A -Optimal Designs for Third-Degree Kronecker Model Mixture Experiments

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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 study investigates A -optimal designs in the Third- Degree Kronecker model. Based on the completeness result, the considerations are restricted to weighted centroid designs. First, the coefficient matrix and the associated parameter subsystem of interest using the unit vectors and a characterization of the feasible weighted centroid design for a maximal parameter subsystem is obtained. The parameter subspace of interest in this study is non-maximal parameter subsystem which is subspace of the full parameter space. Optimal designs of mixture experiments are derived by employing the Kronecker model approach and applying the various optimality criteria.

Keywords: *Mixture experiments, Kronecker product, optimal designs, Weighted centroid designs, Optimality criteria, Moment and information matrices, Efficiency.*

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 (1990) lists numerous examples and provides a thorough discussion of both theory and practice. Early seminal work was done by Scheffe' (1958, 1963) 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.

The work done by Draper and Pukelsheim (1998) is being extended to polynomial regression model for third-degree mixture model, whereby the S-polynomial and the expected response takes the form

$$E[Y_t] = f(t)'\theta = \sum_{i=1}^{m} t_i\theta_i + \sum\sum_{i<j}^{m} t_i t_j \theta_{ij} + \sum\sum\sum_{i<j<k}^{m} \theta_{ijk} t_i t_j t_k \quad \ldots \ldots \ldots \text{(1)}$$

and when the regression function is the homogeneous third-degree K-polynomial, the expected response takes the form

$$E[Y_t] = f(t)'\theta = \sum_{i=1}^{m}\sum_{j=1}^{m}\sum_{k=1}^{m} t_i t_j t_k \theta_{ijk} = (t \otimes t \otimes t)'\theta \quad \ldots \ldots \ldots \text{(2)}$$

in which the Kronecker powers $t^{\otimes 3} = (t \otimes t \otimes t)$, $(m^3 \times 1)$ vectors, consists of pure cubic and three-way interactions of components of *t* in lexicographic order of the subscripts and with evident that third-degree restrictions are $\theta_{ijk} = \theta_{ikj} = \theta_{jik} = \theta_{jki} = \theta_{kij} = \theta_{kji}$ for all i, j, and k.

All observations taken in an experiment are assumed to be uncorrelated and to (0,have∞)common. va

Draper and Pukelsheim (1998) put forward several advantages of the Kronecker model, for example, a more compact notation, more convenient invariance properties and the homogeneity of regression terms.

The moment matrix $M(\tau) = \int_{\tau} f(t) f(t)' d\tau$ for the Kronecker model of degree three has all entries homogeneous of degree six. This matrix reflects the statistic

Pukelsheim (1993) gives a review of the general design environment. Klein (2002) showed that the class of weighted centroid designs is essentially c ordering Cheng, S. C. (1995). As a consequence the search for
optimal designs may be restricted to weighted centroid designs for most criteria. For particular criteria applied to mixture experiments Kiefer (1959, 1975, and 1978) and Galil and Kiefer (1977). All these authors have concentrated their work on the second degree Kronecker model. Korir et al (2009) extended the work to Third degree Kronecker model simple designs .The present work now determines optimal designs for a maximal subsystem of parameters in the third degree Kronecker model. The Keifer's φ_p functions will serve as optimality criteria.

Design problem

Consider canonical unit vectors in i.e. $e_1, e_2, \ldots, e_m$ and set $e_{iij} = e_i e_i e_j$, $e_{ijk} = e_i e_j e_k$ for $i<j<k$, $i,j,k=\{1\ 2, \ldots, m\}$.

Defining the matrix

$$K = (K_1 \, ; K_2) \in \Re^{m_3 \times (m+1)}$$

Where,

$$K_1 = \sum_{i=1}^{m} e_{iii} \, e_i',$$

and

$$K_2 = \frac{1}{(m^3 - m)} \sum_{i,\, j=1}^{m} (e \quad + e \quad + e_{jii}) + \sum_{\substack{i,\, j,k=1 \\ i \neq j \neq k}}^{m} (e \quad)$$

Further define

$$L = (K'K)^{-1} K'$$

So that

$$C_k(M(\tau)) = LM(\tau)L'$$

As is evident from model equation (2), the Kronecker model's full parameter vector $\theta \in \Re^{m_3}$ is not estimable. When fitting this model, the parameter subsystem considered in this study can be written as

$$(\theta_{iii})_{1 \le i \le m}$$

$$K'\theta = \frac{1}{m^3 - m} \sum_{\substack{i,j=1 \\ i \ne j}}^{m} (\theta_{iij} + \theta_{iji} + \theta_{jii}) + \sum_{\substack{i,j,k=1 \\ i \ne j \ne k}}^{m} (\theta_{ijk}) \in \Re^{(m+1)}$$

for all $\theta \in \Re^{m^3}$

where $K \in \Re^{m^3 \times (m+1)}$

The parameter subsystem $K'\theta$ of interest is a non-maximal parameter system in model (2).

The amount of information a design t contains on K' is captured by the information matrix

$$C_k(M(\tau)) = \min\{LM(\tau)L'\} ; \Re^{(m+1)\times(m+1)}$$

The information matrix $C_k(M(\tau))$ is the precision linear unbiased estimator for $K'\theta$ under design τ, Pu (1993, chapter 3). In the present case information matrices for $K'\theta$
takes a particular simple form:

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

Thus the information matrices for the moment matrices.

Optimality Criteria

The most prominent optimality criteria in the design of experiments are the determinant criterion, φ_0, the average-variance criterion, $_{-1}$, the smallest eigenvalue criterion, $\varphi_{-\infty}$ and the trace criterion, φ_1.

These are a particular cases of the matrix means φ_p with parameter p

[-∞;1].

The optimality properties of designs are determined by their moment matrices (Pukelsheim 1993, chapter 5). We compute optimal design for the polynomial fit model, the third degree Kronecker model. This involves searching for the optimum in a set

of competing moment matrices. The matrix means φ_p which are information functions (Pukelsheim (1993)) we utilized in this study.

The amount of information inherent to $C_k(M(\ ))$ is provided by Kiefers φ_p-criteria with $C_k(M(\tau))PD(m+1)$.

These are defined by:

$$\varphi_p(C) = \begin{cases} \lambda_{\min}(C) & \text{if } p = -\infty \\ \det(C)^{\frac{1}{s}} & \text{if } p = 0 \\ \left[\frac{1}{s}\operatorname{trace} C^p\right]^{\frac{1}{p}} & \text{if } p \in [-\infty;1] \setminus \{0\} \end{cases}$$

for all C in $PD(m+1)$, the set of positive definite $(m+1)\times(m+1)$ matrices, where $\lambda_{\min}(C)$ refers to the smallest eigenvalue of C. By definition $\varphi_p(C)$ is a scalar measure which is a function of the eigenvalues of C for all p [-∞;1]. (Pukelsheim 2006, chapter 6). The class of φ_p-criteria includes the prominently used T-, D-, A- and E-criteria corresponding to parameter values 1, 0, -1 and -∞ respectively.

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

Maximize $\varphi_p(C_k(M(\tau)))T$ with τ

Subject to $C_k(M(\tau))PD(m+1$

Theorem 1.0

Let $\alpha \in T_m$ be the weight vector of a weighted centroid design $\eta(\alpha)$ which is feasible for $K'\theta$ and let $\partial()\alpha$ be a set of active indices. Furthermore let $C_j = C_k(M(\eta_j))$ for $j=(1, 2, \ldots, m)$ for all p (-∞;1]. Then $\eta(\alpha)$ is $\varphi_p - optimal$ for $K'\theta$ in T if and only if;

$$traceCCM(((())) \eta\alpha^{p-1}_{jk} \quad =traceCMforallj \in \partial((()))()\eta\alpha\alpha^{p}_{k} \quad \leq traceCMotherwise_k((()))\eta\alpha^{p}$$

Klein (2002).

Weighted centroid designs are exchangeable, that is, they are invariant under permutations of ingredients.

Optimal Weighted Centroid Designs

A convex combination, $\eta(\alpha) = \sum_{j=1}^{m} \alpha_j \eta_j$, with $\alpha = (\alpha_1, \ldots, \alpha_m)' \in T_m$, is called a weighted centroid design with weight vector restricted by $\sum_{i=1}^{m} \alpha_i = 1$. These designs were introduced by Scheffe' (1963). Weighted centroid designs are exchangeable, that is they are invariant under permutations Klein (2002).

Klein (2002) summarized the work by Draper and Heiligers (1999) and Draper, Heiligers and Pukelsheim (2000) by putting forward an idea that affirms the importance of weighted centroid design for the Kronecker model. The researcher proved that, in the second degree Kronecker model for mixture experiments with m 2 ingredients, the set of weighted centroid designs is an essentially complete class. That is, for every p [-∞;1] and for every design $\tau \in T$ there exists a weighted centroid design with

$$(\varphi_p \ C_k \ \ M)(\eta) \geq (\varphi_p \ C_k \ \ M)(\tau).$$

Thus for every design $\tau \in T$ there is a weighted centroid design whose moment matrix M( ) improves upon M( ) in the Kiefer ordering Draper, Heiligers and Pukelsheim (1998).

Under the Kiefer ordering, we say a moment matrix M is more informative than a moment matrix N if M is greater than or equal to some intermediate matrix F under the loewner ordering, and F is majorized by N under the group that leaves the problem invariant:

$M >> N \Leftrightarrow M >> F N$ for some matrix F.

For the information matrix obtained, we show that the matrix is an improvement of a given design in terms of increasing symmetry, as well as obtaining a larger moment matrix under the Loewner ordering. These two criteria show that the information matrix obtained is Kiefer optimal for K’ , the parameter subsystem of interest.

Information Matrices

Information matrices for subsystems of mean parameters in a classical linear model are derived. First, the coefficient matrix, K, is obtained, which will be used to identify the linear parameter subsystems $K'\theta$ of interest .Hence this will be utilized in generating the associated information matrices C_k for *m* factors. The information matrices so obtained will be useful in obtaining the optimality criteria. As an illustration the information matrices for three factors can be derived as follows:

Information matrices for three ingredients

The information matrix for three ingredients for a mixture experiment is given by

C_{kk} CMn==((())) α

32ααα+αα
12222
9619219216
ααααα32 +
21222
1929619216
ααααα 32 +
22122
αααα 9
2222
16161616

Proof

First the coefficient matrix, *K*, for m=3 is derived as follows

$$K_1 = \sum_{i=1}^{3} e_{iii}\, e_i' = e_{111}e_1' + e_{222}e_2' + e_{333}e_3' \text{ , and}$$

$$K_2 = \frac{1}{(3^3-3)} \sum_{\substack{i,j=1 \\ i\neq j}}^{3} \left(e_{iij} + e_{iji} + e_{jii}\right) + \sum_{\substack{i,j,k=1 \\ i\neq j\neq k}}^{3} \left(e_{ijk}\right) = e_{112} + e_{121} + e_{211} + e_{113} + e_{131} + e_{311}$$
$$+ e_{221} + e_{212} + e_{122} + e_{223} + e_{232} + e_{322}$$
$$+ e_{331} + e_{313} + e_{133} + e_{332} + e_{323} + e_{233}$$
$$+ e_{123} + e_{132} + e_{213} + e_{231} + e_{312} + e_{321}$$

Define, $e_{iij} = e_i \otimes e_i \otimes e_j$, $e_{ijk} = e_i \otimes e_j \otimes e_k$ i,j=1,2,3 ,

$e_1 = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, e_2 = \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}$, and $e_3 = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}$.

$L = (K'K)^{-1} K'$.

1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0
0 0 0 0 0 0 0 0 0 0 0 0 0 1 0 0 0 0 0 0 0 0 0 0 0 0 0
0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 1

0
1 1 1 1 1 1 1 1 1 1 1 1 0 1 1 1 1 1 1 1 1 1 1 1 1 0

For the design η_1 , the information matrix is given by

$$C_1 = \begin{pmatrix} \frac{1}{3} & 0 & 0 & 0 \\ 0 & \frac{1}{3} & 0 & 0 \\ 0 & 0 & \frac{1}{3} & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} = C_k(M(\eta_1))$$

While that of design η_2 is given by

$$C_2 = LM(\eta_2)L' = \begin{pmatrix} \frac{1}{96} & \frac{1}{192} & \frac{1}{192} & \frac{1}{16} \\ \frac{1}{192} & \frac{1}{96} & \frac{1}{192} & \frac{1}{16} \\ \frac{1}{192} & \frac{1}{192} & \frac{1}{96} & \frac{1}{16} \\ \frac{1}{16} & \frac{1}{16} & \frac{1}{16} & \frac{9}{16} \end{pmatrix} = C_k(M(\eta_2))$$

$$C_k = C_k(M(\eta(\alpha))) = \begin{pmatrix} \frac{32\alpha_1+\alpha_2}{96} & \frac{\alpha_2}{192} & \frac{\alpha_2}{192} & \frac{\alpha_2}{16} \\ \frac{\alpha_2}{192} & \frac{32\alpha_1+\alpha_2}{96} & \frac{\alpha_2}{192} & \frac{\alpha_2}{16} \\ \frac{\alpha_2}{192} & \frac{\alpha_2}{192} & \frac{32\alpha_1+\alpha_2}{96} & \frac{\alpha_2}{16} \\ \frac{\alpha_2}{16} & \frac{\alpha_2}{16} & \frac{\alpha_2}{16} & \frac{9\alpha_2}{16} \end{pmatrix}$$

This is the desired information matrix for three ingredients.

A-optimal design for m=3 ingredients

Lemma 4.1

In the third-degree Kronecker model for mixture experiments with three ingredients, the unique A-optimal weighted centroid design for $K'\theta$ is

$$\eta\left(\alpha^{(A)}\right) = \alpha_1\eta_1 + \alpha_2\eta_2 = 0.691545741\eta_1 + 0.308454259\eta_2$$

The maximum value of the A-criterion for $K'\theta$ in three ingredients is

$$V(\varphi_{-1}) = 0.211881792$$

Proof

The inverse of information matrix for three factors is

$$C_k^{-1} = \begin{bmatrix} a & b & b & c \\ b & a & b & c \\ b & b & a & c \\ c & c & c & d \end{bmatrix}$$

where, $a = \dfrac{192\alpha_1 + \alpha_2}{\alpha_1(64\alpha_1 + \alpha_2)}$, $b = \dfrac{\alpha_2}{\alpha_1(64\alpha_1 + \alpha_2)}$, $c = \dfrac{-1}{3\alpha_1}$, and $d = \dfrac{16\alpha_1 + \alpha_2}{9\alpha_1\alpha_2}$.

, and

from $[C(M(\eta(\alpha)))]^{-2} = [C(m(\eta(\alpha)))^{-1}]^2 = [C(\alpha)]^{-2}$, we get

$$C_k^{-2} = \begin{bmatrix} a^2+2b^2+c^2 & 2ab+b^2+c^2 & 2ab+b^2+c^2 & ac+2bc+cd \\ 2ab+b^2+c^2 & a^2+2b^2+c^2 & 2ab+b^2+c^2 & ac+2bc+cd \\ 2ab+b^2+c^2 & 2ab+b^2+c^2 & a^2+2b^2+c^2 & ac+2bc+cd \\ ac+2bc+cd & ac+2bc+cd & ac+2bc+cd & 3c^2+d^2 \end{bmatrix} \quad (56)$$

For j=1, the design is A-optimal if and only if

$$trace\, C(C(M(\eta(\alpha))))^{-1}_k = trace\, C(M(\eta(\alpha)))^{-2}_k .$$

Thus, we have,

$$C_1 C_k^{-2} = \begin{pmatrix} \frac{a^2+2b^2+c^2}{3} & \frac{2ab+b^2+c^2}{3} & \frac{2ab+b^2+c^2}{3} & \frac{ac+2bc+cd}{3} \\ \frac{2ab+b^2+c^2}{3} & \frac{a^2+2b^2+c^2}{3} & \frac{2ab+b^2+c^2}{3} & \frac{ac+2bc+cd}{3} \\ \frac{2ab+b^2+c^2}{3} & \frac{2ab+b^2+c^2}{3} & \frac{a^2+2b^2+c^2}{3} & \frac{ac+2bc+cd}{3} \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

giving,

$$trace\,C_1 C_k^{-2} = \frac{a^2+2b^2+c^2}{3} + \frac{a^2+2b^2+c^2}{3} + \frac{a^2+2b^2+c^2}{3} + 0$$

$$= a^2+2b^2+c^2$$

Therefore,

$$trace(C_1 C_k M(\eta(\alpha)))\,(trace(C_k M(\eta(\alpha))))^{-2} =$$

$$\Leftrightarrow \left(\frac{192\alpha_1+\alpha_2}{\alpha_1(64\alpha_1+\alpha_2)}\right)^2 \quad \left(\frac{\alpha_2}{\alpha_1(64\alpha_1+\alpha_2)}\right)^2 \quad \left(\frac{-1}{3\alpha_1}\right)^2 \quad \frac{192\alpha_1+\alpha_2}{\alpha_1(64\alpha_1+\alpha_2)} \quad \frac{16\alpha_1+\alpha_2}{9\alpha_1\alpha_2},$$

which reduces to

$$265{,}356\alpha_1^4 - 656208\alpha_1^3 + 321{,}816\alpha_1^2 + 3472\alpha_1 + 28 = 0,$$

Solving this polynomial together with $\alpha_1+\alpha_2 = 1$ for $\alpha_1 \in (0,1)$ yields

$$\alpha_1 = 0.691545741$$

Similarly, for j=2, is

$$C C_{2k}^{-2} = \begin{pmatrix} a'' & b'' & c'' \\ b'' & a'' & c'' \\ c'' & c'' & d'' \end{pmatrix},$$

where $a' = \frac{1}{96}\left(\ldots 222 \ldots\right)$

$b' = \frac{1}{192}\left(\ldots 222 \ldots\right)$

$c' = \frac{1}{48}\left(c b + c e + \ldots + d c d \ldots 293 \ldots 22\right)$, (cacb′=cc++++dcd293,)

$d' = \frac{1}{16}\left(a b + c a b d (4349(2)) \ldots 222\right)$, and (dabc=ab+cabd(4349(2))+++++)

$e' = \frac{3}{16}\left(c b + c e + \ldots + d c d \ldots 293 \ldots 22\right)$. (eacb′=cc++++dcd293)

$$trace\, C C_{2k}^{-2} = a + a + e' = \ldots 3$$

traceCCaaaeae=+++′=+″″3

$$= \frac{13}{96\,16} \ldots 22222 \ldots$$

=+332262293++++++++++abcabcabdacbccdcd

$$= \frac{1}{32}\left\{ \ldots 2222 \ldots \right\}$$

=+++++++abcdabcabd356182122

$$\Rightarrow trace\, C C_{2k}^{-2} = \frac{1}{32}\left\{ \ldots 22222 \ldots \right\} \qquad \ldots \textbf{(60)}$$

⇒=+++++++traceCCabcdabcabd 356182122

Therefore,

$$trace\,C(M(\eta_\alpha)) \; trace\,C(M(\eta_\alpha))^{-2} = \ldots \quad {}_{2}\;{}_{kk}$$

$$\Leftrightarrow \frac{1}{32}\left\{a^2 + 3b^2 + 56c^2 + 18d^2 + 2ab + 12c\left(a + 2b + d\right)\right\} = 3a + d$$

$\Leftrightarrow$ 19216192 αααααααα 12212122 +++ ++++ 2222 356182 αααααααααααα(64)(64)39(64)+++ 112112112112 -1 22 $_2$which

++12323+= -1 3(64)(64)9(αααααααααα++ 1112112121 192216192αααα 1221212 +++ααα 64)9αααα+ 1212 + $16\alpha\alpha_{12}+$

reduces to

$$265{,}356\alpha_2^4 - 405{,}216\alpha_2^3 - 54{,}672\alpha_2^2 + 260{,}096\alpha_2 - 65536 = 0$$

Solving this polynomial $\alpha_1 + \alpha_2 = 1$ for $\alpha_2 \in (0,1)$ yields

$$\alpha_2 = 0.308454259$$

Therefore,

$$\eta\left(\alpha^{(A)}\right) = \alpha_1\eta_1 + \alpha_2\eta_2 = 0.691545741\eta_1 + 0.308454259\eta_2$$

is the unique A-optimal weighted centroid design for m=3 ingredients.

The average-criterion is given by

$$v(\varphi_{-1}) = \left[\frac{1}{s}\,trace\,C(\alpha)^{-1}\right]^{-1},$$

where $s = m + 1$.

$$traceC_k^{-1} = \frac{1024\alpha_1^2 + 5264\alpha_1\alpha_2 + 28\alpha_2^2}{9\alpha_1\alpha_2(64\alpha_1 + \alpha_2)}$$

implies that,

$$\frac{1}{4}\left(\frac{1024\alpha_1^2 + 5264\alpha_1\alpha_2 + 28\alpha_2^2}{9\alpha_1\alpha_2(64\alpha_1 + \alpha_2)}\right)^{-1} = \frac{1}{4}(18.878450980)^{-1} = 0.211881790$$

Therefore, the maximum value of the A-criterion for $K'\theta$ in m=3 ingredients is

$V(\varphi)^{-1} =$ 0.211881792.

A numerical example using artificial sweetener experiment of three components mixture experiment

The A optimal design for three factors can now be applied to three factor numerical example .In these study only pure blends and binary blends are considered where the average score is the response.

Consider the following simplex centroid design for three ingredients as the initial design.

Design points	t_1	t_2	t_3	average score
1	1	0	0	10.40
2	0	1	0	6.16

3	0	0	1	3.90
4	$\frac{1}{2}$	$\frac{1}{2}$	0	14.97
5	$\frac{1}{2}$	0	$\frac{1}{2}$	12.17
6	0	$\frac{1}{2}$	$\frac{1}{2}$	12.27

Where t_1=glycine,t_2=saccharin and t_3=enhancer

$$\eta_1 = \left\{ (1,0,0),\ (0,1,0),\ (0,0,1) \right\}, \quad \eta_2 = \left\{ \left(\tfrac{1}{2},\tfrac{1}{2},0\right),\ \left(\tfrac{1}{2},0,\tfrac{1}{2}\right),\ \left(0,\tfrac{1}{2},\tfrac{1}{2}\right) \right\}$$

Implying that, the unique A-optimal weighted centroid design for $K'\theta$ in m=3 ingredients is $\eta\left(\alpha^{(A)}\right)=\alpha_1\eta_1+\alpha_2\eta_2=0.691545741\eta_1+0.308454259\eta_2$ as shown above. Therefore the corresponding A-optimal for the above designs is as follows.

Design points	t_1	t_2	t_3
1	0.69154574	0	0
2	0	0.69154574	0
3	0	0	0.69154574

4	0.34577287	0.34577287	0
5	0.34577287	0	0.34577287
6	0	0.34577287	

0.34577287

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