DEPARTMENT OF ENGINEERING MANAGEMENT

I-optimal mixture designs

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Abstract

In mixture experiments, the factors under study are proportions of the ingredients of a mixture. The special nature of the factors in a mixture experiment necessitates specific types of regression models, and specific types of experimental designs. Although mixture experiments usually are intended to predict the response(s) for all possible formulations of the mixture and to identify optimal proportions for each of the ingredients, little research has been done concerning their I-optimal design. This is surprising given that I-optimal designs minimize the average variance of prediction and, therefore, seem more appropriate for mixture experiments than the commonly used D-optimal designs, which focus on a precise model estimation rather than precise predictions. In this paper, we provide the first detailed overview of the literature on the I-optimal design of mixture experiments and identify several contradictions. For the second-order, special cubic and qth degree models, we present I-optimal continuous designs and contrast them with the published results. We also study exact I-optimal designs, and compare them in detail to continuous I-optimal designs and to D-optimal designs. One striking result of our work is that the performance of D-optimal designs in terms of the I-optimality criterion very strongly depends on which of the D-optimal design points are replicated.

Keywords: D-optimality, IV-optimality, moments matrix, Q-optimality, simplex-lattice designs, simplex-centroid designs, V-optimality.

1 Introduction

In recent years, prediction-based optimality criteria have gained substantial popularity for generating response surface designs. The best-known prediction-based optimality criteria are the G-optimality criterion, which seeks designs that minimize the maximum prediction variance over the experimental region, and the I-optimality criterion, which seeks designs that minimize the average prediction variance over the experimental region. Within the optimal experimental design community, the I-optimality criterion is often called the V-optimality criterion (see, for instance, Atkinson et al. (2007)), but the names IV- or Q-optimality have been used as well (Borkowski, 2003; Letsinger, Myers and Lentner, 1996). In this paper, we refer to designs that minimize the average prediction variance as I-optimal designs. Concerning the I- and G-optimality criteria, Montgomery (2009) writes that "they would be most likely used for second-order models, as second-order models are often used for optimization, and good prediction properties are essential for optimization." The increasing use of variance dispersion graphs and fraction of design space plots (Giovannitti-Jensen and Myers, 1989; Zahran and Anderson-Cook, 2003) has contributed to the increasing focus on prediction variances when selecting experimental designs.

The generation of I-optimal completely randomized designs is discussed in Haines (1987), Meyer and Nachtsheim (1988, 1995), Hardin and Sloane (1993) and Borkowski (2003). Hardin and Sloane (1993) demonstrate that D-optimal response surface designs perform poorly in terms of the I-optimality criterion, while I-optimal designs perform reasonably well with respect to the D-optimality criterion. This phenomenon is more pronounced when the experimental region is cuboidal than when it is spherical. Goos and Jones (2011) report an example of a completely randomized response surface experiment involving a three-level categorical factor, where the performance of the I-optimal design in terms of the D-optimality criterion is much better than the performance of the D-optimal design in terms of the I-optimality criterion. Jones and Goos (2012) obtain similar results for split-plot response surface designs. The generation of G-optimal completely randomized designs is treated in Borkowski (2003) and Rodríguez et al. (2010). The latter authors point out that, to minimize the maximum variance of prediction, it is often necessary to accept larger prediction variances over most of the region of interest, and suggest using the I-optimality criterion rather than the G-optimality criterion. In this article, we therefore focus on the I-optimality criterion.

Despite the increasing use of the I-optimality criterion for response surface designs, little is known concerning I-optimal mixture designs. This is surprising for two reasons:

1. In his seminal paper on mixture experiments, Scheffé (1958) already suggests using the variance of the predicted response as a starting point to design mixture experiments. In follow-up work, Lambrakis (1968a,b) formally defines an I-optimality criterion for two special types of mixture designs, which are not commonly used, but he did not succeed in constructing I-optimal designs due to the fact that the required analytical expressions were intractable and the available computing power was limited.

2. Mixture designs are special cases of response surface designs, and prediction and optimization are the main goals of most mixture experiments. The two textbooks on the design of mixture experiments (Cornell, 2002; Smith, 2005) mention the I-

optimality criterion briefly, but they do not discuss I-optimal mixture designs in any detail. The reference books by Atkinson, Donev and Tobias (2007) and Goos and Jones (2011) briefly discuss the D-optimality of some commonly used mixture designs, but pay no attention to the I-optimal mixture designs.

The only published results concerning I-optimal mixture designs for Scheffé models can be found in Lambrakis (1968b), Laake (1975) and Liu and Neudecker (1995). I-optimal designs for Becker models are discussed in Liu and Neudecker (1997).

In this paper, we consider the most commonly used model types, the Scheffé models. We introduce these models in Section 2. Next, in Section 3, we define the D- and I-optimality criteria for continuous and for exact designs. We summarize the theoretical results concerning I-optimality produced by Laake (1975) and Liu and Neudecker (1995) and concerning D-optimality in Section 4. In Section 5, we present continuous I-optimal designs for the second-order, special cubic and qth degree Scheffé models and show that they outperform the I-optimal arrangements presented by Laake (1975). In Section 6, we discuss exact I-optimal designs and contrast them with the continuous I-optimal designs. Also, we provide a detailed comparison of exact I-optimal mixture designs and exact D-optimal mixture designs in terms of the variance of prediction using fraction of design space plots. In the paper's final section, we summarize the key results and discuss the usefulness of the SAS procedure OPTEX for generating I-optimal designs.

2 Statistical models and analysis

In models for data from mixture experiments involving q ingredients, the explanatory variables are the q ingredient proportions $x_1, x_2, \ldots, x_q$. A key feature of these explanatory variables is that they sum to one:

$$\sum_{i=1}^{q} x_i = \mathbf{x}'\mathbf{1}_q = 1, \tag{1}$$

where $\mathbf{x}' = (x_1, x_2, \ldots, x_q)$ and $\mathbf{1}_q$ is a q-dimensional vector of ones. This constraint, which is called the *mixture constraint*, defines a simplex-shaped experimental region and has a substantial impact on the models that can be fitted. The first major consequence of the mixture constraint is that a regression model involving linear terms in the ingredient proportions cannot contain an intercept. Otherwise, the model's parameters cannot be estimated uniquely. A second major consequence is that including cross-products of proportions together with squares of proportions should be avoided too, because this also results in the model parameters not being estimable uniquely. To see this, note that

$$x_i^2 = x_i\Big(1 - \sum_{\substack{j=1\\ j\neq i}}^{q} x_j\Big) = x_i - \sum_{\substack{j=1\\ j\neq i}}^{q} x_i x_j, \tag{2}$$

for every proportion x_i. As a result, the square of a proportion is a linear combination of that proportion and its cross-products with each one of the other $q-1$ proportions.

These considerations led Scheffé (1958) to propose the Scheffé mixture models. The first-order Scheffé model is given by

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i, \tag{3}$$

whereas the second-order Scheffé model is given by

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} \beta_{ij} x_i x_j. \tag{4}$$

The special cubic model can be written as

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} \beta_{ij} x_i x_j + \sum_{i=1}^{q-2} \sum_{j=i+1}^{q-1} \sum_{k=j+1}^{q} \beta_{ijk} x_i x_j x_k, \tag{5}$$

and the full cubic model is

$$\begin{aligned} E(Y) = &\sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} \beta_{ij} x_i x_j + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} \gamma_{ij} x_i x_j (x_i - x_j) \\ &+ \sum_{i=1}^{q-2} \sum_{j=i+1}^{q-1} \sum_{k=j+1}^{q} \beta_{ijk} x_i x_j x_k. \end{aligned} \tag{6}$$

Another model that has received attention in the literature (Scheffé, 1963) is the qth degree model

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} \beta_{ij} x_i x_j + \sum_{i=1}^{q-2} \sum_{j=i+1}^{q-1} \sum_{k=j+1}^{q} \beta_{ijk} x_i x_j x_k + \cdots + \beta_{12\ldots q} x_1 x_2 \ldots x_q. \tag{7}$$

The Scheffé models are by far the most commonly used ones in the literature (see, for example, Cornell (2002) and Smith (2005)). In this article, we therefore also focus on Scheffé mixture models.

3 Criteria for selecting designs

In this section, we define the I-optimality criterion, which is central to this paper. We pay special attention to the computation of the I-optimality criterion because popular implementations of the criterion are based on a grid of points covering the design region, and these implementations can be improved upon substantially by taking an analytical approach. We also define the D-optimality criterion because we use D-optimal designs as benchmarks later in this paper.

3.1 Continuous designs and exact designs

In this paper, we distinguish between continuous optimal designs and exact optimal designs. Exact designs are intended for experiments with a finite number of observations, n, and involve n design points, which are not necessarily distinct. Continuous designs involve a set of distinct design points and a weight for each of these points. The weight of each design point in a continuous design indicates the proportion of experimental tests to be performed at that point. Continuous designs have received much attention in the literature on the optimal design of experiments, because their optimality can be established using the general equivalence theorem (Kiefer and Wolfowitz; 1960) and because continuous optimal designs provide good guidance for the optimal selection of design points when the number of runs available is large. For small numbers of runs, the continuous optimal designs generally do not provide good guidance concerning the optimal selection of design points. This has led to a large body of research on exact optimal designs.

Usually, it is not possible to establish the optimality of exact designs, and experimenters have to rely on heuristic optimization algorithms, such as point-exchange and coordinate-exchange algorithms, to find good designs. Even though there is rarely any guarantee that the resulting designs are truly optimal, they are referred to as optimal exact designs. In this section, we define two versions of the D- and I-optimality criteria, one for continuous designs and one for exact designs.

3.2 D-optimality criterion

The most commonly used optimality criterion to select designs is the D-optimality criterion which seeks designs that maximize the determinant of the information matrix. For exact designs, the information matrix is computed as

$$\mathbf{M} = \mathbf{X}'\mathbf{X},$$

where $\mathbf{X}$ is the $n \times p$ model matrix, with n the number of observations and p the number of terms in the model. For continuous designs involving d distinct design points, the information matrix is given by

$$\mathbf{M} = \mathbf{X}_d'\mathbf{W}\mathbf{X}_d,$$

where $\mathbf{X}_d$ is the $d \times p$ model matrix corresponding to the d design points, $\mathbf{W} = \text{diag}(w_1, w_2, \ldots, w_d)$ is a diagonal matrix whose diagonal elements are the weights of the d design points, and $\sum_{i=1}^{d} w_i = 1$.

We use the D-efficiency to compare the quality of two (continuous or exact) designs with information matrices $\mathbf{M}_1$ and $\mathbf{M}_2$. The D-efficiency of a design with information matrix $\mathbf{M}_1$ relative to a design with information matrix $\mathbf{M}_2$ is defined as $(|\mathbf{M}_1|/|\mathbf{M}_2|)^{1/p}$. A D-efficiency larger than one indicates that Design 1 is better than Design 2 in terms of the D-optimality criterion.

3.3 I-optimality criterion

An I-optimal design minimizes the average prediction variance

$$\text{Average variance } = \frac{\int_\chi \mathbf{f}'(\mathbf{x})\mathbf{M}^{-1}\mathbf{f}(\mathbf{x})d\mathbf{x}}{\int_\chi d\mathbf{x}} \tag{8}$$

over the experimental region χ. To calculate this average variance, we exploit the fact that, when calculating the trace of a matrix product, we can cyclically permute the matrices. Therefore,

$$\text{tr}\left[\mathbf{f}'(\mathbf{x})\mathbf{M}^{-1}\mathbf{f}(\mathbf{x})\right] = \text{tr}\left[\mathbf{M}^{-1}\mathbf{f}(\mathbf{x})\mathbf{f}'(\mathbf{x})\right],$$

and

$$\begin{aligned}\int_\chi \mathbf{f}'(\mathbf{x})\mathbf{M}^{-1}\mathbf{f}(\mathbf{x})d\mathbf{x} &= \int_\chi \text{tr}\left[\mathbf{M}^{-1}\mathbf{f}(\mathbf{x})\mathbf{f}'(\mathbf{x})\right]d\mathbf{x},\\ &= \text{tr}\left[\int_\chi \mathbf{M}^{-1}\mathbf{f}(\mathbf{x})\mathbf{f}'(\mathbf{x})d\mathbf{x}\right].\end{aligned}$$

Now, note that, for any given design, the information matrix $\mathbf{M}$ is constant as far as the integration is concerned. Therefore,

$$\int_\chi \mathbf{f}'(\mathbf{x})\mathbf{M}^{-1}\mathbf{f}(\mathbf{x})d\mathbf{x} = \text{tr}\left[\mathbf{M}^{-1}\int_\chi \mathbf{f}(\mathbf{x})\mathbf{f}'(\mathbf{x})d\mathbf{x}\right],$$

so that we can rewrite the formula for the average prediction variance as

$$\text{Average variance } = \frac{1}{\int_\chi d\mathbf{x}} \cdot \text{tr}\left[\mathbf{M}^{-1}\int_\chi \mathbf{f}(\mathbf{x})\mathbf{f}'(\mathbf{x})d\mathbf{x}\right].$$

If we define

$$\mathbf{B} = \int_\chi \mathbf{f}(\mathbf{x})\mathbf{f}'(\mathbf{x})d\mathbf{x}, \tag{9}$$

then

$$\text{Average variance } = \frac{1}{\int_\chi d\mathbf{x}} \cdot \text{tr}\left[(\mathbf{X}'\mathbf{X})^{-1}\mathbf{B}\right]. \tag{10}$$

The matrix $\mathbf{B}$ is called the moments matrix because its elements are proportional to moments of a uniform distribution on the design region χ (which is a special case of the Dirichlet distribution). Assuming the experimental region is the full $(q-1)$-dimensional simplex, the elements of $\mathbf{B}$ can be obtained using the formula

$$\int_{S_{q-1}} x_1^{p_1}x_2^{p_2}\ldots x_q^{p_q}dx_1dx_2\ldots dx_q = \frac{\prod_{i=1}^q \Gamma(p_i+1)}{\Gamma\left(q+\sum_{i=1}^q p_i\right)}, \tag{11}$$

which is given, for instance, in De Groot (1970, page 63), but which also appeared, albeit in a different notation, in early work on the design of mixture experiments (see Lambrakis

1968a, 1968b). As an example, for a special cubic model involving three ingredients, the moments matrix equals

$$\mathbf{B} = \begin{bmatrix} \frac{1}{24}(\mathbf{I}_3 + \mathbf{1}_3\mathbf{1}_3') & \frac{1}{60}\mathbf{1}_3\mathbf{1}_3' - \frac{1}{120}\mathbf{J}_3 & \frac{1}{360}\mathbf{1}_3 \\ \frac{1}{60}\mathbf{1}_3\mathbf{1}_3' - \frac{1}{120}\mathbf{J}_3 & \frac{1}{360}(\mathbf{I}_3 + \mathbf{1}_3\mathbf{1}_3') & \frac{1}{1260}\mathbf{1}_3 \\ \frac{1}{360}\mathbf{1}_3' & \frac{1}{1260}\mathbf{1}_3' & \frac{1}{5040} \end{bmatrix}, \quad (12)$$

where $\mathbf{I}_3$ is the three-dimensional identity matrix, $\mathbf{J}_3$ is the three-dimensional anti-diagonal identity matrix, and $\mathbf{1}_3$ is a three-dimensional column vector of ones. In Equation (12), the expressions on the diagonal correspond to the three main effects, the three second-order terms and the cubic term, respectively. When the experimental region is the $(q-1)$-dimensional simplex, then its volume equals

$$\int_{\chi} d\mathbf{x} = \int_{S_{q-1}} d\mathbf{x} = \frac{1}{\Gamma(q)}.$$

If P_1 is the average variance of prediction of one design and P_2 is the average variance of prediction of a second design, then the I-efficiency of the former design compared to the latter is computed as

$$\text{I-efficiency} = P_2/P_1.$$

An I-efficiency larger than one indicates that Design 1 is better than Design 2 in terms of the average prediction variance.

4 Review of optimal mixture designs

Here, we provide an overview of the published results on D- and I-optimal mixture designs. We refer readers who are interested in mixture designs that are optimal with respect to other criteria to Chan (1995). Key concepts in the overview in this section are two types of mixture designs called simplex-lattice designs and simplex-centroid designs.

4.1 Simplex-lattice and simplex-centroid designs

A $\{q, m\}$ simplex-lattice design for q ingredients involves all possible formulations, the q individual ingredient proportions of which belong to the set $\{0, \frac{1}{m}, \frac{2}{m}, \ldots, 1\}$. In total, there are

$$\binom{m+q-1}{m}$$

points in a $\{q, m\}$ simplex-lattice design. For instance, a $\{3, 1\}$ simplex-lattice design involves three design points, $(1, 0, 0)$, $(0, 1, 0)$ and $(0, 0, 1)$. We call these points pure components. A $\{3, 2\}$ simplex-lattice design involves six design points, the pure components as well as the points $(\frac{1}{2}, \frac{1}{2}, 0)$, $(\frac{1}{2}, 0, \frac{1}{2})$ and $(0, \frac{1}{2}, \frac{1}{2})$. We call the latter points, involving 50% of one ingredient and 50% of another, binary mixtures in this paper. In general, there are q pure components and $\binom{q}{2} = n(n-1)/2$ binary mixtures.

The full simplex-centroid design involves

$$2^q - 1 = q + \binom{q}{2} + \cdots + \binom{q}{r} + \cdots + 1$$

design points in total: the q pure components, the $\binom{q}{2}$ binary mixtures, the $\binom{q}{3}$ permutations of the ternary mixture $(\frac{1}{3}, \frac{1}{3}, \frac{1}{3}, 0, \ldots, 0)$, the $\binom{q}{4}$ permutations of the quaternary mixture $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4}, \frac{1}{4}, 0, \ldots, 0)$, ..., the q permutations of the mixture $(\frac{1}{q-1}, \frac{1}{q-1}, \ldots, \frac{1}{q-1}, 0)$, and, finally, the mixture $(\frac{1}{q}, \frac{1}{q}, \ldots, \frac{1}{q})$ involving an equal proportion of all q components. For every number of ingredients q, there is only one simplex centroid design, but there is a family of simplex-lattice designs. The simplex-centroid design involves the overall centroid, and the centroids of all lower dimensional simplices.

An important fraction of the simplex-centroid design consists of the pure components, the binary mixtures and the ternary mixtures. That fraction involves

$$q + \binom{q}{2} + \binom{q}{3}$$

design points. In the remainder of this paper, we will refer to this design as the $\{q, 3\}$ simplex-centroid design.

4.2 D-optimal designs

D-optimal continuous designs for the four Scheffé mixture models (3)–(6) discussed in Section 2 are known in case the design region is the $(q-1)$-dimensional simplex. The D-optimal designs have two key features. First, they are minimum support designs, meaning that the number of distinct design points, d, equals the number of model parameters, p. Second, the weight of each design point in the D-optimal design is the same. So, $w_1 = w_2 = \cdots = w_d = 1/p$.

The D-optimality of the $\{q, 1\}$ and $\{q, 2\}$ simplex-lattice designs for the first- and second-order Scheffé models, respectively, was established by Kiefer (1961). Uranisi (1964) showed that the $\{q, 3\}$ simplex-centroid designs are D-optimal for special cubic models. For the full cubic model, the D-optimal design points are neither given by the simplex-centroid design, nor by a simplex-lattice design. The D-optimal design for a q-ingredient full cubic model involves the q pure components, $2\binom{q}{2}$ mixtures involving 27.64% of one ingredient and 72.36% of another ingredient (the exact proportions are given by $(1 \pm 1/\sqrt{5})/2$), and the $\binom{q}{3}$ ternary mixtures. So, a D-optimal design for a full cubic model can be constructed by replacing the proportions 1/3 and 2/3 of the $\{q, 3\}$ simplex-lattice design by 0.2764 and 0.7236, respectively. A general proof for the D-optimality of these designs for the full cubic model is given by Mikaeili (1993).

Table 1: I-optimal weights for a second-order Scheffé model according to Laake (1975), assuming that the optimal design points are the points of the $\{q, 2\}$ simplex lattice design.

i	$q = 3$			$q = 4$			$q = 5$			$q = 6$		
	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i
1	3	0.100723	0.302170	4	0.0560023	0.2240092	5	0.040000	0.200000	6	0.032792	0.196754
2	3	0.232610	0.697831	6	0.1293318	0.7759908	10	0.080000	0.800000	15	0.053550	0.803246
d	6			10			15			21		

For qth degree models, the D-optimal points are the $2^q - 1$ points of the full simplex-centroid designs. We verified that the design with weight $(2^q - 1)^{-1}$ for each of these points satisfies the general equivalence theorem. To the best of our knowledge, we are the first to establish the D-optimality of the full simplex-centroid design with equal weights for all points for the qth degree model.

Exact D-optimal designs involve the same $d = p$ design points as D-optimal continuous designs. If the budgeted number of runs, n, in a mixture experiment is an integer multiple of the number of points of the continuous D-optimal design, then $n/p = n/d$ observations have to be made at each of these points. If n is not an integer multiple of $d = p$, then the $d = p$ design points should be as equireplicated as possible. Which points are replicated most frequently is unimportant, provided the interest is in the D-optimality criterion only.

Due to the general equivalence theorem, continuous D-optimal designs are also G-optimal. This equivalence is, however, not generally valid for exact designs.

4.3 I-optimal designs

A limited number of theoretical results have been published concerning the I-optimal design of mixture experiments. In this section, we provide a tabular overview of the published results and point out a few conflicting results in the literature. The analytical expressions on which the tables are based are given in the appendices.

4.3.1 Second-order model

Laake (1975) analytically derived the I-optimal weights for the designs points in case $q \geq 3$, assuming that the design points are the points of the $\{q, 2\}$ simplex-lattice design. The analytical expressions he obtained for the weights are given in Appendix A. Numerical values for the weights for values of q from 3 to 6 are given in Table 1. The weight w_1 indicates proportion of runs to be conducted with each pure component, whereas the weight w_2 indicates the proportion of runs to be done with each binary mixture. The table also lists the number of pure components (n_1) and the number of binary mixtures (n_2), as well as the proportion of experimental runs involving pure components (n_1w_1) and binary mixtures (n_2w_2). The table's last line contains the number of distinct design points, d, which equals $n_1 + n_2$ in this case.

Table 2: I-optimal weights for a special cubic Scheffé model according to Laake (1975), assuming that the optimal design points are the points of the $\{q, 3\}$ simplex-centroid design.

i	$q=3$			$q=4$			$q=5$			$q=6$		
	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i
1	3	0.0925292	0.2775876	4	0.0416667	0.1666668	5	0.0211965	0.1059825	6	0.0121070	0.072642
2	3	0.1482750	0.4448250	6	0.0555556	0.3333336	10	0.0275082	0.2750820	15	0.0176834	0.265251
3	1	0.2775875	0.2775875	4	0.1250000	0.5000000	10	0.0618935	0.6189350	20	0.0331053	0.662106
d	7			14			25			41		

Unlike in the D-optimal design for the second-order Scheffé model, each pure component has a weight of less than $1/d$, while each binary mixture has a weight of more than $1/d$. Roughly 20% of the experimental effort involves pure components, whereas 80% involves binary mixtures when $q \geq 4$.

Prior to Laake (1975), Lambrakis (1968a) considered the I-optimal assignment of weights to two types of design points, for a second-order model. One type of design point has one ingredient with proportion 1/2 and all other ingredients with proportion $(1/2)(q-1)^{-1}$. The other type of design point has two ingredients with proportion 1/3 each and all other ingredients with proportion $(1/3)(q-2)^{-1}$. In terms of the I-optimality criterion, the designs advocated by Laake (1975) are far superior to those proposed by Lambrakis (1968a).

4.3.2 Special cubic model

Laake (1975) also presents I-optimal weights for the special cubic model, assuming the design points are the points of the $\{q, 3\}$ simplex-centroid design. The analytical expressions he obtained for the weights are given in Appendix B. Numerical values for the weights for values of q from 3 to 6 are given in Table 2. The weight w_1 indicates proportion of runs to be conducted with each pure component, whereas the weight w_2 indicates the proportion of runs to be done with each binary mixture. Finally, the weight w_3 specifies the proportion of runs to be done with each ternary mixture. The table also lists the number of pure components (n_1), the number of binary mixtures (n_2), and the number of ternary mixtures (n_3), as well as the proportion of experimental runs involving each type of design point (n_iw_i). The table's final line gives the number of distinct design points considered, $d = n_1 + n_2 + n_3$.

The key difference between the D-optimal continuous designs for the special cubic Scheffé model and the designs suggested by Laake (1975) is that the latter attach substantially more weight to the ternary mixtures. This can be seen from the fact that n_3w_3, the proportion of runs using ternary mixtures in Table 2, is substantially larger than n_3/d, which is the proportion of runs using ternary mixtures in the D-optimal continuous designs.

Table 3: I-optimal weights for a full cubic Scheffé model according to Laake (1975), assuming that the optimal design points are the points of the $\{q,3\}$ simplex-lattice design.

i	$q=3$			$q=4$			$q=5$			$q=6$		
	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i
1	3	0.041578	0.124735	4	0.017869	0.071478	5	0.011696	0.058481	6	0.009266	0.055596
2	6	0.110830	0.664980	12	0.046426	0.557113	20	0.023538	0.470760	30	0.013899	0.416973
3	1	0.210285	0.210285	4	0.092852	0.371409	10	0.047076	0.470760	20	0.026372	0.527432
d	10			20			35			56		

Table 4: I-optimal weights for qth degree Scheffé model according to Liu and Neudecker (1995), assuming that the optimal design points are the points of the full simplex-centroid design.

i	$q=3$			$q=4$			$q=5$			$q=6$		
	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i
1	3	0.1383110	0.4149330	4	0.0741078	0.2964312	5	0.0423470	0.211735	6	0.0249868	0.1499208
2	3	0.1344982	0.4981047	6	0.0603546	0.3621276	10	0.0309554	0.309554	15	0.0168995	0.2534925
3	1	0.1815725	0.1815725	4	0.0637672	0.2550688	10	0.0282260	0.2822600	20	0.0141234	0.2824680
				1	0.0863725	0.0863725	5	0.0309064	0.154532	15	0.0135286	0.2029290
							1	0.0419194	0.0419194	6	0.0151131	0.0906786
										1	0.0205124	0.0205124
d	7			15			31			63		

4.3.3 Full cubic model

The final general results presented by Laake (1975) involve the I-optimal weights for the full cubic Scheffé model, assuming the design points are the points of the $\{q,3\}$ simplex-lattice design. The analytical expressions he obtained for the weights w_1 of the pure components, w_2 of the mixtures involving 1/3 of one ingredient and 2/3 of another ingredient, and w_3 of the mixtures involving 1/3 of three different ingredients are given in Appendix C. Numerical values for the weights are given in Table 3.

4.3.4 qth degree polynomial model

Liu and Neudecker (1995) consider Scheffé's qth degree polynomial model and, assuming that the optimal points are those from the full simplex-centroid design, analytically derive weights that minimize the I-optimality criterion and claim that the resulting design satisfy the general equivalence theorem. Numerical values for the weights w_1 of the pure components, w_2 of the binary mixtures, w_3 of the ternary mixtures, w_4 of the quaternary mixtures, w_5 of the quinary mixtures, w_6 of the senary mixtures in case $q \geq 3$ are displayed in Table 4. The corresponding analytical expressions are given in Appendix D. Numerical values for the weights when $q = 2$, in which case the qth degree model is a second-order model in two ingredients, are not shown in the table. The analytical expressions in Appendix D give $w_1 = 0.3$ and $w_2 = 0.4$ in this case.

It is interesting to compare the results of Laake (1975) to those of Liu and Neudecker (1995) in case $q = 3$, because, in that case, the qth degree model reduces to the special cubic model. It is easy to see that the weights in the leftmost panel of Table 4 are different from those in the leftmost panel of Table 2. The average variance of prediction for Laake's design is 0.5363, whereas that of the Liu and Neudecker design amounts to 0.5902. The design proposed by Laake (1975) for the special cubic model in three ingredients thus is substantially better than that proposed by Liu and Neudecker (1995).

Laake (1975) also studied I-optimal designs for the qth degree model for $q = 4$, and reports the relative proportions of pure components, binary mixtures, ternary mixtures and quaternary mixtures required by the I-optimal design: w_2/w_1 should be about 1.3, w_3/w_1 should be about 2.1, and w_4/w_1 should be about 3.84. Once more, we see that the results of Liu and Neudecker (1995) are not in line with those of Laake (1975). As a matter of fact, their ratios w_2/w_1, w_3/w_1 and w_4/w_1 equal 0.81, 0.86 and 1.17. Again, Laake's design outperforms that of Liu and Neudecker.

We discuss our results for the qth degree model in the next section. The special case where $q = 2$ and the qth degree model is a second-order model is visited in Section 5.3, while the case where $q = 3$ and the qth degree model is a special cubic model is discussed in Section 5.4. Designs for q values larger than 3 are given in Section 5.5.

5 I-optimal continuous designs

In this section, we present I-optimal continuous designs and contrast them with the results reported in the literature and reviewed in Section 4. The designs were obtained by using the sequential algorithm of Wynn (1972) (see also Wu and Wynn (1978)) and verifying numerically that the general equivalence theorem is satisfied (see Sections 9.4 and 11.2 of Atkinson et al. (2007)).

5.1 General equivalence theorem for I-optimality

As explained in, for example, Section 9.2 of Atkinson et al. (2007), the general equivalence theorem provides a methodology to check the optimality of a given continuous optimal design, for any convex and differentiable design optimality criterion. The best-known special case of the general equivalence theorem states that the maximum prediction variance of D-optimal continuous designs equals the number of model parameters. A corollary of that special case is that D-optimal and G-optimal continuous designs are equivalent.

The equivalence theorem can, however, also be applied to the I-optimality criterion. Atkinson et al. (2007) explain that the I-optimality criterion is a special case of the L-optimality criterion. Therefore, a continuous design with information matrix $\mathbf{M}$ is I-optimal if and only if

$$\mathbf{f}'(\mathbf{x})\mathbf{M}^{-1}\mathbf{B}\mathbf{M}^{-1}\mathbf{f}(\mathbf{x}) \leq \mathrm{tr}(\mathbf{M}^{-1}\mathbf{B}), \tag{13}$$

where $\mathbf{B}$ is the moments matrix introduced in Section 3.3, for each point $\mathbf{x}$ in the design region χ. In this expression, $\mathbf{f}'(\mathbf{x})$ represents the model expansion of $\mathbf{x}$. The equality in

(13) holds only for the points of the design.

The equivalence theorem is not constructive, but it can be used to check the optimality of given designs. We have used the theorem to verify the I-optimality of the designs for first-order, second-order, special cubic and qth degree models we present in this section. We also used the equivalence theorem to show that most of the continuous designs presented by Laake (1975) are not I-optimal.

Except for the first-order model, we took a numerical approach, in which we randomly generated 10000 points from a uniform distribution over the design region χ. For each of these points, we computed the expression on the left-hand side of the inequality in (13) and noted that it is smaller than the right-hand side. For all the design points, we also computed the left-hand side of (13) and noted that it is equal to the inequality's right-hand side.

5.2 First-order model

The I-optimal continuous designs for the first-order Scheffé model involve the points of the $\{q, 1\}$ simplex-lattice designs with equal weight, $1/q$, for each of them. This can be verified using the general equivalence theorem. First, note that $\mathbf{M} = q^{-1}\mathbf{I}_q$ for a $\{q, 1\}$ simplex-lattice design with weight $1/q$ for each of the q design points, and that the moments matrix $\mathbf{B}$ equals $c(\mathbf{I}_q + \mathbf{1}_q\mathbf{1}_q')$. Hence, the right-hand side of (13) equals $cq\ \mathrm{tr}(\mathbf{I}_q + \mathbf{1}_q\mathbf{1}_q') = 2cq^2$. Since, for the first-order model, $\mathbf{f}(\mathbf{x}) = \mathbf{x}$, and since $\mathbf{x}'\mathbf{1}_q = \mathbf{1}_q'\mathbf{x} = 1$, the left-hand side of (13) simplifies to

$$\mathbf{f}'(\mathbf{x})\mathbf{M}^{-1}\mathbf{B}\mathbf{M}^{-1}\mathbf{f}(\mathbf{x}) = cq^2\mathbf{x}'(\mathbf{I}_q + \mathbf{1}_q\mathbf{1}_q')\mathbf{x} = cq^2(\mathbf{x}'\mathbf{x} + 1).$$

Since the elements of $\mathbf{x}$ all lie between zero and one, and sum to one, $\mathbf{x}'\mathbf{x}$ is always smaller than or equal to one. The equality only holds if $\mathbf{x}$ represents a pure component (in which case one element of $\mathbf{x}$ is one and all others are zero, $\mathbf{x}'\mathbf{x} = 1$ and $cq^2(\mathbf{x}'\mathbf{x}+1) = 2cq^2$). For any other point in the simplex, $\mathbf{x}'\mathbf{x} < 1$ and $cq^2(\mathbf{x}'\mathbf{x}+1) < 2cq^2$. The $\{q, 1\}$ simplex-lattice designs with weight $1/q$ for each point therefore satisfies the general equivalence theorem.

5.3 Second-order model

While for continuous D-optimal designs for second-order Scheffé models, the points are those from the $\{q, 2\}$ simplex-lattice design, the points of the continuous I-optimal designs for $q \geq 3$ are those from the $\{q, 3\}$ simplex-centroid design. The I-optimal weights for the pure components, the binary mixtures and the ternary mixtures in the $\{q, 3\}$ simplex-centroid design are shown in Table 5. The I-efficiencies of the designs proposed by Laake (1975) relative to the I-optimal designs we obtained are displayed in Table 6, along with the average prediction variances of Laake's designs and the I-optimal designs. Clearly, the fact that Laake (1975) did not consider the ternary mixtures has a detrimental effect

Table 5: I-optimal weights for the points of the $\{q, 3\}$ simplex-centroid design for a second-order Scheffé model.

i	$q=3$			$q=4$			$q=5$			$q=6$		
	n_i	w_i	$n_i w_i$	n_i	w_i	$n_i w_i$	n_i	w_i	$n_i w_i$	n_i	w_i	$n_i w_i$
1	3	0.100163	0.300489	4	0.051509	0.206037	5	0.031422	0.157109	6	0.021828	0.130967
2	3	0.201553	0.604659	6	0.094692	0.568149	10	0.049318	0.493183	15	0.027706	0.415592
3	1	0.094852	0.094852	4	0.056453	0.225814	10	0.034971	0.349708	20	0.022672	0.453441
d	7			14			25			41		

Table 6: Average prediction variances of Laake's designs for the second-order Scheffé model and the continuous I-optimal designs displayed in Table 5, as well as the I-efficiencies of Laake's designs relative to the I-optimal ones.

q	Laake's design	I-optimal design	I-efficiency
3	3.2856	3.2406	0.9863
4	4.5550	4.3081	0.9458
5	5.9524	5.3290	0.8953
6	7.3805	6.2976	0.8533

on the I-efficiency, especially when there are five or six ingredients.

The continuous I-optimal design for $q = 2$ has weight 1/4 for the point $(1, 0)$, 1/2 for the point $(0.5, 0.5)$, and 1/4 for the point $(0, 1)$. Hence, according to our computations, the I-optimal weight for the two pure components is $w_1 = 1/4$ and the I-optimal weight for the binary mixture is $w_2 = 1/2$. These weights differ from those produced by Liu and Neudecker (1995), $w_1 = 0.3$ and $w_2 = 0.4$. The I-efficiency of Liu and Neudecker's design relative to the I-optimal design is 96%.

The continuous I-optimal design for $q = 3$ matches the one continuous I-optimal mixture design reported in Wang et al. (2012) for the second-order Scheffé model in a series of proof-of-concept examples for a particle swarm optimization algorithm for finding D-, A- and I-optimal mixture designs for various kinds of models.

The insight that the continuous I-optimal designs for the second-order Scheffé model involve design points other than pure components and binary mixtures is new to the literature. This finding is of importance to researchers who use a point-exchange algorithm for constructing exact I-optimal designs. The output quality of such an algorithm to a large extent depends on the user-specified set of candidate design points. The fact that the continuous I-optimal designs contain ternary mixtures implies that the set of candidate points for constructing exact designs should also involve ternary mixtures when the interest is in the second-order model.

Table 7: I-optimal weights for the points of the full simplex-centroid design for a special cubic Scheffé model.

i	$q=3$			$q=4$			$q=5$			$q=6$		
	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i	n_i	w_i	n_iw_i
1	3	0.0925	0.2775	4	0.0426	0.1704	5	0.0227	0.1135	6	0.0134	0.0804
2	3	0.1483	0.4449	6	0.0557	0.3342	10	0.0248	0.2480	15	0.0126	0.1890
3	1	0.2776	0.2776	4	0.0991	0.3964	10	0.0409	0.4090	20	0.0189	0.3780
4				1	0.0990	0.0990	5	0.0413	0.2065	15	0.0186	0.2790
5							1	0.0233	0.0233	6	0.0121	0.0726
6										1	0.0013	0.0013
d	7			14			25			41		

5.4 Special cubic model

While the D-optimal continuous designs' points for special cubic Scheffé models are those from the $\{q, 3\}$ simplex-centroid design, the I-optimal continuous designs' points for $q \geq 3$ are those from the full simplex-centroid design. The I-optimal weights for the pure components, the binary mixtures, the ternary mixtures and any other required higher-order mixtures of the full simplex-centroid design are shown in Table 7. For q values ranging from 4 to 6, 9.9% up to 35.3% of the observations has to be taken at points that are not pure components, binary mixtures or ternary mixtures. The I-efficiencies of the designs proposed by Laake (1975) relative to the I-optimal designs we obtained are displayed in Table 8, along with the average prediction variances for the two sets of designs. Clearly, the fact that Laake (1975) only considered pure components, binary mixtures and ternary mixtures has a negative impact on the I-efficiency of the designs he obtained, especially when $q = 6$.

Note that, for $q = 3$, the weights of our I-optimal continuous design are identical to those obtained by Laake (1975) and to those of Wang et al. (2012), who present exactly one I-optimal design for a special cubic model. Our weights are therefore different from those produced by the analytical expressions in Liu and Neudecker (1995).

The result that the I-optimal continuous designs involve design points other than pure components, binary mixtures and ternary mixtures is also new to the literature and important to researchers using point-exchange algorithms for constructing I-optimal exact designs. The fact that the I-optimal continuous designs contain quaternary, quinary and senary mixtures implies that the set of candidate points when constructing exact designs for the special cubic model using a point-exchange algorithm should also involve these types of mixtures.

5.5 qth degree model

The two simplest qth degree models occur when q takes the value 2 or 3. We discussed the case where $q = 2$ in Section 5.3, and the case where $q = 3$ in Section 5.4. Therefore, in this section, we focus on situations where $q \geq 4$. It turns out that the I-optimal design points for the qth degree model are identical to those for the special cubic model, but the

Table 8: Average prediction variances of Laake's designs for the special cubic Scheffé model and the continuous I-optimal designs displayed in Table 7, as well as the I-efficiencies of Laake's designs relative to the I-optimal ones.

q	Laake's design	I-optimal design	I-efficiency
3	3.7543	3.7543	1.0000
4	6.1690	5.8607	0.9500
5	10.0687	8.4022	0.8345
6	15.9957	11.3257	0.7080

Table 9: I-optimal weights for the points of the full simplex-centroid design for a qth degree Scheffé model.

i	$q=4$			$q=5$			$q=6$		
	n_i	w_i	$n_i w_i$	n_i	w_i	$n_i w_i$	n_i	w_i	$n_i w_i$
1	4	0.0414	0.1656	5	0.0208	0.1040	6	0.0113	0.0678
2	6	0.0541	0.3246	10	0.0226	0.2260	15	0.0104	0.1560
3	4	0.0875	0.3500	10	0.0317	0.3170	20	0.0127	0.2540
4	1	0.1598	0.1598	5	0.0519	0.2595	15	0.0187	0.2805
5				1	0.0935	0.0935	6	0.0309	0.1854
6							1	0.0552	0.0552
d	15			31			63		

required weights are different. More specifically, the weights for the quaternary, quinary and senary mixtures are larger in the I-optimal designs for the qth degree model than in the I-optimal designs for the special cubic model. The I-optimal weights for the pure components, the binary mixtures, the ternary mixtures and any other required higher-order mixtures of the full simplex-centroid design are shown in Table 9.

The I-optimal weights we found are substantially different from those obtained by Liu and Neudecker (1995) and shown in Table 4. The I-efficiencies of Liu and Neudecker's designs relative to the I-optimal designs we obtained are displayed in Table 10, along with the average prediction variances of Liu and Neudecker's designs and the I-optimal designs. Our I-optimal weights for $q = 4$ are in line with the relative proportions of pure components, binary, ternary and quaternary mixtures specified by Laake (1975). Our results for $q = 5$ and $q = 6$ are new.

6 Exact designs

In this section, we study to what extent exact designs differ from continuous designs in case the I-optimality criterion is utilized. More specifically, we investigate whether the design points of the exact designs equal those of the continuous designs. For D-optimal mixture designs for Scheffé models, it is known that, for any number of runs n, the design points of the exact designs are identical to those for the continuous designs. We also study the performance of I-optimal exact designs under the D-optimality criterion and of D-optimal exact designs under the I-optimality criterion, to see whether Hardin and

Table 10: Average prediction variances of Liu and Neudecker's designs for the qth degree model and the continuous I-optimal designs discussed in Sections 5.3 (for $q = 2$) and 5.4 (for $q = 3$) and displayed in Table 9 (for $q \geq 4$), as well as the I-efficiencies of Liu and Neudecker's designs relative to the I-optimal ones.

q	L. & N.'s design	I-optimal design	I-efficiency
2	2.2222	2.1333	0.9600
3	4.1315	3.7543	0.9087
4	7.8432	6.1840	0.7885
5	13.0765	9.8691	0.7547
6	20.6764	15.4908	0.7492

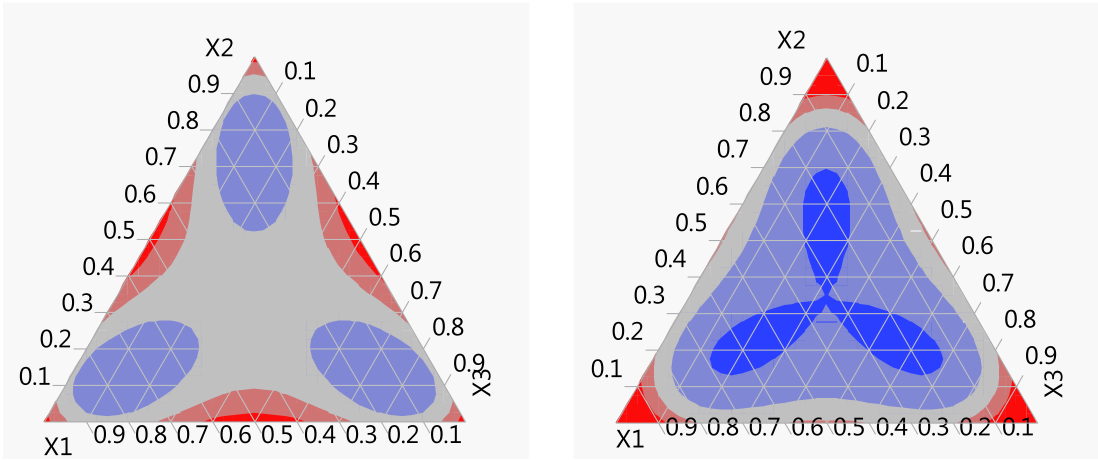
Sloane's (1993) conclusion for ordinary response surface designs, namely that D-optimal response surface designs perform considerably worse in terms of the I-optimality criterion than I-optimal designs fare in terms of the D-optimality criterion, also holds for mixture experiments. The exact designs discussed in this section were all obtained using 1,000,000 runs of the mixture coordinate-exchange algorithm of Piepel et al. (2005). Unlike point-exchange algorithms, this algorithm does not require specification of a candidate set. This is a major advantage when the location of the points of an optimal design is uncertain.

6.1 Three ingredients

6.1.1 30 runs

In this section, we compare D- and I-optimal 30-run mixture designs for estimating a second-order Scheffé model, assuming there are three ingredients ($q = 3$). The I-optimal design involves three replicates of the three pure components and the ternary mixture, and six replicates of the three binary mixtures. In this case, the exact I-optimal design thus has seven distinct design points, which match those of the continuous I-optimal design. The 30-run D-optimal design does not involve the ternary mixture and has five replicates of the pure components and the binary mixtures. The D-optimal design therefore does not collect information in the center of the design region, unlike the I-optimal design. As a consequence, the D-optimal design results in higher prediction variances in the design region's center.

Figure 1 compares the prediction variances produced by the two designs over the entire design region. Dark blue surfaces indicate small prediction variances, whereas dark red surfaces indicate areas with a large prediction variance. From the figure, it is clear that the I-optimal design leads to smaller prediction variances over most of the design region. Figure 2 compares the Fraction of Design Space (FDS) plots for the two designs. The FDS plot also shows that the I-optimal design (represented by the solid blue line) outperforms the D-optimal one (represented by the solid red line) for large portions of the design region and that the I-optimal design has a median prediction variance below 0.10, whereas



(a) D-optimal design

(b) I-optimal design

Figure 1: Comparison of the prediction variances produced by the 30-run 3-ingredient D- and I-optimal designs for a second-order Scheffé model. Dark blue areas have small prediction variances, while dark red areas have large prediction variances.

that of the D-optimal design is about 0.125. The plot also shows that the maximum prediction variance is largest for the I-optimal design. The I-optimal design results in large prediction variances in the corners of the design region, because it has fewer replicates of the pure components and therefore collects less information in the design region's corners.

The D-efficiency of the I-optimal design relative to the D-optimal design equals 89.02%, whereas the I-efficiency of the D-optimal design relative to the I-optimal design equals 85.28%. The D-optimal design thus fares slightly worse in terms of the I-optimality criterion than the I-optimal design fares in terms of the D-optimality criterion. This is in line with the results of Hardin and Sloane (1993) for ordinary response surface designs.

6.1.2 6, 7 and 8 runs

When $n = 6$ and the second-order Scheffé model is used, it is impossible for the exact I-optimal design to contain the points of the continuous I-optimal design, simply because the latter design involves seven distinct points. It turns out that the I-optimal 6-point design equals the D-optimal 6-point design, i.e. the $\{3, 2\}$ simplex-lattice design.

When $n = 7$, the exact I-optimal design is the simplex-centroid design, and, hence, involves the same design points as the continuous I-optimal design. There are six different exact D-optimal designs when $n = 7$. All of these involve the six points of the $\{3, 2\}$ simplex-lattice design, but they differ in the point they replicate. The designs also differ with respect to their performance in terms of the I-optimality criterion. The three D-optimal designs that replicate a pure component have an average prediction variance of 0.62, whereas the three D-optimal designs that replicate a binary mixture have an average

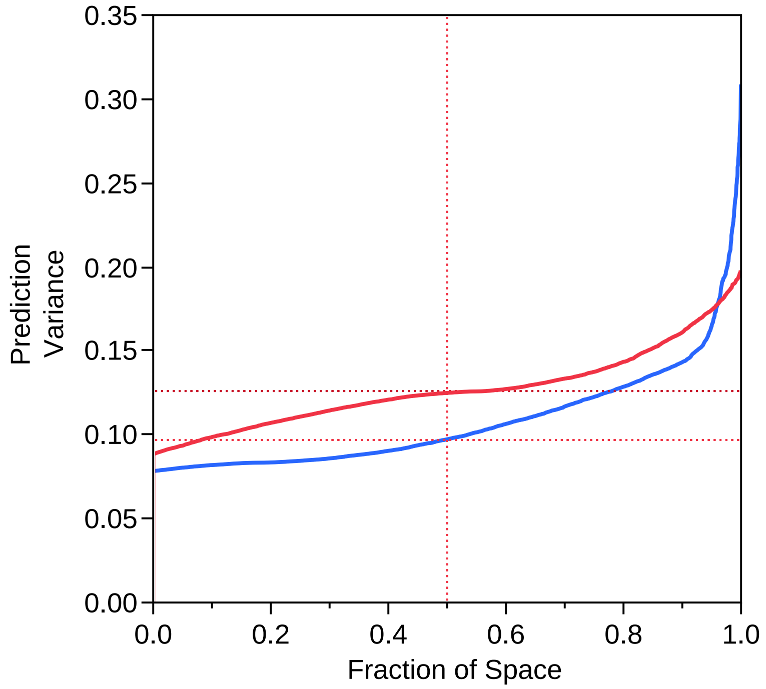


Figure 2: FDS plots for the 30-run 3-ingredient D- and I-optimal designs for a second-order Scheffé model. The D- and I-optimal designs are represented by the solid red and blue line, respectively.

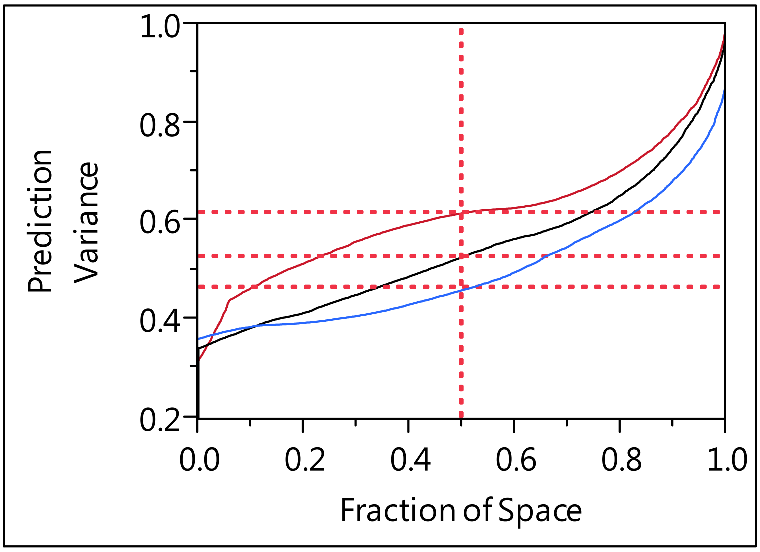


Figure 3: FDS plots for the 7-run 3-ingredient D- and I-optimal designs for a second-order Scheffé model.

prediction variance of 0.54. The I-optimal design's average variance of prediction is 0.50. The performance of the two types of D-optimal designs and the I-optimal design is compared in Figure 3. The red line corresponds to a D-optimal design involving a replicated pure component. The black line corresponds to a D-optimal design with a replicated binary mixture. Finally, the blue line corresponds to the I-optimal design. The dashed horizontal lines in the figure indicate the median prediction variances for the three designs.

The D-efficiency of the I-optimal design relative to the D-optimal designs is 96.64%. The I-efficiency of the D-optimal designs depends on which design point is replicated. In case a pure component is replicated, the I-efficiency of the resulting D-optimal design is 81.00%. In case a binary mixture is replicated, the resulting I-efficiency is 91.67%. Thus, there is a substantial difference in I-efficiency between the two types of D-optimal designs.

When $n = 8$, the I-optimal design has a surprising look, in that it involves four design points that do not belong to the simplex-centroid design. Two of these design points deviate substantially from those of the simplex-centroid design. All eight design points are listed in Table 11 and graphically displayed in Figure 4. The figure shows that the 8-run I-optimal design exhibits symmetry, in spite of the fact that it does not involve points of the simplex-centroid design only. In case $n = 8$, there are three different types of D-optimal designs:

- A type-1 D-optimal design replicates two of the three pure components.
- A type-2 D-optimal design replicates one of the pure components and one of the binary mixtures.

Table 11: 8-run I-optimal 3-ingredient design for a second-order Scheffé model.

Run	x_1	x_2	x_3
1	1.0000	0.0000	0.0000
2	0.5120	0.4880	0.0000
3	0.5000	0.0000	0.5000
4	0.4712	0.0576	0.4712
5	0.2725	0.4550	0.2725
6	0.0000	1.0000	0.0000
7	0.0000	0.4933	0.5067
8	0.0000	0.0000	1.0000

- A type-3 D-optimal design replicates two of the three binary mixtures.

The performance of these three types of D-optimal designs is compared graphically to that of the I-optimal design in Figure 5. The red, black and green lines correspond to the D-optimal designs of type 1 (with two replicated pure components), type 2 (with one replicated pure component and one replicated binary mixture) and type 3 (with two replicated binary mixtures), respectively. The blue line corresponds to the I-optimal design. The plot shows that there is a large difference in performance in terms of prediction variance between the three types of D-optimal designs. The dashed horizontal lines indicate that the type-3 D-optimal design with two replicated binary mixtures (and no replicated pure components) almost has the same median prediction variance as the I-optimal design. The type-1 D-optimal design with two replicated pure components (and no replicated binary mixtures), however, has a median variance of prediction that is about 50% larger than that of the I-optimal design.

When computing the relative efficiencies of the four competing types of designs, we see that the I-optimal design from Figure 4 has a D-efficiency of 94.14% relative to the D-optimal designs. The type-3 D-optimal design (with two replicated binary mixtures and no replicated pure components) performs very well in terms of the I-optimality criterion: it has an I-efficiency of 95.12%. The type-1 D-optimal design (with no replicated binary mixtures and two replicated pure components) performs poorly in terms of the I-optimality criterion, with an I-efficiency of merely 72.00%. The type-2 D-optimal design (with one replicated binary mixture and one replicated pure component) has an I-efficiency of 82.58%. The D-optimal designs' performance in terms of I-efficiency thus increases with the number of replicated binary mixtures.

6.2 Four ingredients

6.2.1 Second-order model

The patterns we observed for three ingredients and a second-order model persist in case the number of ingredients is increased. We illustrate this with a 15-run example involving

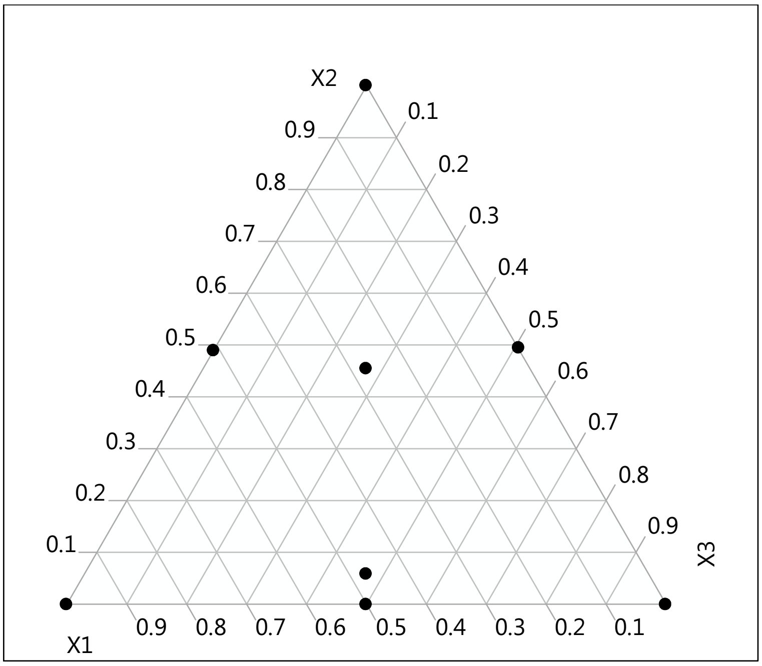


Figure 4: 8-run I-optimal 3-ingredient design for a second-order Scheffé model.

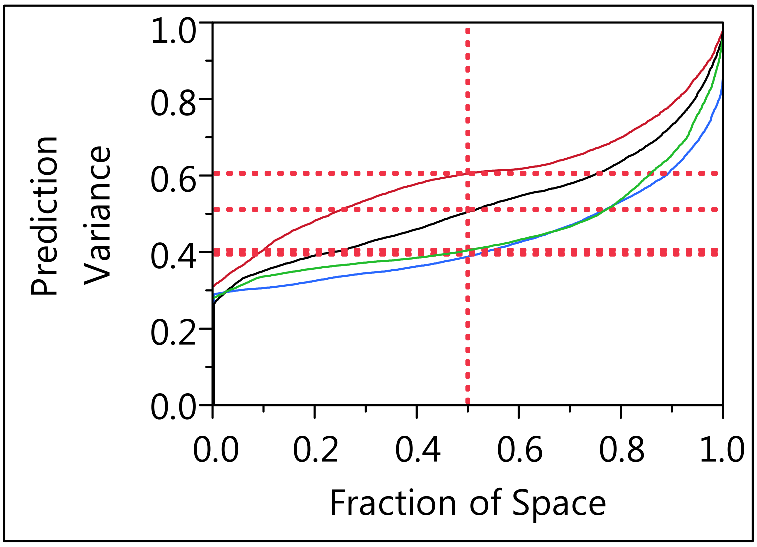


Figure 5: FDS plots for the 8-run 3-ingredient D- and I-optimal designs for a second-order Scheffé model. The red, black and green lines correspond to the D-optimal designs of type 1, 2 and 3, respectively. The blue line corresponds to the I-optimal design.

Table 12: 15-run I-optimal 4-ingredient design for a second-order Scheffé model.

Run	x_1	x_2	x_3	x_4
1	1.0000	0.0000	0.0000	0.0000
2	0.5000	0.5000	0.0000	0.0000
3	0.5000	0.5000	0.0000	0.0000
4	0.0000	1.0000	0.0000	0.0000
5	0.2876	0.2876	0.4248	0.0000
6	0.5050	0.0000	0.4950	0.0000
7	0.0000	0.5000	0.5000	0.0000
8	0.0000	0.0000	1.0000	0.0000
9	0.3331	0.0000	0.3335	0.3335
10	0.0000	0.3326	0.3337	0.3337
11	0.2885	0.2885	0.0000	0.4230
12	0.0000	0.5066	0.0000	0.4934
13	0.5000	0.0000	0.0000	0.5000
14	0.0000	0.0000	0.5000	0.5000
15	0.0000	0.0000	0.0000	1.0000

four ingredients. An I-optimal design for this scenario is shown in Table 12. All D-optimal designs for this scenario involve the four pure components and the six binary mixtures, and replicate five of these ten design points. There are five types of D-optimal designs, which can be characterized by the number of pure components that are replicated. At one end of the spectrum, there is one type of D-optimal design which replicates each of the four pure components, and one of the binary mixtures. We refer to this type of design as a type-1 D-optimal design. Of all D-optimal designs, type-1 designs are those that have the largest possible number of pure component replicates and the smallest possible number of replicates of binary mixtures. At the other end of the spectrum, there is a type of D-optimal design which replicates none of the pure components and five of the six binary mixtures. We refer to this type of design as a type-5 D-optimal design. We do not discuss the three other types of D-optimal designs, with one to three replicated pure components, because their performance in terms of variance of prediction lies in between that of the type-1 and type-5 D-optimal designs.

These two extreme types of D-optimal designs perform quite differently in terms of the I-optimality criterion. This is shown in Figure 6, where the red line corresponds to a type-1 D-optimal design (with four pure component replicates), the black line corresponds to a type-5 D-optimal design (with no pure component replicates), and the blue line corresponds to the I-optimal design in Table 12. The I-efficiency of the type-1 D-optimal design amounts to 67.80% only, while that of the type-5 D-optimal design is 93.04%. The I-efficiency of the D-optimal design thus increases substantially with the number of replicates of binary mixtures. The D-efficiency of the I-optimal design equals 90.51%.

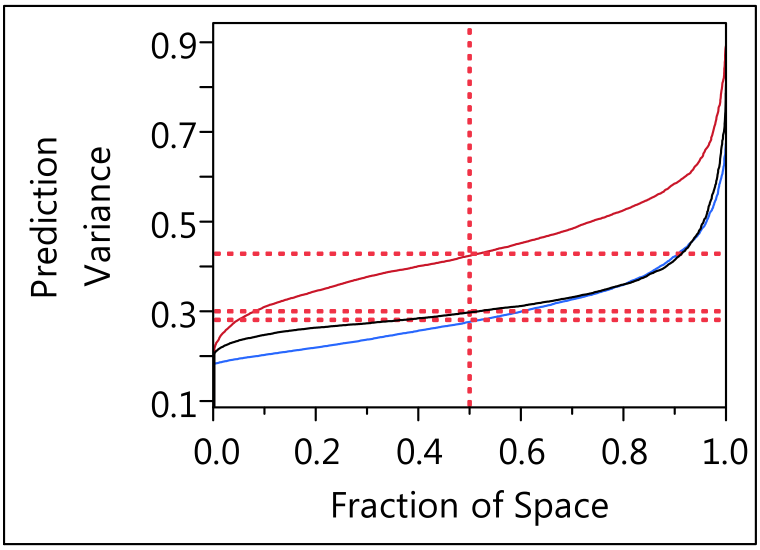


Figure 6: FDS plots for the 15-run 4-ingredient D- and I-optimal designs for a second-order Scheffé model. The red and black lines correspond to the type-1 and type-5 D-optimal designs, respectively. The blue line corresponds to the I-optimal design.

An important feature of the I-optimal design in Table 12 is that it involves design points that are different from those of the $\{q, 2\}$ simplex-lattice and $\{q, 3\}$ simplex-centroid designs. As the points are of a kind typically not included in candidate sets for point-exchange algorithms for design construction, it is impossible to obtain this design by means of a point-exchange algorithm.

6.2.2 Special cubic model

We also computed exact I-optimal designs for the special cubic model involving four ingredients. In some cases, the exact I-optimal designs' points match those of the continuous I-optimal designs, while, in other cases, the exact designs involve a few points that do not occur in the continuous I-optimal design.

For $n = 16$, for instance, the exact I-optimal 4-ingredient design we obtained involves one run with each pure component, each binary mixture and each ternary mixture, and two runs with the quaternary mixture. Hence, for $n = 16$, the points of the exact I-optimal design match those of the continuous I-optimal design. There are various types of 16-run D-optimal design for the special cubic model, all of which involve the four pure components, the six binary mixtures and the four ternary mixtures, and which do not involve the quaternary mixture. One type of D-optimal design involves two replicates of two different pure components. Another type involves two replicates of two binary mixtures and yet another type involves two replicates of two ternary mixtures. All of these types of D-optimal designs perform differently in terms of the I-optimality criterion, the best type being that with two replicated ternary mixtures. The performance of the D-optimal

Table 13: 17-run I-optimal 4-ingredient design for a special cubic model.

Run	x_1	x_2	x_3	x_4
1	1.0000	0.0000	0.0000	0.0000
2	0.5000	0.5000	0.0000	0.0000
3	0.0000	1.0000	0.0000	0.0000
4	0.3358	0.3284	0.3358	0.0000
5	0.5000	0.0000	0.5000	0.0000
6	0.0000	0.5000	0.5000	0.0000
7	0.0000	0.0000	1.0000	0.0000
8	0.2897	0.3021	0.2041	0.2041
9	0.1653	0.3034	0.2656	0.2656
10	0.3338	0.0000	0.3331	0.3331
11	0.3334	0.0000	0.3333	0.3333
12	0.3351	0.3299	0.0000	0.3351
13	0.0000	0.3298	0.3351	0.3351
14	0.0000	0.5000	0.0000	0.5000
15	0.0000	0.0000	0.5000	0.5000
16	0.5000	0.0000	0.0000	0.5000
17	0.0000	0.0000	0.0000	1.0000

designs in terms of the I-optimality criterion strongly increases with the number of replicates of the ternary mixtures, and increases also with the number of replicates of the binary mixtures.

For $n = 17$, the exact I-optimal 4-ingredient design we obtained involves one run with each pure component and each binary mixture, essentially two runs with one of the ternary mixtures and one run with each of the remaining ternary mixtures. Additionally, there are two runs involving nonzero, unequal proportions for each of the four ingredients. The 17-run I-optimal design is displayed in Table 13. This design shows that it is not always sufficient to consider the points of the continuous I-optimal design for constructing an exact I-optimal design for a special cubic model.

6.3 Five ingredients

A 20-run I-optimal design for a second-order Scheffé model involving five ingredients is shown in Table 14. The design, which results in an average prediction variance of 0.2850, involves all pure components and binary mixtures. Its other points are unusual, in the sense that they do not occur in the continuous I-optimal design for the 5-ingredient second-order model. Two of the unusual points involve three proportions of just over 0.25 and one proportion just below 0.25. The three other unusual points involve one proportion of about 0.385 and two proportions of about 0.3075. The former two points are close to quaternary mixtures contained within the continuous I-optimal design, while the latter

Table 14: 20-run I-optimal 5-ingredient design for a second-order Scheffé model.

Run	x_1	x_2	x_3	x_4	x_5
1	1.0000	0.0000	0.0000	0.0000	0.0000
2	0.5000	0.5000	0.0000	0.0000	0.0000
3	0.0000	1.0000	0.0000	0.0000	0.0000
4	0.5000	0.0000	0.5000	0.0000	0.0000
5	0.0000	0.5000	0.5000	0.0000	0.0000
6	0.0000	0.0000	1.0000	0.0000	0.0000
7	0.2531	0.2532	0.2532	0.2405	0.0000
8	0.5000	0.0000	0.0000	0.5000	0.0000
9	0.0000	0.0000	0.5000	0.5000	0.0000
10	0.0000	0.5000	0.0000	0.5000	0.0000
11	0.0000	0.0000	0.0000	1.0000	0.0000
12	0.2540	0.2540	0.2536	0.0000	0.2384
13	0.0000	0.0000	0.3897	0.3051	0.3051
14	0.3875	0.0000	0.0000	0.3063	0.3063
15	0.0000	0.3852	0.0000	0.3074	0.3074
16	0.5000	0.0000	0.0000	0.0000	0.5000
17	0.0000	0.5000	0.0000	0.0000	0.5000
18	0.0000	0.0000	0.0000	0.5000	0.5000
19	0.0000	0.0000	0.5000	0.0000	0.5000
20	0.0000	0.0000	0.0000	0.0000	1.0000

three points are close to the ternary mixtures. It is therefore tempting to conclude that the coordinate-exchange algorithm got stuck in a locally optimal design. However, replacing the five unusual points by the corresponding ternary and quaternary points leads to a slightly higher average prediction variance, 0.2865.

We also investigated exact I-optimal designs for the qth degree model. For this model, we did not come across instances where the design involves points other than those of the full simplex-centroid design. For instance, for five ingredients and 36 runs, the I-optimal design involves the five pure components, the ten binary mixtures and the ten ternary mixtures once, giving 25 runs already. Two of the quaternary mixtures appear once in the designs, while the other three appear twice. Finally, the quinary mixture appears thrice, so that the total number of runs equals 36. The design has an average variance of prediction of 0.2919. D-optimal designs with 36 runs all involve one or two observations at each of the 31 points of the full simplex-centroid design. In each D-optimal design, exactly five design points are replicated. Depending on which points are replicated, the D-optimal designs perform differently in terms of the I-optimality criterion. The type-1 D-optimal design that replicates each of the five pure components has an average prediction variance of 0.3800. Therefore, its I-efficiency is 76.82%. The type-2 D-optimal design that replicates each of the five quaternary mixtures has an average variance of pre-

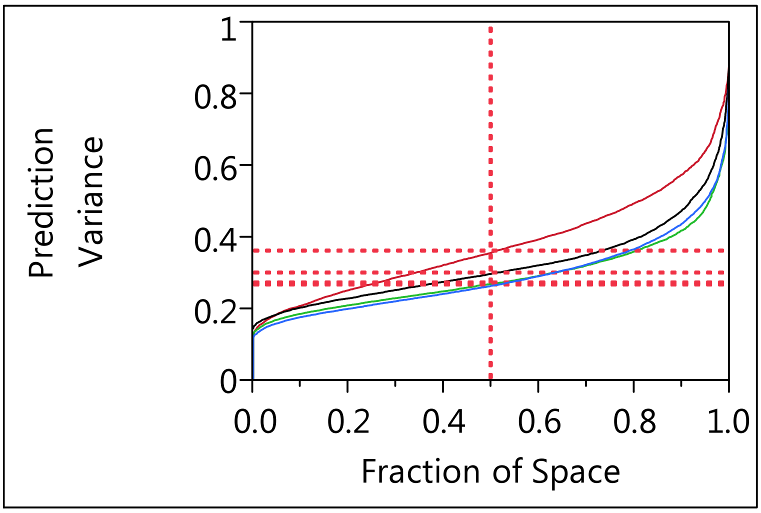


Figure 7: FDS plots for the 36-run 5-ingredient D- and I-optimal designs for a qth degree model. The red, black and green lines correspond to the D-optimal designs of type 1, 2 and 3, respectively. The blue line corresponds to the I-optimal design.

diction of 0.3243 and, therefore, an I-efficiency of 90.01%. Finally, the type-3 D-optimal design that replicates the quinary mixture and four of the five quaternary mixture (and which is the best D-optimal design in terms of I-efficiency) has an average prediction variance of 0.2928 and an I-efficiency of 99.69%. In this scenario, there are many more than three types of D-optimal designs. We do not discuss all of them in detail because their performances in terms of I-efficiency lie in between those of the type-1 and type-2 designs. In any case, the larger the number of replicates of quinary and quaternary mixtures, the better the D-optimal designs' performance in terms of the I-optimality criterion.

The I-optimal design has a D-efficiency of 99.05% relative to the D-optimal designs. As a result, D-optimal designs generally fare worse when evaluated under the I-optimality criterion than I-optimal designs fare under the D-optimality criterion. There are, however, major differences in relative I-efficiency between the various types of D-optimal designs, also for the qth degree model. The relative performance of different types of 36-run D-optimal designs and the 36-run I-optimal design are visualized in Figure 7. In the figure, the red, black and green lines correspond to the type-1, type-2 and type-3 D-optimal designs and the blue line corresponds to the I-optimal design. While the blue and green lines are nearly indistinguishable, the median variance of prediction for the I-optimal design (blue) is 0.2660 as opposed to 0.2712 for the best D-optimal design (green).

7 Discussion

In this article, we provide the first thorough literature review concerning the I-optimal design of mixture experiments. It turns out that the continuous designs proposed in the literature are suboptimal, either because they are based on too few distinct design points (Laake, 1975) or on the wrong design points (Lambrakis, 1968a) or because the weights of the design points are wrong (Liu and Neudecker, 1995). We used Wynn's algorithm to find verifiably I-optimal continuous designs for second-order Scheffé models, special cubic Scheffé models and for qth degree models, and the general equivalence theorem to confirm the I-optimality. Because we were unable to find a reference to D-optimal designs for the qth degree model, we also obtained D-optimal continuous designs for that model.

An important result of our work is that the points of the $\{q, 3\}$ simplex-centroid design are relevant if the goal of the experimenter is to minimize the average variance of prediction across the design region for a second-order Scheffé model. This insight is surprising, because, following the work of Scheffé (1958) and a known result concerning D-optimality (Kiefer, 1961), experimenters associate the $\{q, 2\}$ simplex-lattice design rather than the $\{q, 3\}$ simplex-centroid design with the second-order Scheffé model. Another important result of our work is that all points of the full simplex-centroid design are relevant if the goal is to minimize the average variance of prediction for a special cubic model. This insight is also surprising, because, following the work of Scheffé (1958) and another known result concerning D-optimality (Uranisi, 1964), experimenters associate the $\{q, 3\}$ simplex-centroid design rather than the full simplex-centroid design with the special cubic model.

Our computational results also show that the use of point-exchange algorithms is bound to lead to suboptimal designs if the interest is in I-optimality, even in case the design region is simplex-shaped. This was demonstrated by the I-optimal designs in Tables 11, 12, 13 and 14. For the construction of I-optimal designs for mixture experiments, we strongly recommend using an algorithm that is not based on a set of candidate design points, such as the mixture coordinate-exchange algorithm of Piepel et al. (2005).

In the course of the research that led to this paper, we also employed the SAS procedure OPTEX for generating I-optimal mixture designs, inspired by the online documentation which states that the I-optimality criterion is equivalent to the A-optimality criterion when the candidate set is orthogonally coded. In our view, the use of the SAS procedure OPTEX has two weaknesses when it comes to generating I-optimal designs. First, it is based on a point-exchange algorithm. It will therefore produce suboptimal designs in many situations. Second, even assuming the candidate set contains all the points required to construct an I-optimal design, the SAS procedure OPTEX will fail to do so in most practical instances, because it does not take into account the correct moments matrix **B**. Instead, the OPTEX procedure implicitly works with an approximate moments matrix based on the candidate set specified. A simple example of a situation in which OPTEX fails is one where a candidate set involving the points of the $\{q, 3\}$ simplex-centroid design is specified and a 1,000-run I-optimal design for a second-order model is desired. In that case, SAS produces designs that do not include the ternary mixture, even though the continuous I-optimal design has a weight of 9.5% for the ternary mixture (see Table 5) and the 1,000-run I-optimal design should, therefore, involve 95 replicates of the ternary mixture. Using a larger candidate set, for instance by including the ternary mixture several times in the candidate set, leads to a different design, including the ternary point. For instance, including twelve copies of the ternary mixture in the candidate set leads SAS to produce a 1,000 design with 222 replicates of the ternary mixture, more than twice as many as is I-optimal. This shows that the OPTEX procedure is unreliable when it comes to generating I-optimal designs.

In this paper, we adopted a computational approach to identifying continuous I-optimal designs for the second-order Scheffé model, the special cubic model and the qth degree model, and to verify that the resulting designs satisfy the general equivalence theorem. An interesting topic for future research would be to derive general analytical expressions for the weights in the continuous I-optimal designs, and to prove their optimality analytically.

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Appendices

Appendix A. Laake's weights for the second-order model

The I-optimal weights obtained by Laake (1975) for the second-order Scheffé model, assuming that only the points of the $\{q, 2\}$ simplex-lattice design are used, can be calculated from

$$w_1 = \frac{\sqrt{a_1}}{q\sqrt{a_1} + \binom{q}{2}\sqrt{b_1}} \tag{14}$$

and

$$w_2 = \frac{\sqrt{b_1}}{q\sqrt{a_1} + \binom{q}{2}\sqrt{b_1}}, \tag{15}$$

where

$$a_1 = \frac{2(q^2 - 7q + 17)}{(3 + q)!}$$

and

$$b_1 = \frac{64}{(3 + q)!}.$$

The resulting weighted simplex-lattice designs do not satisfy the general equivalence theorem in Section 5.1, indicating that they are not I-optimal continuous designs.

Appendix B. Laake's weights for the special cubic model

The I-optimal weights obtained by Laake (1975) for the special cubic Scheffé model, assuming that only the points of the $\{q, 3\}$ simplex-centroid design are used, can be calculated from

$$w_1 = \frac{\sqrt{a_2}}{q\sqrt{a_2} + \binom{q}{2}\sqrt{b_2} + \binom{q}{3}\sqrt{c_2}}, \tag{16}$$

$$w_2 = \frac{\sqrt{b_2}}{q\sqrt{a_2} + \binom{q}{2}\sqrt{b_2} + \binom{q}{3}\sqrt{c_2}} \tag{17}$$

and

$$w_3 = \frac{\sqrt{c_2}}{q\sqrt{a_2} + \binom{q}{2}\sqrt{b_2} + \binom{q}{3}\sqrt{c_2}}, \tag{18}$$

where

$$a_2 = \frac{q^4 - 10q^3 + 59q^2 - 218q + 1608}{2(5+q)!},$$

$$b_2 = \frac{16(16q^2 - 144q + 392)}{(5+q)!}$$

and

$$c_2 = \frac{8(27)^2}{(5+q)!}.$$

For $q = 3$, the resulting weighted simplex-centroid design satisfies the general equivalence theorem in Section 5.1. For $q \geq 4$, the resulting designs do not, indicating that they are not continuous I-optimal designs.

Appendix C. Laake's weights for the full cubic model

The I-optimal weights obtained by Laake (1975) for the full cubic Scheffé model, assuming that only the points of the $\{q, 3\}$ simplex-lattice design are used, can be calculated from

$$w_1 = \frac{\sqrt{a_3}}{q\sqrt{a_3} + \binom{q}{2}\sqrt{b_3} + \binom{q}{3}\sqrt{c_3}}, \tag{19}$$

$$w_2 = \frac{\sqrt{b_3}}{q\sqrt{a_3} + \binom{q}{2}\sqrt{b_3} + \binom{q}{3}\sqrt{c_3}} \tag{20}$$

and

$$w_3 = \frac{\sqrt{c_3}}{q\sqrt{a_3} + \binom{q}{2}\sqrt{b_3} + \binom{q}{3}\sqrt{c_3}}, \tag{21}$$

where

$$a_3 = \frac{8q^4 - 104q^3 + 784q^2 - 3088q + 5280}{4(5+q)!},$$

$$b_3 = \frac{81(q^2 - 9q + 38)}{(5+q)!}$$

and

$$c_3 = \frac{8(27)^2}{(5+q)!}.$$

The resulting weighted simplex-lattice designs do not satisfy the general equivalence theorem in Section 5.1, indicating that they are not continuous I-optimal designs.

Appendix D. Liu and Neudecker's weights for the qth degree model

The weights obtained by Liu and Neudecker (1995) for the qth degree Scheffé model, assuming that the points of the full simplex-centroid design are used, can be calculated from

$$w_k = \sqrt{a_k}\left[\sum_{t=1}^{q}\sqrt{a_t\binom{q}{t}}\binom{q}{k}\right]^{-1} \tag{22}$$

for each design point that is a permutation of the vector $(k^{-1}\mathbf{1}'_k, \mathbf{0}'_{q-k})$, where

$$a_k = \sum_{\substack{i=1\\ i\le j}}^{q}\sum_{j=k}^{q}\left[\frac{(-1)^{i+j}ijk^{i+j-2}\binom{q}{i}}{(q+i+j-1)!}\right]\left[\sum_{m=0}^{\min(i-k,q-j)} 2^{i-m}\binom{i}{m}\binom{q-i}{j-i+m}\binom{i-m}{k}\right].$$

and

$$a_q = \frac{2^q q^{2q}}{(3q-1)!}.$$

The resulting weighted simplex-centroid designs do not satisfy the general equivalence theorem in Section 5.1, indicating that they are not continuous I-optimal designs. This is in contrast with what is claimed in Liu and Neudecker (1995).

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