac{1}{2} & \frac{1}{2} & 0 & \frac{1}{3}\end{pmatrix}'$, $t_2 = \begin{pmatrix}0 & 1 & 0 & \frac{1}{2} & 0 & \frac{1}{2} & \frac{1}{3}\end{pmatrix}'$ and $t_3 = \begin{pmatrix}0 & 0 & 1 & 0 & \frac{1}{2} & \frac{1}{2} & \frac{1}{3}\end{pmatrix}'$ such that: for weights α_1, α_2, $\alpha_3 \geq 0$ with $\alpha_1 + \alpha_2 + \alpha_3 = 1$, the design

$$\eta = \alpha_1\eta_1 + \alpha_2\eta_2 + \alpha_{32\ d\ elementary\ centroid}\eta\ _3$$

with weights.

$$\alpha_1 = 3(\mu_4 - \mu_{22} + \mu_{211}),\ \alpha_2 = 24(\mu_{31} + \mu_{22} - 2\mu_{211}) \text{ and } \alpha_3 = 81\mu_{211} \text{where } \mu_4 = \int t_1^4 d\tau,\ \mu_{31} = \int t_1^3 t_2 d\tau,\ \mu_{22} = \int t_1^2 t_2^2 d\tau,\ \mu_{211} = \int t_1^2 t_2 t_3 d\tau, \tag{11}$$

We know that, the moment matrix

$$M(\eta) = \alpha_1 M(\eta_1) + \alpha_2 M(\eta_2) + M(\eta_3) \tag{12}$$

$$\Rightarrow \mu_4 = 0.162477954$$

$$\mu_{22} = 0.010692239$$

$$\mu_{31} = 0.010692239$$

$$\mu_{211} = 0.00176366843 \tag{13}$$

$$\therefore \alpha_1 = 0.428571433$$

$$\alpha_2 = 0.428571387$$

and

$$\alpha_3 = 0.142857142 \tag{14}$$

Alternatively,

$$M(\eta) = \begin{pmatrix} \mu_4 & \mu_{31} & \mu_{31} & \mu_{31} & \mu_{22} & \mu_{211} & \mu_{31} & \mu_{211} & \mu_{22} \\ \mu_{31} & \mu_{22} & \mu_{211} & \mu_{22} & \mu_{31} & \mu_{211} & \mu_{211} & \mu_{211} & \mu_{211} \\ \mu_{31} & \mu_{211} & \mu_{22} & \mu_{211} & \mu_{211} & \mu_{211} & \mu_{22} & \mu_{211} & \mu_{31} \\ \mu_{31} & \mu_{22} & \mu_{211} & \mu_{22} & \mu_{31} & \mu_{211} & \mu_{211} & \mu_{211} & \mu_{211} \\ \mu_{22} & \mu_{31} & \mu_{211} & \mu_{31} & \mu_4 & \mu_{31} & \mu_{211} & \mu_{31} & \mu_{22} \\ \mu_{211} & \mu_{211} & \mu_{211} & \mu_{211} & \mu_{31} & \mu_{22} & \mu_{211} & \mu_{22} & \mu_{31} \\ \mu_{31} & \mu_{211} & \mu_{22} & \mu_{211} & \mu_{211} & \mu_{211} & \mu_{22} & \mu_{211} & \mu_{31} \\ \mu_{211} & \mu_{211} & \mu_{211} & \mu_{211} & \mu_{31} & \mu_{22} & \mu_{211} & \mu_{22} & \mu_{31} \\ \mu_{22} & \mu_{211} & \mu_{31} & \mu_{211} & \mu_{22} & \mu_{31} & \mu_{31} & \mu_{31} & \mu_4 \end{pmatrix} \tag{15}$$

Where $\mu_4 = \int t_1^4 d\tau,\ \mu_{31} = \int t_1^3 t_2 d\tau,\ \mu_{22} = \int t_1^2 t_2^2 d\tau,$ $\mu_{211} = \int t_1^2 t_2 t_3 d\tau,$

$$\Rightarrow \mu_4 = 0.162477954$$

$$\mu_{22} = 0.010692239$$

$$\mu_{31} = 0.010692239$$

$$\mu_{211} = 0.00176366843$$

3.3. Information Matrix

Pukelsheim (1993) gave the definition of an information matrix as:

For a design ξ with the moment matrix M, the information matrix for $K'\theta$ with KxS coefficient matrix K of full column S, is defined to be $C_K(M)$ where the mapping C_K from the cone NND(K) into the space sym(S) is given by

$$C_k(A) = \min_{L \in R^{S\times K};\, LK = I_S} LAL' \quad \text{for all } A \in \text{NND(K)}$$

Where the minimum is taken according to Loewner ordering over all the left inverses L of K.

Now to obtain the information matrix, we utilize the equation

$$D_c = HCH' \tag{16}$$

Where D_C is the coefficient matrix of the slope obtained from the $C = C_k(M)$ and H obtained by getting the differentials of the elements of the design matrix;

$$\text{M}(\tau) = t_1^2 + t_1t_2 + t_1t_3 + t_2t_1 + t_2^2 + t_2t_3 + t_3t_1 + t_3t_2 + t_3^2 \tag{17}$$

$$\partial y/\partial t_i(t_1^2 + \frac{1}{2}(t_1t_2 + t_2t_1) + \frac{1}{2}(t_1t_3 + t_3t_1) + t_2^2 + \frac{1}{2}(t_2t_3 + t_3t_2) + t_3^2) = H, \text{ i} = 1,2,3. \tag{18}$$

This gives;

$$H = \begin{pmatrix} 2t_1 & t_2 & t_3 & 0 & 0 & 0 \\ 0 & t_1 & 0 & 2t_2 & t_3 & 0 \\ 0 & 0 & t_1 & 0 & t_2 & 2t_3 \end{pmatrix}$$

$C = LML'$, where L is the left inverse of K given by;
$L = (K'\text{K})^{-1}K'$

From equation (23), we have;

$$C = \begin{pmatrix} 0.7356\mu_2 + 0.1856\mu_{11} & 0.0928\mu_2 + 0.1144\mu_{11} & 0.0928\mu_2 + 0.1144\mu_{11} \\ 0.0928\mu_2 + 0.1144\mu_{11} & 0.7356\mu_2 + 0.1856\mu_{11} & 0.0928\mu_2 + 0.1144\mu_{11} \\ 0.0928\mu_2 + 0.1144\mu_{11} & 0.0928\mu_2 + 0.1144\mu_{11} & 0.7356\mu_2 + 0.1856\mu_{11} \end{pmatrix}$$

Draper and Pukelsheim (1998), expressed the lower order moments in terms of fourth order moments, such that:

$$\mu_{11} = 2\mu_{31} + 2\mu_{22} + 5\mu_{211} \tag{19}$$

$$\mu_2 = \mu_4 + 2\mu_{31} + 2\mu_{11} \tag{20}$$

Substituting μ_{11} in equation (20) we get

$$\mu_2 = \mu_4 + 6\mu_{31} + 4\mu_{22} + 10\mu_{211} \tag{21}$$

But,

$$\mu_4 = 0.00176366843 \tag{22}$$

Hence,

$$\mu_{11} = 0.287037028 \tag{23}$$

4. Optimality Tests

A-, D- and E-optimal criteria were compute using the derived information matrices D_c.

4.1. A-Optimality

Invariance under reparameterization loses its appeal if the parameters of interest have a definite physical meaning. The average variance criterion save the situation by providing a reasonable alternative. If the coefficients matrix is partitioned into its columns, $K = (c_1, c_2, ..., c_s)$ then the inverse $\frac{1}{\phi_{-1}}$ can be represented as $\frac{1}{\phi_{-1(C_k(A))}} = \frac{1}{s} trace\, C_k(A)^{-1}$

$$= \frac{1}{s} trace\, K' A^{-} K$$

$$= \frac{1}{s} \sum_{j \le s} C_j' A^{-} C_j$$

This corresponds to the average of the standardized variances of the optimal estimates of the scalar parameter systems $c_1'\theta, ..., c_s'\theta$ formed from the columns of K.

We then take recourse to the following average variance criterion as given by Pukelsheim (1993, pg 135).

$$\phi_{-1}(C) = \left(\frac{1}{s} traceC^{-1}\right)^{-1} = 0.0585.$$

4.2. D-Optimality

For the comparison of different criteria and for applying the theory of information functions, the version $\phi_0(C) = (\det C)^{\frac{1}{s}}$ is appropriate the maximisation of the determinant of the information matrices is the same as minimizing the determinant of the dispersion matrices because of the formula $(\det C)^{-1} = \det(C^{-1})$. We therefore take recourse to the formula given in Pukelsheim (1993 pg 135) $\phi_0(C) = (\det C)^{\frac{1}{s}}$ to obtain D- optimal value of 0.07749.

4.3. E-Optimality

The criteria $\phi_{-\infty}$ evaluation of the smallest Eigen value also gains in understanding by a passage to variance. It is the same as minimizing the largest Eigen value of the dispersion matrix.

$$\frac{1}{\phi_{-\infty(C_k(A))}} = \lambda_{\max}\left(C_k(A)^{-1}\right) = \max_{Z \in R^s : \|Z\|=1} Z'K'A^{-}KZ$$

The Eigen value criterion $\phi_{-\infty}$ is one extreme member of the matrix means ϕ_p corresponding to the parameter $p = -\infty$. It is one of the four particular members of the one dimensional family of matrix means ϕ_p that submit itself to the principles that a reasonable criteria must meet as presented in Pukelsheim (1993, chapter 5) we therefore express it in the form $\phi_{-\infty}(C) = \lambda_{\min}(C)$ to give, $\lambda_{\min} = 0.0329$ *and* $\lambda_{\max} = 0.199075$.

5. Conclusion

This paper established the equivalence relations for the existence of optimal designs for the D-, A- and E- optimality criteria for the feasible weighted centroid designs. A quadratic subspace of H-invariant symmetric matrix containing the information matrices (D_C) was derived and used to obtain the optimal design for mixture experiment. Derivations of D-, A- and E-optimal weighted centroid designs were obtained from the information matrix giving 0.07749, 0.0585 and 0.0329 optimal values respectively.

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