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DESIGNS WITH OPTIMAL VALUES IN THE SECOND-DEGREE KRONECKER MODEL MIXTURE EXPERIMENTS

Mike R. Cherutich* **and Gregory K. Kerich**

Department of Mathematics and Computer Science
Moi University
P. O. Box 1125, Eldoret, Kenya
e-mail: mcherutich85@gmail.com

Abstract

The goal of every experimenter is to obtain a design that gives maximum information. Similarly, the performance of a design is measured by the amount of information it contains. This paper investigates mixture experiments in the second-degree Kronecker model. The parameter subspace of interest in this study is maximal parameter subsystem which is subspace of the full parameter space. Previous studies in this area have not been able to show how a design can be improved based on the same parameter subspace. This paper attempts to show an improvement of such designs by first obtaining a proper coefficient matrix. Optimal designs of mixture experiments are derived by employing the Kronecker model approach and applying the various optimality criteria. Results of A- and D-optimal designs for $m = 2, 3$ ingredients are given. The results obtained are higher than those presented in the previous studies. Finally, the efficiencies of these designs are then calculated.

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*Corresponding author

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1. Introduction

A mixture experiment is an experiment which involves mixing of proportions of two or more components to make different compositions of an end product. Consequently, many practical problems are associated with the investigation of mixture ingredients of m factors, assumed to influence the response through the proportions in which they are blended together. The definitive text, Cornell [1] lists numerous examples and provides a thorough discussion of both theory and practice. Early seminal work was done by Scheffe' [14, 15] in which he suggested and analyzed canonical model forms when the regression function for the expected response is a polynomial of degree one, two, or three.

The m component proportions, $t_1, \ldots, t_m$ form the column vector of experimental conditions, $t = (t_1, \ldots, t_m)'$ with $t_i \geq 0$ and further subject to the simplex restriction,

$$\sum_{i=1}^{m} t_i = 1. \tag{1}$$

For the second-degree model, Draper and Pukelsheim [3] proposed a representation involving the Kronecker square $t \otimes t$. Its regression function is $f : T_m \to \Re^{m^2}$, $t = (t_1, \ldots, t_m)' \to t \otimes t = t_i t_j$, $i, j = 1, \ldots, m$ with the lexicographical order of the subscripts. This representation yields the model equation

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

where Y_t, the observed response under the experimental conditions $t \in T$, is taken to be a scalar random variable and $\Theta = (\theta_{11}, \theta_{22}, \ldots, \theta_{mm})' \in \Re^{m^2}$ is an unknown parameter.

Mixture experiments were first discussed in Quenouille [13]. Later on, Scheffe' [14, 15] made a systematic study and laid a strong foundation.

Draper and Pukelsheim [3] proposed a set of mixture models referred to as *K*-models.

Gaffke and Heiligers [5] and Pukelsheim [11] gave a review of the general design environment. Klein [8] showed that the class of weighted centroid designs is essentially complete for $m \geq 2$ ingredients, for Kiefer ordering. As a consequence, the search for optimal designs may be restricted to weighted centroid designs for most criteria.

Klein [8] and Kinyanjui et al. [6] showed how invariance results can be applied to analytical derivations of optimal designs. The spectral analysis of invariant symmetric matrices yielded both eigenvalues and eigenvectors.

2. General Design Problem

The statistical properties of a design τ are reflected by the moment matrix

$$M(\tau) = \int_{T_m} f(t) f(t)' d\tau \in NND(m^2).$$

The amount of information which the design *T* contains on the parameter system $K'\theta$ is captured by the information matrix,

$$C_k(M(\tau)) = (K'K)^{-1} K'M(\tau)K(K'K) \in NND(s) \text{ for } K'\theta.$$

The problem of finding a design with maximum information on the parameter subsystem $K'\theta$ can now be formulated as

$$\text{Maximize } \phi_p(C_k(M(\tau))) \text{ with } t \in T$$

$$\text{Subject to } C_k(M(\tau)) \in PD(s), \tag{3}$$

where *T* denotes the set of all designs T_m. The side condition $C_k(M(\tau)) \in PD(s)$ is equal to the existence of an unbiased linear estimator for $K'\theta$ under τ, Pukelsheim [11, 12]. In this case, the design τ is called *feasible* for $K'\theta$. Any design solving problem (3) for a fixed $p \in (-\theta, 1]$ is called ϕ_p-*optimal* for $K'\theta$ in *T*. For all $p \in (-\infty, 1]$, the existence of ϕ_p-*optimal* design for $K'\theta$ is guaranteed by Theorem 7.13 in Pukelsheim [11].

But from a result of Draper et al. [2] on weighted centroid designs, the design problem reduces to

$$\text{Maximize } (\phi_p \circ C_k \circ M \circ \eta) \text{ with } \alpha \in T_m$$

$$\text{Subject to } C_k(M(\eta(\alpha))) \in PD(s). \tag{4}$$

3. Derivation of the Optimal Designs

By considering the maximal parameter subsystem, the following form of parameter subsystem becomes of interest in this paper,

$$K'\theta = \begin{Bmatrix} (\theta_{ii})_{1 \le i \le m} \\ \dfrac{1}{\binom{m}{2}}(\theta_{ij} + \theta_{ji}),\ _{1 \le i < j \le m} \end{Bmatrix} \in \Re^{\binom{m+1}{2}} \text{ for all } \theta \in \Re^{m^2} \text{ and } m \ge 2. \tag{5}$$

In this formula, the scaling down factor $2\binom{m}{2}$ is motivated by the fact that it coincides with the number of the interaction terms in the model.

When fitting model (2) to a set of observations, a parameter subsystem, say $K'\theta$, of interest is chosen with $K \in \Re^{m^2 \times s}$.

Definition 3.1. We define the K matrix as

$$K = (K_1, K_2) \in \Re^{m^2 \times \binom{m+1}{2}}, \tag{6}$$

where

$$K_1 = \sum_{i=1}^{m} e_{ii} e_i' \quad \text{and} \quad K_2 = \frac{1}{2\binom{m}{2}} \sum_{\substack{i, j=1 \\ i<j}}^{m} (e_{ij} + e_{ji}) E_{ij}'.$$

The amount of information which the design T contains on the parameter system $K'\theta$ is captured by the information matrix for $K'\theta$,

$$C_k(M(\tau)) = (K'K)^{-1} K' M(\tau) K (K'K)^{-1} \in NND(s).$$

Defining $L = (K'K)^{-1}K'$, we get the following information matrix:

$$C_k(M(\tau)) = LM(\tau)L'. \tag{7}$$

A convex combination

$$\eta(\alpha) = \sum_{j=1}^{m} \alpha_j \eta_j, \text{ with } \alpha = (\alpha_1, \alpha_2, ..., \alpha_m)' \in T \tag{8}$$

is called a *weighted centroid design* with weight vector α, where $\sum_{i=1}^{m} \alpha_i = 1$.

In this study, we denote the set of all weighted centroid designs by $\eta(\alpha)$.

A necessary and sufficient condition for ϕ_p*-optimality* of a weighted centroid design $\eta(\alpha)$ with weight vector $\alpha = (\alpha_1, ..., \alpha_m)' \in T_m$ follows from the Kiefer-Wolfowitz equivalence theorem in Pukelsheim [11] and given by Klein [7]. Suppose $\eta(\alpha)$ satisfies the side condition $C_k(M(\eta(\alpha))) \in PD(s)$ and C_j written as $C_j = C_k(M(\eta_j))$ for $j = 1, ..., m$. Then, $\eta(\alpha)$ solves problem (4) with $p \in (-\infty, 1]$ if and only if

$$\operatorname{trace} C_j C_k(M(\eta(\alpha)))^{p-1} \begin{cases} = \operatorname{trace} C_k(M(\eta(\alpha)))^p & \text{for all } j \in \delta(\alpha), \\ \leq \operatorname{trace} C(M(\eta(\alpha)))^p & \text{otherwise} \end{cases} \tag{9}$$

with $\delta(\alpha) := \{j \backslash \alpha_j > 0\}$. The case $p = -\infty$, that is, E-optimality, has a similar optimality condition, Klein [7].

Klein [8] in Lemma 3.1 shows that an H-invariant symmetric matrix has seven distinct entries at most.

Then any matrix $C = sym(s, H)$ can be uniquely represented in the form

$$C = \begin{pmatrix} C_{11} & C_{12} \\ C_{12}' & C_{22} \end{pmatrix}. \tag{10}$$

3.1. *A*-optimal weighted centroid designs

Utilizing condition (9), a weighted centroid design $\eta(\alpha)$ is *A*-optimal for $K'\theta$ if and only if

$$\operatorname{trace}(C_j C(\alpha)^{-2}) \begin{cases} = \operatorname{trace}(C(\alpha)^{-1}) & \text{for } j \in \{1, 2\}, \\ < \operatorname{trace}(C(\alpha)^{-1}) & \text{otherwise.} \end{cases} \tag{11}$$

3.1.1. *A*-optimal weighted centroid design with two ingredients

Theorem 3.1.1. *In the second-degree Kronecker model for mixture experiments with* $m = 2$ *ingredients*, *the unique A-optimal design for* $K'\theta$ *is*

$$\eta(\alpha^A) = 0.673368365\,\eta_1 + 0.326631635\,\eta_2.$$

The maximum value of the A-criterion for $K'\theta$ *in m ingredients is* $v(\phi_{-1}) =$ 0.320064674.

Proof. Let $\alpha = (\alpha_1, \alpha_2, 0, ..., 0)' \in T_m$ be a weight vector with $\partial(\alpha) = \{1, 2\}$. Suppose $\eta(\alpha)$ is *A*-optimal for $K'\theta$ in *T*. Let $C(\alpha) = C_k(M(\eta(\alpha)))$.

By applying equations (6), (7) and (8), and substituting the moment matrix for $m = 2$ (*M* as in Klein [7]) when $j = 1$ and when $j = 2$, we get

$$C_k = C_k(M(\eta(\alpha))) = \begin{bmatrix} \frac{8\alpha_1 + \alpha_2}{16} & \frac{\alpha_2}{16} & \frac{\alpha_2}{4} \\ \frac{\alpha_2}{4} & \frac{8\alpha_1 + \alpha_2}{16} & \frac{\alpha_2}{4} \\ \frac{\alpha_2}{4} & \frac{\alpha_2}{4} & \alpha_2 \end{bmatrix}, \tag{12}$$

the corresponding information matrix for $m = 2$ ingredients.

Utilizing condition (11) for $j = 1$, the inverse of equation (12), and $C(\alpha)^{-2} = [C(\alpha)^{-1}]^2$, and putting the simplex restriction, $\alpha_1 + \alpha_2 = 1$, we obtain $13\alpha_1^2 - 34\alpha_1 + 17 = 0$.

Upon solving, we get $\alpha_1 = 0.673368365$ since $\alpha_1 \in (0, 1)$.

Similarly, for $j = 2$, we obtain $13\alpha_2^2 + 8\alpha_2 - 4 = 0$.

Upon solving, we get $\alpha_2 = 0.326631634$ since $\alpha_1 \in (0, 1)$.

Thus the A-optimal weighted centroid design is

$$\eta(\alpha^A) = \alpha_1\eta_1 + \alpha_2\eta_2 = 0.673368365\,\eta_1 + 0.326631635\,\eta_2.$$

To obtain the optimal value for 2 ingredients, we adopt the definition of average-variance criterion $v(\phi_{-1}) = \left(\frac{1}{s}\operatorname{trace} C(\alpha)^{-1}\right)^{-1}$, where $s = \binom{m+1}{2}$ as provided in Pukelsheim [11]. That is, for m-2 ingredients, we have

$$v(\phi_{-1}) = \left(\frac{1}{\binom{m+1}{2}}\operatorname{trace} C(\alpha)^{-1}\right)^{-1} = \left(\frac{1}{3}\,\frac{4\alpha_1 + 17\alpha_2}{4\alpha_1\alpha_2}\right)^{-1}$$

$$= (3.124368545)^{-1} = 0.320064674.$$

3.1.2. *A*-optimal weighted centroid design with three ingredients

Theorem 3.1.2. *In the second-degree Kronecker model for mixture experiments with $m = 3$ ingredients, the unique A-optimal design for $K'\theta$ is*

$$\eta(\alpha^A) = 0.606470182\,\eta_1 + 0.393529818\,\eta_2.$$

The maximum value of the A-criterion for $K'\theta$ in m ingredients is $v(\phi_{-1}) =$ 0.232298577.

Proof. Let $\alpha = (\alpha_1, \alpha_2, 0, \ldots, 0)' \in T_m$ be a weight vector with $\partial(\alpha) = \{1, 2\}$. Suppose $\eta(\alpha)$ is A-optimal for $K'\theta$ in T. Let

$$C(\alpha) = C_k(M(\eta(\alpha))).$$

By applying equations (6), (7) and (8), and substituting the moment matrix for $m = 3$ (M as in Klein [7]) when $j = 1$ and when $j = 2$, we get

$$C_k = C_k(M(n(\alpha)))$$

$$= \begin{bmatrix} \frac{8\alpha_1 + \alpha_2}{24} & \frac{\alpha_2}{48} & \frac{\alpha_2}{48} & \frac{\alpha_2}{8} & \frac{\alpha_2}{8} & 0 \\ \frac{\alpha_2}{48} & \frac{8\alpha_1 + \alpha_2}{24} & \frac{\alpha_2}{48} & \frac{\alpha_2}{8} & 0 & \frac{\alpha_2}{8} \\ \frac{\alpha_2}{48} & \frac{\alpha_2}{48} & \frac{8\alpha_1 + \alpha_2}{24} & 0 & \frac{\alpha_2}{8} & \frac{\alpha_2}{8} \\ \frac{\alpha_2}{8} & \frac{\alpha_2}{8} & 0 & \frac{3\alpha_2}{4} & 0 & 0 \\ \frac{\alpha_2}{8} & 0 & \frac{\alpha_2}{8} & 0 & \frac{3\alpha_2}{4} & 0 \\ 0 & \frac{\alpha_2}{8} & \frac{\alpha_2}{8} & 0 & 0 & \frac{3\alpha_2}{4} \end{bmatrix}, \quad (13)$$

the corresponding information matrix for $m = 3$ ingredients.

Utilizing condition (11) for $j = 1$, the inverse of equation (13), and $C(\alpha)^{-2} = [C(\alpha)^{-1}]^2$, and putting the simplex restriction, $\alpha_1 + \alpha_2 = 1$, we obtain $11\alpha_1^2 - 38\alpha_1 + 19 = 0$.

Upon solving, we get $\alpha_1 = 0.606470182$ since $\alpha_1 \in (0, 1)$.

Similarly, for $j = 2$, we obtain, $11\alpha_2^2 + 16\alpha_2 - 8 = 0$.

Upon solving, we get $\alpha_2 = 0.393529818$ since $\alpha_1 \in (0, 1)$.

Thus the A-optimal weighted centroid design is

$$\eta(\alpha^A) = \alpha_1\eta_1 + \alpha_2\eta_2 = 0.606470181\,\eta_1 + 0.393529818\,\eta_2.$$

To obtain the optimal value for 3 ingredients, we adopt the definition of average-variance criterion $v(\phi_{-1}) = \left(\frac{1}{s}\operatorname{trace} C(\alpha)^{-1}\right)^{-1}$, where $s = \binom{m+1}{2}$ as provided in Pukelsheim [11]. That is, for m-3 ingredients, we have

$$v(\phi_{-1}) = \left(\frac{1}{\binom{m+1}{2}}\operatorname{trace} C(\alpha)^{-1}\right)^{-1} = \left(\frac{1}{6}\,\frac{8\alpha_1 + 19\alpha_2}{2\alpha_1\alpha_2}\right)^{-1}$$

$$= (4.304804668)^{-1} = 0.232298577.$$

3.2. *D*-optimal weighted centroid designs

Utilizing condition (9), a weighted centroid design $\eta(\alpha)$ is *D*-optimal for $K'\theta$ if and only if

$$\operatorname{trace}(C_j C(\alpha)^{-1}) \begin{cases} = \operatorname{trace}(C(\alpha)^0) & \text{for } j \in \{1, 2\}, \\ < \operatorname{trace}(C(\alpha)^0) & \text{otherwise.} \end{cases} \tag{14}$$

3.2.1. *D*-optimal weighted centroid design with two ingredients

Theorem 3.2.1. *In the second-degree Kronecker model for mixture experiments with* $m = 2$ *ingredients, the unique D-optimal design for* $K'\theta$ *is*

$$\eta(\alpha^A) = \frac{2}{3}\eta_1 + \frac{1}{3}\eta_2.$$

The maximum value of the A-criterion for $K'\theta$ *in m ingredients is* $v(\phi_{-1}) = \frac{1}{3}$.

Proof. Using the inverse of equation (12) and the respective information matrix when $j = 1$ together with condition (14), we get

$$\operatorname{trace}(C_1 C(\alpha)^{-1}) = \operatorname{trace}(C(\alpha)^0) \Leftrightarrow \frac{2}{\alpha_1} = 3, \text{ meaning that } \alpha_1 = \frac{2}{3}.$$

Similarly, utilizing the inverse of equation (12) and the respective information matrix when $j = 2$ together with condition (14), we get

$$\operatorname{trace}(C_2 C(\alpha)^{-1}) = \operatorname{trace}(C(\alpha)^0) \Leftrightarrow \frac{1}{\alpha_1} = 3, \text{ meaning that } \alpha_1 = \frac{1}{3}.$$

Substituting these values to equation (12) and taking the determinant, we get $\det(C(\alpha)) = 0.037037037$.

The maximum value of the *D*-criterion for $m = 2$ ingredients is

$$V(\phi_0) = (\det(C(\alpha)))^{\frac{1}{3}} = (0.037037037)^{\frac{1}{3}} = \frac{1}{3}.$$

3.2.2. *D*-optimal weighted centroid design with three ingredients

Theorem 3.2.2. *In the second-degree Kronecker model for mixture experiments with* $m = 3$ *ingredients*, *the unique D-optimal design for* $K'\theta$ *is*

$$\eta(\alpha^{A}) = \frac{1}{2}\eta_1 + \frac{1}{2}\eta_2.$$

The maximum value of the A-criterion for $K'\theta$ *in m ingredients is* $v(\phi_{-1}) = \frac{1}{4}$.

Proof. Using the inverse of equation (13) and the respective information matrix when $j = 1$ together with condition (14), we get

$$\operatorname{trace}(C_1 C(\alpha)^{-1}) = \operatorname{trace}(C(\alpha)^{0}) \Leftrightarrow \frac{3}{\alpha_1} = 6, \text{ meaning that } \alpha_1 = \frac{1}{2}.$$

Similarly, utilizing the inverse of equation (13) and the respective information matrix when $j = 2$ together with condition (14), we get

$$\operatorname{trace}(C_2 C(\alpha)^{-1}) = \operatorname{trace}(C(\alpha)^{0}) \Leftrightarrow \frac{3}{\alpha_2} = 6, \text{ meaning that } \alpha_2 = \frac{1}{2}.$$

Substituting these values to equation (13) and taking the determinant, we get

$$\det(C(\alpha)) = 0.0002441406.$$

The maximum value of the *D*-criterion for $m = 3$ ingredients is

$$V(\phi_0) = (\det(C(\alpha)))^{\frac{1}{6}} = (0.0002441406)^{\frac{1}{6}} = \frac{1}{4}.$$

4. Efficiency

The problem in equation (4) calls for maximizing information as measured by the information function ϕ, in the set M of competing moment matrices. The *optimal value* of this problem is, by definition,

$$V(\phi) = \sup_{M \in \mathrm{M}} \phi(C_K(M)).$$

A moment matrix $M \in \mathrm{M}$ is said to be *formally* ϕ*-optimal for* $K'\theta$ in M

when $\phi(C_K(M))$ attains the optimal value $V(\phi)$. If, in addition, the matrix *M* lies in the feasibility cone $A(K)$, then *M* is called ϕ-*optimal for* $K'\theta$ in *M*, Pukelsheim [12].

However, an optimal design is not an end in itself, but an aid to identifying efficient practical designs. The appropriate notation for efficiency is as follows:

Definition. The ϕ-*efficiency* of a design $\xi \in E$ is defined by

$$\phi - eff\,\xi = \frac{\phi(C_K(M(\xi)))}{V(\phi)}.$$

It is a number between 0 and 1, and gives the extent (often quoted in percent) to which the design ξ exhausts the maximum information $V(\phi)$ for $K'\theta$ in *M*, Pukelsheim [12].

We now turn to the designs, we obtained in Section 3 to illustrate their efficiencies.

Example 4.1. The *A*- and *D*- efficiencies for $m = 3$ ingredients.

The maximum value of the *A*-criterion for $K'\theta$ in three ingredients is 0.0641. Thus,

$$\phi - eff\,\xi = \frac{\phi(C_K(M(\xi)))}{V(\phi_{-1})} = \frac{0.0641}{0.2323} = 0.2759.$$

Similarly, the maximum value of the *D*-criterion for $K'\theta$ in three ingredients is 0.0833. Thus,

$$\phi - eff\,\xi = \frac{\phi(C_K(M(\xi)))}{V(\phi_0)} = \frac{0.0833}{0.2500} = 0.3332.$$

5. Discussions and Conclusions

The parameter subsystem considered in this study is the maximal parameter subsystem. The optimality criteria used were the average-variance and the determinant criteria. It is worth noting that maximizing the average-variance and the determinant of the information matrix is appropriate. The optimal values obtained have been found to be larger than the ones obtained in the previous studies. This is attributed to the scaling factor which we included while developing the coefficient matrix. These large values indicate that the information matrices of these designs carry large information. This is always the goal of every experimenter and it is the main result of this paper. Furthermore, the efficiencies also indicate that these designs perform about three times more than the earlier designs. This therefore is an appealing result for these designs. The details on the quadratic subspace can be found in the previous studies.

Finally, in this paper, results for two and three ingredients were derived, a general result for any m ingredients is worth investigating. An obvious question is to change the regression function from the Kronecker square to the Kronecker cube $f(t) = t \otimes t \otimes t$.

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Appendix I

Parameter subsystems of interest:

When $m = 2$,

$$K'\theta = \begin{Bmatrix} \theta_{11} \\ \theta_{22} \\ \dfrac{\theta_{12} + \theta_{21}}{2} \end{Bmatrix}.$$

When $m = 3$,

$$K'\theta = \begin{Bmatrix} \theta_{11} \\ \theta_{22} \\ \theta_{33} \\ \dfrac{\theta_{12} + \theta_{21}}{6} \\ \dfrac{\theta_{13} + \theta_{31}}{6} \\ \dfrac{\theta_{23} + \theta_{32}}{6} \end{Bmatrix}.$$