KIEFER ORDERING OF SECOND-DEGREE MIXTURE DESIGNS FOR FOUR INGREDIENTS

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1 Introduction

Many practical problems are associated with the investigation of mixture ingredients $t_1, t_2, \ldots, t_m$ of m factors, with $t_i \geq 0$ and being further restricted by $\sum_{i=1}^m t_i = 1$. The definitive text Cornell (1990) lists numerous examples and provides a thorough discussion of both theory and practice. Early seminal work was done by Scheffé (1958, 1963) in which he suggested (1958, page 347) and analyzed canonical model forms when the regression function for the expected response $y = y(t)$ is a polynomial of degree one, two, or three. We shall refer to these as the S-polynomials, or S-models; for example, the second-degree S-polynomial has the form

$$y(t) = \sum_{1 \leq i \leq m} \beta_i t_i + \sum_{1 \leq i < j \leq m} \beta_{ij} t_i t_j. \qquad (1)$$

In this paper, we use the alternative representation of mixture models introduced in Draper and Pukelsheim (1998b, 1999). Our versions are based on the Kronecker algebra of vectors and matrices, and give rise to homogeneous model functions and moment matrices. We refer to the corresponding expressions as the K-models, or K-polynomials. We emphasize, however, that our results on the Kiefer ordering of experimental designs for second-degree mixture models are the same whether the S-model or the K-model is employed.

Our notation is the same as in the previous papers Draper and Pukelsheim (1998b, 1999). We consider multifactor experiments, for m deterministic ingredients that are assumed to influence the response only through the percentages or proportions in which they are blended together. For $i = 1, \ldots, m$, let $t_i \in [0,1]$ be the proportion of ingredient i in the mixture. As usual, we assemble the individual components to form the column vector of experimental conditions, $t = (t_1, \ldots, t_m)'$. It ranges over the experimental domain $\mathcal{T}$, the standard probability simplex in the space $\mathbb{R}^m$. Let $\mathbf{1}_m = (1, \ldots, 1)' \in \mathbb{R}^m$ be the unity vector, whence $\mathbf{1}_m' t = t_1 + \cdots + t_m$ is the sum of the components of a vector t. Therefore, in our case, the experimental domain is $\mathcal{T} = \{t \in [0,1]^m : \mathbf{1}_m' t = 1\}$.

Under experimental conditions $t \in \mathcal{T}$, the experimental response Y_t is taken to be a scalar random variable. Replications under identical experimental conditions, or responses from distinct experimental conditions are assumed to be of equal (unknown) variance σ^2, and uncorrelated. When the regression function is a second-degree K-polynomial, the expected response takes the form

$$\mathrm{E}[Y_t] = y(t) = \sum_{i=1}^m \sum_{j=i}^m t_i t_j \theta_{ij} = (t \otimes t)'\theta. \qquad (2)$$

An experimental design τ on the experimental domain $\mathcal{T}$ is a probability measure having a finite number of support points. If τ assigns weights $w_1, w_2, \ldots$ to its points of support in $\mathcal{T}$, then the experimenter is directed to draw proportions $w_1, w_2, \ldots$ of all observations under the respective experimental conditions. We associate with τ its (second-degree K-) moment matrix,

$$M(\tau) = \int_{\mathcal{T}} (t \otimes t)(t \otimes t)' \, \mathrm{d}\,\tau. \qquad (3)$$

The Kronecker square $(t \otimes t)$ in (2) repeats the mixed products $t_i t_j = t_j t_i$, $i < j$, and thus overparameterizes the quadratic response function y, (while the corresponding Scheffé setup (1) is based on a minimal parameterization). As a consequence, (2) is a non-parsimonious representation of (1), as,

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for $t \in \mathcal{T}$,

$$t'\beta = (t'\beta) \otimes (t'\mathbf{1}_m) = (t \otimes t)'(\beta \otimes \mathbf{1}_m) = (t \otimes t)'\theta.$$

Nevertheless we propose to utilize the K-model (2) instead of (1), since the Kronecker algebra is powerful enough to outweigh the overparameterization, and, more importantly, in the K-model the moment matrices from (3) have all entries homogeneous of degree four. This homogeneity is a distinctive advantage over the S-model for which some of the entries of the moment matrix are homogeneous of degree two, others of degree three, and the rest of degree four. A closely related emphasis on proper standardization is put forward by Dette (1997) though the motivation, rescaling the experimental domain, is different. In our work, the experimental domain $\mathcal{T}$ is the simplex and stays fixed.

Given an arbitrary design τ, we obtain an exchangeable (permutation invariant) design $\overline{\tau}$ by averaging over the permutation group,

$$\overline{\tau} = \frac{1}{m!}\sum\nolimits_{R \in \mathrm{Perm}(m)} \tau \circ R^{-1}.$$

If the original design τ itself is exchangeable then it is reproduced, $\overline{\tau} = \tau$. Otherwise the average $\overline{\tau}$ is an improvement over τ, in that it exhibits more symmetry and balancedness. In terms of matrix majorization (relative to the congruence action that is induced on the moment matrices M), the moment matrix of the averaged design $\overline{\tau}$ is majorized by the moment matrix of τ, $M(\overline{\tau}) \prec M(\tau)$. The terminology "is majorized by" is standard, even though for design purposes the emphasis is reversed: $M(\overline{\tau})$, being more balanced, is superior to $M(\tau)$. As a consequence, the design $\overline{\tau}$ yields better values than τ, under a large class of optimality criteria (Pukelsheim 1993, page 349). For a recent review of invariance and optimality of polynomial regression designs see Gaffke and Heiligers (1996).

Symmetry and balancedness have always been a prime attribute of good experimental designs, and comprise the first step of the Kiefer design ordering. The second step concerns the usual Loewner matrix ordering. Lemma 1 in Heiligers (1991) and Theorem 2 in Heiligers (1992) imply that any second-degree mixture design which is not a weighted centroid design can be improved upon, in the sense of the Kiefer ordering, by a weighted centroid design. Here we show *how* to derive the improving design from the properties of the starter design.

We shall restrict ourself to the K-model (2) with $m = 4$ ingredients, only. We show that the class of weighted centroid designs is minimal complete, see Theorem 4. As a consequence, the search for optimal designs may be restricted to weighted centroid designs, for most criteria. For particular criteria, this was observed already by Kiefer (1959, 1975, 1978), and Galil and Kiefer (1977). Related results on Kiefer ordering completeness of rotatable designs on the ball are reviewed by Draper and Pukelsheim (1998a). The setting of Cheng (1995) is slightly different, in that his permutations act on the regression vector $x = t \otimes t$, rather than on the experimental conditions t.

While for models with two or three factors Kiefer comparability of exchangeable moment matrices is described by one parameter only, cf. Draper and Pukelsheim (1999), the corresponding result in the four factor model is more complicated as it involves two dependent parameters, see Lemma 1, below. This is also true for $m \geq 5$ factors—these models, however, have to cope with some ambiguity introducing additional complications. Setups with five or more ingredients are discussed in Draper, Heiligers and Pukelsheim (1998); that paper also provides a complementary, geometric view of the present Complete Class Theorem 4.

Our approach is an extension of Draper and Pukelsheim (1998a, 1999). In view of the initial symmetrization step it suffices to search for an improvement in the Loewner ordering sense, among exchangeable moment matrices only. We first aim at finding necessary and sufficient conditions for two exchangeable second-degree K-moment matrices to be comparable in the Loewner matrix ordering. The "Comparison Lemma" 1 provides conditions in terms of two moment inequalities, in the spirit of Theorem 2 of Heiligers (1992). Weighted centroid designs effectively remove one degree of freedom, see the "Characterization Lemma" 2. Given a first, poor design we then construct a second, better design, in the "Existence Lemma" 3. The complete class result is stated in Theorem 4.

2 Four Factors

The four-ingredient second-degree model features all possible moments of order four,

$$\mu_4 = \int_{\mathcal{T}} t_i^4 \,\mathrm{d}\,\overline{\tau}, \qquad \mu_{31} = \int_{\mathcal{T}} t_i^3 t_j \,\mathrm{d}\,\overline{\tau},$$

$$\mu_{22} = \int_{\mathcal{T}} t_i^2 t_j^2 \,\mathrm{d}\,\overline{\tau}, \qquad \mu_{211} = \int_{\mathcal{T}} t_i^2 t_j t_k \,\mathrm{d}\,\overline{\tau},$$

$$\mu_{1111} = \int_{\mathcal{T}} t_i t_j t_k t_\ell \,\mathrm{d}\,\overline{\tau},$$

where the subscripts $i, j, k, \ell = 1, \dots, 4$ are pairwise distinct and where $\overline{\tau}$ is some exchangeable design on the simplex $\mathcal{T}$. The associated K-moment matrix $M = M(\overline{\tau})$ is of the generic form

$$M = \mu_4 V_4 + \mu_{31} V_{31} + \mu_{22} V_{22} + \mu_{211} V_{211} + \mu_{1111} V_{1111}$$

with 0-1 matrices V_i of order 16×16, indicating the position of the moments μ_i in M. As usual, let e_i denote the i-th Euclidean unit vector in $\mathbb{R}^4$ with i-th component one and zeros elsewhere. For a concise representation of V_i we use the 16×1 Euclidean unit vectors $e_{ij} = e_i \otimes e_j$ having a single one as the i-th block's j-th element, for $i, j = 1, \dots, 4$,

$$\begin{aligned}
V_4 &= \sum_i e_{ii} e_{ii}', \\
V_{31} &= \sum_{i,j}{}' (e_{ii} e_{ij}' + e_{ij} e_{ii}' + e_{ii} e_{ji}' + e_{ji} e_{ii}'), \\
V_{22} &= \sum_{i,j}{}' (e_{ii} e_{jj}' + e_{ij} e_{ij}' + e_{ij} e_{ji}'), \\
V_{211} &= \sum_{i,j,k}{}' (e_{ii} e_{jk}' + e_{jk} e_{ii}' + e_{ij} e_{ki}' + \\
&\qquad + e_{ji} e_{ik}' + e_{ij} e_{ik}' + e_{ji} e_{ki}'), \\
&\qquad + e_{ji} e_{ik}' + e_{ij} e_{ik}' + e_{ji} e_{ik}'), \\
V_{1111} &= \sum_{i,j,k,\ell}{}' e_{ij} e_{kl}'.
\end{aligned}$$

The sign $\sum'$ means that the summation is restricted to pairwise distinct subscripts. The rank of M is at most 10, implying M has at least six nullvectors.

The simplex restriction entails $\mathbf{1}_{16}' M \mathbf{1}_{16} = \int (\mathbf{1}_4' t)^4 \, d\overline{\tau} = 1$. That is, the elements of M sum to one, $4\mu_4 + 48\mu_{31} + 36\mu_{22} + 144\mu_{211} + 24\mu_{1111} = 1$. Moreover, the lower order moments are functions of the fourth order moments, i.e.,

$$\begin{aligned}
\mu_3 &= \mu_4 + 3\mu_{31}, \\
\mu_{21} &= \mu_{31} + \mu_{22} + 2\mu_{211}, \\
\mu_{111} &= 3\mu_{211} + \mu_{1111}; \\
\mu_2 &= \mu_3 + 3\mu_{21} \\
&= \mu_4 + 6\mu_{31} + 2\mu_{22} + 6\mu_{211}, \\
\mu_{11} &= 2\mu_{21} + 2\mu_{111} \\
&= 2\mu_{31} + 2\mu_{22} + 10\mu_{211} + 2\mu_{1111}.
\end{aligned}$$

Now we consider two exchangeable designs η and $\overline{\tau}$ possessing identical moments of order three. Then the moments of order two are equal, too. For the fourth order moment differences we get, with $\gamma = \mu_4(\eta) - \mu_4(\overline{\tau})$ and $\delta = \mu_{1111}(\eta) - \mu_{1111}(\overline{\tau})$,

$$\begin{aligned}
\mu_4(\eta) - \mu_4(\overline{\tau}) &= \gamma, \\
\mu_{31}(\eta) - \mu_{31}(\overline{\tau}) &= -\tfrac{1}{3}\gamma, \\
\mu_{22}(\eta) - \mu_{22}(\overline{\tau}) &= \tfrac{1}{3}\gamma + \tfrac{2}{3}\delta, \\
\mu_{211}(\eta) - \mu_{211}(\overline{\tau}) &= -\tfrac{1}{3}\delta, \\
\mu_{1111}(\eta) - \mu_{1111}(\overline{\tau}) &= \delta.
\end{aligned} \tag{4}$$

Of course, there is an infinity of ways to parameterize the two degrees of freedom in (4). We find γ and δ a natural choice to work with. Using the indicator matrices V_i, the moment matrices of η and $\overline{\tau}$ differ by

$$\begin{aligned}
\Delta &= M(\eta) - M(\overline{\tau}) \\
&= \gamma V_4 - \frac{\gamma}{3} V_{31} + \frac{\gamma + 2\delta}{3} V_{22} - \frac{\delta}{3} V_{211} + \delta V_{1111}.
\end{aligned}$$

This decomposition has five terms although there are only two degrees of freedom, γ and δ.

There are, however, simpler representations for Δ involving just two matrices A and B, say. A convenient choice is $A = V_4 - \frac{1}{3} V_{31} + \frac{1}{3} V_{22}$, i.e.,

$$A = \frac{1}{3} \begin{pmatrix}
3 & -1 & -1 & -1 & -1 & 1 & 0 & 0 & -1 & 0 & 1 & 0 & -1 & 0 & 0 & 1 \\
-1 & 1 & 0 & 0 & 1 & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
-1 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & -1 & 0 & 0 & 0 & 0 & 0 \\
-1 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & -1 \\
-1 & 1 & 0 & 0 & 1 & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
1 & -1 & 0 & 0 & -1 & 3 & -1 & -1 & 0 & -1 & 1 & 0 & 0 & -1 & 0 & 1 \\
0 & 0 & 0 & 0 & 0 & -1 & 1 & 0 & 0 & 1 & -1 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -1 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & -1 \\
-1 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & -1 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -1 & 1 & 0 & 0 & 1 & -1 & 0 & 0 & 0 & 0 & 0 \\
1 & 0 & -1 & 0 & 0 & 1 & -1 & 0 & -1 & -1 & 3 & -1 & 0 & 0 & -1 & 1 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -1 & 1 & 0 & 0 & 1 & -1 \\
-1 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & -1 \\
0 & 0 & 0 & 0 & 0 & -1 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & -1 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -1 & 1 & 0 & 0 & 1 & -1 \\
1 & 0 & 0 & -1 & 0 & 1 & 0 & -1 & 0 & 0 & 1 & -1 & -1 & -1 & -1 & 3
\end{pmatrix},$$

and $B = \frac{2}{3} V_{22} - \frac{1}{3} V_{211} + V_{1111}$, i.e.,

$$B = \frac{1}{3} \begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 2 & -1 & -1 & 0 & -1 & 2 & -1 & 0 & -1 & -1 & 2 \\
0 & 2 & -1 & -1 & 2 & 0 & -1 & -1 & -1 & -1 & -1 & 1 & -1 & -1 & 1 & -1 \\
0 & -1 & 2 & -1 & -1 & -1 & -1 & 1 & 2 & -1 & 0 & -1 & -1 & 1 & -1 & -1 \\
0 & -1 & -1 & 2 & -1 & -1 & 1 & -1 & -1 & 1 & -1 & -1 & 2 & -1 & -1 & 0 \\
0 & 2 & -1 & -1 & 2 & 0 & -1 & -1 & -1 & -1 & -1 & 1 & -1 & -1 & 1 & -1 \\
2 & 0 & -1 & -1 & 0 & 0 & 0 & 0 & -1 & 0 & 2 & -1 & -1 & 0 & -1 & 2 \\
-1 & -1 & -1 & 1 & -1 & 0 & 2 & -1 & -1 & 2 & 0 & -1 & 1 & -1 & -1 & -1 \\
-1 & -1 & 1 & -1 & -1 & 0 & -1 & 2 & 1 & -1 & -1 & -1 & -1 & 2 & -1 & 0 \\
0 & -1 & 2 & -1 & -1 & -1 & -1 & 1 & 2 & -1 & 0 & -1 & -1 & 1 & -1 & -1 \\
-1 & -1 & -1 & 1 & -1 & 0 & 2 & -1 & -1 & 2 & 0 & -1 & 1 & -1 & -1 & -1 \\
2 & -1 & 0 & -1 & -1 & 2 & 0 & -1 & 0 & 0 & 0 & 0 & -1 & -1 & 0 & 2 \\
-1 & 1 & -1 & -1 & 1 & -1 & -1 & -1 & -1 & -1 & 0 & 2 & -1 & -1 & 2 & 0 \\
0 & -1 & -1 & 2 & -1 & -1 & 1 & -1 & -1 & 1 & -1 & -1 & 2 & -1 & -1 & 0 \\
-1 & -1 & 1 & -1 & -1 & 0 & -1 & 2 & 1 & -1 & -1 & -1 & -1 & 2 & -1 & 0 \\
-1 & 1 & -1 & -1 & 1 & -1 & -1 & -1 & -1 & -1 & 0 & 2 & -1 & -1 & 2 & 0 \\
2 & -1 & -1 & 0 & -1 & 2 & -1 & 0 & -1 & -1 & 2 & 0 & 0 & 0 & 0 & 0
\end{pmatrix}.$$

The matrices A and B have the same rank six, both possessing three non-zero eigenvalues (with respective multiplicities one, two, and three):

$$\begin{aligned} \lambda_1[A] &= \tfrac{8}{3}, & \lambda_1[B] &= \tfrac{8}{3}, \\ \lambda_2[A] &= \tfrac{2}{3}, & \lambda_2[B] &= \tfrac{14}{3}, \\ \lambda_3[A] &= \tfrac{4}{3}, & \lambda_3[B] &= -\tfrac{4}{3}. \end{aligned}$$

Moreover, the eigenspaces $\mathcal{E}_i$ associated with the i-th eigenvalue of A and of B coincide; upon setting $u_{ijk\ell} = (e_i - e_j) \otimes (e_k - e_\ell)$ we have

$$\begin{aligned} \mathcal{E}_1 &= \operatorname{span}\{\mathbf{1}_{16} - 4\textstyle\sum_{i=1}^4 e_{ii}\}, \\ \mathcal{E}_2 &= \operatorname{span}\{(u_{1234} + u_{3412}),\ (u_{1324} + u_{2413})\}, \\ \mathcal{E}_3 &= \operatorname{span}\{(u_{1212} - u_{3434}),\ (u_{1313} - u_{2424}), \\ &\qquad (u_{1414} - u_{2323})\}. \end{aligned}$$

In summary, the representation for $\Delta = M(\eta) - M(\overline{\tau})$ takes the form

$$\Delta = \gamma A + \delta B, \tag{5}$$

and has eigenvalues

$$\begin{aligned} \lambda_1[\Delta] &= \tfrac{8}{3}(\gamma + \delta), \\ \lambda_2[\Delta] &= \tfrac{2}{3}(\gamma + 7\delta), \\ \lambda_3[\Delta] &= \tfrac{4}{3}(\gamma - \delta), \\ \lambda_4[\Delta] &= 0. \end{aligned} \tag{6}$$

The comparison of two exchangeable moment matrices in the Loewner ordering is now reduced to the comparison of individual moments. Let $\mu_{(3)} = (\mu_2, \mu_{11}, \mu_3, \mu_{21}, \mu_{111})'$ be the vector of moments up to and including order three.

LEMMA 1. *Let η and $\overline{\tau}$ be two exchangeable designs on the simplex $\mathcal{T}$. Then we have, with $\gamma = \mu_4(\eta) - \mu_4(\overline{\tau})$ and $\delta = \mu_{1111}(\eta) - \mu_{1111}(\overline{\tau})$,*

$$M(\eta) \geq M(\overline{\tau})$$

if and only if

$$\mu_{(3)}(\eta) = \mu_{(3)}(\overline{\tau}), \quad \textit{and} \quad -\tfrac{1}{7}\gamma \leq \delta \leq \gamma.$$

PROOF. For the direct part, assume that $\Delta = M(\eta) - M(\overline{\tau})$ is nonnegative definite. Then $(\mathbf{1}_4 \otimes \mathbf{1}_4)'\Delta(\mathbf{1}_4 \otimes \mathbf{1}_4) = 0$ forces $\Delta(\mathbf{1}_4 \otimes \mathbf{1}_4) = 0$. This implies equality of second order moments. Now we get

$$\begin{aligned} (e_1 \otimes \mathbf{1}_4)'M(\eta)(e_1 \otimes \mathbf{1}_4) &= \int_{\mathcal{T}} t_1^2 \, d\eta = \mu_2 \\ &= \int_{\mathcal{T}} t_1^2 \, d\overline{\tau} = (e_1 \otimes \mathbf{1}_4)'M(\overline{\tau})(e_1 \otimes \mathbf{1}_4). \end{aligned}$$

This yields $\Delta(e_1 \otimes \mathbf{1}_4) = 0$, that is, $\int (t \otimes t) t_1 \, d\eta = \int (t \otimes t) t_1 \, d\overline{\tau}$. Hence the third order moments of η and $\overline{\tau}$ are equal as well, giving $\mu_{(3)}(\eta) = \mu_{(3)}(\overline{\tau})$. By (6), nonnegative definiteness of Δ entails nonnegativity of $\gamma+\delta$, $\gamma-\delta$ and $\gamma+7\delta$, that is, $-\frac{1}{7}\gamma \leq \delta \leq \gamma$. For the converse part, equality of third order moments implies the representation (5). According to (6), the assumption on γ and δ immediately implies $\Delta \geq 0$. □

By Lemma 1, comparability of exchangeable designs η and $\overline{\tau}$ ensures that the difference $\Delta = M(\eta) - M(\overline{\tau})$ lies in $\mathcal{D} = \{\gamma A + \delta B : \gamma \geq 0, -\frac{\gamma}{7} \leq \delta \leq \gamma\}$, which is a two-dimensional subcone in the set of nonnegative definite 16×16 matrices. Thus, for any fixed $\overline{\tau}$, the moment matrices of those exchangeable designs η which improve upon $\overline{\tau}$ w.r.t. the Kiefer ordering are obtained by intersecting the set $\mathcal{M}$ of all moment matrices with the shifted cone $M(\overline{\tau}) + \mathcal{D}$; Figure 1 illustrates this geometry.

Of course, most of the designs η associated with $\mathcal{M} \cap (M(\overline{\tau}) + \mathcal{D})$ can be further improved upon by a third exchangeable design ζ, which then is also a further improvement on the starter design $\overline{\tau}$. We are particularly interested here in describing those exchangeable designs η which improve upon $\overline{\tau}$ and which can not be further improved upon. These turn out to be among the weighted centroid designs, introduced next.

There are four *elementary centroid designs*: the vertex points design η_1, the edge midpoints design η_2, the face centroids design η_3, and the overall centroid design η_4,

$$\begin{aligned} \eta_1(e_i) &= \tfrac{1}{4} && \text{for } 1 \leq i \leq 4, \\ \eta_2\big(\tfrac{1}{2}(e_i + e_j)\big) &= \tfrac{1}{6} && \text{for } 1 \leq i < j \leq 4, \\ \eta_3\big(\tfrac{1}{3}(e_i + e_j + e_k)\big) &= \tfrac{1}{4} && \text{for } 1 \leq i < j < k \leq 4, \\ \eta_4(\overline{e}) &= 1; \end{aligned}$$

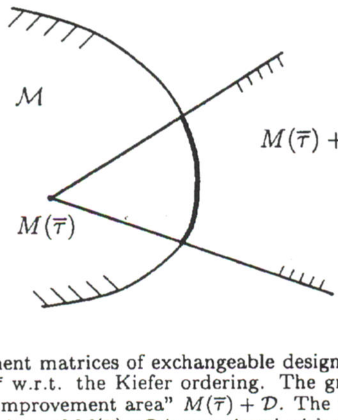


FIG. 1. Moment matrices of exchangeable designs which improve upon $\overline{\tau}$ w.r.t. the Kiefer ordering. The gray area describes the "improvement area" $M(\overline{\tau}) + \mathcal{D}$. The thick line in the east boundary of $M(\overline{\tau}) + \mathcal{D}$ is associated with the weighted centroid designs which improve upon $\overline{\tau}$.

Figure 2 illustrates these designs. The moments of order four of these designs are

$$
\begin{array}{rclrcl}
\mu_4(\eta_1) &=& \frac{1}{4}, & \mu_4(\eta_2) &=& \frac{1}{32}, \\
\mu_4(\eta_3) &=& \frac{1}{108}, & \mu_4(\eta_4) &=& \frac{1}{256}, \\
\mu_{31}(\eta_1) &=& 0, & \mu_{31}(\eta_2) &=& \frac{1}{96}, \\
\mu_{31}(\eta_3) &=& \frac{1}{162}, & \mu_{31}(\eta_4) &=& \frac{1}{256}, \\
\mu_{211}(\eta_1) &=& 0, & \mu_{211}(\eta_2) &=& 0, \\
\mu_{211}(\eta_3) &=& \frac{1}{324}, & \mu_{211}(\eta_4) &=& \frac{1}{256}, \\
\mu_{1111}(\eta_1) &=& 0, & \mu_{1111}(\eta_2) &=& 0, \\
\mu_{1111}(\eta_3) &=& 0, & \mu_{1111}(\eta_4) &=& \frac{1}{256};
\end{array}
$$

we have $\mu_{31}(\eta_j) = \mu_{22}(\eta_j)$, for all $j = 1, 2, 3, 4$.

For weights $\alpha_1, \alpha_2, \alpha_3, \alpha_4 \geq 0$ summing to one, the design $\eta = \alpha_1\eta_1 + \alpha_2\eta_2 + \alpha_3\eta_3 + \alpha_4\eta_4$ is called a *weighted centroid design.*

LEMMA 2. *Let $\overline{\tau}$ be an exchangeable design on the simplex $\mathcal{T}$. Then we have*

$$\mu_{31}(\overline{\tau}) \geq \mu_{22}(\overline{\tau}),$$

with equality if and only if $\overline{\tau}$ is a weighted centroid design.

PROOF. The function

$$\psi(t_1, t_2, t_3, t_4) = \sum_{i<j} t_i t_j (t_i - t_j)^2$$

is nonnegative on the simplex $\mathcal{T}$, and integrates under $\overline{\tau}$ to $12(\mu_{31} - \mu_{22})$. This proves $\mu_{31} \geq \mu_{22}$. We have $\int \psi \, d\overline{\tau} = 0$ if and only if every support point $t = (t_1, t_2, t_3, t_4)'$ of $\overline{\tau}$ satisfies $t_i t_j (t_i - t_j)^2 = 0$. Because of exchangeability, $\overline{\tau}$ must then be a weighted centroid design. □

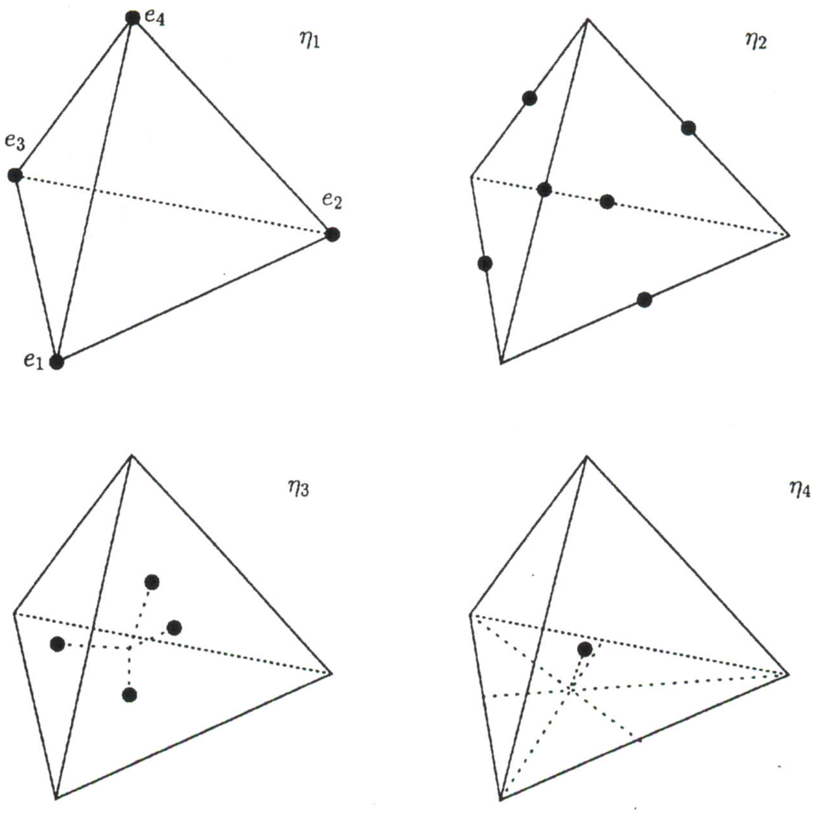


FIG. 2. Elementary centroid designs $\eta_1, \eta_2, \eta_3, \eta_4$ in a three-dimensional space. The individial designs assign equal weights to the associated support points, represented by the dots. The weights total 1 in each case.

Let $\eta = \alpha_1\eta_1 + \alpha_2\eta_2 + \alpha_3\eta_3 + \alpha_4\eta_4$ be a weighted centroid design. When we calculate the difference of line two and three in (4), the contribution of η vanishes due to $\mu_{31}(\eta) = \mu_{22}(\eta)$. Suppressing the dependence on $\overline{\tau}$ of the remaining moments, we get

$$\mu_{31} - \mu_{22} = \frac{2}{3}(\gamma + \delta). \qquad (7)$$

From this, we determine γ in terms of δ and the moments of $\overline{\tau}$, i.e., $\gamma = \frac{3}{2}(\mu_{31} - \mu_{22}) - \delta$. The restrictions $-\frac{1}{7}\gamma \leq \delta \leq \gamma$ provide initial bounds for δ,

$$-\frac{1}{4}(\mu_{31} - \mu_{22}) \leq \delta \leq \frac{3}{4}(\mu_{31} - \mu_{22}). \qquad (8)$$

In order to find a set of weights for $\eta = \alpha_1\eta_1 + \alpha_2\eta_2 + \alpha_3\eta_3 + \alpha_4\eta_4$ to improve upon $\overline{\tau}$, we refer to (4) and equate fourth order moments,

$$\begin{aligned}
\mu_4 + \gamma &= \tfrac{1}{4}\alpha_1 + \tfrac{1}{32}\alpha_2 + \tfrac{1}{108}\alpha_3 + \tfrac{1}{256}\alpha_4, \\
\mu_{31} - \tfrac{1}{3}\gamma &= \tfrac{1}{96}\alpha_2 + \tfrac{1}{162}\alpha_3 + \tfrac{1}{256}\alpha_4, \\
\mu_{211} - \tfrac{1}{3}\delta &= \tfrac{1}{324}\alpha_3 + \tfrac{1}{256}\alpha_4, \\
\mu_{1111} + \delta &= \tfrac{1}{256}\alpha_4;
\end{aligned} \qquad (9)$$

The solutions are, inserting $\gamma = \frac{3}{2}(\mu_{31} - \mu_{22}) - \delta$,

$$(\alpha_1, \alpha_2, \alpha_3, \alpha_4)' = 4\begin{pmatrix} 1 & -3 & 3 & -1 \\ 0 & 24 & -48 & 24 \\ 0 & 0 & 81 & -81 \\ 0 & 0 & 0 & 64 \end{pmatrix} \begin{pmatrix} \mu_4 + \frac{3}{2}(\mu_{31} - \mu_{22}) - \delta \\ \frac{1}{2}(\mu_{31} + \mu_{22}) + \frac{1}{3}\delta \\ \mu_{211} - \frac{1}{3}\delta \\ \mu_{1111} + \delta \end{pmatrix}.$$

In final terms we obtain

$$\begin{aligned}
\alpha_1 &= 4\,(\mu_4 - 3\mu_{22} + 3\mu_{211} - \mu_{1111} - 4\delta), \\
\alpha_2 &= 48\,(\mu_{31} + \mu_{22} - 4\mu_{211} + 2\mu_{1111} + 4\delta), \\
\alpha_3 &= 108\,(3\mu_{211} - 3\mu_{1111} - 4\delta), \\
\alpha_4 &= 256\,(\mu_{1111} + \delta).
\end{aligned} \qquad (10)$$

In addition to the initial bounds (8), the requirements $\alpha_j \geq 0$ in (10) enforce further bounds on δ. Overall, we get the range $\delta_{\min}(\overline{\tau}) \leq \delta_{\max}(\overline{\tau})$ where

$$\delta_{\max}(\overline{\tau}) = \min\left\{\frac{3}{4}(\mu_{31} - \mu_{22}),\ \frac{3}{4}(\mu_{211} - \mu_{1111}),\ \frac{1}{4}(\mu_4 - 3\mu_{22} + 3\mu_{211} - \mu_{1111})\right\},$$

and

$$\delta_{\min}(\overline{\tau}) = -\min\left\{\frac{1}{4}(\mu_{31} - \mu_{22}),\ \mu_{1111},\ \frac{1}{4}(\mu_{31} + \mu_{22} - 4\mu_{211} + 2\mu_{1111})\right\}.$$

The following lemma shows that $\delta_{\min}(\overline{\tau}) \leq 0 \leq \delta_{\max}(\overline{\tau})$. In particular, $\delta = 0$ is always a feasible choice. The lemma says that, for every exchangeable design $\overline{\tau}$, there indeed exists a weighted centroid design $\eta(\delta)$ improving upon $\overline{\tau}$.

LEMMA 3. *Let $\overline{\tau}$ be an exchangeable design on the simplex $\mathcal{T}$, with fourth order moments $\mu_4, \mu_{31}, \mu_{22}, \mu_{211}, \mu_{1111}$. Then we have $\delta_{\min}(\overline{\tau}) \leq 0 \leq \delta_{\max}(\overline{\tau})$, and for every $\delta \in [\delta_{\min}(\overline{\tau}), \delta_{\max}(\overline{\tau})]$ the weighted centroid design $\eta(\delta) = \alpha_1\eta_1 + \alpha_2\eta_2 + \alpha_3\eta_3 + \alpha_4\eta_4$, with weights from (10), satisfies*

$$M(\eta(\delta)) \geq M(\overline{\tau}),$$

with equality if and only if $\delta = 0$ and $\overline{\tau} = \eta(0)$.

PROOF. The relation $\alpha_1 + \alpha_2 + \alpha_3 + \alpha_4 = 4\mu_4 + 48\mu_{31} + 36\mu_{22} + 144\mu_{211} + 24\mu_{1111} = 1$ is the simplex restriction formula. In order to show that the weights α_j are nonnegative, we start with the special case $\delta = 0$.

Clearly, we have $\alpha_4 = 256\mu_{1111} \geq 0$. We also have $\alpha_3 = 324(\mu_{211} - \mu_{1111}) \geq 0$, since the nonnegative function $162 t_1 t_2 (t_3 - t_4)^2$ integrates to α_3. For α_2, the inequality $\mu_{31} \geq \mu_{22}$ from Lemma 2 yields

$$\begin{aligned}
\alpha_2 &\geq 96\,(\mu_{22} - 2\mu_{211} + \mu_{1111}) \\
&= 24 \int (t_1 - t_2)^2 (t_3 - t_4)^2 \, d\overline{\tau} \geq 0.
\end{aligned}$$

For α_1, we use the symmetric function

$$\begin{aligned}
\psi(t_1, t_2, t_3, t_4) &= t_1^4 + t_2^4 + t_3^4 + t_4^4 \\
&- 2t_1^2t_2^2 - 2t_1^2t_3^2 - 2t_1^2t_4^2 - 2t_2^2t_3^2 - 2t_2^2t_4^2 - 2t_3^2t_4^2 \\
&+ t_1^2t_2t_3 + t_1^2t_2t_4 + t_1^2t_3t_4 + t_1t_2^2t_3 + t_1t_2^2t_4 \\
&+ t_2^2t_3t_4 + t_1t_2t_3^2 + t_1t_3^2t_4 + t_2t_3^2t_4 + t_1t_2t_4^2 \\
&+ t_1t_3t_4^2 + t_2t_3t_4^2 - 4t_1t_2t_3t_4.
\end{aligned}$$

Because of homogeneity, ψ is nonnegative on the simplex $\mathcal{T}$ if and only if it is nonnegative on the quadrant $[0,\infty)^4$. In the interior $(0,\infty)^4$, the gradient vanishes only along the equiangular line, $t_1 = t_2 = t_3 = t_4$, where ψ attains the minimum value zero. By continuity, ψ stays nonnegative on all boundaries. This ensures $\alpha_1 = \int \psi \, d\overline{\tau} \geq 0$.

Hence, in the special case when $\delta = 0$, the weights α_j are nonnegative, and $\delta_{\min}(\overline{\tau}) \leq 0 \leq \delta_{\max}(\overline{\tau})$. Generally then, as long as δ stays in the range $[\delta_{\min}(\overline{\tau}), \delta_{\max}(\overline{\tau})]$, the weights α_j remain nonnegative. Therefore the weighted centroid design $\eta(\delta)$ is well-defined. It fulfills $\mu_{(3)}(\eta(\delta)) = \mu_{(3)}(\overline{\tau})$. We verify equation (7), whence the bounds on the range of δ secure $-\frac{1}{7}\gamma \leq \delta \leq \gamma$. Now Lemma 1 yields $M(\eta(\delta)) \geq M(\overline{\tau})$.

If equality holds then $\Delta = 0$ in (5). Hence $\gamma = \delta = 0$ and, from (7), $\mu_{31} = \mu_{22}$. By Lemma 2, $\overline{\tau}$ is a weighted centroid design. Denote the weights of $\overline{\tau}$

by $\beta_1, \beta_2, \beta_3, \beta_4$. That $\eta(0)$ and $\bar{\tau}$ are identical now follows from

$$\begin{aligned}
&(\alpha_1, \alpha_2, \alpha_3, \alpha_4)' \\
&= 4\begin{pmatrix} 1 & -3 & 3 & -1 \\ 0 & 24 & -48 & 24 \\ 0 & 0 & 81 & -81 \\ 0 & 0 & 0 & 64 \end{pmatrix} \begin{pmatrix} \mu_4 + \frac{3}{2}(\mu_{31} - \mu_{22}) \\ \frac{1}{2}(\mu_{31} + \mu_{22}) \\ \mu_{211} \\ \mu_{1111} \end{pmatrix} \\
&= (\beta_1, \beta_2, \beta_3, \beta_4)'
\end{aligned}$$
□

Our examples share a common characteristic: Let $\epsilon(t_1, t_2, t_3, t_4)$ be the one-point design with support point $t = (t_1, t_2, t_3, t_4)' \in \mathcal{T}$. By averaging over all permutations we obtain the exchangeable design $\bar{\epsilon}(t_1, t_2, t_3, t_4)$, assigning equal weight to the distinct permutations of t. Because of exchangeability we may assume the components of t to be ordered, $t_1 \geq t_2 \geq t_3 \geq t_4$. In Examples 1 and 2, t depends on a real support parameter r.

EXAMPLE 1. Lemma 3 is illustrated using the one-parameter family of exchangeable designs $\tau_r = \bar{\epsilon}(\frac{1}{2} - r, \frac{1}{2} - r, r, r)$ for $r \in [0, \frac{1}{4}]$, assigning equal weight 1/6 to each of the six permutations of $(\frac{1}{2} - r, \frac{1}{2} - r, r, r)'$. This family includes the edge midpoints design, $r = 0$, and the overall centroid design, $r = \frac{1}{4}$. The improving design $\eta(0)$ from Lemma 3 has weights

$$\begin{aligned}
\alpha_1(r) &= \tfrac{1}{2} r(1-2r)(1-4r)^2, \\
\alpha_2(r) &= (1 - 6r + 12r^2)(1-4r)^2, \\
\alpha_3(r) &= 27\,\alpha_1(r), \\
\alpha_4(r) &= 64\, r^2(1-2r)^2.
\end{aligned}$$

The bounds $\delta_{\min}$ and $\delta_{\max}$ for δ are conveniently expressed in terms of the preceding weights as

$$\begin{aligned}
\delta_{\max}(\tau_r) &= \tfrac{1}{16}\alpha_1(r), \\
\delta_{\min}(\tau_r) &= \begin{cases} -\frac{1}{48}\alpha_1(r) & \text{for } r \in [\frac{1}{4} - \frac{\sqrt{3}}{8}, \frac{1}{4}], \\ -\frac{1}{256}\alpha_4(r) & \text{for } r \in [0, \frac{1}{4} - \frac{\sqrt{3}}{8}]. \end{cases}
\end{aligned}$$

The edge midpoints design has $\alpha_2(0) = 1$, and the overall centroid design has $\alpha_4(\frac{1}{4}) = 1$, as one would expect. With $\delta = 0$, the value of γ is $\frac{1}{8}\alpha_1(r)$. □

EXAMPLE 2. Our second example is provided by the designs $\tau_r = \bar{\epsilon}(1 - 3r, r, r, r)'$ for $r \in [0, \frac{1}{3}]$, assigning weight 1/4 to the four permutations of $(1-3r, r, r, r)'$. This family includes the vertex points design, $r = 0$, the overall centroid design, $r = \frac{1}{4}$, and the face centroids design, $r = \frac{1}{3}$. The weights of the improving design $\eta(0)$ from Lemma 3 are

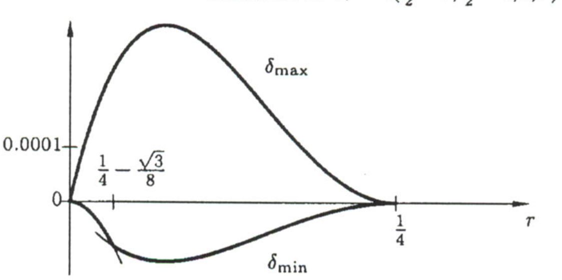
$$\begin{aligned}
\alpha_1(r) &= (1-r)(1-3r)(1-4r)^2, \\
\alpha_2(r) &= 12\, r(1-3r)(1-4r)^2, \\
\alpha_3(r) &= 81\, r^2(1-4r)^2, \\
\alpha_4(r) &= 256\, r^3(1-3r).
\end{aligned}$$

With $\delta = 0$, the value of γ is $\frac{1}{32}\alpha_2(r)$. The bounds $\delta_{\max}$ and $\delta_{\min}$ are

$$\begin{aligned}
\delta_{\max}(\tau_r) &= \begin{cases} \frac{1}{16}\alpha_1(r) & \text{for } r \in [\frac{1}{4}, \frac{1}{3}], \\ \frac{1}{432}\alpha_3(r) & \text{for } r \in [0, \frac{1}{4}]; \end{cases} \\
\delta_{\min}(\tau_r) &= \begin{cases} \frac{1}{192}\alpha_2(r) & \text{for } r \in [\frac{1}{8}, \frac{1}{3}], \\ \frac{1}{256}\alpha_4(r) & \text{for } r \in [0, \frac{1}{8}]. \end{cases}
\end{aligned}$$

The graphs of $\delta_{\max}(\tau_r)$ and $\delta_{\min}(\tau_r)$ are shown in Figure 3. □

EXAMPLE 1: $\tau_r = \bar{\epsilon}(\frac{1}{2} - r, \frac{1}{2} - r, r, r)$



EXAMPLE 2: $\tau_r = \bar{\epsilon}(1 - 3r, r, r, r)$

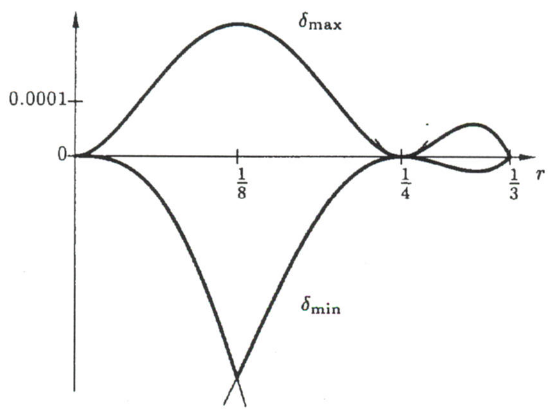


FIG. 3. Range $[\delta_{\min}(\tau_r), \delta_{\max}(\tau_r)]$, for various designs τ_r. The range contains the moment parameter δ. It thus reflects how many weighted centroid designs $\eta(\delta)$ improve upon the given exchangeable design τ_r.

In both examples, each of the two terms of which $\delta_{\min}$ is the minimum is needed, for some r. This is not so for $\delta_{\max}$. In Example 1, the three terms of which $\delta_{\max}$ is the minimum are identical, so any one of them suffices. In Example 2, the minimum in the definition of $\delta_{\max}$ is determined by the second and the third terms. In other examples, though, the first term may become relevant. For instance, this happens in the family of designs τ_r supported by the 12 permutations of $(0.4 - r, 0.3, 0.3, r)'$, for $r \in [0, 0.4]$. In general, each of the three terms of which $\delta_{\max}$ is defined to be the minimum has a role to play.

In conclusion, the complete class result for the Kiefer ordering in the case of four ingredients takes the following form. The class is formed by all convex combinations of the vertex points design η_1, the edge midpoints design η_2, the face centroids design η_3, and the overall centroid design η_4. The theorem refers to K-models and S-models alike.

THEOREM 4. *In the four-ingredient second-degree mixture model, the set of weighted centroid designs*

$$\mathcal{C} = \{\alpha_1\eta_1 + \alpha_2\eta_2 + \alpha_3\eta_3 + \alpha_4\eta_4 : (\alpha_1, \alpha_2, \alpha_3, \alpha_4)' \in \mathcal{T}\}$$

is convex, and constitutes a minimal complete class of designs for the Kiefer ordering.

PROOF. Completeness follows as in Theorems 6.4 and 7.4 of Draper and Pukelsheim (1999). For minimal completeness, the last paragraph in the proof of Lemma 3 shows that any two weighted centroid designs η and τ satisfy $M(\eta) \geq M(\tau)$ only if $\eta = \tau$. □

We append some remarks elucidating the various roles played by the simplex $\mathcal{T}$. In its primal meaning, as the experimental domain that underlies mixture models, $\mathcal{T}$ is exchangeable. In its dual meaning, as a set parameterizing the convex complete class $\mathcal{C}$ of Theorem 4, $\mathcal{T}$ is, of course, not exchangeable: α_1 belongs to the vertex design η_1, α_2 to the edge midpoints designs η_2 which is different, etc.

This helps in understanding the distinct geometric properties of the solutions of the system (10). As δ varies over its range $[\delta_{\min}(\overline{\tau}), \delta_{\max}(\overline{\tau})]$, let $\alpha(\delta) = (\alpha_1, \alpha_2, \alpha_3, \alpha_4)'$ denote the weight vector that uniquely solves (10). Evidently we have

$$\alpha(\delta) = \alpha(0) + \delta d,$$

where $d = 16(-1, 12, -27, 16)'$. This means that the set of all such weight vectors forms a segment on the line through $\alpha(0)$ in the direction given by d. The design $\overline{\tau}$ enters in that it determines the base point, $\alpha(0)$, and the length of the segment, $\delta_{\max}(\overline{\tau}) + \delta_{\min}(\overline{\tau})$. The direction d, however, always stays the same, because it has to be understood relative to the fixed orientation of the parameter space $\mathcal{T}$. In the geometry of these weight vectors, the range for δ ensures that $\alpha(0) + \delta d$ stays in the parameter space $\mathcal{T}$, besides securing (8).

The geometry translates into the moment space. For a given value of δ, let $\mu(\eta(\delta)) = (\mu_4(\eta(\delta)), \mu_{31}(\eta(\delta)), \mu_{211}(\eta(\delta)), \mu_{1111}(\eta(\delta)))'$ denote the fourth order moment vector of the weighted centroid design $\eta(\delta)$ of Lemma 3. From (9) we obtain the line segment

$$\mu(\eta(\delta)) = \mu(\eta(0)) + \delta e,$$

where again the direction $e = \frac{1}{3}(-3, 1, -1, 3)'$ does not depend on $\overline{\tau}$. The present line segment assembles those moment vectors $\mu(\eta(\delta))$ that improve upon $\overline{\tau}$, in the Loewner ordering sense of having $M(\eta(\delta)) \geq M(\overline{\tau})$. In the geometry of these moment vectors, the bounds on δ secure (4), and also imply that $\mu(\eta(0)) + \delta e$ lies in the moment polytope spanned by $\mu(\eta_j)$ for $j = 1, 2, 3, 4$.

The structure of the moment polytope, in the general case of five or more ingredients, is discussed in more detail in Draper, Heiligers and Pukelsheim (1998).

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