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KIEFER ORDERING OF SIMPLEX DESIGNS FOR SECOND-DEGREE MIXTURE MODELS WITH FOUR OR MORE INGREDIENTS

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For mixture models on the simplex, we discuss the improvement of a given design in terms of increasing symmetry, as well as obtaining a larger moment matrix under the Loewner ordering. The two criteria together define the Kiefer design ordering. For the second-degree mixture model, we show that the set of weighted centroid designs constitutes a convex complete class for the Kiefer ordering. For four ingredients, the class is minimal complete. Of essential importance for the derivation is a certain moment polytope, which is studied in detail.

1. Introduction. Many practical problems are associated with the investigation of mixture ingredients of m factors, assumed to influence the response only 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 Scheffé (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 individual proportions $t_1, \dots, t_m$ of the mixture ingredients form the column vector of experimental conditions, $t = (t_1, \dots, t_m)'$, with $t_i \geq 0$ and further restricted by $\Sigma_{i=1}^m t_i = 1$. Let $1_m = (1, \dots, 1)' \in \mathbb{R}^m$ be the unity vector, whence $1_m' t$ is the sum of the components of t. Therefore, the experimental domain is the simplex $\mathscr{T} = \{t \in [0,1]^m : 1_m' t = 1\}$.

Under experimental conditions $t \in \mathscr{T}$, the 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. An experimental design τ is a probability measure on the experimental domain $\mathscr{T}$ with a finite number of support points. If τ assigns weights $w_1, w_2, \dots$ to its points of support in $\mathscr{T}$, then the experimenter is directed to draw proportions $w_1, w_2, \dots$ of all observations under the respective experimental conditions.

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We are concerned throughout with a quadratic regression function, represented as a homogeneous second-degree Kronecker polynomial. The expected response thus takes the form $E[Y_t] = \Sigma_{i=1}^{m}\Sigma_{j=i}^{m} t_i t_j \theta_{ij} = (t \otimes t)'\theta$; see Draper and Pukelsheim (1998b, 1999). We emphasize, however, that our results on the Kiefer ordering of experimental designs for second-degree mixture models do not depend on the actual parameterization of the response function; see Draper and Pukelsheim [1998a, page 210].

Use of the Kronecker representation ensures that each entry in the moment matrix $M(\tau) = \int_{\mathscr{T}} (t \otimes t)(t \otimes t)'\, d\tau$ is homogeneous of degree four. With four or more ingredients, all five possible moments of order four actually occur. In a nonmixture context, Dette (1997) achieves homogeneity by using "standardized" optimality criteria; this may offer an alternative approach. For reviews of the general design environment, see Pukelsheim [(1993), Chapter 14] and Gaffke and Heiligers (1996). For particular optimality criteria applied to mixture models, see Kiefer (1959, 1975, 1978) [included in Kiefer (1985)] and Galil and Kiefer (1977). Related work on Kiefer ordering completeness of rotatable designs on the ball is reviewed by Draper and Pukelsheim (1998a). The setting of Cheng (1995) is different; his permutations act on the $m^2 \times 1$ regression vector $x = t \otimes t$, rather than on the $m \times 1$ vector t of experimental conditions.

In Section 2 we investigate Kiefer ordering of designs in the second-degree model for $m \geq 4$ ingredients. Theorem 2.4 states that the class of weighted centroid designs is complete for $m \geq 5$ (minimal complete for $m = 4$). The two and three ingredient cases, $m = 2, 3$, provide a much simpler structure, described in Draper and Pukelsheim (1999). The present completeness results can also be deduced from Lemma 1 in Heiligers (1991) and Theorem 2 in Heiligers (1992). Here, we achieve much more by showing how to compute all improvement weighted centroid designs from moments of the starter design. In Section 3 we discuss a certain moment polytope which plays an essential role in our derivations. This leads to Theorem 3.2 which states that, for $m \geq 5$, the subclass of weighted centroid designs requiring only four components is essentially complete. In Section 4 we illustrate various consequences of our results.

2. Four or more factors.

Given an arbitrary mixture design τ, we obtain an exchangeable (permutation invariant) design $\overline{\tau}$ by averaging over the permutation group. If the original design τ itself is exchangeable, then it is reproduced, $\overline{\tau} = \tau$. Otherwise $\overline{\tau}$ is an improvement over τ, in that it exhibits more symmetry and balancedness. The relevant Kiefer improvements upon the starter design τ are thus the exchangeable designs η which Loewner improve upon the exchangeable design $\overline{\tau}$.

The m-ingredient ($m \geq 4$) second-degree model features all possible moments of order four, $\mu_4 = \int t_i^4\, d\overline{\tau}$, $\mu_{31} = \int t_i^3 t_j\, d\overline{\tau}$, $\mu_{22} = \int t_i^2 t_j^2\, d\overline{\tau}$, $\mu_{211} = \int t_i^2 t_j t_k\, d\overline{\tau}$, and $\mu_{1111} = \int t_i t_j t_k t_l\, d\overline{\tau}$, where the subscripts $i, j, k, l = 1, \ldots, m$ are pairwise distinct and where $\overline{\tau}$ is some exchangeable design on the simplex

$\mathscr{T}$. For the Kronecker model, $\overline{\tau}$ has a moment matrix $M = \int_{\mathscr{T}} (t \otimes t)(t \otimes t)' \, d\overline{\tau}$ of the form $M = \mu_4 V_4 + \mu_{31} V_{31} + \mu_{22} V_{22} + \mu_{211} V_{211} + \mu_{1111} V_{1111}$. The zero–one matrices V_i are of order $m^2 \times m^2$ and indicate the position of the moments μ_i in M. For a concise representation of V_i we use the $m^2 \times 1$ Euclidean unit vectors $e_{ij} = e_i \otimes e_j$ having a single one as the ith block's jth element, for $i, j = 1, \ldots, m$,

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

The sign Σ' means that the summation is restricted to pairwise distinct subscripts. The rank of M is at most $\binom{m+1}{2}$, implying M has at least $\binom{m}{2}$ nullvectors.

The simplex restriction entails $1_{m^2}' M 1_{m^2} = \int (1_m' t)^4 \, d\overline{\tau} = 1$. Hence, the elements of M sum to one, so that $m\mu_4 + 4m(m-1)\mu_{31} + 3m(m-1)\mu_{22} + 6m(m-1)(m-2)\mu_{211} + m(m-1)(m-2)(m-3)\mu_{1111} = 1$. In terms of fourth-order moments, the third- and second-order moments are

$$\begin{aligned}
\mu_3 &= \mu_4 + (m-1)m_{31}, \\
\mu_{21} &= \mu_{31} + \mu_{22} + (m-2)m_{211}, \\
\mu_{111} &= 3\mu_{211} + (m-3)\mu_{1111}; \\
\mu_2 = \mu_3 + (m-1)\mu_{21} &= \mu_4 + 2(m-1)\mu_{31} + (m-1)\mu_{22} \\
&\quad + (m-1)(m-2)\mu_{211}, \\
\mu_{11} = 2\mu_{21} + (m-2)\mu_{111} &= 2\mu_{31} + 2\mu_{22} + 5(m-2)\mu_{211} \\
&\quad + (m-2)(m-3)\mu_{1111}.
\end{aligned}$$

Let η and $\overline{\tau}$ be two exchangeable designs on the simplex $\mathscr{T}$ having identical moments up to order three, $\mu_{(3)}(\eta) = \mu_{(3)}(\overline{\tau})$. The fourth-order moment differences take the following forms, upon defining $\gamma = \mu_4(\eta) - \mu_4(\overline{\tau})$ and $\delta = \mu_{1111}(\eta) - \mu_{1111}(\overline{\tau})$:

$$\begin{aligned}
\mu_{31}(\eta) - \mu_{31}(\overline{\tau}) &= -\frac{1}{m-1}\gamma, \\
\mu_{22}(\eta) - \mu_{22}(\overline{\tau}) &= \frac{1}{m-1}\gamma + \frac{(m-2)(m-3)}{3}\delta, \qquad (1) \\
\mu_{211}(\eta) - \mu_{211}(\overline{\tau}) &= -\frac{m-3}{3}\delta.
\end{aligned}$$

The properties of the moment matrix difference $\Delta = M(\eta) - M(\overline{\tau})$ become visible from

$$(2)\qquad \begin{aligned} 3m(m-1)(m-2)\Delta = {} & \big(3\gamma - (m-1)(m-3)\delta\big)C \\ & + \big(3\gamma + (m-1)(m^2-3m+3)\delta\big)D, \end{aligned}$$

where $C = (m-1)(m^2-3m+3)V_4 - (m^2-3m+3)V_{31} + (2m-3)V_{22} + (m-3)V_{211} - 3V_{1111}$ and $D = (m-1)(m-3)V_4 - (m-3)V_{31} + (m-1)(m-3)V_{22} - (m-3)V_{211} - 3V_{1111}$. With contrasts $c_i = e_i - (1/m)1_m$, we define $u_{ij} = c_i \otimes c_j$ and

$$v_{ij} = u_{ij} + u_{ji} + \frac{2}{m-2}(u_{ii} + u_{jj}) + \frac{\sqrt{2(m-2)(m-3)} - 2}{(m-1)(m-2)} \sum_{k=1}^{m} u_{kk} = v_{ji}.$$

It transpires that $C = m^3 \Sigma_i u_{ii} u_{ii}'$ and $D = m(m-2)\Sigma_{i<j} v_{ij} v_{ij}'$. Hence the matrices C and D are nonnegative definite. For the special case $\delta = 0$ in (2) we get $(m-1)\Delta = \gamma E$, where $E = \Sigma_{i<j} w_{ij} w_{ij}' \geq 0$ and $w_{ij} = (e_i - e_j) \otimes (e_i - e_j) = w_{ji}$.

Lemma 2.1 provides a condition for when the moment matrices of two exchangeable designs η and $\overline{\tau}$ are Loewner comparable, that is, the moments of order up to and including three are equal, and γ and δ obey two inequalities.

LEMMA 2.1. *Let η and $\overline{\tau}$ be two exchangeable designs on the simplex $\mathscr{T}$, and let $\gamma = \mu_4(\eta) - \mu_4(\overline{\tau})$ and $\delta = \mu_{1111}(\eta) - \mu_{1111}(\overline{\tau})$. Then $M(\eta) \geq M(\overline{\tau})$ if and only if $\mu_{(3)}(\eta) = \mu_{(3)}(\overline{\tau})$ and $-3/((m-1)(m^2-3m+3))\gamma \leq \delta \leq 3/((m-1)(m-3))\gamma$.*

PROOF. Let $\Delta = M(\eta) - M(\overline{\tau}) \geq 0$. Then $(1_m \otimes 1_m)'\Delta(1_m \otimes 1_m) = 0$ forces $\Delta(1_m \otimes 1_m) = 0$, implying equality of second-order moments. Now we get $(e_1 \otimes 1_m)' M(\eta)(e_1 \otimes 1_m) = \int t_1^2 \, d\eta = \mu_2 = \int t_1^2 \, d\overline{\tau} = (e_1 \otimes 1_m)' M(\overline{\tau})(e_1 \otimes 1_m)$. This forces $\Delta(e_1 \otimes 1_m) = 0$, that is, $\int (t \otimes t) t_1 \, d\eta = \int (t \otimes t) t_1 \, d\overline{\tau}$. Hence $\mu_{(3)}(\eta) = \mu_{(3)}(\overline{\tau})$, and (2) holds. Thus $\Delta \geq 0$ implies the inequalities stated in the lemma. Conversely, $\Delta \geq 0$ follows from (2). $\square$

There are m *elementary centroid designs* η_j, for $j = 1, \ldots, m$. Each places equal weight $1/\binom{m}{j}$ on the points having j out of their m components equal to $1/j$ and zeros elsewhere. The moments of order four are

$$\begin{aligned} \mu_4(\eta_j) &= \frac{1}{j^3 m}, \\ \mu_{31}(\eta_j) = \mu_{22}(\eta_j) &= \frac{1}{j^3 m} \frac{j-1}{m-1}, \\ \mu_{211}(\eta_j) &= \frac{1}{j^3 m} \frac{j-1}{m-1} \frac{j-2}{m-2}, \\ \mu_{1111}(\eta_j) &= \frac{1}{j^3 m} \frac{j-1}{m-1} \frac{j-2}{m-2} \frac{j-3}{m-3}. \end{aligned}$$

For given weights $\alpha_1, \dots, \alpha_m \geq 0$ summing to one, the design $\eta = \Sigma_{j=1}^m \alpha_j \eta_j$ is called a *weighted centroid design*. These designs are characterized by the moment identity $\mu_{31} = \mu_{22}$, as follows.

LEMMA 2.2. *Let $\bar{\tau}$ be an exchangeable design on the simplex $\mathscr{T}$. Then we have $\mu_{31}(\bar{\tau}) \geq \mu_{22}(\bar{\tau})$. Equality holds if and only if $\bar{\tau}$ is a weighted centroid design.*

PROOF. The function $\psi(t_1, \dots, t_m) = \Sigma_{i<j} t_i t_j (t_i - t_j)^2$ is nonnegative on $\mathscr{T}$ and integrates under $\bar{\tau}$ to $m(m-1)(\mu_{31} - \mu_{22})$, so that $\mu_{31} \geq \mu_{22}$. The integral vanishes if and only if $t_i t_j (t_i - t_j)^2 = 0$ for every support point, whence $\bar{\tau}$ is a weighted centroid design. □

When η is a weighted centroid design, we use the equality $\mu_{31}(\eta) = \mu_{22}(\eta)$ to write γ and δ in (1) solely in terms of moments of $\bar{\tau}$. Suppressing the dependence on $\bar{\tau}$, we get

$$\mu_{31} - \mu_{22} = \frac{2}{m-1}\gamma + \frac{(m-2)(m-3)}{3}\delta. \tag{3}$$

Substituting $\gamma = ((m-1)/2)(\mu_{31} - \mu_{22}) - \binom{m-1}{3}\delta$ into the two inequalities in Lemma 2.1 provides an initial set of bounds for δ,

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

In order to find weights for $\eta = \Sigma_{j=1}^m \alpha_j \eta_j$ to improve upon $\bar{\tau}$, we refer to (1) and equate fourth order moments:

$$\begin{aligned}
\mu_4(\eta) &\equiv \frac{1}{m}\sum_{j=1}^m \frac{1}{j^3}\alpha_j = \mu_4 + \gamma,\\
\mu_{31}(\eta) &\equiv \frac{1}{m(m-1)}\sum_{j=2}^m \frac{j-1}{j^3}\alpha_j = \mu_{31} - \frac{1}{m-1}\gamma,\\
\mu_{211}(\eta) &\equiv \frac{1}{m(m-1)(m-2)}\sum_{j=3}^m \frac{(j-1)(j-2)}{j^3}\alpha_j = \mu_{211} - \frac{m-3}{3}\delta,\\
\mu_{1111}(\eta) &\equiv \frac{1}{m(m-1)(m-2)(m-3)}\sum_{j=4}^m \frac{(j-1)(j-2)(j-3)}{j^3}\alpha_j = \mu_{1111} + \delta.
\end{aligned} \tag{5}$$

The system becomes more transparent when we multiply each equation by the number of times the corresponding moment can arise, and again substitute for γ. We then define, for $j = 1, \dots, m$, the 4×1 vectors

$$a_j = \begin{pmatrix} m\mu_4(\eta_j) \\ m(m-1)\mu_{31}(\eta_j) \\ m(m-1)(m-2)\mu_{211}(\eta_j) \\ m(m-1)(m-2)(m-3)\mu_{1111}(\eta_j) \end{pmatrix}$$

(6)
$$= \frac{1}{j^3}\begin{pmatrix} 1 \\ j-1 \\ (j-1)(j-2) \\ (j-1)(j-2)(j-3) \end{pmatrix},$$

$$b(\bar{\tau}) = \begin{pmatrix} m\mu_4(\bar{\tau}) + \frac{m(m-1)}{2}(\mu_{31}(\bar{\tau}) - \mu_{22}(\bar{\tau})) \\ \frac{m(m-1)}{2}(\mu_{31}(\bar{\tau}) + \mu_{22}(\bar{\tau})) \\ m(m-1)(m-2)\mu_{211}(\bar{\tau}) \\ m(m-1)(m-2)(m-3)\mu_{1111}(\bar{\tau}) \end{pmatrix},$$

$$c = \frac{1}{6}m(m-1)(m-2)(m-3)\begin{pmatrix} -1 \\ 1 \\ -2 \\ 6 \end{pmatrix}.$$

Clearly, $a_j = b(\eta_j)$. The equations in (5) thus give rise to the system

$$\begin{pmatrix} a_1, \ldots, a_m \end{pmatrix}\begin{pmatrix} \alpha_1 \\ \vdots \\ \alpha_m \end{pmatrix} = b(\bar{\tau}) + \delta c. \tag{7}$$

System (7) is always solvable since the coefficient matrix $A_m = (a_1, \ldots, a_m)$ has full row rank four. The vector $z = (1, 7, 6, 1)'$ satisfies $z'c = 0$. The simplex restriction entails $z'b(\bar{\tau}) = 1 = z'a_j$ and $z'A_m = 1_m'$. Hence premultiplication of (7) by z' yields $\Sigma_{j=1}^m \alpha_j = 1$, and so every set of solutions of (7) necessarily sums to one. Our task is to find nonnegative solutions which thus qualify as weights of a weighted centroid design η. In fact, we achieve much more by giving *all* possible nonnegative solutions.

The geometry that underlies (7) entails further bounds on δ. Let

$$\mathscr{P}(m) = \mathrm{conv}\{a_1, \ldots, a_m\} \subset \mathbb{R}^4 \tag{8}$$

be the convex hull of the vectors $a_1, \ldots, a_m$. This is a polytope in $\mathbb{R}^4$, with vertices a_j consisting of the scaled fourth-order moments of the elementary centroid designs η_j. We call $\mathscr{P}(m)$ the *moment polytope* of our problem; it will turn out to be of essential importance for our arguments. For a given design $\bar{\tau}$ we now define

(9)
$$\delta_{\max}(\bar{\tau}) = \min\left\{\frac{3}{m(m-3)}(\mu_{31} - \mu_{22}), \sup\{\delta \in \mathbb{R}\colon b(\bar{\tau}) + \delta c \in \mathscr{P}(m)\}\right\},$$
$$\delta_{\min}(\bar{\tau}) = -\min\left\{\frac{3}{m(m-3)}(\mu_{31} - \mu_{22}), \sup\{\delta \in \mathbb{R}\colon b(\bar{\tau}) - \delta c \in \mathscr{P}(m)\}\right\}.$$

The values $\delta \in [\delta_{\min}(\bar{\tau}), \delta_{\max}(\bar{\tau})]$ are such that the line segment $b(\bar{\tau}) + \delta c$ stays in the moment polytope $\mathscr{P}(m)$, while at the same time observing (4).

The following existence statement provides the basis for the completeness Theorem 2.4.

LEMMA 2.3. *Let $\overline{\tau}$ be an exchangeable design on the simplex $\mathscr{T}$. Then we have $\delta_{\min}(\overline{\tau}) \leq 0 \leq \delta_{\max}(\overline{\tau})$. For every $\delta \in [\delta_{\min}(\overline{\tau}), \delta_{\max}(\overline{\tau})]$, there exists a weighted centroid design $\eta(\delta)$ satisfying $M(\eta(\delta)) \geq M(\overline{\tau})$. Equality holds if and only if $\overline{\tau}$ is a weighted centroid design (which implies $\delta = 0$).*

PROOF. If $b(\overline{\tau})$ lies in $\mathscr{P}(m)$ then the suprema in (9) are nonnegative. They are also finite, since $\mathscr{P}(m)$ is bounded. This proves $\delta_{\min}(\overline{\tau}) \leq 0 \leq \delta_{\max}(\overline{\tau})$.

We need to prove that indeed $b(\overline{\tau})$ lies in $\mathscr{P}(m)$, for every exchangeable design $\overline{\tau}$. Let $\varepsilon(t)$ denote the one-point design in $t \in \mathscr{T}$, and $\overline{\varepsilon}(t)$ its average over the permutation group. Since $\overline{\tau}$ lies in the convex hull of the one-point designs $\overline{\varepsilon}(t)$, it suffices to show that $\operatorname{conv}\{b(\overline{\varepsilon}(t)): t \in \mathscr{T}\} \subseteq \mathscr{P}(m)$. This is achieved by investigating the associated support functions. For $x \in \mathbb{R}^4$ we define $f(x) = \max_{t \in \mathscr{T}} x'b(\overline{\varepsilon}(t))$ and consider all vectors $s \in \mathscr{T}$ attaining the maximum in this definition. Thus

$$
\begin{aligned}
f(x) &= x'b(\overline{\varepsilon}(s)) \\
&= x_1\bigg(\sum_i s_i^4 + \tfrac{1}{4}\sum_{i,j}{}' s_i s_j (s_i - s_j)^2\bigg) + \frac{x_2}{4}\sum_{i,j}{}' s_i s_j (s_i + s_j)^2 \\
&\quad + x_3 \sum_{i,j,k}{}' s_i^2 s_j s_k + x_4 \sum_{i,j,k,l}{}' s_i s_j s_k s_l.
\end{aligned}
\tag{10}
$$

Again, Σ' means that the summation is restricted to pairwise distinct subscripts. From all the possible vectors in (10) we choose a particular one, again denoted by s, for which the number of positive components is a minimum, say j. We claim that

$$
f(x) \leq \max\{x'a_1, \dots, x'a_m\} \quad \text{for all } x \in \mathbb{R}^4. \tag{11}
$$

In case $j = 1$, we obtain $\overline{\varepsilon}(s) = \eta_1$. Then $b(\overline{\varepsilon}(s)) = a_1$ proves (11). In case $j > 1$ we select two arbitrary components of s that are positive. Because of exchangeability, there is no loss of generality in assuming $s_1 > 0$ and $s_2 > 0$. We define the polynomial

$$
Q(t) = x'b\big(\overline{\varepsilon}(t, s_1 + s_2 - t, s_3, \dots, s_m)\big), \qquad t \in [0, s_1 + s_2].
$$

By construction, Q has local maxima at the interior points s_1 and s_2. If Q is constant then $(0, s_1 + s_2, s_3, \dots, s_m)'$ fulfills (10) while having only $j - 1$ positive components. This contradicts our assumption on the minimality of j. Nor can Q be linear. Furthermore the term t^4 arises only within the two sums that accompany x_1 in (10). The coefficient of t^4 is easily found to vanish. Therefore the polynomial Q has degree two or three and cannot have two distinct local maxima, forcing $s_1 = s_2$. Since the choice of s_1 and s_2 was arbitrary, we conclude that all positive components of s are equal to $1/j$. This entails $\overline{\varepsilon}(s) = \eta_j$, and $b(\overline{\varepsilon}(s)) = a_j$ proves (11). Hence $b(\overline{\tau})$ lies in the moment polytope $\mathscr{P}(m)$.

If equality holds, then $\Delta = 0$ in (2) and $\gamma = \delta = 0$. From (3) we get $\mu_{31} = \mu_{22}$ whence, by Lemma 2.2, $\bar{\tau}$ is a weighted centroid design. Conversely, if $\bar{\tau}$ is a weighted centroid design, then $\mu_{31} = \mu_{22}$ and $\delta_{\max}(\bar{\tau}) = \delta_{\min}(\bar{\tau}) = 0$. The only choice is $\delta = 0$. Thus (3) forces $\gamma = 0$, entailing $\Delta = 0$ in (2). □

Even for $\delta = 0$, the weighted centroid design $\eta(0)$ in Lemma 2.3 is not unique except for $m = 4$. For this reason, minimal completeness must be weakened to completeness for $m \geq 5$. Because the Kiefer ordering does not depend on the form in which the model is specified, it is not necessary to distinguish between the Kronecker model or any other alternative model form of second order.

THEOREM 2.4. *In the second-degree mixture model for $m \geq 4$ ingredients, the set of weighted centroid designs $\mathscr{C} = \{\alpha_1\eta_1 + \cdots + \alpha_m\eta_m : (\alpha_1, \ldots, \alpha_m)' \in \mathscr{T}\}$ is convex and constitutes a complete class of designs for the Kiefer ordering. For $m = 4$, the class is minimal complete.*

PROOF. In view of Lemma 2.3, the results follow as in Theorems 6.4 and 7.4 of Draper and Pukelsheim (1999). □

3. The moment polytope. In practice, one does not need the full complement of m terms indicated in Theorem 2.4. The reason is that the moment polytope $\mathscr{P}(m)$ in (8) lies in a three-dimensional affine subspace, so that any point in it can be represented as a convex combination of no more than four vertices, by the Carathéodory Theorem. Hence four weights suffice; see Theorem 3.2. The following geometric argument describes the structure of $\mathscr{P}(m)$.

LEMMA 3.1. *The moment polytope $\mathscr{P}(m)$ in (8) has two disjoint sets of faces, each consisting of $m - 2$ triangles. Set I consists of the triangles with vertices a_i, a_{i+1}, a_m, for $i = 1, \ldots, m-2$, each inducing on the moment vector $b(\bar{\tau}) + \delta c$ in (6) the inequality*

$$\begin{aligned} 0 \leq\ & i(i-1)\mu_4 + \tfrac{1}{2}(i-1)(i-2)(m-2)\mu_{31} - \tfrac{1}{2}(i-1)(im+2m-4)\mu_{22} \\ & + (2i+m-5)(m-2)\mu_{211} - (m-2)(m-3)\mu_{1111} \\ & - \tfrac{1}{6}i(i+1)m(m-2)(m-3)\delta. \end{aligned}$$

Set II consists of the triangles with vertices a_1, a_j, a_{j+1}, for $j = 2, \ldots, m-1$, each with associated moment inequality

$$\begin{aligned} 0 \leq\ & \tfrac{1}{2}(j-1)(j-2)(\mu_{31} + \mu_{22}) - 2(j-2)(m-2)\mu_{211} \\ & + (m-2)(m-3)\mu_{1111} + \tfrac{1}{6}j(j+1)(m-2)(m-3)\delta. \end{aligned}$$

PROOF. Since $m \geq 4$, the two sets are disjoint. There are only three degrees of freedom in the moment polytope, and we choose to omit the first component. We distinguish the resulting quantities with a tilde so that, for example, a_j in (6) turns into $\tilde{a}_j = j^{-3}(j-1, (j-1)(j-2), (j-1)(j-2)(j-3))'$.

For $1 \le i < j < k \le m$, we study the hyperplane $\tilde{\mathscr{H}}_{ijk}$ in $\mathbb{R}^3$ generated by $\tilde{a}_i, \tilde{a}_j, \tilde{a}_k$. A vector orthogonal to $\tilde{\mathscr{H}}_{ijk}$ is

$$\tilde{z}_{ijk} = \begin{pmatrix} 4(i+j+k) - 6(ij+ik+jk) + 7ijk \\ 5(i+j+k) - 6(ij+ik+jk) + 6ijk \\ i+j+k-(ij+ik+jk)+ijk \end{pmatrix}.$$

For a generating vertex, that is, for $l = i, j, k$, we have $\tilde{z}_{ijk}{}'\tilde{a}_l = (i-1)(j-1)(k-1)$. For an arbitrary vertex $\tilde{a}_l$, that is, $l = 1, \ldots, m$, its position relative to the hyperplane $\tilde{\mathscr{H}}_{ijk}$ is indicated by the sign of the difference of the inner products,

$$d(\tilde{a}_l) \equiv \tilde{z}'_{ijk}\tilde{a}_l - \tilde{z}'_{ijk}\tilde{a}_i = \frac{1}{l^3}(l-i)(l-j)(l-k). \tag{12}$$

If $i > 1$ and $k < m$, we insert 1 and m for l and get $d(\tilde{a}_1) < 0 < d(\tilde{a}_m)$. Hence the vertices $\tilde{a}_1$ and $\tilde{a}_m$ lie on opposite sides of $\tilde{\mathscr{H}}_{ijk}$. The hyperplane $\tilde{\mathscr{H}}_{ijk}$ bisects $\tilde{\mathscr{P}}(m)$, rather than being supporting to it. If $k = m$ and it happens that $i < l < j$, or if $i = 1$ and it happens that $j < l < k$, then again $\tilde{\mathscr{H}}_{ijk}$ bisects $\tilde{\mathscr{P}}(m)$. Otherwise, we have either $j = i+1$ and $k = m$, in which case $d(\tilde{a}_l) \le 0$ for all $l = 1, \ldots, m$. Or, we have $i = 1$ and $k = j+1$, in which case $d(\tilde{a}_l) \ge 0$ for all $l = 1, \ldots, m$. Either way, the hyperplane $\tilde{\mathscr{H}}_{ijk}$ is supporting to $\tilde{\mathscr{P}}(m)$. The intersections of the supporting hyperplanes with the polytope yield the two sets of triangular faces stated in the assertion.

The moment inequalities arise from $d(\tilde{b} + \delta\tilde{c})$ being less than or equal to 0 or greater than or equal to 0 in the two cases, respectively. For Set I this leads to $0 \le (i-1)i(m-1) - \tilde{z}_{i,\,i+1,\,m}{}'\tilde{b} - \delta\tilde{z}_{i,\,i+1,\,m}{}'\tilde{c}$. We convert the first term into a linear combination of fourth-order moments and collect terms to establish the first inequality of the assertion. For Set II we get $0 \le \tilde{z}_{1,\,j,\,j+1}{}'\tilde{b} + \delta\tilde{z}_{1,\,j,\,j+1}{}'\tilde{c}$. Collecting terms and factorizing the coefficients leads to the second inequality. □

The inequalities of Lemma 3.1 help to recalculate the bounds (9) for δ. Given the fourth-order moments of a design $\bar{\tau}$, we define

$$\begin{aligned} g(i) = {} & \frac{6}{i(i+1)m(m-2)(m-3)} \\ & \times \Big(i(i-1)\mu_4 + \frac{1}{2}(i-1)(i-2)(m-2)\mu_{31} - \frac{1}{2}(i-1)(im+2m-4)\mu_{22} \\ & \qquad + (2i+m-5)(m-2)\mu_{211} - (m-2)(m-3)\mu_{1111} \Big), \end{aligned}$$

$$h(j) = \frac{6}{j(j+1)(m-2)(m-3)} \times \left(\frac{1}{2}(j-1)(j-2)(\mu_{31}+\mu_{22}) - 2(j-2)(m-2)\mu_{211} + (m-2)(m-3)\mu_{1111} \right).$$

The largest δ for which $b(\overline{\tau}) + \delta c$ stays in $\mathscr{P}(m)$ is equal to the smallest value of $g(i)$, for $i = 1, \dots, m-2$. In the opposite direction, the relevant minimum is that of $h(j)$, for $j = 2, \dots, m-1$. To find the minimizers $i(\overline{\tau})$ and $j(\overline{\tau})$ for g and h, we use a recursion involving third-order moments of $\overline{\tau}$,

$$g(i+1) = g(i) + \frac{12}{i(i+1)(i+2)m(m-2)(m-3)} \times \Big((\mu_3 + \mu_{21})i - (m-2)(\mu_{21} - \mu_{111}) \Big),$$

$$h(j+1) = h(j) + \frac{12}{j(j+1)(j+2)(m-2)(m-3)} \times \Big(\mu_{21} j - (\mu_{21} + (m-2)\mu_{111}) \Big).$$

For convex combinations $\overline{\tau}$ of the vertex design η_1 and the overall centroid design η_m we set $i(\overline{\tau}) = 1$. For all other designs $\overline{\tau}$, we can show that $\mu_3 - \mu_{21} \geq \mu_{21} - \mu_{111} = \frac{1}{2}\int (t_1 - t_2)^2 t_3 \, d\overline{\tau} > 0$. Hence $(m-2)(\mu_{21} - \mu_{111})/(\mu_3 - \mu_{21})$ lies in the half-open interval $(0, m-2]$. Now g is minimized by rounding this number to the smallest integer above it or equal to it, $i(\overline{\tau}) = \lceil (m-2)(\mu_{21} - \mu_{1111})/(\mu_3 - \mu_{21}) \rceil \in \{1, \dots, m-2\}$. As for minimizing h, we set $j(\overline{\tau}) = 2$ in the case $\mu_{111} = 0$. In all other cases we have $\mu_{21} \geq \mu_{111} > 0$. Hence $1 + (m-2)\mu_{111}/\mu_{21}$ lies in the half-open interval $(1, m-1]$. Now h is minimized at $j(\overline{\tau}) = \lceil 1 + (m-2)\mu_{111}/\mu_{21} \rceil \in \{2, \dots, m-1\}$. Altogether, (9) reduces to

$$\delta_{\max}(\overline{\tau}) = \min\left\{ \frac{3}{m(m-3)}(\mu_{31} - \mu_{22}), g\big(i(\overline{\tau})\big) \right\},$$
(13)
$$\delta_{\min}(\overline{\tau}) = -\min\left\{ \frac{3}{m(m-1)}(\mu_{31} - \mu_{22}), h\big(j(\overline{\tau})\big) \right\}.$$

Now we are in a position to indicate which choices of four weights suffice. The moment polytope $\mathscr{P}(m)$ contains the $m-2$ tetrahedra $\mathscr{P}_j(m) = \operatorname{conv}\{a_1, a_j, a_{j+1}, a_m\}$, of which any two may share boundary points, but no interior points. Thus, we can select a subclass of the set $\mathscr{C}$ in Theorem 2.4 that is essentially complete, as follows.

THEOREM 3.2. *In the second-degree mixture model for five or more ingredients, the set* $\mathscr{C}' = \bigcup_{j=2}^{m-2} \operatorname{conv}\{\eta_1, \eta_j, \eta_{j+1}, \eta_m\}$ *constitutes an essentially complete class of exchangeable designs for the Kiefer ordering.*

4. Examples and discussion. Our results suggest a simple practical procedure for Kiefer improving an arbitrary mixture design τ. First, compute the fourth-order moments of the exchangeable design $\overline{\tau}$. Then, applying Lemma 2.3 with $\delta_{\min}$ and $\delta_{\max}$ from (13) gives the one-parameter family of those exchangeable design $\eta(\delta)$ which improve upon $\overline{\tau}$ and which cannot be further improved upon. According to Theorem 3.2, improvement designs can be found which involve not more than four elementary centroid designs; more can be used if desired. We now comment upon some specific practical aspects of our design improvement procedures.

1. It is possible that the improvement procedure will reduce the initial number of support points by relocating some of them. A simple $m = 3$ example appears in Draper and Pukelsheim [(1999), page 341]. The initial design is the $\{3, 3\}$ simplex lattice with ten support points. The improvement design consists of seven support points, keeping the three vertex locations, with weight $\alpha_1 = \frac{11}{30}$, replacing the six points of type $(\frac{1}{3}, \frac{2}{3}, 0)$ and so on, with the three points $(\frac{1}{2}, \frac{1}{2}, 0)$ and so on, with weight $\alpha_2 = \frac{16}{30}$, and keeping the overall centroid location, with weight $\alpha_3 = \frac{3}{30}$.
2. When the initial design has fewer support points than parameters, the design improvement procedure can provide additional needed support points. For $m = 4$, consider the design τ_r which assigns weight $\frac{1}{6}$ to each of the six permutations of $(\frac{1}{2}-r, \frac{1}{2}-r, r, r)'$, for $r \in [0, \frac{1}{4})$. (For $r = 0$ this gives the edge midpoints design.) Six support points are inadequate for estimating the ten parameters of the second-order model. The improvement design $\eta(0)$ from Lemma 2.3 has positive weights,

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

   and thus 15 support points. With $\delta = 0$, the value of γ in Lemma 2.1 is $\frac{1}{8}\alpha_1(r)$. The bounds $\delta_{\max}$ and $\delta_{\min}$ for δ in (13), in terms of the preceding weights, are

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

   As a result of the improvement procedure, the rank of the moment matrix has increased from six to fifteen. Further discussion for four ingredients is contained in Draper, Heiligers and Pukelsheim (2000).
3. When $m \leq 4$ and the initial design uses some or all of the elementary centroid designs, there will be no improvement through our methods because the initial design is simply reproduced.

4. When $m \geq 5$, improvement will occur if the initial design contains more than four elementary designs. The moment matrix will be unchanged; the improvement lies in a reduction in the number of support points needed. For example, let $\overline{\tau} = \frac{1}{5}(\eta_1 + \eta_2 + \eta_3 + \eta_4 + \eta_5)$ be the average of the elementary centroid designs for $m = 5$. In view of $\delta_{\max}(\overline{\tau}) = 0 = \delta_{\min}(\overline{\tau})$, the only possible value is $\delta = 0$, and (3) entails $\gamma = 0$, implying that the moment matrix does not change; see (2). Relying on Theorem 3.2, we solve (7) with one of the five weights $\alpha_j = 0$ set equal to zero. For $j = 5, 2$, and 1, the resulting solutions contain a negative component. For $j = 4$ and $j = 3$, respectively, we obtain two legitimate solutions, with weights

$$\alpha_1 = \frac{257}{1280} = 0.201, \quad \alpha_2 = \frac{224}{1280} = 0.175,$$
$$\alpha_3 = \frac{418}{1280} = 0.326, \quad \alpha_5 = \frac{381}{1280} = 0.298;$$

and

$$\alpha_1 = \frac{161}{810} = 0.199, \quad \alpha_2 = \frac{194}{810} = 0.239,$$
$$\alpha_4 = \frac{418}{810} = 0.516, \quad \alpha_5 = \frac{37}{810} = 0.046.$$

Both solutions, and all of their convex combinations, reproduce the moment matrix $M(\overline{\tau})$. However, the first choice leaves a design with 26 support points, while the second choice leaves 21. Designs formed by convex combinations all employ the initial 31 support points, with weights depending on the combination used.

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