

ON THE DESIGN OF MIXTURE EXPERIMENTS

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ABSTRACT

A mixture experiment is one in which the level of each quantitative factor in a particular treatment is a proportion of the total treatment. Because of these properties the factor space is a regular simplex with one less dimension than the number of factors.

The variance function of the expected mean response is examined in the 3-component case and under linear, quadratic and cubic mixture models. A function, utilising the generalised variance of the D-optimum design, is given whereby upper and lower bounds can be calculated for the maximum possible variance of the expected mean response for any given design space, design and model.

D-optimum designs, that is, designs with minimum generalised variances, were generated for the same design spaces and mathematical models. A generalisation is given for designing D-optimum experiments for a simplex design space of more than three components under a linear model.

A method was devised for designing mixture experiments that are as near D-optimum as possible for a given experiment size, design space and model. The method was tested on the same design spaces and models.

This last method was extended to enable incomplete block mixture experiments to be designed such that the generalised variance of the estimable parameters is minimised while at the same time minimising the effect of blocking on these estimations. Incomplete block mixture

designs were generated for a number of block sizes and numbers of blocks when the design space is the 3-component full simplex and the above three mathematical models are used.

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1. INTRODUCTION

A mixture experiment is one in which the levels of each quantitative factor in a particular treatment is a proportion of the total treatment. Because of these properties the factor space is a regular simplex with one less dimension than the number of factors.

Mixture experiments arise in a number of branches of science and technology, for example, testing the tensile strength of an alloy or testing the baking quality of a cake batter. In such situations the investigator cannot make use of the usual factorial experimental methods because the sum of the levels of the factors, the components of the alloy or batter, applied to an experimental unit must be unity.

Early examples of mixture experiments in the statistical literature are found in the publications of Plackett and Hewlett (1952), Quenouille (1953), Claringbold (1955) and Hewlett and Plackett (1956). Scheffé (1958) wrote an important paper which forms the basis of much of the recent research. Subsequently a number of papers have appeared on both the design (Scheffé, 1963; McLean and Anderson, 1966; Becker, 1970; Nigam, 1970, and others) and the analysis (Scheffé, 1963; Becker, 1968; Cox, 1971, and others) of mixture experiments.

A review of the literature on the design of mixture experiments is given in Chapter 2. In Chapter 3 the design of mixture experiments, under several designs and mathematical models is examined in terms of the nature of the variance function of the expected mean response.

The design of D-optimum mixture experiments using the results of Kiefer and Wolfowitz (1959) are investigated in Chapter 4. The D-optimum designs are generated, by using the method of Wynn (1970),

for the same design space and model combinations as those used in Chapter 3. Suggestions of methods of generating designs which are as near D-optimum as possible for a fixed number of observations are made in Section 5.1 and in Section 5.3 these are tested on the same design space and model combinations used in Chapters 3 and 4.

The design of relatively efficient incomplete block mixture experiments is considered in Chapter 6. The method given in Section 6.1 is used to generate incomplete block designs for several block sizes and numbers of blocks for the 3-component full simplex design space under linear, quadratic and cubic models.

2. PRELIMINARIES

2.1 Notation and definitions

The application of any mixture, $\mathbf{x}_j = (x_{1j}, \dots, x_{qj})$ in $\mathcal{X}$ the continuous design space defined by the constraints $x_{ij} \geq 0$, $x_{1j} + \dots + x_{qj} = 1$ will result in a response $y(\mathbf{x}_j)$, a random variable, such that $\eta(\mathbf{x}) = E\{y(\mathbf{x}_j)\} = \boldsymbol{\theta}' f(\mathbf{x}_j)$, (2.1.1) where $f(\mathbf{x}_j)$ is the $k \times 1$ column vector of functions $f_i(\mathbf{x})$ evaluated at $\mathbf{x}_j$ and $\boldsymbol{\theta}$ is a $k \times 1$ column vector of unknown real parameters. Assume also that $\text{var}\{y(\mathbf{x}_j)\} = \sigma^2$, $\text{cov}\{y(\mathbf{x}_1), y(\mathbf{x}_2)\} = 0$ for $\mathbf{x}_1 \neq \mathbf{x}_2$; $\mathbf{x}_j, \mathbf{x}_1, \mathbf{x}_2$ in $\mathcal{X}$.

Define a discrete design D_n with n design points $\mathbf{x}_i$ in $\mathcal{X}$ ($i = 1, \dots, n$) not necessarily distinct. For this discrete design D_n in $\mathcal{X}$ there exists a corresponding $n \times k$ matrix X_n , whose j th row is the vector $f(\mathbf{x}_j)$. The matrix, X_n , corresponding to a discrete design, D_n , will always be given the same subscript. A design measure, denoted by ξ and referred to in future merely as a measure, is a member of the set, $\mathcal{D}$, of all formal probability measures defined on $\mathcal{X}$.

For a measure ξ on $\mathcal{X}$ let

$$m_{ij}(\xi) = \int_{\mathcal{X}} f_i(\mathbf{x}) f_j(\mathbf{x}) \xi(d\mathbf{x}). \quad (2.1.2)$$

Let $M(\xi) = \{m_{ij}(\xi)\}$ be the $k \times k$ information matrix. The covariance matrix of the best linear estimates of $\boldsymbol{\theta}$ is given by $\frac{1}{n} \sigma^2 M^{-1}(\xi)$. This implies that the more diagonally dominant $M(\xi)$ is, the more independent will the estimates of θ_i ($i = 1, \dots, k$) be; a diagonal $M(\xi)$ means that

the θ_i can be interpreted separately. A simple measure of the magnitude of the information matrix $M(\xi)$ is its determinant, $|M(\xi)|$.

The notation $|A|$ will throughout denote the determinant of A .

For any discrete design D_n a measure, ξ_n , may be formed by attaching a mass of $1/n$ to each point of D_n ; this is called the associated measure of D_n . Therefore, from this definition,

$$m_{ij}(\xi_n) = \frac{1}{n} \sum_{l=1}^n f_i(x_l) f_j(x_l)$$

and

$$M(\xi_n) = \frac{1}{n} (X_n' X_n). \quad (2.1.3)$$

2.2. Mathematical models and parameterisation

Several mathematical models appear in the literature designed to provide meaningful interpretations to responses in mixture experiments. One of the earliest, and most probably one of the simplest, is the model suggested by Scheffé (1958) and extended by Gorman and Hinman (1962) and Lambrakis (1969a). A response $\eta(x)$ can generally be represented as a $\{q, m\}$ polynomial, i.e. a polynomial function of degree m in the q components, of the form

$$\eta(x) = \beta_0 + \sum_{i=1}^q \beta_i x_i + \sum_{1 \leq i < j \leq q} \beta_{ij} x_i x_j + \dots + \sum_{1 \leq i_1 < \dots < i_m \leq q} \beta_{i_1 \dots i_m} x_{i_1} \dots x_{i_m} \quad (2.2.1)$$

with $\binom{m+q}{m}$ parameters. All these coefficients are, however, not unique since by substituting $x_q = 1 - \sum_{i=1}^{q-1} x_i$ in (2.2.1), it reduces to a

polynomial in $(q-1)$ variables $x_1, \dots, x_{q-1}$ with only $\binom{m+q-1}{m}$ coefficients.

Scheffé (1958) suggested an alternative parameterisation, i.e.

the canonical polynomial,

$$\eta(\mathbf{x}) = \sum_{r=1}^{m \leq q} \sum_{1 \leq i_1 < \dots < i_r \leq q} \beta_{i_1 \dots i_r} x_{i_1} \dots x_{i_r}, \quad (2.2.2)$$

with $\binom{m+q-1}{m}$ parameters, all estimable. In particular, for $m = 3$, he proposed using

$$\begin{aligned} \eta(\mathbf{x}) = & \sum_{i=1}^q \beta_i x_i + \sum_{1 \leq i < j \leq q} \beta_{ij} x_i x_j + \sum_{1 \leq i < j \leq q} \gamma_{ij} x_i x_j (x_i - x_j) \\ & + \sum_{1 \leq i < j < k \leq q} \beta_{ijk} x_i x_j x_k, \end{aligned} \quad (2.2.3)$$

with $\gamma_{ij} = \beta_{iij} - \beta_{ijj}$.

Gorman and Hinman (1962) extended (2.2.3) to $m = 4$ and

Lambrakis (1969a) gives the generalised form of the m^{th} order canonical polynomial as

$$\begin{aligned} \eta(\mathbf{x}) = & \sum_{i=1}^q \beta_i x_i + \sum_{r=2}^m \sum_{1 \leq i < j \leq q} \alpha_{ij}^{(r)} x_i x_j (x_i - x_j)^{r-2} \\ & + \sum_{r=3}^m \sum_{1 \leq i_1 < \dots < i_r \leq q} \alpha_{i_1, \dots, i_r}^{s_1, \dots, s_r} x_{i_1}^{s_1} \dots x_{i_r}^{s_r}, \end{aligned} \quad (2.2.4)$$

where $\sum_{i=1}^r s_i = m$. The main drawback of (2.2.4) is that the constant

term normally included in a mathematical model is confounded with the estimates of the β_i ($i = 1, \dots, q$). For example, the usual 3-component linear model is

$$\eta(\underline{x}) = \mu + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 ,$$

where $\beta_1 + \beta_2 + \beta_3 = 0$, but because $x_1 + x_2 + x_3 = 1$ this polynomial can also be written as

$$\eta(\underline{x}) = (\beta_1 + \mu)x_1 + (\beta_2 + \mu)x_2 + (\beta_3 + \mu)x_3 ,$$

the canonical polynomial estimating $\beta_i^x = \beta_i + \mu$. This could lead to different estimates of the β_i in two replicate experiments on the same system and will rule out sequential experimentation. Other drawbacks according to Cox (1971) are that (i) the absence of squared terms from the second degree polynomial makes it meaningless to consider the direction and magnitude of the curvature of the response to a particular component, and (ii) the interpretation of the β_i is in terms of expected responses to 'pure' mixtures and the β_{ij} in terms of a comparison of a binary mixture with $x_i = x_j = 1/2$ ($i \neq j$) with the corresponding pure mixtures. A pure mixture consists of only one component, x_i say, i.e. $x_i = 1, x_j = 0$ ($i \neq j = 1, \dots, q$). These interpretations are impossible if the experimenter is forced to use only mixtures with more than a few components present.

To overcome some of the drawbacks of the parameterisation of the canonical polynomial, Cox (1971) suggested fitting the model in the form.

$$\eta(\underline{x}) = \beta_0 + \sum_{i=1}^q \beta_i z_i + \sum_{1 \leq i < j \leq q} \beta_{ij} z_i z_j + \dots , \quad (2.2.5)$$

where $z_i = x_i / (1 - s_i)$, and $\underline{s} = (s_1, \dots, s_q)$ represents some standard reference mixture. In some cases one would wish to take $s_1 = \dots = s_q = 1/q$ and in other s will be some convenient reference mixture, usually near the centre of the region of main interest.

Suppose that for any change, Δ_i say, in x_i the other $(q-1)$ components are changed proportionally to the standard mixture. Thus if x_i is decreased by Δ_i , every x_j ($i \neq j; j=1, \dots, q$) is increased by

$$\frac{\Delta_i s_j}{(1 - s_i)}. \text{ For the first degree model namely } \eta_1(\underline{x}) = \beta_0 + \sum_{i=1}^q \beta_i x_i,$$

the change is thus

$$\Delta \eta(\underline{x}) = - \frac{\beta_i \Delta_i}{(1 - s_i)} + \frac{\Delta_i}{(1 - s_i)} \sum_{j=1}^q \beta_j s_j \quad (j \neq i).$$

It is therefore convenient to measure changes in x_i in terms of $(1 - s_i)$.

By imposing the constraints

$$\sum_{j=1}^q \beta_j s_j = 0;$$

$$\sum_{k=1}^q \beta_{jk} s_k = 0 \quad (j = 1, \dots, q);$$

and similarly for higher degree interactions, $\eta(s_1, \dots, s_q) = \beta_0$. The constant term is thus the height of the response surface at s and is independent of the design. This parameterisation is thus advantageous in sequential 'search' experimentation where the design changes from run to run.

Becker (1968) criticised the polynomial regression function

since it cannot account satisfactorily for components which are inert or have an additive effect. He arrived at the conclusion that models homogenous of degree one are more appropriate for mixtures.

If $\eta(\underline{x}) = \eta(x_1, \dots, x_q)$ represents the response to a q -component mixture and a further component, x_{q+1} , has an additive effect with respect to any mixture of the other q components, the response of the $(q+1)$ -component mixture could be represented by

$$\begin{aligned}\eta(\underline{x}) &= \beta_{q+1} x_{q+1} + (1 - x_{q+1}) f\{x_1/(1 - x_{q+1}), \dots, x_q/(1 - x_{q+1})\} \\ &= \beta_{q+1} x_{q+1} + \sum_{i=1}^q x_i f\left\{x_1 / \sum_{i=1}^q x_i, \dots, x_q / \sum_{i=1}^q x_i\right\}.\end{aligned}\quad (2.2.6)$$

Becker (1968) proposes three alternative models which are all homogenous of degree one with additive properties viz.

$$\eta(\underline{x}) = \sum_{i=1}^q \beta_i x_i + \sum_{1 \leq i < j \leq q} \beta_{ij} \min(x_i, x_j) + \dots + \beta_{1,2,\dots,q} \min(x_1, x_2, \dots, x_q); \quad (2.2.7)$$

$$\begin{aligned}\eta(\underline{x}) &= \sum_{i=1}^q \beta_i x_i + \sum_{1 \leq i < j \leq q} \beta_{ij} x_i x_j / (x_i + x_j) + \sum_{1 \leq i < j < k \leq q} \beta_{ijk} x_i x_j x_k / (x_i + x_j + x_k)^2 + \\ &\dots + \beta_{1,2,\dots,q} x_1 x_2 \dots x_q / \left(\sum_{i=1}^q x_i\right)^{q-1};\end{aligned}\quad (2.2.8)$$

$$\begin{aligned}\eta(\underline{x}) &= \sum_{i=1}^q \beta_i x_i + \sum_{1 \leq i < j \leq q} \beta_{ij} (x_i x_j)^{1/2} + \sum_{1 \leq i < j < k \leq q} \beta_{ijk} (x_i x_j x_k)^{1/3} + \\ &\dots + \beta_{1,2,\dots,q} (x_1 x_2 \dots x_q)^{1/q}.\end{aligned}\quad (2.2.9)$$

The term $x_{i_1} x_{i_2} \dots x_{i_r} / (x_{i_1} + \dots + x_{i_r})$ ($r = 2, \dots, q$) in (2.2.8) is defined as zero when $(x_{i_1} + \dots + x_{i_r}) = 0$. This model (2.2.8), can be considered as a form of Scheffé's polynomial (2.2.2) adjusted so as to allow additive effects. Becker (1968) acknowledges that (2.2.7), although being completely additive, has the restriction that it is unsatisfactory for response surfaces with a maximum (or minimum) not at the centroid of the design. A major drawback of these models, however, lies in the difficulty in interpreting the response function when it is used in the analysis. This is especially the case when dealing with models (2.2.7) and (2.2.9). Similar to the Scheffé model (2.2.2) these do not make provision for a constant term.

A number of authors have suggested different transformations of the form $z_i = g(x_1, x_2, \dots, x_q)$ ($i = 1, \dots, (q-1)$) to overcome the $\sum_{i=1}^q x_i = 1$ constraint and thus regain the ability to use normal regression methods utilizing the z_i . Draper and Lawrence (1965a, b) suggested the sequential transformation $z_{iT} = (I - Z(Z'Z)^{-1}Z')x_i$ ($i = 1, \dots, q$),

(2.2.10)

where Z is a matrix of $n \times l$ column vectors already transformed, x_i is the next column vector to be transformed and z_{iT} is the transformed vector orthogonal to vectors already in Z . This transformation destroys the main reason for doing a mixture experiment because the independent variables are no longer measured as proportions, or simple transformations of proportions, but as complex transformations of them. The problem thus lies in the interpretation of the resultant regression coefficients. It could happen that a two-factor interaction has a large and meaningful effect and would most probably be noticed in the original

model. Under the transformation model, however, a large proportion of its effect could have been absorbed by the main effects and other interactions that might appear before it in the model.

Claringbold (1955) suggested shifting the origin to the centroid of the design, scaling to eliminate fractions and then rotating the axes such that the design is orthogonal to the q^{th} axis. This method would be useful if it is known that the q^{th} component merely acts as a dilutant for the others.

Box and Tildwell (1962) suggested non-linear transformations of the form $z_{ij} = x_{ij}^{\alpha_i}$ ($i = 1, \dots, q; j = 1, \dots, n$) for general regression problems if there is reason to believe that the transformed model is valid. They, however, give a method for computing confidence intervals for the α_i which should give an indication whether a transformation would be of any use.

Other transformations suggested by Kenworthy (1963); Thompson and Myers (1968); Becker (1969, 1970); and Box and Hunter (1954) all suffer from the disadvantage that they may be useful if the underlying mechanism is known but could be extremely misleading otherwise.

In the case where one of the components, x_1 say, merely acts as a dilutant of the others, or is suspected to act as such, Cox (1971) suggested fitting the model

$$\eta(\underline{x}) = \alpha(x_1 - s_1) + p\{x_2/(1-x_1), \dots, x_q/(1-x_1)\}, \quad (2.2.11)$$

where $p\{.\}$ may be polynomial in the new arguments and would amount to a mixture with $(q-1)$ components

$$x_{i-1}^x = x_i/(1-x_1) \quad (i = 2, \dots, q).$$

This could thus be treated as a series of mixture experiments performed at different levels of another variable x_1 . A test for $\alpha = 0$ would then show if x_1 is after all a dilutant or whether it has an additive effect. A form of blocking on the x_i^x , using a different value for x_1 for every block would reduce the size of the experiment appreciably.

2.3. General Designs for mixture experiments

Many different designs for randomized complete block mixture experiments have been suggested. Two of the earliest, and simplest designs were the $\{q, m\}$ simplex-lattice and the simplex-centroid designs of Scheffé (1958, 1963).

The $\{q, m\}$ simplex-lattice consists of $\binom{m+q-1}{m}$ points in the simplex $0 \leq x_i \leq 1, (i = 1, \dots, q), \sum_{i=1}^q x_i = 1$, representing all possible mixtures in which the proportion x_i of the i^{th} component has the $(m+1)$ equally spaced values $x_i = 0, 1/m, 2/m, \dots, 1$.

The simplex-centroid design consists of a subset of the $\{q, q\}$ simplex-lattice. It contains the $(2^q - 1)$ mixtures with equal proportions, namely

q pure-component mixtures, $(x_i = 1, x_j = 0; i \neq j; i, j = 1, 2, \dots, q)$;

$\binom{q}{2}$ binary mixtures of equal proportions,

$(x_i = 1/2, x_j = 1/2, x_k = 0; i \neq j \neq k; i, j, k = 1, 2, \dots, q)$;

$\binom{q}{3}$ ternary mixtures of equal proportions,

$(x_i = x_j = x_k = 1/3, x_l = 0; i \neq j \neq k \neq l; i, j, k, l = 1, 2, \dots, q); \dots;$ and

1 q -nary mixture $(x_i = 1/q; i = 1, 2, \dots, q)$.

Subsequently Murty and Das (1968) evolved the symmetric-simplex design. This design consists of a subset of the simplex-lattice design subject to the following constraints

$$\sum_{u=1}^n x_{iu}^2 = A, \quad \sum_{u=1}^n x_{iu} x_{ju} = B, \quad \sum_{u=1}^n x_{iu}^2 x_{ju} = C, \quad \sum_{u=1}^n x_{iu} x_{ju} x_{ku} = D,$$

$$\sum_{u=1}^n x_{iu}^2 x_{ju}^2 = E, \quad \sum_{u=1}^n x_{iu}^3 x_{ju} x_{ku} = F, \quad \sum_{u=1}^n x_{iu} x_{ju} x_{ku} x_{lu} = G,$$

($i, j, k, l, i, j, k, l = 1, \dots, q$) where A, B, C, D, E, F and G are constants.

The reason for these constraints is to facilitate the numerical analysis arising out of the symmetry of the design. With the present powerful computer techniques and generally available package programs for solving the normal equations imposing these constraints seems unnecessary. It could be that these constraints lead to estimates with desirable statistical properties, although this aspect of the design has not been discussed by Murty and Das (1968).

Scheffé (1963) extended the simplex-centroid design to include 'process' variables which is equivalent to imposing a full normal factorial set of treatment combinations at each of the $2^q - 1$ points of the simplex-centroid design. Thus an unreplicated design with $q = 3$ and two process variables at two levels each will contain $(2^3 - 1)(2^2) = 28$ observations. Such experiments could be carried out as split-plot experiments with either the 'process' treatment combinations or the mixture treatments as main plots depending on the circumstances. If no interaction between the process and mixture variables can be assumed, the size of the

experiment could be reduced by some form of blocking and using a split-block design.

As a generalisation of the $\{q, m\}$ simplex-lattice, Lambrakis (1968a) introduced the $\{q_1, \dots, q_p; m_1, \dots, m_p\}$ multiple-lattice design consisting of p simplex-lattice designs $\{q_i, m_i\}$ ($i = 1, \dots, p$).

In this generalisation the i^{th} component becomes a major component with a fixed proportion $c_i > 0$ and is a mixture of q_i sub components in which the proportion x_{ij} of the j^{th} sub component takes values in the

simplex $x_{ij} \geq 0$ ($j = 1, \dots, q_i$); $\sum_{j=1}^{q_i} x_{ij} = 1$. Thus the proportions

$X_{ij} = c_i x_{ij}$ of the major components take values in the multiple simplex $X_{ij} \geq 0$ ($i = 1, \dots, p; j = 1, \dots, q_i$);

$$\sum_{i=1}^p \sum_{j=1}^{q_i} X_{ij} = 1.$$

The $\{q_1, q_2, \dots, q_p; m_1, \dots, m_p\}$ multiple-lattice then consists of

$\prod_{i=1}^p \binom{m_i + q_i - 1}{m_i}$ points in the multiple-simplex.

Lambrakis (1969a) derived the estimating regression function of the double-lattice design and derived the estimated regression function of any multiple-lattice design (Lambrakis (1969b)). Since the c_i are fixed this design will be of limited applicability. It will in any case lead to very large experiments and interpretation of the fitted regression function will be difficult because of the very large number of interdependent regression coefficients in the resulting equation.

Nigam (1970) gives a method of designing mixture experiments through mutually orthogonal Latin squares. The procedure is as follows. Suppose there is a set of $(q-1)$ mutually orthogonal Latin squares of order q with symbols p_i ($i = 1, \dots, q$). Let the symbols p_i take non-negative integral values not necessarily all different. Then if each element of each of the $(q-1)$ mutually orthogonal Latin squares is divided by $\sum_{i=1}^q p_i$, the set forms a mixture design in q components with $q(q-1)$ observations. Because he restricts his designs to the symmetrical-simplex of Murty and Das (1968) he restricts his choice of p_i to two cases namely (i) all the p 's are zero except two, say p_i and p_j ; and (ii) all the p 's are zero except three, say p_i, p_j and p_k .

For example suppose we have $q = 5$ components in the mixture. Then there exists a set of 4 mutually orthogonal Latin squares of size 5. Let $p_i = 1, p_j = 2, p_k = 3, p_l = p_m = 0$ be the 5 symbols of these Latin squares. By dividing each element by 6, the total for every row, a 5 component mixture design in four blocks and 20 mixtures results. The four squares and the corresponding mixtures are given in Table 2.1.

By deleting the two restrictions above but imposing the restriction $\sum_{i=1}^q p_i = m$, a constant, and by varying the p_i in successive runs a $\{q, m\}$ simplex-lattice design can be generated. The simplex-centroid design results when $p_{i_1} = p_{i_2} = \dots = p_{i_r}, p_{i_{r+1}} = \dots = p_{i_q} = 0$ ($r = 1, \dots, q$). An additional advantage is that every $q \times q$ Latin square produces a set of q mixtures, $x_j = (x_{1j}, \dots, x_{qj})$ ($j = 1, \dots, q$), such

TABLE 2.1. The set of 4 mutually orthogonal Latin squares of size 5
and the mixture design with 20 observations obtained from it

Latin square No.	Observation no.	Latin Square	Mixture
1	1	0 1 3 0 2	(0, 1/6, 1/2, 0, 1/3)
	2	1 2 0 0 3	(1/6, 1/3, 0, 0, 1/2)
	3	2 3 0 1 0	(1/3, 1/2, 0, 1/6, 0)
	4	3 0 1 2 0	(1/2, 0, 1/6, 1/3, 0)
	5	0 0 2 3 1	(0, 0, 1/3, 1/2, 1/6)
2	6	0 3 0 2 1	(0, 1/2, 0, 1/3, 1/6)
	7	1 0 0 3 2	(1/6, 0, 0, 1/2, 1/3)
	8	2 0 1 0 3	(1/3, 0, 1/6, 0, 1/2)
	9	3 1 2 0 0	(1/2, 1/6, 1/3, 0, 0)
	10	0 2 3 1 0	(0, 1/3, 1/2, 1/6, 0)
3	11	0 0 2 1 3	(0, 0, 1/3, 1/6, 1/2)
	12	1 0 3 2 0	(1/6, 0, 1/2, 1/3, 0)
	13	2 1 0 3 0	(1/3, 1/6, 0, 1/2, 0)
	14	3 2 0 0 1	(1/2, 1/3, 0, 0, 1/6)
	15	0 3 1 0 2	(0, 1/2, 1/6, 0, 1/3)
4	16	0 2 1 3 0	(0, 1/3, 1/6, 1/2, 0)
	17	1 3 2 0 0	(1/6, 1/2, 1/3, 0, 0)
	18	2 0 3 0 1	(1/3, 0, 1/2, 0, 1/6)
	19	3 0 0 1 2	(1/2, 0, 0, 1/6, 1/3)
	20	0 1 0 2 2	(0, 1/6, 0, 1/3, 1/2)

that $\sum_{j=1}^q x_{ij} = 1$. In the cases where $p_{i_1} = p_{i_2} = \dots = p_{i_r}$; $p_{i_{r+1}} = \dots = p_{i_q} = 0$

the design will however be replicated $r!(q-r)!/(q-2)!$ times.

All these designs are, however, usually unrealistic in the sense that in practice there often exist some constraints upon the components so that the feasible region for experimentation is relatively restricted.

These restrictions may be of the form (i) $0 \leq a_i \leq x_i \leq b_i \leq 1$, where any or both the inequalities $0 < a_i$ and $b_i < 1$ may hold; or (ii) a mixture must consist of at least $p > 1$ components at non-zero levels; or

$$(iii) \sum_{j=1}^r x_{i_j} \geq k > 0 \text{ or } \sum_{j=1}^r x_{i_j} \leq l < 1.$$

An alternative to the $\{q, m\}$ simplex-lattice was suggested by Gammon (1963) which is similar to the normal $\{q, m\}$ simplex-lattice except that instead of the pure mixtures, $x_i = 1$, say, the $(q-1)$ -nary mixtures g_i , such that $x_i = 0$, $x_j = 1/(q-1)$, ($i \neq j$; $i, j = 1, 2, \dots, q$) are used. Lambrakis (1969a) gives the canonical polynomial for analysing this design.

As a possible solution to the situation where one of the components, x_1 say, cannot appear in a pure form, and specifically if $x_1 \leq h \leq 1$, whereas there are no constraints on the other x_j , ($j = 2, \dots, q$), Scheffé (1958) suggested the use of a pseudocomponent. This pseudocomponent is formed by replacing the first component throughout the simplex-lattice design by a mixture consisting of the proportion h of the first component and proportions p_i of the other components ($i > 1$, $\sum_{i=2}^q p_i = 1-h$), the p_i being selected by the experimenter. In other words the experiment

is designed in the usual way following say a simplex-lattice design.

Suppose the constraint $0 \leq x_i \leq h < 1$ is imposed. Let the i^{th} component in a particular mixture be present in the proportion s_i and the other $(q-1)$ components in the proportions s_j ($i \neq j$; $i, j = 1, \dots, q$). This i^{th} component is then replaced by a pseudocomponent consisting of a proportion $h s_i$ of the i^{th} component plus proportions of $p_j s_i$ ($i \neq j$; $i, j = 1, \dots, q$) of the other j components, where
$$\sum_{i \neq j=1}^q p_j = 1-h.$$

The j^{th} component is thus present in the proportion $(s_j + p_j s_i)$ ($i \neq j$; $i, j = 1, \dots, q$) in the mixture. This method could then be extended to more than one pseudocomponent if more than one of the components is restricted to values smaller than 1.

About the only really viable design method for experiments constrained to a subregion of the whole simplex is the extreme vertices design suggested by McLean and Anderson (1966). Their method consists of (i) placing a design point on each of the extreme vertices bounding the design space; and (ii) placing a design point on the centroids of each boundary $2, 3, \dots, r$ ($r \leq q-2$)-dimensional face. The centroids of 2-dimensional faces are found by isolating sets of vertices for which each of $(q-3)$ factor levels remains constant within a given set and by averaging the factor levels for each of the three remaining factors. All remaining centroids are found in a similar fashion using all vertices which have $(q-r-1)$ factor levels constant within a set for an r -dimensional face, where $3 \leq r \leq q-2$. The dimensionality of the hyper-polyhedron formed by the extreme vertices will of course always be $(q-1)$. McLean and Anderson (1966) suggest that the size of the design could be reduced

by (i) deleting certain centroids; or (ii) computing the normalized distance between design points and randomly omitting points that are less than a certain minimum distance from other design points. One method of normalization is to define the distance between two points x_i

$$\text{and } x_j \text{ as } d_{ij} = \left\{ \sum_{r=1}^q \left(\frac{x_{ir} - x_{jr}}{b_r - a_r} \right)^2 \right\}^{1/2}, \text{ where } 0 \leq a_r \leq x_r \leq b_r \leq 1.$$

This method assumes that sensitivity for each factor is inversely proportional to the length of its constrained interval. Computing and inspection of these normalized distances as a routine procedure should prevent the unnoticed occurrence of clustering vertices as warned against by Gorman (1966). These occur when a small change in one constraint would cause a number of vertices to collapse into one vertex.

This method of McLean and Anderson (1966) could, however, lead to designs with relatively few design points. For example the design for the full space ($0 \leq x_i \leq 1$, $i = 1, 2, \dots, q$) for $q = 4$ would only contain 8 design points, one at each $x_i = 1$, $x_j = 0$ ($i \neq j$; $i, j = 1, \dots, 4$) and one each at $x_i = x_j = x_k = 1/3$, $x_l = 0$ ($i \neq j \neq k \neq l$; $i, j, k, l = 1, 2, \dots, 4$). For $q = 3$ the only design points would be at the vertices. In such cases design points could be placed in the middle of bounding lines and at the centroid of the 2-dimensional design space.

In the case of relatively narrowly constrained designs, the normal equations for estimating the regression function tend to become very difficult to handle computationally. The experimenter should investigate whether his mathematical model can in fact be easily fitted to the design although he may have appreciably more design points than

parameters to be estimated. A judicious choice of the parameterisation of the model is important as rounding errors become critical.

Lambrakis (1968b) suggests a p -component mixture design in which every mixture has exactly p components, for example $x_i = 1/2$, $x_j = 1/2 (p-1)^{-1}$ ($i \neq j$; $i, j = 1, 2, \dots, p$) or $x_i = x_j = 1/3$, $x_k = 1/3(p-2)^{-1}$ ($i \neq j \neq k$; $i, j, k = 1, \dots, p$).

2.4. Optimality of designs

Kiefer and Wolfowitz (1959) defined a measure ξ^* as D-optimum if

$$|M(\xi^*)| = \max_{\xi \in \mathcal{D}} |M(\xi)|. \quad (2.4.1)$$

Defining

$$d(x, \xi) = f(x)' M^{-1}(\xi) f(x), \quad (2.4.2)$$

Kiefer and Wolfowitz (1960) showed that D-optimality of ξ^* as defined by (2.4.1) is equivalent to

$$\inf_{\xi \in \mathcal{D}} \sup_{x \in \mathcal{X}} d(x, \xi) = \sup_{x \in \mathcal{X}} d(x, \xi^*) \quad (2.4.3)$$

and also to

$$\sup_{x \in \mathcal{X}} d(x, \xi^*) = k, \quad (2.4.4)$$

where k is the dimension of $f(x)$.

Kiefer and Wolfowitz (1960) discuss the statistical meaning of D-optimality. First, let $|\frac{1}{n} (X' X)_n|$ be proportional to the generalised variance, $n^{-1} \sigma^2 |M(\xi_n)|^{-1}$, of the parameters forming $\mathcal{Q}$ in (2.1.1). Suppose the responses $y(x_j)$ ($j = 1, \dots, n$) are independently distributed random normal variables. Maximising $|M(\xi_n)|$ is then equivalent to

maximising the Gaussian curvature of the power surface of the usual F test, or χ^2 test if σ^2 is known, at the null hypothesis $\varrho = \varrho_0$, where ϱ_0 is some fixed value of ϱ .

The combination of (2.1.3) and (2.4.2) gives

$$d(\underline{x}, \xi_n) = n f(\underline{x})' (X_n' X_n)^{-1} f(\underline{x}) \quad (2.4.5)$$

and $\sigma^2 f(\underline{x})' (X_n' X_n)^{-1} f(\underline{x})$ is the variance of the usual least squares estimate of the expected response, $\hat{\eta}(\underline{x})$ at $\underline{x}$. The equivalence of (2.4.1) and (2.4.3) means that if D_n is such that ξ_n is D-optimum, the design minimises the maximum over $\mathcal{X}$ of the variance function derived from using that design.

Wynn (1970) has shown that, given an initial design D_{n_0} with n_0 points $\underline{x}_1, \dots, \underline{x}_{n_0}$ such that $X_{n_0}' X_{n_0}$ is non-singular, it is possible to obtain convergence to a D-optimum measure by the successive addition of points. The procedure for doing this is as follows: first find a point $\underline{x}_{n_0+1}$ in $\mathcal{X}$ which maximises the variance function obtained by using D_{n_0} ; that is, choose $\underline{x}_{n_0+1}$ such that

$$\max_{\underline{x} \in \mathcal{X}} d(\underline{x}, \xi_{n_0}) = d(\underline{x}_{n_0+1}, \xi_{n_0}).$$

Form a new design D_{n_0+1} with n_0+1 points to maximise the new variance function obtained from using D_{n_0+1} and continue this process to obtain a sequence of designs $D_{n_0} \subset D_{n_0+1} \subset \dots \subset D_n \subset \dots$, where D_n is obtained from D_{n-1} by adding a point in $\mathcal{X}$ at which maximum variance is attained.

Formally this can be stated as his Theorem 1, namely: as

$n \rightarrow \infty$, $\lim \det \{M(\xi_n)\} = \det \{M(\xi^*)\}$, where ξ^* is a D-optimum measure.

There exist upper and lower bounds for the ratio $R = |M(\xi^*)| / |M(\xi_n)|$, where $M(\xi_n)$ is the information matrix of a given design D_n , and $M(\xi^*)$ the information matrix of the D-optimum design for that design space and model. Wynn (1970) gives a lower bound as

$$R \geq \left\{ \frac{\bar{d}(\xi)}{k} \right\}^k \left\{ \frac{k-1}{\bar{d}(\xi) - 1} \right\}^{k-1},$$

where $\bar{d}(\xi) = \sup_{x \in \mathcal{X}} d(x, \xi)$. This can be rearranged to give

$$\bar{d}(\xi) \leq k R \left[\frac{k \{\bar{d}(\xi) - 1\}}{\bar{d}(\xi)(k-1)} \right]^{k-1} \leq k R \{k/(k-1)\}^{k-1} \quad (2.4.6)$$

Kiefer (1961b) gives the upper bound as

$$R \leq \exp \{ \bar{d}(\xi) - k \}.$$

Rearranging this gives

$$\bar{d}(\xi) \geq k + \ln R. \quad (2.4.7)$$

Combining (2.4.6) and (2.4.7) and dividing by n gives

$$k/n + \frac{1}{n} \ln R \leq \sup_{x \in \mathcal{X}} \text{var} \{ \hat{\eta}(x) \} \leq \frac{k}{n} R \{k/(k-1)\}^{k-1}.$$

The maximum $\text{var} \{ \hat{\eta}(x) \}$ for x in $\mathcal{X}$ is therefore not determined by the shape of the design space but how near the design is to the D-optimum, as will be demonstrated in Section 3.3.

Box and Draper (1959) divide the mean squared deviation averaged over the region into two parts, one due to pure error (V),

and the other due to bias introduced by fitting the incorrect model (B), namely

$$\frac{n\Omega}{\sigma^2} \int_R [\hat{E}\{\hat{y}(\underline{x})\} - \eta(\underline{x})]^2 d\underline{x} = \frac{n\Omega}{\sigma^2} \int_R V\{\hat{y}(\underline{x})\} d\underline{x} + \frac{n\Omega}{\sigma^2} \int_R [\hat{E}\{\hat{y}(\underline{x})\} - \eta(\underline{x})]^2 d\underline{x} \quad (2.4.8)$$

or $\overline{\text{MSD}} = V + B$.

Here $\Omega^{-1} = \int_R d\underline{x}$; $\hat{y}(\underline{x})$ = value of estimated response at the point $\underline{x}$, where this value is obtained by the least squares fitting of the polynomial of degree d_1 using a design containing n points; $\eta(\underline{x})$ is the true response at this point given exactly by the polynomial of higher degree d_2 ; σ^2 is the experimental error variance; and V is the variance function as given by Box and Hunter (1957). Depending on the relative sizes of the V and B components, one would try to minimise either one or the other, or their sum, in an effort to achieve the minimisation of (2.4.8).

Draper and Lawrence (1965a,b) considered the problem of minimising the mean squared deviation averaged over the experimental region ($\overline{\text{MSD}}$) when fitting a linear model and the true model is taken to be a second order polynomial for 3 and 4 component mixtures.

In 1970 Becker extended the work of Draper and Lawrence (1965a,b) to the general p -component case. Becker (1970) as well as Draper and Lawrence (1965a,b) transforms the q -component mixture polynomials to $(q-1)$ variable polynomials and does the appropriate analysis. They give designs for the case where the bias component is relatively large, the designs being close to or the same as simplex-lattice or simplex-centroid designs.

2.5. Incomplete block designs

Since mixture designs tend to become very large a method of forming incomplete block designs would be very advantageous. Nigam (1970) gives a few methods for achieving some form of blocking. He constrains his designs to the form given by Murty and Das (1968) that is, to the symmetric-simplex type and they are all concerned with experiments on the full simplex. Two of his methods require relatively large block sizes. The one method which obtains designs through suitable choice of mixture points on a simplex, requires $3\binom{q}{3}$ points per block which are concentrated in small clusters on the 3-dimensional faces of the simplex plus suitable points added at the interior and exterior in common to each block to make a uniform exploration of the simplex. Thus only two large blocks result. The other method which generates designs through mutually orthogonal Latin squares, requires $q(q-1)/2$ observations per block. This method requires that q be odd and again only results in two blocks. The efficiency of these two designs relative to randomised complete block designs with the same observations are unknown.

Nigam's (1970) third method relies on using s^{q-1} confounded factorial designs and then transforming the $(q-1)$ variable regression function to a q -component mixture response function.

There does not exist any published method of designing incomplete block mixture experiments when the design space is restricted to a $(q-1)$ dimensional subspace of the whole $(q-1)$ dimensional simplex.

3. GENERAL DESIGN, ACCURACY OF PREDICTIONS

3.1. General design considerations

When fitting a response surface one not only wishes to estimate the expected mean response, $\hat{\eta}(\underline{x})$, at a point, $\underline{x}$, in the design space, $\mathcal{X}$, but also to know something about the accuracy of the estimate.

The variance of the expected mean response at a point $\underline{x}$ in $\mathcal{X}$ using the design D_n , with corresponding matrix X_n , is given by

$$\text{var} \{ \hat{\eta}(\underline{x}) \} = \sigma^2 f(\underline{x})' (X_n' X_n)^{-1} f(\underline{x}).$$

Without loss of generality assume $\sigma^2 = 1$. Then

$$\text{var} \{ \hat{\eta}(\underline{x}) \} = f(\underline{x})' (X_n' X_n)^{-1} f(\underline{x}). \quad (3.1.1)$$

The nature of $\text{var} \{ \hat{\eta}(\underline{x}) \}$ can be investigated at the design stage, since (3.1.1) does not rely on measured responses. It would be of interest to see how $\text{var} \{ \hat{\eta}(\underline{x}) \}$ behaves under various mathematical models and designs. This may be done by drawing contours of equal variance over the region $\mathcal{X}$ for specific design and model combinations and comparing these.

In particular the position of the minimum and maximum value of $\text{var} \{ \hat{\eta}(\underline{x}) \}$ would be of interest.

Initially the $\text{var} \{ \hat{\eta}(\underline{x}) \}$ should be plotted for some simple general designs and models and then some more complicated situations examined. The reference set might consist of a selection of simplex-lattice designs with the models polynomials, for example, linear, quadratic, etc. in the parameters. The simplest non-trivial case, and virtually the only one manageable graphically is the 3-component

mixture which can be represented as an equilateral triangle in the plane. For $q > 3$ one would concentrate mainly on examining the $|M(\xi_n)|$ and comparing these for different designs.

Gorman and Hinman (1962) published figures showing the contours of $\text{var} \{ \hat{\eta}(\underline{x}) \}$ for the full simplex when $q = 3$ for the quadratic, special cubic, cubic and quartic models. For the quadratic model they used the $\{3, 2\}$ simplex-lattice design with 6 observations, for the cubic model the $\{3, 3\}$ simplex-lattice design with 10 observations was used and for the quartic model the $\{3, 4\}$ simplex-lattice design. The design for the special cubic consisted of the $\{3, 2\}$ simplex-lattice design plus a seventh observation at $(1/3, 1/3, 1/3)$, the centroid of the design space.

The results of Gorman and Hinman (1962) are extended to consider what happens when the design space is not the full simplex and when the number of observations is greater than the number of estimable parameters in the model.

Since $x_1 + x_2 + \dots + x_q = 1$, $\text{var} \{ \hat{\eta}(\underline{x}) \}$ is independent of the parameterisation, although for computational reasons some choices of parameterisation may be preferable to others. For example for $q = 3$ in the quadratic case for a given design D_n , the three parameterisations

$$(i) f^{\underline{x}}(\underline{x})' = (x_1, x_2, x_3, x_1 \cdot x_2, x_1 \cdot x_3, x_2 \cdot x_3),$$

with corresponding matrix $X_n^{\underline{x}}$;

$$(ii) f^{\underline{xx}}(\underline{x})' = \{1, (x_1 - \bar{x}_1), (x_2 - \bar{x}_2), (x_1 - \bar{x}_1)^2, (x_1 - \bar{x}_1)(x_2 - \bar{x}_2), (x_2 - \bar{x}_2)^2\},$$

with matrix $X_n^{\underline{xx}}$; and

$$(iii) f^{XXX}(\underline{x})' = \{1, (x_1 - x_3), (x_2 - x_3), (x_1 - x_3)^2, (x_1 - x_3)(x_2 - x_3), (x_2 - x_3)^2\}$$

with matrix X_n^{XXX} , can all be derived from the general parameterisation

$$f(\underline{x})' = (1, x_1, x_2, x_3, x_1^2, x_1 x_2, \dots, x_3^2), \quad (3.1.2)$$

with matrix X_n . For (3.1.2) define $\beta_1 + \beta_2 + \beta_3 = 0$, $\sum_{k=1}^3 \beta_{ik} = \sum_{k=1}^3 \beta_{kj} = 0$,

where the β_i are the coefficients of the x_i terms and the β_{ij} the coefficients of the $x_i x_j$ terms ($i, j = 1, 2, 3$) in $\eta(\underline{x}) = \beta' f(\underline{x})$. The above four functions would yield the same var $\{ \hat{\eta}(\underline{x}) \} = f(\underline{x})' (X_n' X_n)^{-1} f(\underline{x})$ for any $\underline{x}$ in $\mathcal{X}$. Thus, also

$$\left| \frac{1}{n} (X_n' X_n) \right| \propto \left| \frac{1}{n} (X_n^X X_n^X) \right| \propto \left| \frac{1}{n} (X_n^{XX} X_n^{XX}) \right| \propto \left| \frac{1}{n} (X_n^{XXX} X_n^{XXX}) \right|$$

for different designs and n . The parameterisation of Cox (1971) and

(3.1.2) are equivalent for computing var $\{ \hat{\eta}(\underline{x}) \}$.

Suppose the quadratic canonical polynomial model (2.2.2) is fitted to a 3-component design. This model has 6 estimable parameters namely $\beta' = (\beta_1, \beta_2, \beta_3, \beta_{12}, \beta_{13}, \beta_{23})$. Since the investigator is usually not only interested in predicting the response $\hat{\eta}(\underline{x})$ but also in estimating the k estimable parameters in β , he would wish to get an indication of the relative precision by which they are estimated. Now $\text{var}(\beta) = n \sigma^2 (X_n' X_n)^{-1}$. Assume $\sigma^2 = 1$. Ideally $(X_n' X_n)$ should be diagonal and the diagonal terms equal, the parameters then being estimated independently and with equal accuracy.

A measure of the independence of the β is given by

$$W = |M(\xi_n)| / \prod_{i=1}^k m_{ii}(\xi_n), \quad (3.1.3)$$

where $m_{ii}(\xi_n)$ is the diagonal element of the i^{th} row of $M(\xi_n)$. A value of $W = 1$ indicates complete independence among the β 's. The value of W , however, depends on the parameterisation of the model, and can be made unity by a suitable transformation.

Let

$$M(\xi_n) = \begin{bmatrix} M_{11}(\xi_n), & M_{21}(\xi_n) \\ M_{12}(\xi_n), & M_{22}(\xi_n) \end{bmatrix}, \quad (3.1.4)$$

where $M_{11}(\xi_n)$ is the $s \times s$ information matrix, $s \leq k$, of a subset of the k estimable parameters, $M_{22}(\xi_n)$ is the $(k-s) \times (k-s)$ information matrix of the remaining $(k-s)$ parameters and $nM_{12}(\xi_n) = nM_{21}'(\xi_n)$ is the $s \times (k-s)$ matrix of sums of products between the set of s and the set of $(k-s)$ parameters. The corresponding partition of the inverse is

$$M^{-1}(\xi_n) = \begin{bmatrix} M^{11}(\xi_n), & M^{21}(\xi_n) \\ M^{12}(\xi_n), & M^{22}(\xi_n) \end{bmatrix}. \quad (3.1.5)$$

If $s = 1$, then $M_{11}(\xi_n) = m_{11}(\xi_n)$ and $M^{11}(\xi_n) = m^{11}(\xi_n)$. Then a measure of the correlation between any parameter, β_1 say in the quadratic canonical polynomial model, and the other $(k-1)$ parameters is given by

$$\rho_{1.2\dots k} = + \sqrt{1 - |M(\xi_n)| / \{m_{11}(\xi_n) |M_{22}(\xi_n)|\}}. \quad (3.1.6)$$

But since

$$\begin{aligned} |M(\xi_n)| &= |M_{22}(\xi_n)| |M_{11}(\xi_n) - M_{12}(\xi_n) M_{22}^{-1}(\xi_n) M_{21}(\xi_n)| \\ &= |M_{22}(\xi_n)| |M^{11}(\xi_n)|, \end{aligned}$$

$$|M^{11}(\xi_n)| = |M(\xi_n)| / |M_{22}(\xi_n)|.$$

Rewrite (3.1.6) as

$$\rho_{1,2,\dots,k} = + \sqrt{1 - m^{11}(\xi_n)/m_{11}(\xi_n)}.$$

It can easily be shown that under a linear model the minimum $f(\underline{x})' (X_n' X_n)^{-1} f(\underline{x}) = 1/n$ and lies at the centroid of the design. The same generalisation however cannot be made for higher order models.

3.2. Design spaces, designs and mathematical models

Since the design space for the simplest non-trivial mixture experiment, that with three components, can be represented in two dimensions as an equilateral triangle it was decided to concentrate on mixtures with three components. The computations for mixtures with a larger number of components would be tedious and difficult to represent visually. Results for $q = 3$ for the full simplex should however be an indication of what will happen in the case of higher dimensions. The results for the bounding faces of the tetrahedron of the 4 component mixture are the same or proportional to those of the three component triangle as each bounding face is a 3-component mixture with the fourth component absent.

In order to extend the results of Gorman and Hinman (1962) the following three design spaces were considered:

- (i) Full simplex; $0 \leq x_i \leq 1$ for all $i = 1, 2, 3$;

(ii) constrained space, triangular region;

$$0.2 \leq x_1 \leq 0.9; \quad 0.1 \leq x_2 \leq 0.8; \quad 0 \leq x_3 \leq 0.7; \text{ and}$$

(iii) constrained space, quadrilateral region; $0.2 \leq x_1 \leq 0.9$;

$0.1 \leq x_2 \leq 0.8$; $0 \leq x_3 \leq 0.2$. These three spaces are shown in Figure 3.1.

The designs used were

(a) for space (i): the Scheffé (1958) $\{3,1\}$, $\{3,2\}$ and $\{3,3\}$ simplex-lattice designs with respectively 3, 6 and 10 distinct design points.

The $\{3,1\}$ design is used in Figure 3.2, the $\{3,2\}$ design in Figures 3.3 and 3.4 and the $\{3,3\}$ design in Figures 3.5, 3.6 and 3.7, the dots in all the figures representing the design points;

(b) for space (ii): three suitably scaled designs derived from those for space (i), again with 3, 6 and 10 design points, the 3 point design being used in Figure 3.8, the 6 point design in Figures 3.9 and 3.10 and the 10 point design in Figures 3.11, 3.12 and 3.13;

(c) for space (iii): adapted McLean and Anderson (1966) designs with 3, 4, an irregular design with 6 points and 14 points. The 3 point design is used in Figure 3.14, the 4 point in Figure 3.15, the 6 point in Figures 3.16 and 3.17 and the 14 point design in Figure 3.18. Because of the shape of the design space no 10 point design was found with a non-singular $M(\xi_{10})$ matrix. The design in fact used was arrived at by means of the method described in Section 5.1.

At each design point only one observation was taken, in other words a design with n observations, will have these observations taken at n distinct points in the design space.

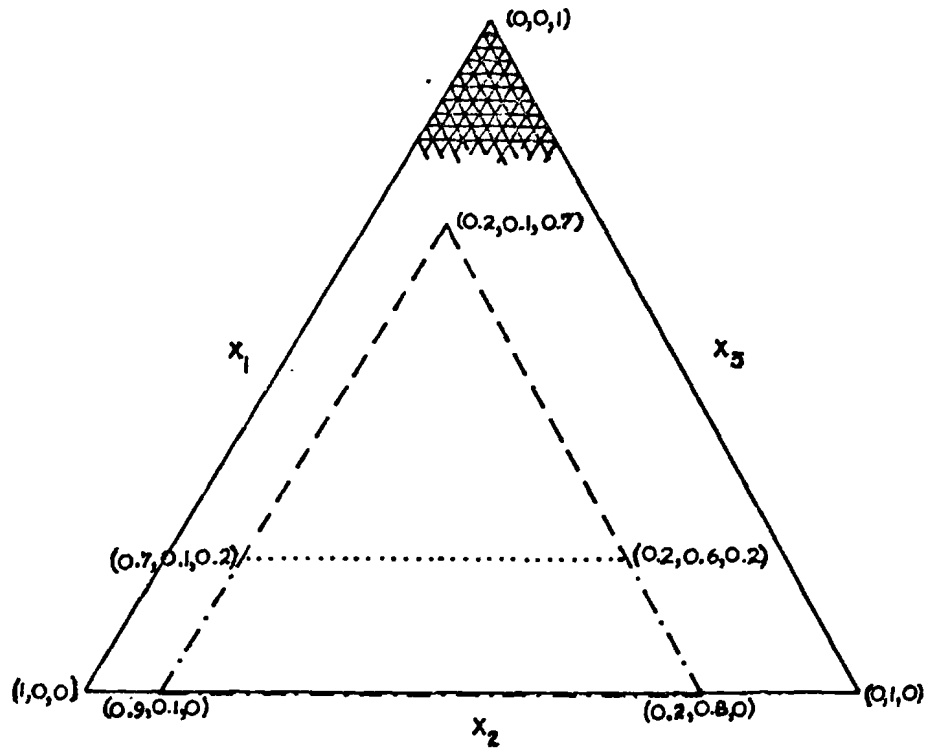


Figure 3.1 Design spaces for 3-component mixtures and a portion of the grid on which the values of $\text{var}\{\hat{\eta}(\underline{x})\}$ were calculated. Boundary of space (i):—; space (ii):----; space (iii):.....

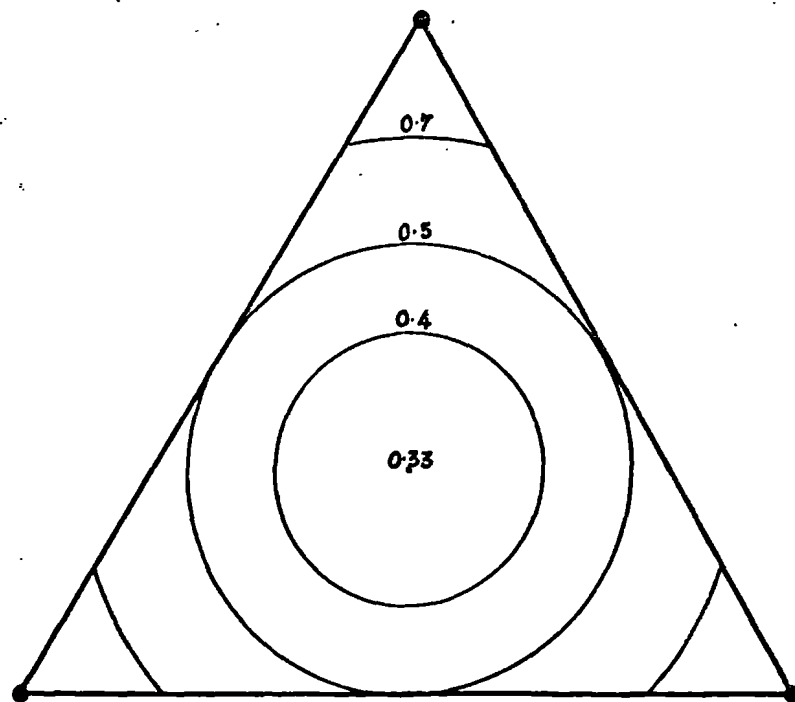


Figure 3.2 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (I) to a 3-point design in design space (i).

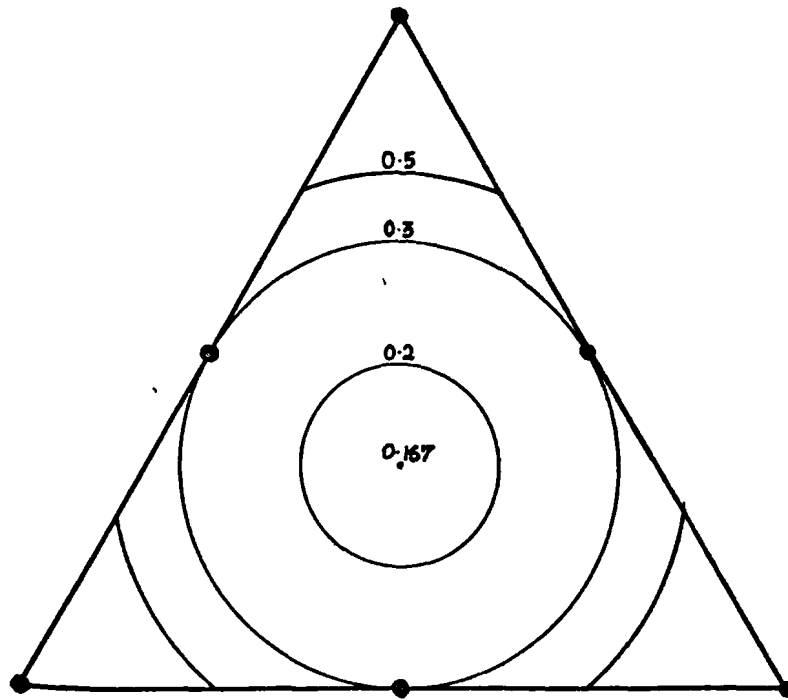


Figure 3.3 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (I) to a 6-point design in design space (i).

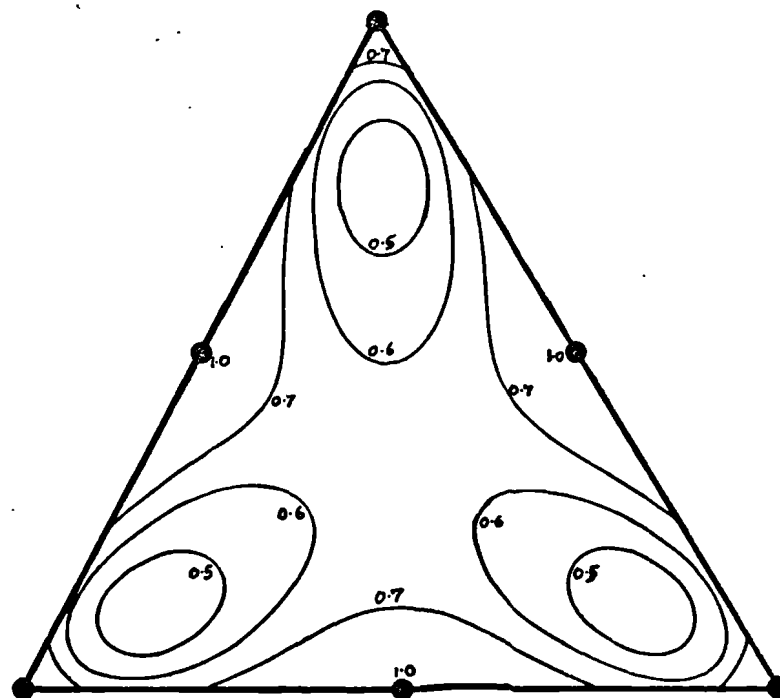


Figure 3.4 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (II) to a 6-point design in design space (i).

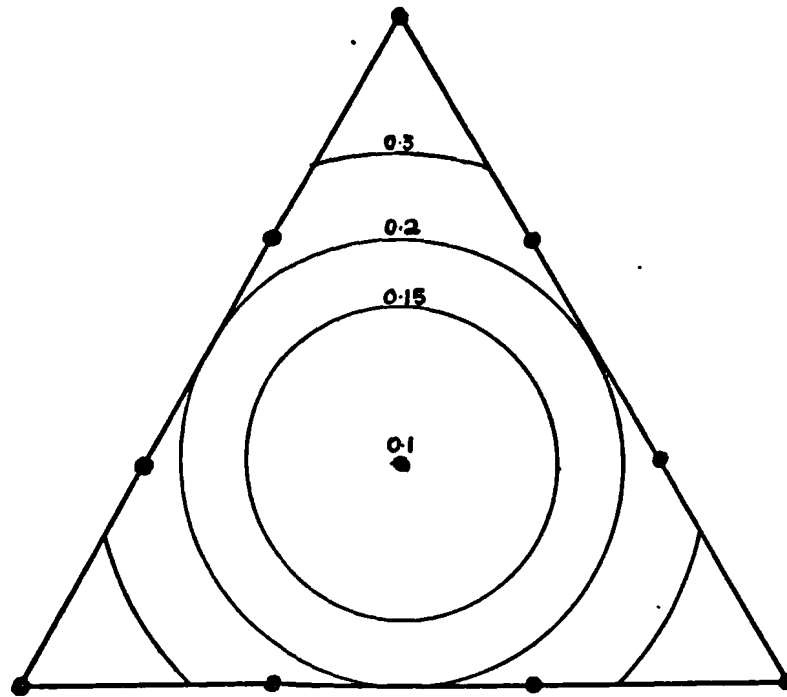


Figure 3.5 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (I) to a 10-point design in design space (i).

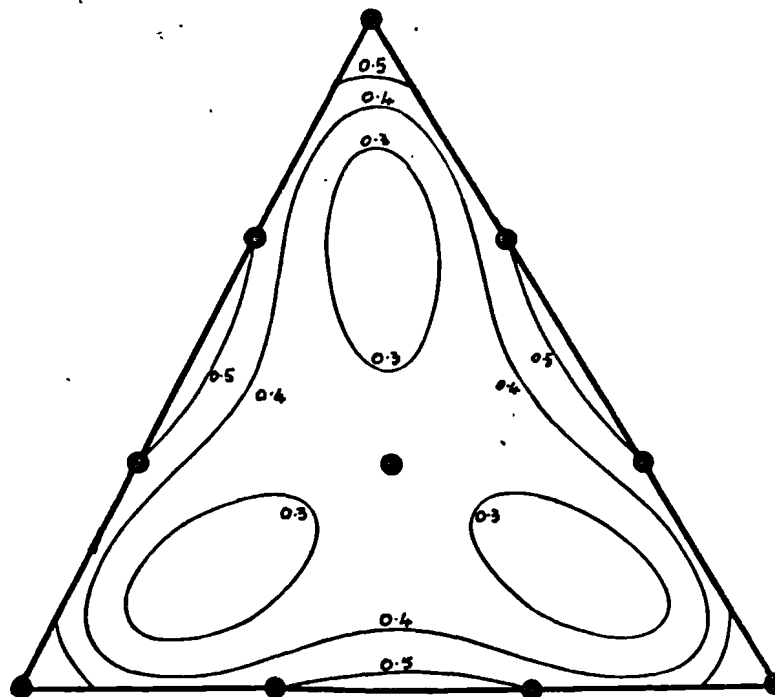


Figure 3.6 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (II) to a 10-point design in design space (i).

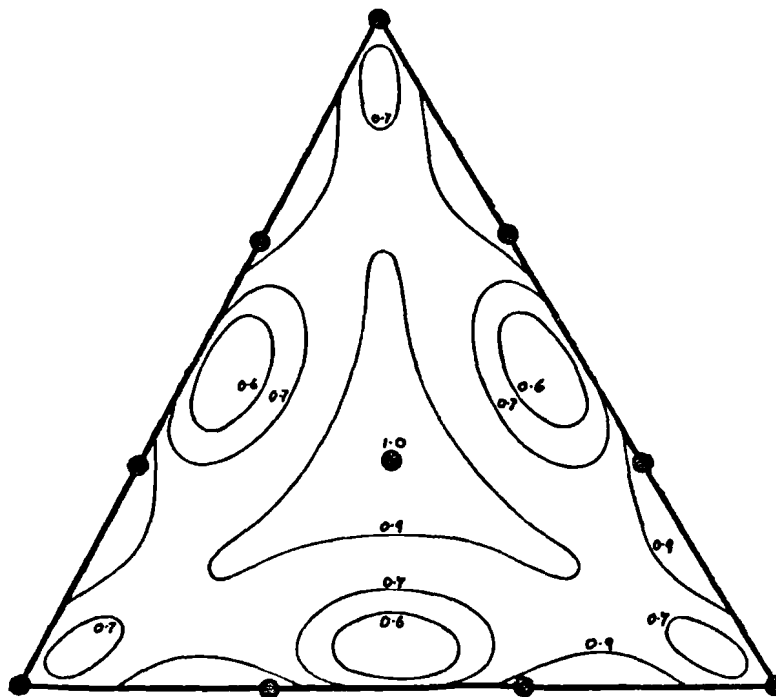


Figure 3.7 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (III) to a 10-point design in design space (i).

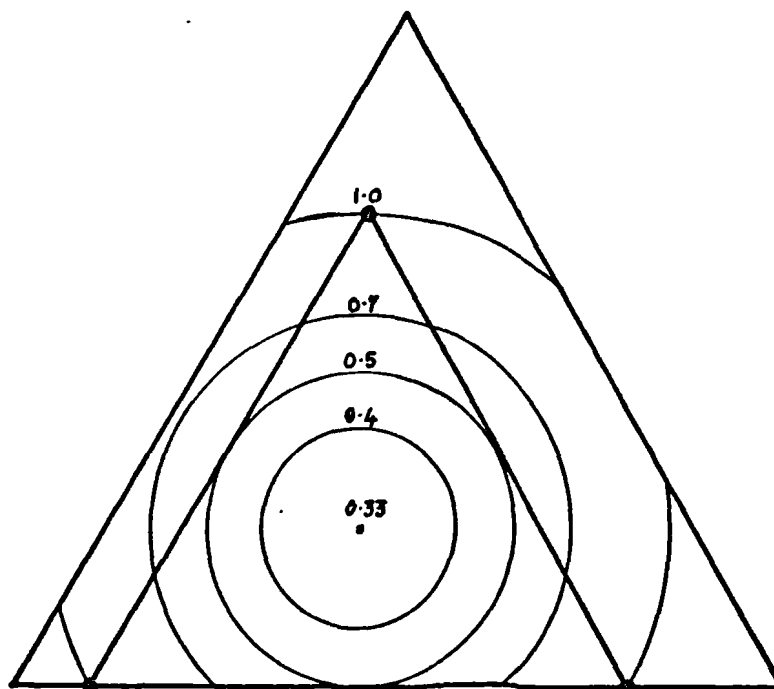


Figure 3.8 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (I) to a 3-point design in design space (ii).

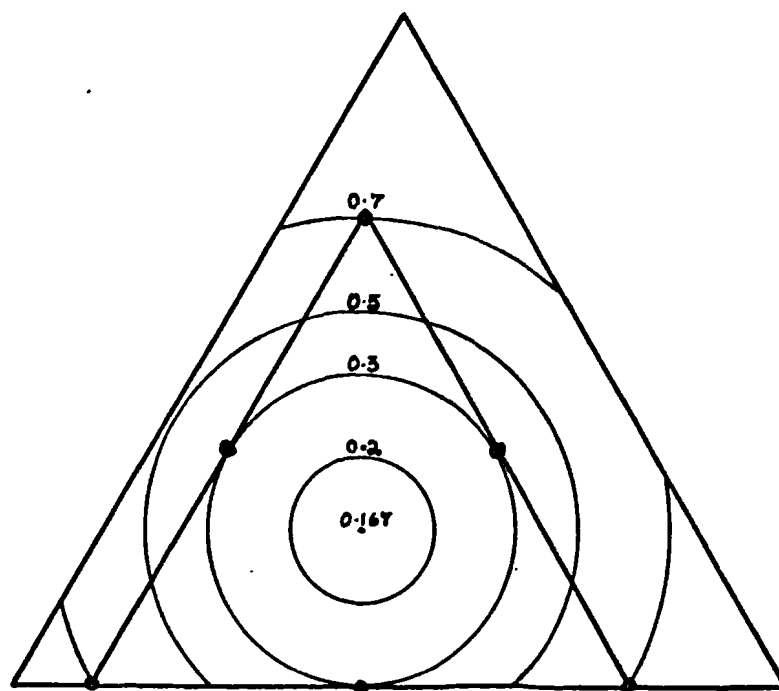


Figure 3.9 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (I) to a 6-point design in design space (ii).

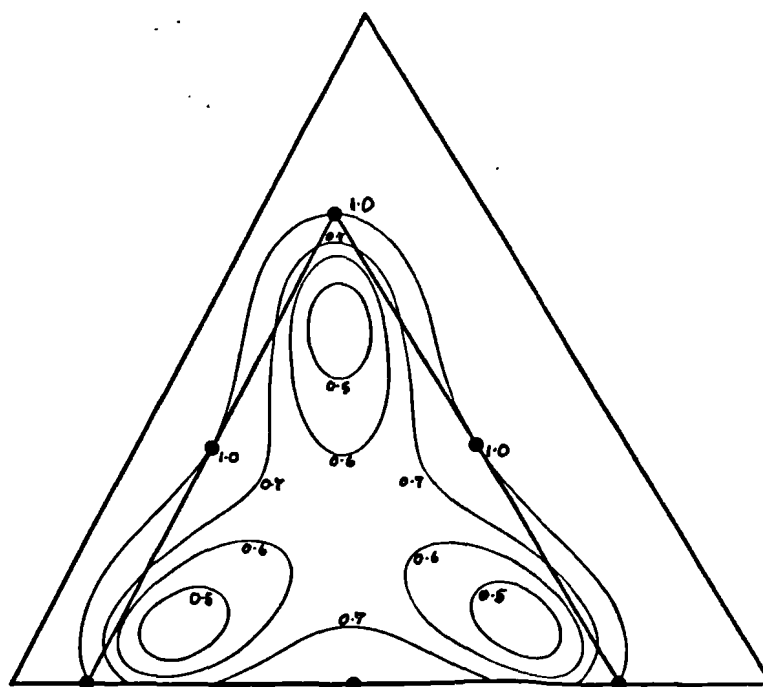


Figure 3.10 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (II) to a 6-point design in design space (ii).

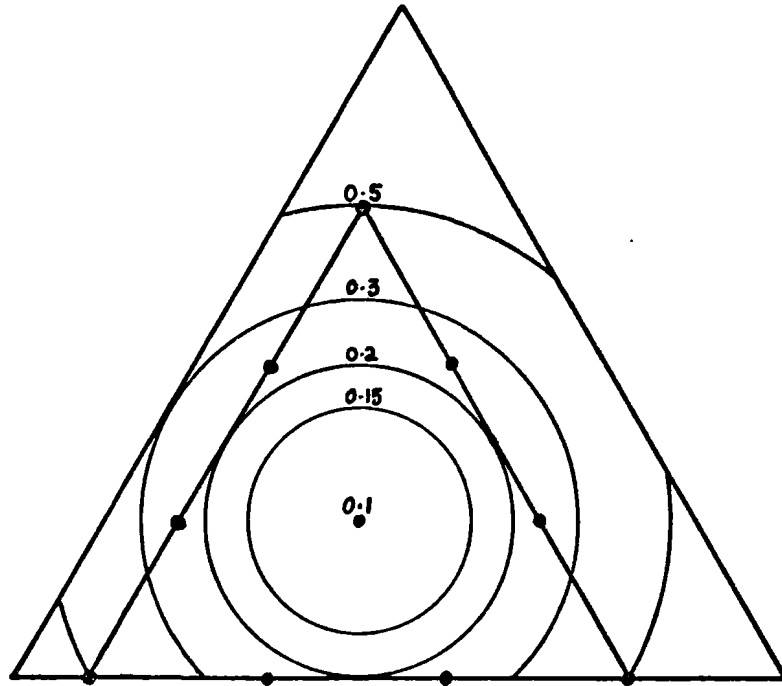


Figure 3.11 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (I) to a 10-point design in design space (ii).

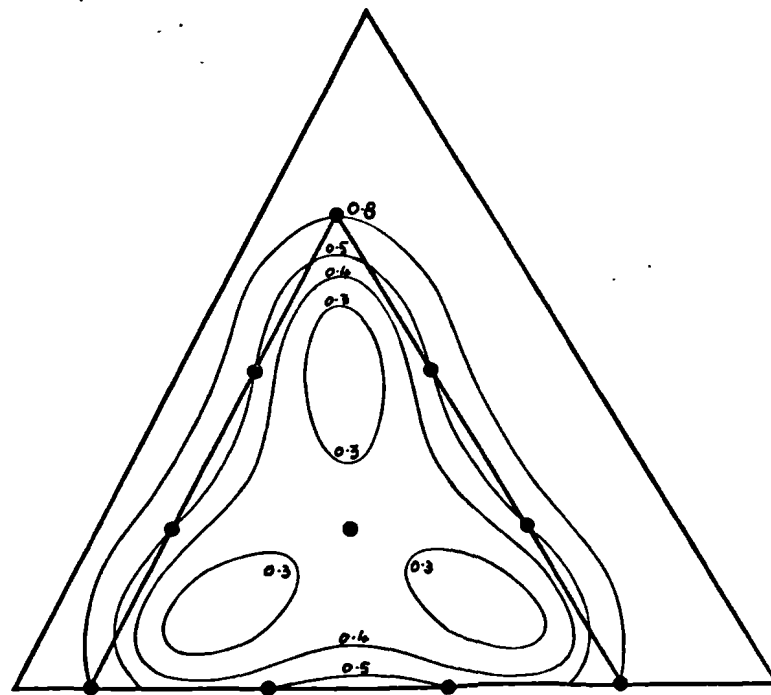


Figure 3.12 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (II) to a 10-point design in design space (ii).

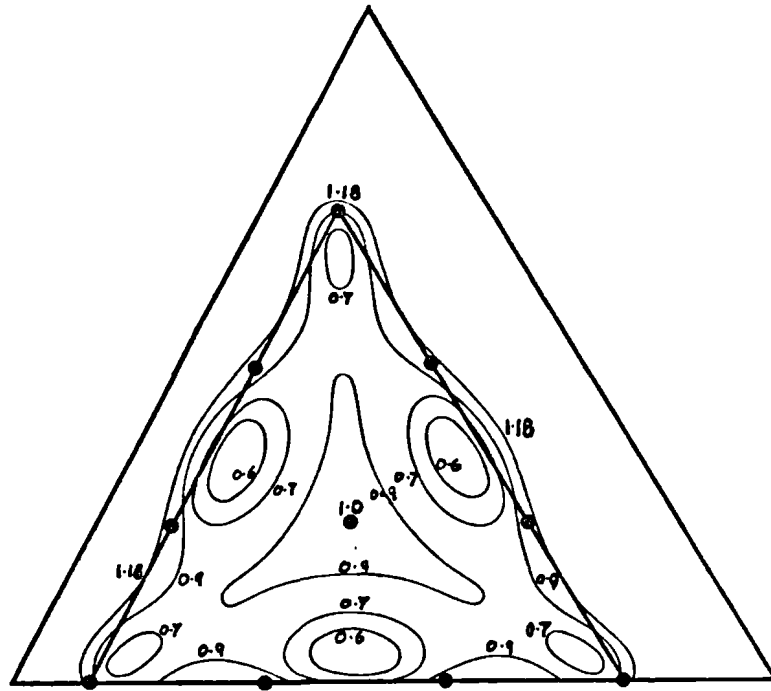


Figure 3.13 Contours of equal var $\{\hat{\eta}(\underline{x})\}$ for fitting model (III) to a 10-point design in design space (ii).

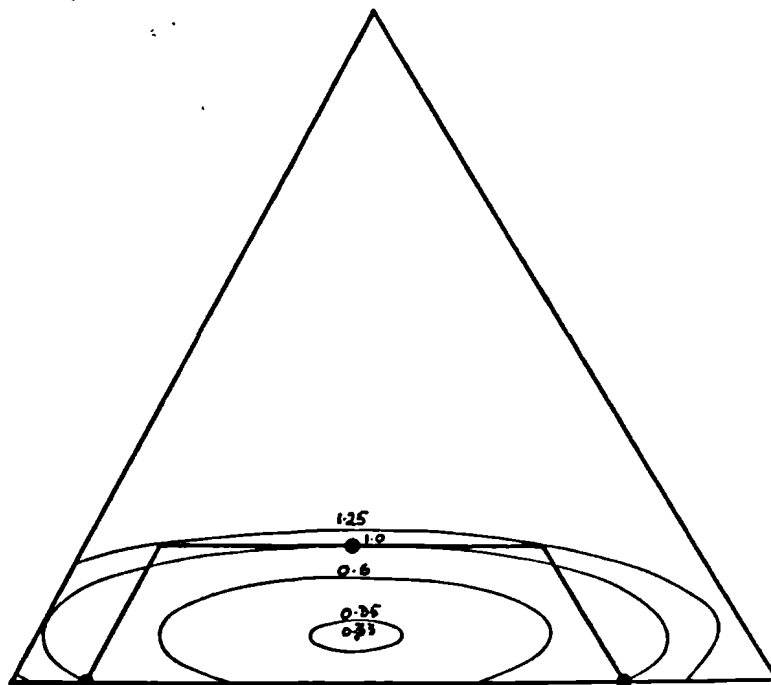


Figure 3.14 Contours of equal var $\{\hat{\eta}(\underline{x})\}$ for fitting model (I) to a 3-point design in design space (iii).

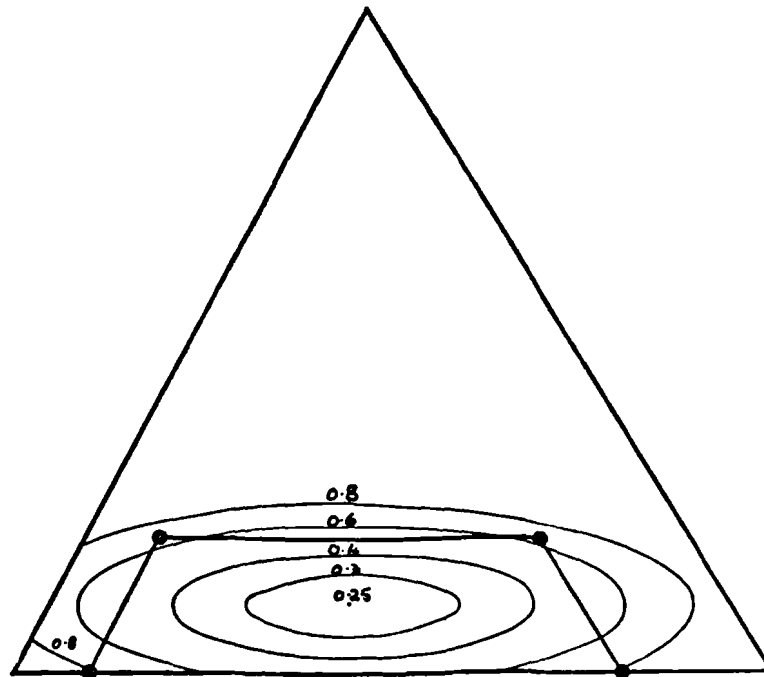


Figure 3.15 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (I) to a 4-point design in design space (iii).

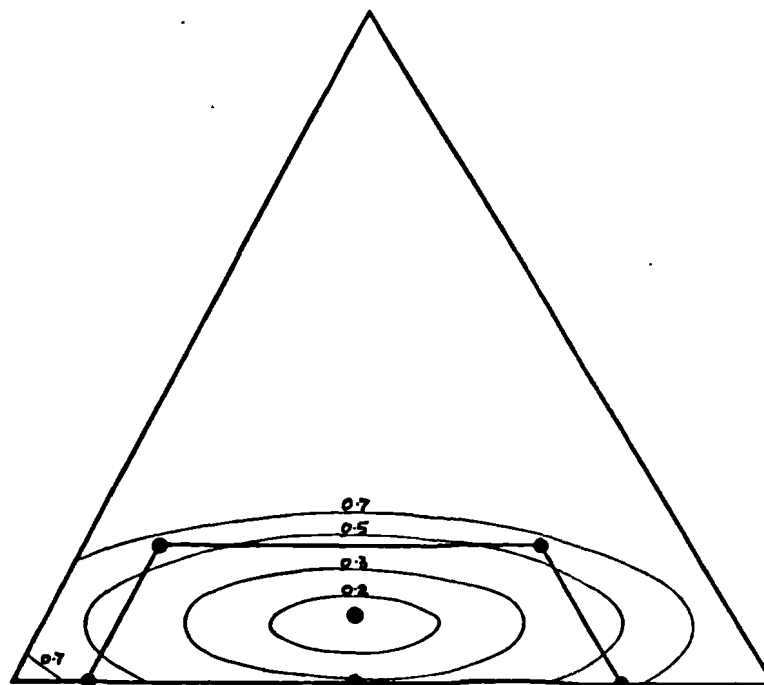


Figure 3.16 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (I) to a 6-point design in design space (iii).

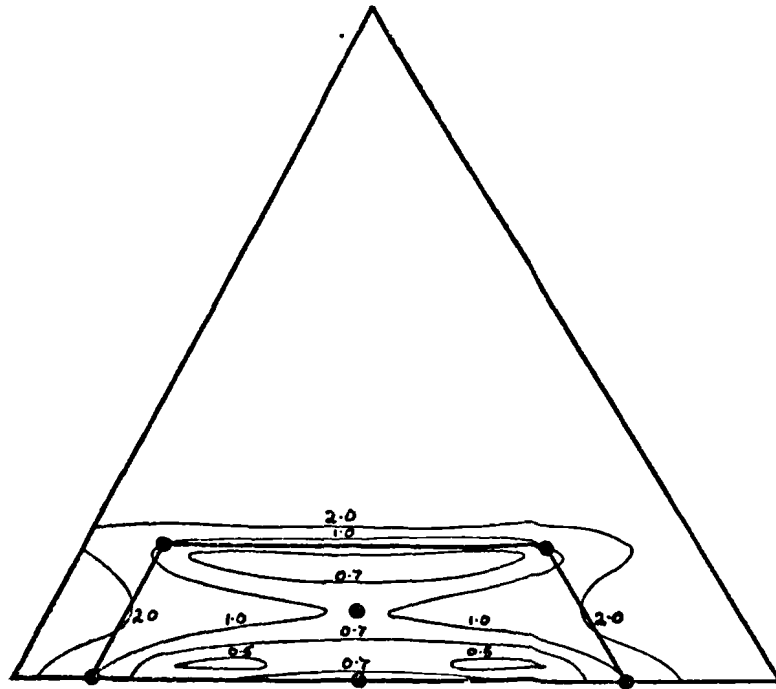


Figure 3.17 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (II) to a 6-point design in design space (iii).

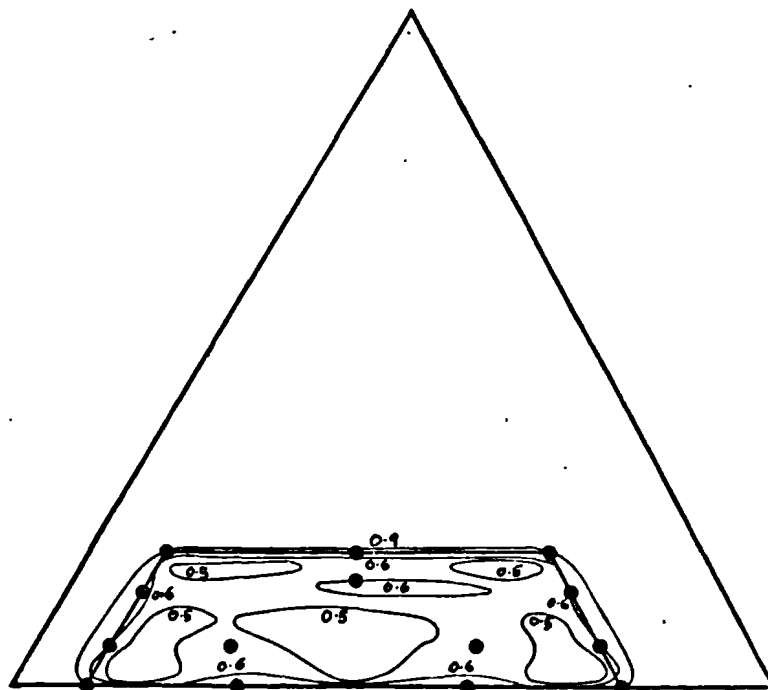


Figure 3.18 Contours of equal $\text{var}\{\hat{\eta}(\underline{x})\}$ for fitting model (III) to a 14-point design in design space (iii).

The mathematical models used were:

(I) linear:

$$\eta(\underline{x}) = \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3$$

(II) quadratic:

$$\eta(\underline{x}) = \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{11} x_1^2 + \beta_{12} x_1 x_2 + \beta_{22} x_2^2; \text{ and}$$

(III) cubic:

$$\eta(\underline{x}) = \sum_{i=1}^3 \beta_i x_i + \sum_{i=1}^2 \sum_{j=1}^2 \beta_{ij} x_i x_j + \sum_{i=1}^2 \sum_{j=1}^2 \sum_{l=1}^2 \beta_{ijl} x_i x_j x_l$$

3.3. Results

The method used to arrive at contours of var $\{\hat{\eta}(\underline{x})\}$ for the above models consisted of

- (i) computing var $\{\hat{\eta}(\underline{x})\}$ for each of 1326 points on a triangular grid. This allowed for spacings of 0.02 in each of the three components. A portion of this grid is shown in Figure 3.1, the nodes of the grid-lines forming the grid points;
- (ii) printing these values as suitably scaled integers on a grid forming an equilateral triangle with 51 points on each side. The scaling is usually with respect to either the maximum or minimum var $\{\hat{\eta}(\underline{x})\}$ within the design space for that design and model combination; and
- (iii) manually connecting points of equal value to give the contours of equal var $\{\hat{\eta}(\underline{x})\}$.

Table 3.1 contains values of the maximum var $\{\hat{\eta}(\underline{x})\}$ and the points in the design space where this is achieved and minimum

$\text{var } \{\hat{\eta}(\underline{x})\}$ and its locations. Table 3.2 contains the $\text{var } \{\hat{\eta}(\underline{x})\}$ at the centroid of the design space and the $|M(\xi_n)|$ value. These values are given for each design and model combination defined in Section 3.2. The last column in each table gives the number of the figure in which the variance contours for that design and model combination are shown.

As is expected, the linear model (I) has the minimum $\text{var } \{\hat{\eta}(\underline{x})\} = 1/n$ at the centroid of the design. The values of the maximum and minimum $\text{var } \{\hat{\eta}(\underline{x})\}$ and their relative locations in the design spaces are the same for spaces (i) and (ii). The $|M(\xi_n)|$, however, differ, that for space (i) being the larger. The ratios of the $|M(\xi_n)|$ for similar designs and the same models in the two spaces is discussed in Section 4.4.

In Figures 3.2 to 3.7 the contours of equal $\text{var } \{\hat{\eta}(\underline{x})\}$ for design space (i) are shown for all viable combinations of the three models and the three designs specified. Design points are shown as dots in the figures. Similarly the variance contours for design space (ii) are shown in Figures 3.8 to 3.13 and those for design space (iii) in Figures 3.14 to 3.18. The design with 14 points in the design space (iii) were, however, only used in conjunction with model III.

It is noticeable that corresponding designs in design spaces (i) and (ii) are similar in appearance and generally one would expect this to hold for all triangular design spaces. This makes the problem of design for any simplex space much simpler, designs for simplex subspaces being scaled down versions of designs for the whole simplex.

The variance contours, however, change considerably when

TABLE 3.1. Maximum and minimum values of var $\{f(x)\}$ and the points at which they occur, the number, n of design points and the figure number of each design and model combination considered.

Design space	n	Model no.	Maximum		Minimum		Fig. no.
			Points	var $\{f(x)\}$	Points	var $\{f(x)\}$	
(i) Full simplex	3	(I)	(0, 0, 1)	1.0000	(0.33, 0.33, 0.33)	0.3333	3.2
$0 \leq x_1 \leq 1$ (i=1, 2, 3)	6	(I)	(0, 1, 0)				
			(1, 0, 0)				
			(0, 0, 1)	0.7000	(0.33, 0.33, 0.33)	0.1667	3.3
			(0, 1, 0)				
			(1, 0, 0)				
	6	(III)	(0, 0, 1)	1.0000	(0.12, 0.12, 0.76)	0.4423	3.4
			(0, 1, 0)				
			(1, 0, 0)				
			(0, 0, 1)				
			(0, 5, 0.5)				
			(0.5, 0, 0.5)				
			(0.5, 0.5, 0)				
	10	(I)	(0, 0, 1)	0.5000	(0.33, 0.33, 0.33)	0.1000	3.5
			(0, 1, 0)				
			(1, 0, 0)				
			(0, 0, 1)				
	10	(II)	(0, 1, 0)	0.8857	(0.16, 0.16, 0.68)	0.2552	3.6
			(0, 1, 0)		(0.16, 0.68, 0.16)		
			(1, 0, 0)		(0.68, 0.16, 0.16)		
	10	(III)	(0, 0.24, 0.76)	1.1770	(0.06, 0.47, 0.47)	0.4972	3.7
			(0.24, 0, 0.76)		(0.47, 0.06, 0.47)		
			(0, 0.76, 0.24)		(0.47, 0.47, 0.06)		
			(0.76, 0, 0.24)				
			(0.24, 0.76, 0)				
			(0.76, 0.24, 0)				
(ii) Constrained, triangular;	3	(I)	(0.2, 0.1, 0.7)	1.0000	(0.43, 0.33, 0.23)	0.3333	3.8
$0.2 \leq x_1 \leq 0.9$	6	(I)	(0.2, 0.8, 0)				
$0.1 \leq x_2 \leq 0.8$			(0.9, 0.1, 0)				
$0 \leq x_3 \leq 0.7$			(0.2, 0.1, 0.7)	0.7000	(0.43, 0.33, 0.23)	0.1667	3.9
			(0.2, 0.8, 0)				
			(0.9, 0.1, 0)				
	6	(III)	(0.2, 0.1, 0.7)	1.0000	(0.28, 0.18, 0.54)	0.4424	3.10
			(0.2, 0.8, 0)		(0.28, 0.64, 0.08)		
			(0.9, 0.1, 0)		(0.74, 0.18, 0.08)		
			(0.2, 0.45, 0.35)				
			(0.55, 0.10, 0.35)				
			(0.55, 0.45, 0)				
	10	(I)	(0.2, 0.1, 0.7)	0.5000	(0.43, 0.33, 0.23)	0.1000	3.11
			(0.2, 0.8, 0)				
			(0.9, 0.1, 0)				
	10	(II)	(0.2, 0.1, 0.7)	0.8857	(0.3, 0.2, 0.5)	0.2567	3.12
			(0.2, 0.8, 0)		(0.3, 0.6, 0.1)		
			(0.9, 0.1, 0)		(0.7, 0.2, 0.1)		
	10	(III)	(0.2, 0.26, 0.54)	1.1773	(0.24, 0.43, 0.33)	0.4974	3.13
			(0.36, 0.1, 0.54)		(0.53, 0.43, 0.04)		
			(0.2, 0.64, 0.16)		(0.53, 0.14, 0.33)		
			(0.74, 0.1, 0.16)				
			(0.36, 0.64, 0)				
			(0.74, 0.26, 0)				
(iii) Constrained, quadrilateral;	3	(I)	(0.2, 0.6, 0.2)	1.2551	(0.52, 0.42, 0.06)	0.3333	3.14
$0.2 \leq x_1 \leq 0.9$	4	(I)	(0.7, 0.1, 0.2)	0.8311	(0.5, 0.4, 0.1)	0.2500	3.15
$0.1 \leq x_2 \leq 0.8$	6	(I)	(0.9, 0.1, 0)	0.6414	(0.51, 0.41, 0.08)	0.1667	3.16
$0 \leq x_3 \leq 0.2$	6	(II)	(0.9, 0.1, 0)	2.0815	(0.34, 0.64, 0.02)	0.4601	3.17
			(0.2, 0.7, 0.1)				
			(0.8, 0.1, 0.1)				
	14	(III)	(0.2, 0.8, 0)	0.9077	(0.24, 0.74, 0.02)	0.3947	3.18
			(0.9, 0.1, 0)		(0.84, 0.14, 0.02)		

TABLE 3.2. The number, n of design points, the $\text{var}\{\hat{\eta}(\underline{x})\}$ at the centroid of the design space, the value of $|M(\xi_n)|$ and the figure number of each design and model combination considered

Design Space	n	Model no.	Centroid	$\text{var}\{\hat{\eta}(\underline{x})\}$	$ M(\xi_n) $	Fig. no.
(i) Full simplex	3	(I)	(0.33, 0.33, 0.33)	0.3333	0.037037	3.2
$0 \leq x_i \leq 1$ ($i=1, 2, 3$)	6	(I)	(0.33, 0.33, 0.33)	0.1667	0.014468	3.3
	6	(II)	(0.33, 0.33, 0.33)	0.6296	0.5233×10^{-8}	3.4
	10	(I)	(0.33, 0.33, 0.33)	0.1000	0.009259	3.5
	10	(II)	(0.33, 0.33, 0.33)	0.3143	0.1912×10^{-8}	3.6
	10	(III)	(0.33, 0.33, 0.33)	1.0000	0.2729×10^{-23}	3.7
(ii) Constrained, triangular,	3	(I)	(0.43, 0.33, 0.23)	0.3333	0.8893×10^{-2}	3.8
$0.2 \leq x_1 \leq 0.9$,	6	(I)	(0.43, 0.33, 0.23)	0.1667	0.3474×10^{-2}	3.9
$0.1 \leq x_2 \leq 0.8$,	6	(II)	(0.43, 0.33, 0.23)	0.6296	0.1739×10^{-10}	3.10
$0 \leq x_3 \leq 0.7$.	10	(I)	(0.43, 0.33, 0.23)	0.1000	0.2223×10^{-2}	3.11
	10	(II)	(0.43, 0.33, 0.23)	0.3143	0.6355×10^{-11}	3.12
	10	(III)	(0.43, 0.33, 0.23)	0.1000	0.1738×10^{-29}	3.13
(iii) Constrained, quadrilateral;	3	(I)	(0.5, 0.4, 0.1)	0.3750	0.7259×10^{-3}	3.14
$0.2 \leq x_1 \leq 0.9$,	4	(I)	(0.5, 0.4, 0.1)	0.2500	0.9250×10^{-3}	3.15
$0.1 \leq x_2 \leq 0.8$,	6	(I)	(0.5, 0.4, 0.1)	0.1724	0.4968×10^{-3}	3.16
$0 \leq x_3 \leq 0.2$	6	(II)	(0.5, 0.4, 0.1)	1.0000	0.6304×10^{-14}	3.17
	14	(III)	(0.5, 0.4, 0.1)	0.4320	0.5145×10^{-37}	3.18

we are constrained to a non-triangular subspace like space (iii) above. At this stage the investigator should consider the particular space he is interested in. He should pay particular attention to the parameterisation as some forms are difficult to handle computationally because elements of the inverse matrix sometimes are of the order 10^5 and rounding errors become important. Some of the effect of this can be seen in Figure 3.18 where the contours of equal values of $\text{var} \{ \hat{\eta}(\underline{x}) \}$ are asymmetrical although the design is symmetrical with respect to the design space. The National Physical Laboratory (1961, p. 5) considers that the largest coefficient in every row or column of $(X_n' X_n)$ should lie between 0.1 and 1.0. The parameterisation of Cox (1971) as given in Section 2.2 will in some cases lead to covariance matrices of this nature.

If one component is constrained to a much narrower range than any of the other it might be necessary to consider this one as a process variable as defined by Scheffé (1963). The experiment would then be a $(q-1)$ -component mixture experiment with one process variable.

4. D-OPTIMUM MIXTURE DESIGNS

4.1. Introduction

To ascertain to what extent the existing methods of designing mixture experiments, as used in Section 3.2, can be expected to yield approximate D-optimum designs it is desirable to generate a series of D-optimum or approximate D-optimum designs for the design spaces and models of Section 3.2.

The method given by Wynn (1970), and discussed in Section 2.4, is used here. Each iteration of the computer routine to maximise

$|M(\xi_n)|$ consisted of the following steps:

(i) for a given design D_n , variances of the expected mean response were computed on the same grid as in Section 3.3 to ascertain the maximum value of $\text{var} \{ \hat{\gamma}(\underline{x}_0) \}$ and its location, $\underline{x}_0$;

(ii) the point $\underline{x}_0$ was added to the design D_n to yield a design D_{n+1} , and the matrix $M(\xi_{n+1}) = M(\xi_n) + f(\underline{x}_0) f(\underline{x}_0)'$ computed;

(iii) the augmented matrix $M^{-1}(\xi_{n+1})$ was computed by means of the formula given by Plackett (1950), namely

$$M^{-1}(\xi_{n+1}) = M^{-1}(\xi_n) - M^{-1}(\xi_n) f(\underline{x}_0) \left\{ 1 + f(\underline{x}_0)' M^{-1}(\xi_n) f(\underline{x}_0) \right\}^{-1} f(\underline{x}_0)' M^{-1}(\xi_n)$$

and the $|M(\xi_{n+1})|$ computed by using the formula given by Scheffe' (1959b p.417), namely

$$|M(\xi_{n+1})| = |M(\xi_n)| \cdot \left\{ 1 + f(\underline{x}_0)' M^{-1}(\xi_n) f(\underline{x}_0) \right\}.$$

Usually the experimental design points at which the observations should be taken become apparent after only a few iterations, but it is not so easy to determine the proportions in which the observations

should be taken at those points. For each design space and model combination, 100 iterations were made, starting with an evenly spread design, which ascertained the points at which the observations should be made. Then, each of these new points formed a new design and up to 500 iterations were made. This was done to obtain a closer approximation to the optimum design measure ξ^* . By continually checking whether a new maximum $|M(\xi_n)|$ had been reached, the best design, in terms of D-optimality, having up to 500 observations, was arrived at, i.e. a design with at most 500 observations was found such that the observations would be distributed over the experimental points in proportions as near as possible to ξ^* . The reason for using two series of iterations was that, although the design points could be ascertained with only a few iterations the evenly spaced background design tended to obscure the true proportions in which observations should be taken at those points. Instead of the second series of iterations some quadratic minimisation routine could perhaps be used to ascertain what the proportions of observations at the different design points should be.

For a D-optimum design, $|M(\xi_n)|$, under a linear model and for the q -component full simplex, is given by

$$|M(\xi_n)| = 1/q^q,$$

and the design points are at the q vertices of the $(q-1)$ -dimensional simplex and have equal measure of $1/q$. This follows directly from the fact that, when using the first order canonical polynomial model (2.2.2) with $f(x)' = (x_1, \dots, x_q)$ and the above vertex design D_q with $x_i = 1$,

$x_j = 0$ ($i \neq j$, $i, j = 1, \dots, q$), $(X_n' X_n) = I$. Thus $|X_n' X_n| = 1$, the maximum possible value.

If the design space is a $(q-1)$ -dimensional subspace of the full $(q-1)$ -dimensional simplex, and is itself a simplex, the D-optimum design for fitting a linear model is similar to that for the full space, i. e. design points with equal measure $1/q$ are taken at each of the q vertices of the subspace. For this subspace $|M(\xi_n)|$ will be smaller than that for the full simplex. If the design space is not a simplex, no generalisation about the nature of the D-optimum design can be made, the designs being space specific.

Kiefer (1961a) shows that the D-optimum design for fitting a quadratic model to a q -component full simplex design space consists of assigning a measure $\frac{2}{q(q+1)}$ to each of the $\frac{1}{2}q(q+1)$ points of the $\{q, 2\}$ simplex-lattice design.

4.2. Three component mixtures

The same design spaces and mathematical models are considered in order to compare the methods of constructing designs used in Section 3.3 with those for constructing D-optimum designs.

Table 4.1 contains, for every design space and model combination of Section 3.2, the number of distinct design points n_d , the number of observations $n_o \leq 500$ giving the maximum value of $|M(\xi_{n_o})|$. For comparative purposes *except* from Table 3.2, namely the number of observations used there, n_s , and the value of $|M(\xi_{n_s})|$, are also given in Table 4.1.

Table 4.2 shows the design points and numbers of observations

TABLE 4.1. The number of distinct design points, n_d , the number of observations $n_o \leq 500$ giving the maximum value of $|M(\xi_{n_o})|$, the number of observations n_s and the value of $|M(\xi_{n_s})|$ of the smallest viable design used in Section 3.3 for every design space and model combination of Section 3.2

Design space	Model no.	n_d	n_o	$ M(\xi_{n_o}) $	n_s	$ M(\xi_{n_s}) $
(i) Full simplex	(I)	3	3	0.037037	3	0.037037
	(II)	6	6	0.5233×10^{-8}	6	0.5233×10^{-8}
	(III)	10	10	0.4486×10^{-23}	10	0.2729×10^{-23}
(ii) Constrained, triangular.	(I)	3	3	0.8893×10^{-2}	3	0.8893×10^{-2}
	(II)	6	6	0.1739×10^{-10}	6	0.1739×10^{-10}
	(III)	10	10	0.2841×10^{-29}	10	0.1738×10^{-29}
(iii) Constrained, quadrilateral.	(I)	4	146	0.9482×10^{-3}	3	0.7259×10^{-3}
	(II)	8	486	0.9474×10^{-14}	6	0.1641×10^{-14}
	(III)	14	496	0.6406×10^{-37}	14	0.1111×10^{-37}

TABLE 4.2. Relative numbers of observations to be taken at each distinct design point to give D-optimum designs for fitting the three mathematical models to design space (i)

Design point	Relative number of observations		
	Model (I)	Model (II)	Model (III)
(0, 0, 1)	1	1	1
(0, 1, 0)	1	1	1
(1, 0, 0)	1	1	1
(0, 0.5, 0.5)	0	1	0
(0.5, 0, 0.5)	0	1	0
(0.5, 0.5, 0)	0	1	0
(0, 0.28, 0.72)	0	0	1
(0.28, 0, 0.72)	0	0	1
(0, 0.72, 0.28)	0	0	1
(0.72, 0, 0.28)	0	0	1
(0.28, 0.72, 0)	0	0	1
(0.72, 0.28, 0)	0	0	1
(0.33, 0.33, 0.33)	0	0	1
Total	3	6	10

TABLE 4.3. Relative numbers of observations to be taken at each distinct design point to give D-optimum designs for fitting the three mathematical models to design space (ii)

Design point	Relative number of observations		
	Model (I)	(Model (II)	Model (III)
(0.2, 0.1, 0.7)	1	1	1
(0.2, 0.8, 0)	1	1	1
(0.9, 0.1, 0)	1	1	1
(0.2, 0.45, 0.35)	0	1	0
(0.55, 0.10, 0.35)	0	1	0
(0.55, 0.45, 0)	0	1	0
(0.2, 0.29, 0.51)	0	0	1
(0.4, 0.1, 0.5)	0	0	1
(0.2, 0.6, 0.2)	0	0	1
(0.71, 0.1, 0.19)	0	0	1
(0.4, 0.6, 0)	0	0	1
(0.71, 0.29, 0)	0	0	1
(0.43, 0.33, 0.23)	0	0	1
Total	3	6	10

TABLE 4.4. Relative numbers of observations to be taken at each distinct design point to give D-optimum designs for fitting the three mathematical models to design space (iii)

Design point	Relative number of observations		
	Model (I)	Model (II)	Model (III)
(0.2, 0.6, 0.2)	31	75	45
(0.7, 0.1, 0.2)	31	75	45
(0.2, 0.8, 0)	42	75	47
(0.9, 0.1, 0)	42	75	46
(0.2, 0.7, 0.1)	0	39	0
(0.5, 0.4, 0.1)	0	40	0
(0.8, 0.1, 0.1)	0	39	0
(0.55, 0.45, 0)	0	68	0
(0.45, 0.35, 0.2)	0	0	31
(0.47, 0.37, 0.16)	0	0	26
(0.2, 0.66, 0.14)	0	0	32
(0.76, 0.1, 0.14)	0	0	32
(0.36, 0.58, 0.08)	0	0	21
(0.66, 0.26, 0.08)	0	0	20
(0.2, 0.74, 0.06)	0	0	30
(0.84, 0.1, 0.06)	0	0	30
(0.4, 0.6, 0)	0	0	42
(0.7, 0.3, 0)	0	0	42
Total	146	486	496

to be taken at each distinct point to obtain the maximum value of $|M(\xi_n)|$ for design space (i) of Section 3.2, the full simplex. The designs are given for each of the three mathematical models of Section 3.2. Similarly Tables 4.3 and 4.4 show the respective designs for design spaces (ii) and (iii) of Section 3.2.

Table 4.1 shows that designs with a high $|M(\xi_n)|$ usually require observations to be taken at only a few distinct points. Moreover, in the case of the simplex design spaces (i) and (ii), the D-optimum designs require equal proportions of observations to be taken at each distinct design point; thus $n_o = n_d$. However, in irregular design spaces, for example space (iii) the proportions may be highly irregular. The efficiency, in terms of the $|M(\xi_n)|$ of the designs of Section 3.2 for space (iii) does not compare very well with the efficiency of D-optimum designs. The largest proportions of the observations should, however, still be taken at the vertices of the bounding surface and smaller proportions at other points in the space. These other design points are determined from the specific model and space combination. The irregular proportions of observations to be taken at the design points is the main reason for the large numbers of observations required to reach a near optimum design. If simpler approximations to these proportions are taken, designs result which require only a relatively few observations and nevertheless have a relatively high value of $|M(\xi_n)|$. For example in model (I) a design with one observation at each of the four vertices has $|M(\xi_4)| = 0.9250 \times 10^{-3}$; taking 3 observations instead of 31 at (0.2, 0.6, 0.2) and (0.7, 0.1, 0.2) and 4 instead of 42 at (0.2, 0.8, 0)

and $(0.9, 0.1, 0)$ yields a $|M(\xi_{14})| = 0.948105 \times 10^{-3}$ whereas the design with 146 observations has a $|M(\xi_{146})| = 0.948176 \times 10^{-3}$. The same holds for model (II) for with $n = 8$, one observation at each design point, the $|M(\xi_8)| = 0.8001 \times 10^{-14}$; with $n = 13$, an additional observation at each vertex and at $(0.55, 0.45, 0)$, the $|M(\xi_{13})| = 0.9443 \times 10^{-14}$; and the design with $n = 486$ has a $|M(\xi_{486})| = 0.9447 \times 10^{-14}$.

Some of the intuitively most acceptable experimental designs for experiments covering the full simplex is the simplex-lattice designs suggested by Scheffé (1958); see Section 2.3. For models (I) and (II) of Section 3.2 simplex-lattice designs also lead to D-optimum designs. However, for the cubic model (III), the simplex-lattice design does not lead to a D-optimum design. The D-optimum design has some of its design points at $P(0, 0.28, 0.72)$, whereas the corresponding points for the simplex-lattice design lie at $P(0, 1/3, 2/3)$, where $P(a, b, c) = \{(a, b, c), (a, c, b), (b, a, c), (b, c, a), (c, a, b), (c, b, a)\}$ the set of all distinct permutations of (a, b, c) , for example if $c = a$ then $P(a, b, a) = \{(a, b, a), (a, a, b), (b, a, a)\}$. Figure 3.7 and Table 3.1 imply that something of this nature would be the case as the maximum $\text{var} \{\hat{\eta}(\underline{x})\} = 1.1770$ is at $P(0, 0.24, 0.72)$ when using the simplex-lattice design. Kiefer and Wolfowitz (1960) show that D-optimum designs have the maximum $\text{var} \{\hat{\eta}(\underline{x})\} = k/n$ lying at the design points, where k is the number of estimable parameters in the model and n the number of observations in the design.

Section 3.3 showed that when using similar designs for spaces (i) and (ii), the contours of equal $\text{var} \{\hat{\eta}(\underline{x})\}$ for space (ii) are scaled

down versions of those for space (i). This similarity also holds for D-optimum designs. The design points are in the same relative positions and the $|M(\xi^x)|$ also vary proportionately. Thus, for purposes of design, one need only consider the full simplex and scale down suitably to cope with other triangular design spaces.

A reasonable approach to the problem of an initial design for generating a D-optimum design would be to start with an evenly spread design and to ascertain where local maxima of $\text{var} \{ \hat{\eta}(\underline{x}) \}$ lie for this design; then either (i) use these points as an initial design for generating a D-optimum design; or (ii) repeat the first step using the design arrived at instead of the evenly spread design. This will be to ascertain whether a slight shift in the design points might not be desirable to use in step (i). By performing step (ii) the investigator should early in the process of generating a D-optimum design get a fairly good indication of the nature of the D-optimum design. This, in fact, might be his only possibility if $q > 3$. For example, if $q = 4$, even just computing $\text{var} \{ \hat{\eta}(\underline{x}) \}$ on the same grid as in Section 3.3, that is, in steps of 0.02 in every component, would result in a three-dimensional grid containing 45,526 points. A more efficient method than this of ascertaining where the maximum $\text{var} \{ \hat{\eta}(\underline{x}) \}$ lies in the design space would be very useful for these more complex cases.

4.3. Four component mixtures

For the D-optimum designs for fitting linear, quadratic and cubic models in the 4-component full simplex case, the mathematical models used were

$$(Ia) \text{ linear : } \eta(\underline{x}) = \sum_{i=1}^4 \beta_i x_i ;$$

$$(IIa) \text{ quadratic : } \eta(\underline{x}) = \sum_{i=1}^4 \beta_i x_i + \sum_{i=1}^3 \sum_{j=1}^3 \beta_{ij} x_i x_j ; \text{ and}$$

$$(IIIa) \text{ cubic : } \eta(\underline{x}) = \sum_{i=1}^4 \beta_i x_i + \sum_{i=1}^3 \sum_{j=1}^3 \beta_{ij} x_i x_j + \sum_{i=1}^3 \sum_{j=1}^3 \sum_{l=1}^3 \beta_{ijl} x_i x_j x_l .$$

From Section 4.2 the D-optimum designs for first and second order models, (Ia) and (IIa), for the 4-component full simplex space are known. The D-optimum design for the linear model (Ia) is the $\{4, 1\}$ simplex-lattice design consisting of 4 design points, one at each vertex of the tetrahedron. The D-optimum design for the quadratic model (IIa) is the $\{4, 2\}$ simplex-lattice consisting of 10 design points, one at each vertex and one each at $P(0, 0, 1/2, 1/2)$.

Some doubt may exist about the nature of the 4-component D-optimum design for the cubic model, (IIIa), because the 3-component simplex-lattice and D-optimum designs do not coincide. Owing to the computational difficulty involved in generating D-optimum designs for 4-component mixtures, it was decided to compute $|M(\xi_n)|$ for a few different designs. These designs were chosen to lie on and around the expected D-optimum design. It was expected that the D-optimum design would in any case include points at the vertices and at the centroids of the 4 faces of the tetrahedron. These 8 points were thus taken to be common to all the designs investigated. The designs differed in the

values of $a > 0$, $b > 0$, $a \neq b$ in $P(a, b, 0, 0)$. The designs, in terms of the $P(a, b, 0, 0)$ points, were

(a) $P(0.6667, 0.3333, 0, 0)$;

(b) $P(0.7, 0.3, 0, 0)$;

(c) $P(0.72, 0.28, 0, 0)$;

(d) $P(0.75, 0.25, 0, 0)$; and

(e) $P(0.8, 0.2, 0, 0)$,

where design (a) is the $\{4, 3\}$ simplex-lattice design and (c) the design expected to have the largest $|M(\xi_{20})|$ in accordance with the results of the previous Section 4.3.

For the above five designs and the designs for the first and second order models the values of $|M(\xi_n)|$ are given in Table 4.5.

As expected, the design with the largest value of $|M(\xi_{20})|$ is that which is a direct extension of the corresponding 3-component design, namely design (c). It is thus expected that this design can be extended to mixture experiments involving the full simplex of any number of components for third order models. Suitable scaling would make these D-optimum designs, for first, second and third order models over the full simplex, applicable to any subspace of triangular, tetrahedral or some higher dimensional simplex form.

No generalisations of the nature of D-optimum designs can be made if the region is not a simplex. It is in fact not even certain that at least some of the design points in a D-optimum design would necessarily lie at each of the vertices of the bounding surface of the design space. Such cases would have to be dealt with individually. If a $(q-1)$

TABLE 4.5. The numbers of observations, n and values of $|M(\xi_n)|$
for the different four component mixture designs under the
three models (Ia), (IIa) and (IIIa)

Design	n	Model no.	$ M(\xi_n) $
{4, 1} simplex-lattice	4	(Ia)	0.3906×10^{-2}
{4, 2} simplex-lattice	10	(Ia)	0.9438×10^{-3}
	10	(IIa)	0.5960×10^{-17}
(a) $P(0, 0, 0.33, 0.67)$,	20	(Ia)	0.3970×10^{-3}
{4, 3} simplex-lattice	20	(IIa)	0.6824×10^{-18}
	20	(IIIa)	0.1398×10^{-48}
(b) $P(0, 0, 0.3, 0.7)$	20	(Ia)	0.4490×10^{-3}
	20	(IIa)	0.5181×10^{-18}
	20	(IIIa)	0.2772×10^{-48}
(c) $P(0, 0, 0.28, 0.72)$	20	(Ia)	0.4873×10^{-3}
	20	(IIa)	0.4211×10^{-18}
	20	(IIIa)	0.3203×10^{-48}
(d) $P(0, 0, 0.25, 0.75)$	20	(Ia)	0.5563×10^{-3}
	20	(IIa)	0.2880×10^{-18}
	20	(IIIa)	0.2756×10^{-48}
(e) $P(0, 0, 0.2, 0.8)$	20	(Ia)	0.7091×10^{-3}
	20	(IIa)	0.1226×10^{-18}
	20	(IIIa)	0.7630×10^{-49}

dimensional non-simplex subspace of the full simplex can be transformed to a $(q-1)$ cube by a linear transformation, the method given by Kiefer (1961) can be used to obtain D-optimum designs.

4.4. Comparability of the values of $|M(\xi_n)|$.

To enable comparison of $|M(\xi_n)|$ for different models with different numbers, k , of estimable parameters, to be made, several authors have calculated $|M(\xi_n)|^{1/p}$, taking $p = k$. For the parameterisation of models (I), (II) and (III) of Section 3.2, however, taking $p = 2, 5$ and 9 respectively do not yield comparable answers. Neither does the use of $p = 3, 9$ and 19 for models (Ia), (IIa) and (IIIa) of Section 4.3. By setting $p = 2, 8$ and 20 respectively for models (I), (II) and (III) and $p = 3, 15$ and 45 respectively for models (Ia), (IIa) and (IIIa), comparable values of $|M(\xi_n)|^{1/p}$ are obtained.

It seems that for a q -component mixture experiment with a m^{th} order polynomial model of the form of (2.2.2)

$$p = \sum_{r=1}^m r \binom{q-1}{r},$$

where $\binom{q-1}{r}$ is the number of estimable r^{th} order terms in the model.

Table 4.6 shows the values of k , p , $|M(\xi_n)|^{1/k}$, and $|M(\xi_n)|^{1/p}$ for the D-optimum designs for all the 3- and 4-component design space and model combinations of Sections 3.2 and 4.3.

For similar designs in similar design spaces and under the same mathematical model the value of $|M(\xi_n)|$ should be proportional to the squared volume of the design. Comparison of the $|M(\xi_n)|$ for

the linear model for similar designs for design spaces (i) and (ii) yields a ratio of

$$\begin{aligned} \left\{ |M(\xi_n)| \text{ for space (i)} / |M(\xi_n)| \text{ for space (ii)} \right\} &= 4.165 \\ &= \left\{ \text{area of space (i)} / \text{area of space (ii)} \right\}^4. \end{aligned}$$

For the quadratic model the ratio is

$$\begin{aligned} \left\{ |M(\xi_n)| \text{ for space (i)} / |M(\xi_n)| \text{ for space (ii)} \right\} &= 300.9 \\ &= (\text{ratio of areas})^{16}, \end{aligned}$$

and for the cubic model

$$\begin{aligned} \left\{ \text{ratio of the } |M(\xi_n)| \text{ values} \right\} &= .1579 \times 10^7 \\ &= (\text{ratio of the areas})^{40}. \end{aligned}$$

Therefore, the ratio of the $|M(\xi_n)|^{1/p}$ is equal to the squared ratio of the areas. The ratio of $\left\{ |M(\xi_n)|^{1/p} \text{ for space (i)} / |M(\xi_n)|^{1/p} \text{ for space (iii)} \right\}$ is equal to 1.99 for model (I), 5.2 for model (II) and 4.9 for model (III) whereas the squared ratio of the areas is 13.03. These differences occur because of the differences in the shape of the design space and consequently the nature of the D-optimum design.

TABLE 4.6. Values of $k, p, |M(\xi_n)|, |M(\xi_n)|^{1/k}, |M(\xi_n)|^{1/p}$ for the D-optimum designs of all the 3- and 4-component design space and model combinations of Sections 3.2 and 4.3.

Design Space	Model no.	k	p	$ M(\xi_n) $	$ M(\xi_n) ^{1/k}$	$ M(\xi_n) ^{1/p}$
(i) Full Simplex	I	2	2	0.037037	0.19245	0.19245
3-component	II	5	8	0.5233×10^{-8}	0.02207	0.092155
	III	9	20	0.4486×10^{-23}	0.002546	0.06802
(ii) 3-component, I	2	2	2	0.8893×10^{-2}	0.09430	0.09430
simplex	II	5	8	0.1739×10^{-10}	0.007048	0.04519
	III	9	20	0.2841×10^{-29}	0.0005213	0.03332
(iii) 3-component, I	2	2	2	0.9482×10^{-2}	0.09737	0.09737
quadrilateral	II	5	8	0.9474×10^{-14}	0.001568	0.01766
	III	9	20	0.6406×10^{-37}	0.7373×10^{-4}	0.01393
4-component,	Ia	3	3	0.3906×10^{-2}	0.1575	0.1575
full simplex	IIa	9	15	0.5960×10^{-17}	0.01218	0.07107
	IIIa	19	45	0.3203×10^{-48}	0.002803	0.08363

5. DESIGNS FOR FIXED NUMBERS OF OBSERVATIONS

5.1. Statement of the problem

In practice the investigator often wishes to design an experiment with a maximum value of $|M(\xi_n)|$ for a fixed number of observations, n say. The D-optimum design would most probably require $n^x \neq n$ observations. Here some methods of designing near D-optimum experiments for fixed numbers of observations are suggested.

Addition to a D-optimum design

If the desired experiment is of size n , where $n > \ell n^x$, where ℓ is an integer and n^x the size of the D-optimum design, a design with a maximum $|M(\xi_n)|$ can easily be obtained. Start with the D-optimum design, D_{n^x} , repeated ℓ times to yield a design $D_{\ell n^x}$ and then add observations, one at a time such that

$$f(x_{\ell n^x + i})' (X'_{\ell n^x + i} X_{\ell n^x + i})^{-1} f(x_{\ell n^x + i}),$$

where $x_{\ell n^x + i}$ is in $\mathcal{X} \setminus \{i = 1, \dots, (n - \ell n^x)\}$, is a maximum. This is essentially the method of Wynn (1970).

Sometimes the D-optimum design requires a large number of observations distributed in irregular proportions over the design points. If a design D_p , with $p < n$ observations and a non-singular $X'_p X_p$ matrix, can be found with proportions close to that of the D-optimum design, it would be advantageous to use the design D_p as a starting design and adding observations to it following the procedure of Wynn (1970) until a D_n design is reached. As will be shown in Section

5.3, if n should be less than the number of distinct design points in the D-optimum design, this method could lead to designs with less than the maximum value of $|M(\xi_n)|$.

Replacement of observations

If $n < n^x$, i.e. the D-optimum design requires more observations than available for experimentation, the following procedure could be followed to maximise $|M(\xi_n)|$.

Let X_n be the $k \times n$ matrix of $f(x_i)$, ($i = 1, \dots, n$), with rank $k \leq n$ corresponding to any arbitrary design D_n which consists of the observations x_i in $\mathcal{X}$ ($i = 1, \dots, n$). Then $|M(\xi_n)|$ is maximised by successively replacing observations in D_n with others in $\mathcal{X}$. An observation, x_i say, in D_n is replaced by another, x_j say, in $\mathcal{X}$ if

$$\Delta_{ij} = \left\{ 1 + f(x_j)' (X_n' X_n)^{-1} f(x_j) \right\} \left\{ 1 - f(x_i)' (X_n' X_n)^{-1} f(x_i) \right\} + \left\{ f(x_j)' (X_n' X_n)^{-1} f(x_i) \right\}^2 > 1. \quad (5.1.1)$$

This process is continued until no further $\Delta_{ij} > 1$ can be found. The following Lemma 1 shows why the Δ_{ij} should be taken larger than 1.

Lemma 1. If W is a symmetric positive definite $k \times k$ matrix and A is a $k \times 2$ matrix then

$$\det(W + AA') = \det(I + A'W^{-1}A) \det(W) \quad (5.1.2)$$

Proof: Wynn (1970) has proved that if a is a $k \times 1$ column vector then $\det(W + aa') = (1 + a'W^{-1}a) \det(W)$.

Let $A = (\underline{a}_1, \underline{a}_2)$; thus $AA' = \underline{a}_1 \underline{a}_1' + \underline{a}_2 \underline{a}_2'$.

Now

$$\begin{aligned} \det(W_o + \underline{a}_1 \underline{a}_1') &= (1 + \underline{a}_1' W_o^{-1} \underline{a}_1) \det(W_o) \\ &= \det(W_1), \end{aligned}$$

$$\begin{aligned} \det(W_1 + \underline{a}_2 \underline{a}_2') &= (1 + \underline{a}_2' W_1^{-1} \underline{a}_2) \det(W_1) \\ &= \left[1 + \underline{a}_2' \left\{ W_o^{-1} - W_o^{-1} \underline{a}_1 (1 + \underline{a}_1' W_o^{-1} \underline{a}_1)^{-1} \underline{a}_1' W_o^{-1} \right\} \underline{a}_2 \right] (1 + \underline{a}_1' W_o^{-1} \underline{a}_1) \det(W_o) \\ &= \left\{ (1 + \underline{a}_2' W_o^{-1} \underline{a}_2) - \frac{\underline{a}_2' W_o^{-1} \underline{a}_1 \underline{a}_1' W_o^{-1} \underline{a}_2}{(1 + \underline{a}_1' W_o^{-1} \underline{a}_1)} \right\} (1 + \underline{a}_1' W_o^{-1} \underline{a}_1) \det(W_o) \\ &= \left\{ (1 + \underline{a}_2' W_o^{-1} \underline{a}_2)(1 + \underline{a}_1' W_o^{-1} \underline{a}_1) - (\underline{a}_2' W_o^{-1} \underline{a}_1)^2 \right\} \det(W_o) \\ &= \det(I + A' W_o^{-1} A) \det(W_o). \end{aligned}$$

Therefore $\Delta_{ij} = \det \{ I + A' (X_n' X_n)^{-1} A \}$, if $\underline{a}_1 = f(\underline{x}_j)$ and

$\underline{a}_2 = \sqrt{1} f(\underline{x}_i)$, giving $\underline{a}_1 \underline{a}_1' = f(\underline{x}_j) f(\underline{x}_j)'$ and $\underline{a}_2 \underline{a}_2' = -f(\underline{x}_i) f(\underline{x}_i)'$.

Therefore, $\Delta_{ij} > 1$ implies that $|W + AA'| > |W|$ which means an increase in the value of $|M(\xi_n)|$ and hence in the relative D-optimality of the design.

Lemma 2. Given any design D_n , with corresponding matrix X_n of rank $r \leq k \leq n$, the average variance of the expected mean response at any design point $\underline{x}_i$ say in D_n is equal to r/n . In other words

$$\sum_{i=1}^n f(\underline{x}_i)' (X_n' X_n)^{-} f(\underline{x}_i) = r, \quad (5.1.3)$$

where $(X_n' X_n)^{-}$ is a generalised inverse of $X_n' X_n$.

Proof. The matrix $X_n (X_n' X_n)^{-} X_n'$ is idempotent and

$$\text{rank} \{X_n (X_n' X_n)^{-} X_n'\} = \text{tr} \{X_n (X_n' X_n)^{-} X_n'\}.$$

Also

$$\text{rank} \{X_n (X_n' X_n)^{-} X_n'\} = r.$$

Therefore,

$$\begin{aligned} \text{tr} \{X_n (X_n' X_n)^{-} X_n'\} &= \sum_{i=1}^n f(\underline{x}_i)' (X_n' X_n)^{-} f(\underline{x}_i) \\ &= r, \end{aligned}$$

for $\underline{x}_i$ in D_n .

For convenience of notation let $f(\underline{x}_i) = \underline{b}$, the column being deleted from X_n ; $f(\underline{x}_j) = \underline{a}$, the column being added to X_n and $X_n' X_n = W$.

Now

$$\Delta_{ij} = (1 + \underline{a}' W^{-1} \underline{a})(1 - \underline{b}' W^{-1} \underline{b}) + (\underline{a}' W^{-1} \underline{b})^2 > 1$$

if (i) generally the squared correlation between $\underline{a}$ and $\underline{b}$, namely

$$(\underline{a}' W^{-1} \underline{b})^2 / (\underline{a}' W^{-1} \underline{a})(\underline{b}' W^{-1} \underline{b}) > 1 + (\underline{a}' W^{-1} \underline{a})^{-1} - (\underline{b}' W^{-1} \underline{b})^{-1}; \quad (5.1.4)$$

and more specifically,

$$(ii) \quad \underline{a}' W^{-1} \underline{a} > (\underline{b}' W^{-1} \underline{b}) / (1 - \underline{b}' W^{-1} \underline{b}), \quad (5.1.5)$$

if $\underline{b}' W^{-1} \underline{b} < 1$; or

$$(iii) \quad (\underline{a}' W^{-1} \underline{b})^2 > 1, \quad (5.1.6)$$

if $\underline{b}' W^{-1} \underline{b} = 1$.

From Lemma 2 it can be deduced that for some $\underline{b}_i = f(\underline{x}_i)$ in X_n , $\underline{b}_i' W^{-1} \underline{b}_i \leq 1$, the inequality holding if $k < n$ and all the $\underline{b}_i' W^{-1} \underline{b}_i$ equal. The situation can arise when $k = n$ that all the $\underline{b}_i' W^{-1} \underline{b}_i = 1$ but the design is not D-optimum. In such a case some $f(\underline{x}_j)' W^{-1} f(\underline{x}_j) > 1$ can be found such that $\underline{x}_j$ is in $\mathcal{X}$ but not in D_n .

The situation could arise whereby there does not exist a

$\Delta_{ij} > 1$ although there does exist at least some $\tilde{x}_j$ in $\mathcal{X}$, and not in D_n and $\tilde{x}_i$ in D_n such that $f(\tilde{x}_j)' W^{-1} f(\tilde{x}_j) > f(\tilde{x}_i)' W^{-1} f(\tilde{x}_i)$. This means that a local maximum for $|M(\xi_n)|$ has been reached. This will most probably happen when $f(\tilde{x}_i)' W^{-1} f(\tilde{x}_i) = k/n$ for all $\tilde{x}_i$ in D_n . It might be possible to make progress by finding $\max_{\tilde{x}_j \text{ in } \mathcal{X}} f(\tilde{x}_j)' (X_n' X_n)^{-1} f(\tilde{x}_j)$, $\tilde{x}_j$ not in D_n ; and then $\max_{\tilde{x}_i \text{ in } D_n} f(\tilde{x}_i)' (X_n' X_n)^{-1} f(\tilde{x}_i)$ and replacing $\tilde{x}_i$

by $\tilde{x}_j$ provided that $\Delta_{ij} > 0$, i.e. $X_n' X_n$ does not become singular on replacing $\tilde{x}_i$ by $\tilde{x}_j$. It might even be necessary to do a few of these replacements involving different $\tilde{x}_i$ and $\tilde{x}_j$ combinations to get far enough away from this local optimum to be able to make progress in maximising $|M(\xi_n)|$.

5.2. Method of generating designs

The method followed consisted of generating an evenly spaced design D_{n_e} with $n_e \gg n$. The design was then reduced by successively deleting points from it until either

- (a) a design with the required number of observations remained, or
- (b) until a stage is reached whereby the deletion of any observation from the design D_{n_1} , $n_1 > n$, would result in a singular $X_n' X_n$ matrix. The observation deleted at each stage is that non-essential observation in the design with the smallest $\text{var} \{ \hat{\eta}(\tilde{x}) \}$. A non-essential observation in a design D_{n_1} , say, can be deleted from that design without the $X_{n_1-1}' X_{n_1-1}$ matrix becoming singular.

If stage (a) had been reached, the procedure of replacing observations could be directly followed.

If stage (b), however, had been reached one observation is replaced and the deleting process repeated. This two-step process is continued until either no further progress is possible in deleting points or increasing the $|M(\xi_n)|$ or stage (a) above had been reached, after which the normal replacement is followed.

The method of replacing observations consisted of a series of iterations. At each iteration one observation, $\underline{x}_i$ in D_n say, was replaced with another $\underline{x}_j$ in $\mathcal{X}$, if $\underline{x}_i$ is that observation with the smallest var $\{\hat{\eta}(\underline{x}_i)\}$ and with a $\Delta_{ij} > 1$. Normally this would be continued until no further $\Delta_{ij} > 1$, for any $i = 1, \dots, n$, could be found. To find whether a $\Delta_{ij} > 1$ existed for any specific $\underline{x}_i$ in D_n a series of Δ_{ij} was computed on the same grid as that described in Section 3.3, namely a grid with steps of 0.02 in each component.

5.3. Results and conclusions

The same design spaces and mathematical models as described in Section 3.2 were used. Where feasible, designs were computed for $n = k, \dots, 2k+1$ observations, k being the number of estimable parameters in the model. If an n could be found such that $k < n \leq 2k+1$ designs were generated for $n, \dots, 2k+1$ observations. The only instance in which no $n = k$ could be found was for the design space (iii) when the cubic model was fitted. The smallest design with a non-singular information matrix had 14 observations.

The computer routine used made provision for generating one

design with the smallest number of observations that it could find if $n \geq 2k+1$. This situation, and the one above where $n > k$, could arise through inadequacies, both of the parameterisation and the numerical methods used. Single precision arithmetic with 14 significant digits was used throughout. With very near singular matrices the rounding errors become important.

In design spaces where the D-optimum design for a model with k parameters requires a D_k design, one would expect that the methods of Section 5.1 would yield identical $|M(\xi_n)|$. This is then also what happened when applying the second method to design spaces (i) and (ii) of Section 3.2. These results are given in Table 5.1 to 5.3 for space (i) and Tables 5.4 to 5.6 for space (ii). Each table contains the D-optimum design with n observations and its $|M(\xi_n)|$ in the final column and opposite the n^{th} observation and then the observations to be added to get a design with $n+1, \dots, 2n+1$ observations in the successive rows and the value of $|M(\xi_{n+j})|$ next to the $(n+j)$ th observation ($j = 0, \dots, n+1$).

In the case of the 2-dimensional quadrilateral design space (iii) Section 4.2 shows that the D-optimum designs have more distinct design points than there are parameters in the models. The D-optimum design for fitting the linear model (I) has 4 design points whereas the model has only 3 estimable parameters. Similarly the D-optimum designs for fitting models (II) and (III) have 8 and 14 design places and 6 and 10 estimable parameters respectively. It is thus theoretically possible to get designs with fewer observations than there are design points in the D-optimum designs. No design with less than 14 observations and a non-singular $M(\xi_n)$ matrix could however be found for model (III).

TABLE 5.1. Designs for design space (i) under model (I) with a maximum value of $|M(\xi_n)|$ for $n = 3, \dots, 7$ observations

n	Design points	$ M(\xi_n) $
1	(0, 0, 1)	
2	(0, 1, 0)	
3	(1, 0, 0)	0.037037
4	(0, 0, 1)	0.03125
5	(0, 1, 0)	0.03200
6	(1, 0, 0)	0.037037
7	(0, 0, 1)	0.03499

TABLE 5.2. Designs for design space (i) under model (II) with a maximum value of $|M(\xi_n)|$ for $n = 6, \dots, 13$ observations

n	Design points	$ M(\xi_n) $
1	(0, 0, 1)	
2	(0, 1, 0)	
3	(1, 0, 0)	
4	(0, 0.5, 0.5)	
5	(0.5, 0, 0.5)	
6	(0.5, 0.5, 0)	0.5233×10^{-8}
7	(0.5, 0.5, 0)	0.4150×10^{-8}
8	(0, 0, 1)	0.3725×10^{-8}
9	(0, 0.5, 0.5)	0.3675×10^{-8}
10	(0, 1, 0)	0.3906×10^{-8}
11	(0.5, 0, 0.5)	0.4410×10^{-8}
12	(1, 0, 0)	0.5233×10^{-8}
13	(0, 0, 1)	0.4856×10^{-8}

TABLE 5.3. Designs for design space (i) under model (III) with a maximum value of $|M(\xi_n)|$ for $n = 10, \dots, 21$ observations

n	Design points	$ M(\xi_n) $
1	(0, 0, 1)	
2	(0, 0.28, 0.72)	
3	(0.28, 0, 0.72)	
4	(0.33, 0.33, 0.33)	
5	(0, 0.72, 0.28)	
6	(0.72, 0, 0.28)	
7	(0, 1, 0)	
8	(0.28, 0.72, 0)	
9	(0.72, 0.28, 0)	
10	(1, 0, 0)	0.4486×10^{-23}
11	(0.72, 0, 0.28)	0.3459×10^{-23}
12	(0.72, 0.28, 0)	0.2898×10^{-23}
13	(0, 0.72, 0.28)	0.2603×10^{-23}
14	(1, 0, 0)	0.2482×10^{-23}
15	(0.28, 0, 0.72)	0.2490×10^{-23}
16	(0, 0.28, 0.72)	0.2611×10^{-23}
17	(0, 0, 1)	0.2848×10^{-23}
18	(0.28, 0.72, 0)	0.3217×10^{-23}
19	(0, 1, 0)	0.3746×10^{-23}
20	(0.33, 0.33, 0.33)	0.4486×10^{-23}
21	(0, 0, 1)	0.4131×10^{-23}

TABLE 5.4. Designs for design space (ii) under model (I) with a maximum value of $|M(\xi_n)|$ for $n = 3, \dots, 7$ observations

n	Design points	$ M(\xi_n) $
1	(0.2, 0.1, 0.7)	
2	(0.2, 0.8, 0)	
3	(0.9, 0.1, 0)	0.8893×10^{-2}
4	(0.2, 0.8, 0)	0.7503×10^{-2}
5	(0.9, 0.1, 0)	0.7683×10^{-2}
6	(0.2, 0.1, 0.7)	0.8893×10^{-2}
7	(0.2, 0.1, 0.7)	0.8400×10^{-2}

TABLE 5.5. Designs for design space (ii) under model (I) with a maximum value of $|M(\xi_n)|$ for $n = 6, \dots, 13$ observations

n	Design points	$ M(\xi_n) $
1	(0.2, 0.1, 0.7)	
2	(0.2, 0.45, 0.35)	
3	(0.55, 0.1, 0.35)	
4	(0.2, 0.8, 0)	
5	(0.55, 0.45, 0)	
6	(0.9, 0.1, 0)	0.1739×10^{-10}
7	(0.2, 0.1, 0.7)	0.1379×10^{-10}
8	(0.55, 0.1, 0.35)	0.1238×10^{-10}
9	(0.55, 0.45, 0)	0.1221×10^{-10}
10	(0.2, 0.45, 0.35)	0.1298×10^{-10}
11	(0.9, 0.1, 0)	0.1466×10^{-10}
12	(0.2, 0.8, 0)	0.1739×10^{-10}
13	(0.2, 0.45, 0.35)	0.1614×10^{-10}

TABLE 5.6. Designs for design space (ii) under Model (III) with a maximum value of $|M(\xi_n)|$ for $n = 10, \dots, 21$ observations

n	Design points	$ M(\xi_n) $
1	(0.2, 0.1, 0.7)	
2	(0.2, 0.29, 0.51)	
3	(0.4, 0.1, 0.5)	
4	(0.43, 0.33, 0.23)	
5	(0.2, 0.6, 0.2)	
6	(0.71, 0.1, 0.19)	
7	(0.2, 0.8, 0)	
8	(0.4, 0.6, 0)	
9	(0.71, 0.29, 0)	
10	(0.9, 0.1, 0)	0.2841×10^{-29}
11	(0.43, 0.33, 0.23)	0.2190×10^{-29}
12	(0.71, 0.1, 0.19)	0.1836×10^{-29}
13	(0.71, 0.29, 0)	0.1656×10^{-29}
14	(0.2, 0.6, 0.2)	0.1574×10^{-29}
15	(0.9, 0.1, 0)	0.1576×10^{-29}
16	(0.4, 0.1, 0.5)	0.1653×10^{-29}
17	(0.2, 0.29, 0.51)	0.1803×10^{-29}
18	(0.2, 0.1, 0.7)	0.2036×10^{-29}
19	(0.4, 0.6, 0)	0.2372×10^{-29}
20	(0.2, 0.8, 0)	0.2841×10^{-29}
21	(0.2, 0.29, 0.51)	0.2615×10^{-29}

Tables 5.7 to 5.9 are similar to the above and show the results for design space (iii) for all three models. In the case of the cubic model (III) however designs and $|M(\xi_n)|$ are given only for $n = 14, \dots, 21$ observations. Contrary to expectation the design generated for $n = 7$, when fitting the quadratic model (II) to design space (iii) is not the design for $n = 6$ plus another appropriate observation. The reason for this is that the design with a maximum value for $|M(\xi_7)|$ has one design point at none of the 8 distinct design points of the D-optimum design.

There exist three designs with $n = 6$ with the maximum $|M(\xi_6)|$ and two designs for $n = 7$ with the maximum value for $|M(\xi_7)|$. The designs for $n = 6$ are subsets of the design with $n = 8$ observations. A comparison of the var $\{\hat{\eta}(\underline{x})\}$ for all the $\underline{x}_i$ in the 8-point design shows that the 5 design points common to all three of the 6-point designs have a relatively high var $\{\hat{\eta}(\underline{x})\}$, namely var $\{\hat{\eta}(\underline{x})\} = 0.8834$ at $(0.2, 0.6, 0.2)$ and $(0.7, 0.1, 0.2)$; var $\{\hat{\eta}(\underline{x})\} = 0.8820$ at $(0.2, 0.8, 0)$ and $(0.9, 0.1, 0)$; and var $\{\hat{\eta}(\underline{x})\} = 0.7663$ at $(0.55, 0.45, 0)$. The three points $(0.2, 0.7, 0.1)$, $(0.5, 0.4, 0.1)$ and $(0.8, 0.1, 0.1)$ have equal var $\{\hat{\eta}(\underline{x})\} = 0.5679$. Inclusion of any one of these latter three points in the 6 point design yields the same $|M(\xi_6)|$.

The two designs for $n = 7$ have the same 5 point subset as the 6 point designs plus one of the two points $(0.2, 0.7, 0.1)$ or $(0.8, 0.1, 0.1)$, plus an additional point near the centroid of the design space depending on which of the latter two points are included. For example, the point to be used in conjunction with $(0.8, 0.1, 0.1)$ is $(0.43, 0.47, 0.1)$.

TABLE 5.7. Designs for design space (iii) under model (I)
with a maximum value of $|M(\xi_n)|$ for $n = 3, \dots, 7$
observations

n	Design points	$ M(\xi_n) $
1	(0.2, 0.6, 0.2)	
2	(0.2, 0.8, 0)	
3	(0.9, 0.1, 0)	0.7259×10^{-3}
4	(0.7, 0.1, 0.2)	0.9250×10^{-3}
5	(0.2, 0.8, 0)	0.8672×10^{-3}
6	(0.9, 0.1, 0)	0.9111×10^{-3}
7	(0.2, 0.6, 0.2)	0.9190×10^{-3}

TABLE 5.8. Designs for design space (iii) under model (II) with a maximum value of $|M(\xi_n)|$ for $n = 6, \dots, 13$ observations

n	Design points	$ M(\xi_n) $
1	(0.2, 0.6, 0.2)	
2	(0.7, 0.1, 0.2)	
3	(0.2, 0.7, 0.1)	
4	(0.2, 0.8, 0)	
5	(0.55, 0.45, 0)	
6	(0.9, 0.1, 0)	0.6304×10^{-14}
1	(0.2, 0.6, 0.2)	
2	(0.7, 0.1, 0.2)	
3	(0.44, 0.46, 0.1)	
4	(0.8, 0.1, 0.1)	
5	(0.2, 0.8, 0)	
6	(0.56, 0.44, 0)	
7	(0.9, 0.1, 0)	0.7861×10^{-14}
1	(0.2, 0.6, 0.2)	
2	(0.7, 0.1, 0.2)	
3	(0.2, 0.7, 0.1)	
4	(0.5, 0.4, 0.1)	
5	(0.8, 0.1, 0.1)	
6	(0.2, 0.8, 0)	
7	(0.55, 0.45, 0)	
8	(0.9, 0.1, 0)	0.8001×10^{-14}
9	(0.2, 0.6, 0.2)	0.7443×10^{-14}
10	(0.2, 0.8, 0)	0.7476×10^{-14}
11	(0.7, 0.1, 0.2)	0.7927×10^{-14}
12	(0.90, 0.10, 0)	0.8767×10^{-14}
13	(0.55, 0.45, 0)	0.9443×10^{-14}

TABLE 5.9. Designs for design space (iii) under model (III) with
a maximum value of $|M(\xi_n)|$ for $n = 14, \dots, 21$
observations

n	Design points	$ M(\xi_n) $
1	(0.2, 0.6, 0.2)	
2	(0.45, 0.35, 0.2)	
3	(0.7, 0.1, 0.2)	
4	(0.43, 0.37, 0.16)	
5	(0.2, 0.66, 0.14)	
6	(0.76, 0.1, 0.14)	
7	0.2, 0.74, 0.06)	
8	(0.36, 0.58, 0.06)	
9	(0.68, 0.26, 0.06)	
10	(0.84, 0.1, 0.06)	
11	(0.2, 0.8, 0)	
12	(0.4, 0.6, 0)	
13	(0.7, 0.3, 0)	
14	(0.9, 0.1, 0)	0.5145×10^{-37}
15	(0.9, 0.1, 0)	0.4924×10^{-37}
16	(0.2, 0.8, 0)	0.4926×10^{-37}
17	(0.7, 0.1, 0.2)	0.5063×10^{-37}
18	(0.2, 0.6, 0.2)	0.5385×10^{-37}
19	(0.4, 0.6, 0)	0.5541×10^{-37}
20	(0.7, 0.3, 0)	0.5829×10^{-37}
21	(0.76, 0.1, 0.14)	0.5808×10^{-37}

Generally it seems that unique designs only exist in certain cases, namely experiments with (a) numbers of observations equal to or multiples of that in the D-optimum design; (b) numbers of observations equal to the number of distinct design points in the D-optimum design, as was seen for design space (iii); or (c) the design being a close approximation to the D-optimum in terms of proportions of observations to be taken at the distinct design points. An instance of case (c) is to be found in Section 4.2 where the D-optimum design for fitting a linear model (I) to space (iii) requires 146 observations in a ratio of 31:42. However, the design, consisting of 14 observations with a ratio of 3:4 has the maximum value of $|M(\xi_{14})|$ which is also relatively high when compared to that of the above D-optimum design. There is no other 14 point design for fitting model (I) to space (iii) with such a large $|M(\xi_{14})|$.

When the designs are not unique, in the sense of being the only possible design of that size having the maximum value for $|M(\xi_n)|$, the investigator will have to decide which design suits his purposes best. He should choose that design which will give the greatest accuracy in that region in the design space in which he is the most interested. For example the design with 11 observations for fitting model (III) to the full simplex, design space (i), will have the 10-point D-optimum design of Table 4.2 plus an additional observation at any of these 10 points. For example, if the prime interest lies in the centre of the design space the 11th observation should be taken there.

6. INCOMPLETE BLOCK MIXTURE DESIGNS

6.1. Introduction

Some method of designing incomplete block mixture experiments is very desirable because the number of observations in a mixture experiment rapidly increases with the number of components and the degree of the model. Nigam (1970) suggested a few methods of designing incomplete block mixture experiments under certain conditions. These methods are discussed in Section 2.5. His methods lead to experiments with only a few large blocks, which is often unsatisfactory when dealing with block sizes fixed within narrow limits by conditions outside the control of the investigator.

Let the true response $\eta(\underline{x})$ in an incomplete block mixture experiment be represented by

$$\eta(\underline{x}) = \underline{\beta}' f(\underline{x}_1) + \underline{\gamma}' g(\underline{x}_2),$$

where $\underline{x}' = (\underline{x}_1', \underline{x}_2')$, with $\underline{x}_1$ a $q \times 1$ column vector in $\mathcal{X}_1$, the mixture design space, and $\underline{x}_2$, a $b \times 1$ column vector in $\mathcal{X}_2$, the block design space; $\underline{\beta}$ is the $k \times 1$ column vector of estimable mixture parameters and $\underline{\gamma}$ the $(b-1) \times 1$ column vector of block effects. The basic problem lies in designing the experiment such that the block effects have a minimal influence on the mixture parameters.

Given an incomplete block mixture design D_n , with a corresponding matrix X_n , with b blocks, s observations per block, k estimable treatment parameters and n observations the $p \times p$ information matrix $M(\xi_n) = \frac{1}{n} (X_n' X_n)$ can be computed, where $p = k + (b-1)$, $n \geq p$. Suppose $M(\xi_n)$ is of full rank p . As in (3.1.4) the matrix $M(\xi_n)$ can be partitioned into

$$M(\xi_n) = \begin{bmatrix} M_{11}(\xi_n) & M_{12}(\xi_n) \\ M_{21}(\xi_n) & M_{22}(\xi_n) \end{bmatrix} ,$$

where $M_{11}(\xi_n)$ is the $k \times k$ information matrix of the mixture parameters; $M_{22}(\xi_n)$ is the $(b-1) \times (b-1)$ information matrix of the block effects; and $nM_{12}(\xi_n) = nM_{21}'(\xi_n)$ is the $k \times (b-1)$ matrix of sums of products between mixture parameters and block effects. As in (3.1.5) the inverse of $M(\xi_n)$ can be correspondingly partitioned into

$$M^{-1}(\xi_n) = \begin{bmatrix} M^{11}(\xi_n) & M^{21}(\xi_n) \\ M^{12}(\xi_n) & M^{22}(\xi_n) \end{bmatrix} .$$

If the blocking has no influence on the estimation of the parameters, $M_{12}(\xi_n)$ will be zero. This can only happen when the block size $s \geq k$, the number of parameters to be estimated. For $|M_{11}(\xi_n)|$ to be a maximum and $M_{12}(\xi_n) = 0$, however, $s = \ell n_d$, where $\ell = 1, 2, \dots$, and n_d is the size of the D-optimum design. This situation very seldom arises for usually $s < k$. In this latter case, when $s < k$, one would prefer to maximise

$$|M_{11}(\xi_n) - M_{12}(\xi_n) M_{22}^{-1}(\xi_n) M_{21}(\xi_n)| = |M^{11}(\xi_n)|^{-1}. \quad (6.1.2)$$

This determinant is given by Karlin and Studden (1966) as the analogue of $|M(\xi_n)|$ when considering the estimation of k ($k < p$) of the parameters $\theta_1, \dots, \theta_p$. Maximising (6.1.2) would thus mean that the effect of blocks on treatments is minimised. One would, of course, wish to retain the block structure of the experiment and would thus keep $M_{22}(\xi_n)$ constant, only varying the mixtures applied within the blocks.

It is also known that

$$|M(\xi_n)| = |M_{22}(\xi_n)| \cdot |M_{11}(\xi_n) - M_{12}(\xi_n)M_{22}^{-1}(\xi_n)M_{21}(\xi_n)|,$$

and thus

$$|M^{11}(\xi_n)|^{-1} = |M(\xi_n)| |M_{22}(\xi_n)|^{-1}. \quad (6.1.3)$$

Since the number of observations, n , and the number of blocks, b , in the experiment is fixed beforehand the matrix $M_{22}(\xi_n)$ will remain constant. It is thus only necessary to maximise $|M(\xi_n)|$ for fixed n , while retaining the block structure. The second method described in Section 5.1 can thus be used to maximise (6.1.2).

The advantage here is that one is not limited to a specific set of mixtures which has to be subdivided into the blocks as in the usual incomplete block experiments but can vary the mixtures at will. Any possible mixture within the design space is a candidate for inclusion in the experiment. As will be shown in Section 6.3, it is possible to get higher values of $|M_{11}(\xi_n) - M_{12}(\xi_n)M_{22}^{-1}(\xi_n)M_{21}(\xi_n)|$ than with the usual methods.

In the incomplete block mixture experiments there will not necessarily be replications in the usual sense and the error will not be measured as a replication effect but rather as deviations from regression.

Because the maximising of (6.1.2) is lengthy it would be advantageous if some simple methods for designing incomplete block mixture experiments could be devised. One method is to generate incomplete block mixture experiments with a maximum value for $|M(\xi_n)|$ for a selection of different mathematical models; numbers of observations, n ; and numbers of blocks, b , for the 3-component

full simplex design space to ascertain whether any general results emerge.

In cases where the D-optimum design has a small fixed number of design points, n_d say, only block designs with blocks of size $s < n_d$ observations per block were considered. If the block sizes $s = n_d + \ell$, $0 < \ell < n_d$, the method of Section 5.2 will yield a design with $n_d + \ell b$ observations. The first n_d points being the D-optimum design and will be common to all b of the blocks. The first block will have in addition the first ℓ of the additional ℓb observations; the second block, the second ℓ ; and so forth. Although this will not necessarily yield a design with a maximum value for $|M(\xi_n)|$, it will not differ much from the design with the maximum value of $|M(\xi_n)|$. An alternative method is to generate incomplete block designs with blocks of size ℓ and to use these to augment the D-optimum design of size n_d to yield blocks of size $s = n_d + \ell$. This latter method will, however, most probably result in designs with smaller values of $|M(\xi_n)|$ than the preceding method.

6.2. Method of generating designs

The method used was similar to that described in Section 5.2.

The changes are that

(i) a design with a fixed block structure was used as a starting design and no observations were deleted, n , b and k being fixed beforehand, and

(ii) when $f(x_i)' = f(x_{i1}, x_{i2})'$ in X_n is replaced by $f(x_j)' = f(x_{j1}, x_{j2})'$ in $\mathcal{X}$, x_{i2} must be equal to x_{j2} . In other words, if the X_n matrix is

partitioned as $X_n = (X_{n1}, X_{n2})$, where X_{n1} is the $n \times k$ matrix of treatment variables $f(x_{1i})$ ($i = 1, \dots, n$) and X_{n2} is the $n \times (b-1)$ matrix of block variables $f(x_{2i})$ ($i = 1, \dots, n$), the X_{n2} matrix is specified beforehand and does not change whereas the rows of X_{n1} are changed by the replacement of mixtures. An observation, x_i say in D_n is replaced with another, x_j say in $\mathcal{X}$ if x_i is that observation with the smallest $\text{var} \{ \hat{\eta}(x_i) \}$, x_i in D_n which has a $\Delta_{ij} > 1$, where Δ_{ij} is as defined in (5.1.1).

The situation of reaching a local maximum for $|M(\xi_n)|$ never actually arose in the examples examined in Section 5.3. Under the present conditions, however, when maximising $|M(\xi_n)|$ keeping $|M_{22}(\xi_n)|$ constant, local maxima were reached. This factor becomes very important in the more complex cases where quadratic and cubic models are assumed. It was considered that a design with a maximum $|M(\xi_n)|$ had been reached if the increase in $|M(\xi_n)|$ in two successive steps was less than $0.001|M(\xi_n)|$. A replacement as described in Section 5.2 is, however, virtually impossible. In Section 5.2 only the $\text{var} \{ \hat{\eta}(x_i) \}$, $\text{var} \{ \hat{\eta}(x_j) \}$ and $f(x_j)' (X_n' X_n)^{-1} f(x_i)$ functions, where x_i is in D_n and x_j is in $\mathcal{X}$ but not in D_n , had to be considered. In this case, however, all the possible interactions between blocks and treatments will have to be brought into consideration, possibly resulting in some extremely complex computations. In an effort to overcome this, different grid sizes in steps of 0.025, 0.02, 0.0125 and 0.01 in the components were used. This device had limited success. Different starting designs were also used, a second design usually being the end design, obtained from the first, with some subjective changes. This method

proved to be more effective but time consuming. As will be seen, in spite of all this, there are some cases in which no reasonably balanced design has as yet been reached. There also exist some cases where a slightly less efficient design, in terms of $|M^{11}(\xi_n)|^{-1}$, contains more homogenous and intuitively more acceptable mixture proportions than the design with maximum $|M^{11}(\xi_n)|^{-1}$.

The efficiency of an incomplete block mixture design may be expressed relative to the determinant that would have been achieved using the same mixtures but in a complete block experiment, i. e.

$$E = |M^{11}(\xi_n)|^{-1} / |M_{11}(\xi_n)| \quad (6.2.1)$$

An alternative measure of efficiency is to measure $|M^{11}(\xi_n)|^{-1}$ relative to the maximum $|M_{11}(\xi_n)|$ possible for that n . This would give the minimum efficiency of that incomplete block mixture design and can be formulated as

$$E_{\min} = |M^{11}(\xi_n)|^{-1} / \max_{x_1 \in \mathcal{X}_1} |M_{11}(\xi_n)|$$

6.3. Results

For the purposes of this investigation only the 3-component full simplex space was considered. The mathematical models were the same as those of Section 3.2, namely the linear model (I), the quadratic model (II) and the cubic model (III).

The sizes of the designs were constrained to $n \leq 24$ and the block sizes to $s = 2$ for model (I) and $2 \leq s \leq 5$ for models (II) and (III). Designs were generated for all integer values of $b = n/s$ such that $n \geq p$, $p = k + (b-1)$.

The linear model (I)

Because the D-optimum design for the full simplex with model (I) has only $n_d = 3$ distinct design points only incomplete block mixture designs with $s = 2$ observations per block were considered. The method described in Section 6.1 will yield designs with $s = n_d + \ell$ ($\ell = 0, 1, \dots$).

The basic design for $s = 2$ contains three blocks with two mixtures in each, namely

- (i) $(0, 0, 1)$ and $(0, 1, 0)$;
- (ii) $(0, 0, 1)$ and $(1, 0, 0)$; and
- (iii) $(0, 1, 0)$ and $(1, 0, 0)$.

An experiment with any number of blocks can be built up by repetitions of these basic three blocks plus any subset of it to make up the required number of blocks.

Table 6.1 shows the incomplete block designs for fitting model (I) to the full simplex design space for $b = 2, \dots, 12$ blocks with $s = 2$. In this case, the $(b+1)$ block design consists of a b block design plus an additional block. If $b = 2$ for example, use the upper 2 blocks; for $b = 3$, the upper 3; and so forth for any experiment with up to 12 blocks. The values of $|M^{11}(\xi_n)|^{-1}$ and $E = \left\{ |M^{11}(\xi_n)| |M_{11}(\xi_n)| \right\}^{-1}$ for the b block design ($b = n/s$) are given next to the b th block in Table 6.1.

As is to be expected, all designs with a multiple of 3 blocks have the maximum values of $|M^{11}(\xi_n)|^{-1} = 0.02083$ and $E = 0.5624$ for $s = 2$, being balanced incomplete block designs utilising the D-optimum design.

This basic design can be extended to higher dimensions. Thus,

TABLE 6.1. The values of $|M^{11}(\xi_n)|^{-1}$ and E for the incomplete block mixture designs for fitting the linear model (I) to the full simplex for $b = 2, \dots, 12$ blocks with 2 observations per block.

b	n	Design Points	$ M^{11}(\xi_n) ^{-1}$	E
1	1	(0, 0, 1)		
	2	(0, 1, 0)		
2	3	(0, 0, 1)	0.01563	0.5000
	4	(1, 0, 0)		
3	5	(0, 1, 0)	0.02083	0.5624
	6	(1, 0, 0)		
4	7	(0, 0, 1)	0.01953	0.5555
	8	(0, 1, 0)		
5	9	(0, 0, 1)	0.02000	0.5556
	10	(1, 0, 0)		
6	11	(0, 1, 0)	0.02083	0.5624
	12	(1, 0, 0)		
7	13	(0, 0, 1)	0.02041	0.5600
	14	(0, 1, 0)		
8	15	(0, 0, 1)	0.02051	0.5600
	16	(1, 0, 0)		
9	17	(0, 1, 0)	0.02083	0.5624
	18	(1, 0, 0)		
10	19	(0, 0, 1)	0.02063	0.5612
	20	(0, 1, 0)		
11	21	(0, 0, 1)	0.02066	0.5612
	22	(1, 0, 0)		
12	23	(0, 1, 0)	0.02083	0.5624
	24	(1, 0, 0)		

for $q > 3$ and $s < q$, the basic design will consists of $\binom{q}{s}$ blocks. The method for constructing a design with the number of blocks not equal to a multiple of $\binom{q}{s}$ would be to use repetitions of the basic set plus a subset of it to make up the required number of blocks. This subset must be chosen such that the number of times the q different mixtures appear in the subset are as equal as possible. For example for $q = 4$, $s = 2$ and $b = 8$, one would use the basic 6 blocks plus possibly $\{(0, 0, 0, 1), (0, 0, 1, 0)\}$ and $\{(0, 1, 0, 0), (1, 0, 0, 0)\}$ as the two remaining blocks.

The quadratic model (II)

In this case only incomplete block mixture designs with fewer than 6 observations per block were considered because the D-optimum design has only 6 distinct design points. If $s > 6$ the method described in Section 6.1 can be used.

If a large number of blocks are available one method of designing an incomplete block mixture experiment is to use the 6 design points of the D-optimum design as 6 treatments and use the usual incomplete block designs. With a few possible exceptions, these designs will, unfortunately, not have a maximum value of $|M^{11}(\xi_n)|^{-1}$. For example the design generated by the method described in Section 6.2, with $n = 12$, $b = 4$ and $s = 3$ has a $|M^{11}(\xi_n)|^{-1} = 0.1618 \times 10^{-8}$, whereas the partially balanced incomplete block design of the same dimensions has $|M^{11}(\xi_n)|^{-1} = 0.1550 \times 10^{-8}$, an efficiency of about 96% with respect to the generated design. Similarly a partially balanced incomplete block design, utilising the D-optimum design, with $n = 18$, $b = 6$ and $s = 3$, has $|M^{11}(\xi_n)|^{-1} = 0.2943 \times 10^{-8}$, which is about 95% of

that of the generated design. With the development of cyclic incomplete block designs with unequal replication the use of D-optimum designs should become even more useful. This is especially in the more complex cases where the generation of an optimum design by the method of Section 6.2 would be very lengthy. In Table 6.2, the values of $|M(\xi_n)|$, $|M^{11}(\xi_n)|^{-1}$ and E are given for a number of balanced, partially balanced and cyclic incomplete block designs using the 6 design points, given in Table 4.2 for the D-optimum design for the second order model, as 'treatments'. The plans for the balanced incomplete blocks are from Cochran and Cox (1957), their plan numbers being used. The plans for the partially balanced incomplete block designs are from Bose, Clatworthy and Shrikhande (1954), the plan numbers used by Bose et al (1954) being used. The cyclic incomplete block designs are a selection of those given by John (1966). Here, in lieu of plan numbers, the generating sets are given.

In Table 6.3, the values of $|M(\xi_n)|$, $|M^{11}(\xi_n)|^{-1}$ and E and the table in which that design appears are given for all the combinations of sizes, n, of experiments, block numbers b and block sizes s, used to generate incomplete incomplete block mixture designs for the quadratic model (II). It will be noted that for some combinations more than one set of values are given. These are the cases where the designs with the largest values of $|M^{11}(\xi_n)|^{-1}$ contain mixtures which are less homogenous than those of designs with slightly smaller $|M^{11}(\xi_n)|^{-1}$, the latter containing some permutations of only a few basic mixtures. For example the design in Table 6.14(a) with non-vertex observations

TABLE 6.2. Values of $|M(\xi_n)|$, $|M^{11}(\xi_n)|^{-1}$ and E for the balanced (BIB), partially balanced (PBIB) and cyclic (CIB) incomplete block designs utilising the D-optimum design for the quadratic model (II), for different numbers, n of observations and b of blocks and the corresponding block sizes s. Here $|M_{11}(\xi_n)| = 0.5233 \times 10^{-8}$ throughout.

Plan Type and Number	n	b	s	$ M(\xi_n) $	$ M^{11}(\xi_n) ^{-1}$	E
BIB 11.3	30	15	2	0.2091×10^{-24}	0.4069×10^{-9}	0.0778
11.4	30	10	3	0.1715×10^{-16}	0.1715×10^{-8}	0.3277
11.5	60	20	3	0.6541×10^{-32}	0.1715×10^{-8}	0.3277
11.6	60	15	4	0.1588×10^{-23}	0.3090×10^{-8}	0.5905
π	30	6	5	0.3292×10^{-11}	0.4267×10^{-8}	0.8154
PBIB S1	12	3	4	0.9811×10^{-9}	0.2943×10^{-8}	0.5625
R1	18	6	3	0.1260×10^{-11}	0.1633×10^{-8}	0.3121
R2	24	6	4	0.2339×10^{-11}	0.3032×10^{-8}	0.5794
R3	36	12	3	0.2717×10^{-19}	0.1682×10^{-8}	0.3215
R4	48	12	4	0.4976×10^{-19}	0.3081×10^{-8}	0.5888
SR1	12	4	3	0.9690×10^{-10}	0.1550×10^{-8}	0.2963
SR4	30	9	4	0.6413×10^{-15}	0.3067×10^{-8}	0.5862
CIB(0, 1, 2)	18	6	3	0.1108×10^{-11}	0.1436×10^{-8}	0.2743
(0, 1, 3)	18	6	3	0.1260×10^{-11}	0.1633×10^{-8}	0.3121
(0, 1, 2)(0, 2, 4)	24	8	3	0.5915×10^{-14}	0.1550×10^{-8}	0.2963
(0, 1, 3)(0, 2, 4)	24	8	3	0.6290×10^{-14}	0.1649×10^{-8}	0.3151
(0, 1, 3)(0, 2, 4)(0, 2, 4)	30	10	3	0.1556×10^{-16}	0.1556×10^{-8}	0.2973
(0, 1, 2)(0, 1, 3)	36	12	3	0.2721×10^{-19}	0.1685×10^{-8}	0.3220
(0, 1, 2, 3)	24	6	4	0.2343×10^{-11}	0.3036×10^{-8}	0.5802
(0, 1, 2, 4)	24	6	4	0.2339×10^{-11}	0.3031×10^{-8}	0.5794
(0, 1, 2, 4)(0, 1, 3, 4)	36	9	4	0.6411×10^{-15}	0.3066×10^{-8}	0.5860
(0, 1, 2, 3)(0, 1, 3, 4)	36	9	4	0.6413×10^{-15}	0.3067×10^{-8}	0.5862

TABLE 6.3. Values of $|M(\xi_n)|$, $|M^{11}(\xi_n)|^{-1}$ and E and the number of the table in which the design is given for the incomplete block mixture designs generated for the different block sizes s, numbers, b of blocks and n of observations for fitting the quadratic model (II) to the full simplex.

s	b	n	$ M(\xi_n) $	$ M^{11}(\xi_n) ^{-1}$	E	Table No.
2	5	10	0.1948×10^{-11}	0.2435×10^{-9}	0.1281	6.4
	6	12	0.2353×10^{-12}	0.3049×10^{-9}	0.0783	6.5
	7	14	0.2017×10^{-13}	0.3391×10^{-9}	0.0921	6.6
	8	16	0.1462×10^{-14}	0.3833×10^{-9}	0.1146	6.7
	9	18	0.8469×10^{-16}	0.4051×10^{-9}	0.0874	6.8
	10	20	0.4115×10^{-17}	0.4115×10^{-9}	0.0941	6.9
	11	22	0.1784×10^{-18}	0.4207×10^{-9}	0.1011	6.10
	12	24	0.6741×10^{-20}	0.4174×10^{-9}	0.1054	6.11
3	3	9	0.3743×10^{-9}	0.1123×10^{-8}	0.3516	6.12
	4	12	0.1040×10^{-9}	0.1665×10^{-8}	0.3329	6.13a
	4	12	0.1046×10^{-9}	0.1674×10^{-8}	0.3405	6.13b
	5	15	0.1235×10^{-10}	0.1543×10^{-8}	0.3483	6.14a
	5	15	0.1276×10^{-10}	0.1595×10^{-8}	0.3779	6.14b
	6	18	0.1333×10^{-11}	0.1728×10^{-8}	0.3403	6.15
	7	21	0.1055×10^{-12}	0.1759×10^{-8}	0.4031	6.16
	8	24	0.6573×10^{-14}	0.1723×10^{-8}	0.3409	6.17
4	2	8	0.2330×10^{-8}	0.2330×10^{-8}	0.6951	6.18
	3	12	0.1028×10^{-8}	0.3084×10^{-8}	0.6168	6.19
	4	16	0.1873×10^{-9}	0.2868×10^{-8}	0.6452	6.20
	5	20	0.2388×10^{-10}	0.2985×10^{-8}	0.6484	6.21
	6	24	0.2380×10^{-11}	0.3084×10^{-8}	0.6169	6.22
5	2	10	0.3322×10^{-8}	0.3322×10^{-8}	0.8877	6.23
	3	15	0.1299×10^{-8}	0.3898×10^{-8}	0.8633	6.24
	4	20	0.2558×10^{-9}	0.4093×10^{-8}	0.8474	6.25

at all the permutations of (0, 0.45, 0.55) has a $|M^{11}(\xi_n)|^{-1} = 0.1543 \times 10^{-8}$ whereas the design in Table 6.14(b) with non-vertex points at (0.54, 0, 0.46), (0.46, 0, 0.54), (0, 0.56, 0.44), (0, 0.56, 0.44), (0.56, 0.44, 0), (0.58, 0.42, 0), (0, 0.4, 0.6) and (0.36, 0.64, 0) has a larger value of $|M^{11}(\xi_n)|^{-1} = 0.1595 \times 10^{-8}$. Comparison of the tables containing the alternative designs should make the differences, usually small, clear. Tables 6.4 to 6.25 contain the designs generated for all the viable combinations of $n \leq 24$ and $2 \leq s \leq 5$.

Except for the design in Table 6.4 with $n = 10$, $s = 2$ and $b = 5$, all the designs have design points only on the boundary of the design space. In other words, at least one of the components is absent from any mixture.

When $s = 2$ virtually all the non-vertex mixtures are either of the form (0, 0.5, 0.5) or lie between (0, 0.32, 0.68) and (0, 0.4, 0.6). Many of the blocks consist of a point at a vertex, (0, 0, 1) for example, and a point on one of the two bounding lines subtending that vertex of the design space, namely (0, 0.625, 0.375) for block 1 and (0.625, 0, 0.375) for block 2 of the design of Table 6.5 with $s = 2$, $b = 6$ and $n = 12$. This design with $n = 12$, $b = 6$ is thus balanced for these design points as can be seen in Table 6.5.

When $s > 2$ the non-vertex mixtures tend to lie between permutations of (0, 0.4, 0.6) and (0, 0.5, 0.5) with (0, 0.45, 0.55) predominating.

The increase in efficiency, measured in terms of the values

TABLE 6.4. Incomplete block

design for model (II) with

$s = 2$, $b = 5$ and $n = 10$.

TABLE 6.5. Incomplete block

design for model (II) with $s = 2$,

$b = 6$ and $n = 12$.

Block no.	Design points	Block no.	Design points
1	(1, 0, 0) (0.32, 0, 0.68)	1	(0, 0, 1) (0, 0.625, 0.375)
2	(0, 1, 0) (0, 0.32, 0.68)	2	(0, 0, 1) (0.625, 0, 0.375)
3	(0, 0, 1) (0.38, 0.38, 0.24)	3	(0, 1, 0) (0, 0.375, 0.625)
4	(0.66, 0, 0.34) (0.34, 0.66, 0)	4	(0, 1, 0) (0.625, 0.375, 0)
5	(0.66, 0.34, 0) (0, 0.66, 0.34)	5	(1, 0, 0) (0.375, 0, 0.625)
		6	(1, 0, 0) (0.375, 0.625, 0)

TABLE 6.6. Incomplete block

design for model (II) with
 $s = 2$, $b = 7$ and $n = 14$.

TABLE 6.7. Incomplete block

design for model (II) with $s = 2$,
 $b = 8$ and $n = 16$.

Block no.	Design points	Block no.	Design points
1	(0, 0, 1) (0, 0.66, 0.34)	1	(0, 0, 1) (0, 0.65, 0.35)
2	(0, 0, 1) (0.6, 0, 0.4)	2	(0, 0, 1) (0.65, 0, 0.35)
3	(0, 1, 0) (0, 0.36, 0.64)	3	(0, 1, 0) (0, 0.35, 0.65)
4	(0, 1, 0) (0.64, 0.36, 0)	4	(0, 1, 0) (0.65, 0.35, 0)
5	(1, 0, 0) (0.38, 0, 0.62)	5	(1, 0, 0) (0.35, 0, 0.65)
6	(1, 0, 0) (0.34, 0.66, 0)	6	(1, 0, 0) (0.35, 0.65, 0)
7	(0, 0.46, 0.54) (0.54, 0.46, 0)	7	(0, 0.5, 0.5) (0.6, 0, 0.4)
		8	(0.5, 0.5, 0) (0.4, 0, 0.6)

TABLE 6.8 . Incomplete block

design for model (II) with

$s = 2$, $b = 9$ and $n = 18$.

TABLE 6.9. Incomplete block

design for model (II) with $s = 2$,

$b = 10$ and $n = 20$.

Block no.	Design points	Block no.	Design points
1	(0, 0, 1) (0, 0.6, 0.4)	1	(0, 0, 1) (0, 0.6, 0.4)
2	(0, 0, 1) (0.6, 0, 0.4)	2	(0, 0, 1) (0.625, 0, 0.375)
3	(0, 1, 0) (0, 0.4, 0.6)	3	(0, 1, 0) (0, 0.4, 0.6)
4	(0, 1, 0) (0.6, 0.4, 0)	4	(0, 1, 0) (0.625, 0.375, 0)
5	(1, 0, 0) (0.4, 0, 0.6)	5	(1, 0, 0) (0.375, 0, 0.625)
6	(1, 0, 0) (0.4, 0.6, 0)	6	(1, 0, 0) (0.375, 0.625, 0)
7	(0, 0, 1) (0.5, 0.5, 0)	7	(0, 0, 1) (0.5, 0.5, 0)
8	(0, 1, 0) (0.5, 0, 0.5)	8	(0, 1, 0) (0.5, 0, 0.5)
9	(1, 0, 0) (0.5, 0.5, 0)	9	(1, 0, 0) (0, 0.5, 0.5)
		10	(0.5, 0, 0.5) (0.5, 0.5, 0)

TABLE 6.10. Incomplete block

design for model (II) with
 $s = 2$, $b = 11$ and $n = 22$.

Block no.	Design points
1	(0, 0, 1) (0, 0.625, 0.375)
2	(0, 0, 1) (0.625, 0, 0.375)
3	(0, 1, 0) (0, 0.375, 0.625)
4	(0, 1, 0) (0.625, 0.375, 0)
5	(1, 0, 0) (0.375, 0, 0.625)
6	(1, 0, 0) (0.375, 0.625, 0)
7	(0, 0, 1) (0.5, 0.5, 0)
8	(0, 1, 0) (0.5, 0, 0.5)
9	(1, 0, 0) (0, 0.5, 0.5)
10	(0, 0.5, 0.5) (0.5, 0.5, 0)
11	(0.5, 0, 0.5) (0.5, 0.5, 0)

TABLE 6.11. Incomplete block

design for model (II) with $s = 2$,
 $b = 12$ and $n = 24$.

Block no.	Design points
1	(0, 0, 1) (0, 0.625, 0.375)
2	(0, 0, 1) (0.625, 0, 0.375)
3	(0, 1, 0) (0, 0.375, 0.625)
4	(0, 1, 0) (0.625, 0.375, 0)
5	(1, 0, 0) (0.375, 0, 0.625)
6	(1, 0, 0) (0.375, 0.625, 0)
7	(0, 0, 1) (0.5, 0.5, 0)
8	(0, 1, 0) (0.5, 0, 0.5)
9	(1, 0, 0) (0, 0.5, 0.5)
10	(0, 0.5, 0.5) (0.5, 0, 0.5)
11	(0, 0.5, 0.5) (0.5, 0.5, 0)
12	(0.5, 0, 0.5) (0.5, 0.5, 0)

TABLE 6.12. Incomplete block design for model (II) with $s = 3$, $b = 3$ and $n = 9$.

Block no.	Design points
1	(0, 0, 1) (0, 0.575, 0.425) (0.375, 0.625, 0)
2	(0, 1, 0) (0.575, 0.425, 0) (0.625, 0, 0.375)
3	(1, 0, 0) (0.425, 0, 0.575) (0, 0.375, 0.625)

TABLE 6.13a. Incomplete block design for model (II) with $s = 3$, $b = 4$ and $n = 12$.

TABLE 6.13b. Incomplete block design for model (II) with $s = 3$, $b = 4$ and $n = 12$.

Block no.	Design points	Block no.	Design points
1	(0, 0, 1) (0, 1, 0) (0.55, 0, 0.45)	1	(0, 0, 1) (0, 1, 0) (0.56, 0, 0.44)
2	(0, 1, 0) (1, 0, 0) (0, 0.45, 0.55)	2	(0, 1, 0) (1, 0, 0) (0, 0.44, 0.56)
3	(0, 0, 1) (0.55, 0.45, 0) (0, 0.55, 0.45)	3	(0, 0, 1) (0.54, 0.46, 0) (0, 0.58, 0.42)
4	(1, 0, 0) (0.45, 0.55, 0) (0.45, 0, 0.55)	4	(1, 0, 0) (0.46, 0.54, 0) (0.44, 0, 0.56)

TABLE 6.14a. Incomplete block

design for model (II) with
 $s = 3$, $b = 5$ and $n = 15$.

TABLE 6.14b. Incomplete block

design for model (II) with $s = 3$,
 $b = 5$ and $n = 15$.

Block no.	Design points	Block no.	Design points
1	(0, 0, 1)	1	(0, 0, 1)
	(0, 0.55, 0.45)		(0.54, 0, 0.46)
	(0.55, 0, 0.45)		(0, 0.56, 0.44)
2	(0, 1, 0)	2	(0, 1, 0)
	(0, 0.45, 0.55)		(0, 0.4, 0.6)
	(0.55, 0.45, 0)		(0.56, 0.44, 0)
3	(1, 0, 0)	3	(1, 0, 0)
	(0.45, 0, 0.55)		(0.46, 0, 0.54)
	(0.45, 0.55, 0)		(0.36, 0.64, 0)
4	(0, 0, 1)	4	(0, 0, 1)
	(0, 1, 0)		(0, 1, 0)
	(0.55, 0.45, 0)		(0.58, 0.42, 0)
5	(0, 0, 1)	5	(0, 0, 1)
	(1, 0, 0)		(1, 0, 0)
	(0, 0.55, 0.45)		(0, 0.56, 0.44)

TABLE 6.15. Incomplete block

design for model (II) with

$s = 3$, $b = 6$ and $n = 18$.

TABLE 6.16. Incomplete block

design for model (II) with $s = 3$,

$b = 7$ and $n = 21$.

Block no.	Design points	Block no.	Design points
1	(0, 0, 1) (0, 0.55, 0.45) (0.55, 0, 0.45)	1	(0, 0, 1) (0, 0.575, 0.425) (0.6, 0, 0.4)
2	(0, 1, 0) (0, 0.45, 0.55) (0.55, 0.45, 0)	2	(0, 1, 0) (0, 0.425, 0.575) (0.6, 0.4, 0)
3	(1, 0, 0) (0.45, 0, 0.55) (0.45, 0.55, 0)	3	(1, 0, 0) (0.425, 0, 0.575) (0.4, 0.6, 0)
4	(0, 0, 1) (0, 1, 0) (0.5, 0, 0.5)	4	(0, 1, 0) (0.55, 0, 0.45) (0, 0.4, 0.6)
5	(0, 0, 1) (1, 0, 0) (0.5, 0.5, 0)	5	(1, 0, 0) (0.45, 0, 0.55) (0.4, 0.6, 0)
6	(0, 1, 0) (1, 0, 0) (0, 0.5, 0.5)	6	(0, 0, 1) (0, 1, 0) (0.575, 0.425, 0)
		7	(0, 0, 1) (1, 0, 0) (0, 0.6, 0.4)

TABLE 6.17. Incomplete block

design for model (II) with
 $s = 3$, $b = 8$ and $n = 24$.

TABLE 6.18. Incomplete block

design for model (II) with $s = 4$,
 $b = 2$ and $n = 8$.

Block no.	Design points	Block no.	Design points
1	(0, 0, 1) (0, 0.55, 0.45) (0.55, 0, 0.45)	1	(0, 0, 1) (1, 0, 0) (0, 0.6, 0.4)
2	(0, 1, 0) (0, 0.45, 0.55) (0.55, 0.45, 0)	2	(0.4, 0.6, 0) (0, 1, 0) (0.5, 0, 0.5)
3	(1, 0, 0) (0.45, 0, 0.55) (0.45, 0.55, 0)		(0.62, 0.38, 0) (0, 0.38, 0.62)
4	(0, 0, 1) (0, 1, 0) (0.55, 0, 0.45)		
5	(0, 0, 1) (1, 0, 0) (0.45, 0.55, 0)		
6	(0, 1, 0) (1, 0, 0) (0, 0.45, 0.55)		
7	(0, 0, 1) (0, 1, 0) (1, 0, 0)		
8	(0, 0.5, 0.5) (0.5, 0, 0.5) (0.5, 0.5, 0)		

TABLE 6.19. Incomplete block

design for model (II) with

$s = 4$, $b = 3$ and $n = 12$.

Block no.	Design points
1	(0, 0, 1) (0, 1, 0) (0.55, 0, 0.45) (0.55, 0.45, 0)
2	(0, 0, 1) (1, 0, 0) (0, 0.55, 0.45) (0.45, 0.55, 0)
3	(0, 1, 0) (1, 0, 0) (0, 0.45, 0.55) (0.45, 0, 0.55)

TABLE 6.20. Incomplete block

design for model (II) with $s = 4$,

$b = 4$ and $n = 16$.

Block no.	Design points
1	(0, 0, 1) (0, 1, 0) (0.575, 0, 0.425) (0.55, 0.45, 0)
2	(0, 0, 1) (1, 0, 0) (0, 0.575, 0.425) (0.425, 0.575, 0)
3	(0, 1, 0) (1, 0, 0) (0, 0.45, 0.55) (0.425, 0, 0.575)
4	(0, 1, 0) (0.5, 0, 0.5) (0, 0.45, 0.55) (0.55, 0.45, 0)

TABLE 6.21. Incomplete block

design for model (II) with

$s = 4$, $b = 5$ and $n = 20$.

Block no.	Design points
1	(0, 0, 1) (0, 1, 0) (0.55, 0, 0.45) (0.575, 0.425, 0)
2	(0, 0, 1) (1, 0, 0) (0, 0.575, 0.425) (0.425, 0.575, 0)
3	(0, 0, 1) (1, 0, 0) (0, 0.575, 0.425) (0.425, 0.575, 0)
4	(0, 1, 0) (1, 0, 0) (0, 0.425, 0.575) (0.45, 0, 0.55)
5	(0, 1, 0) (0.5, 0, 0.5) (0, 0.425, 0.575) (0.575, 0.425, 0)

TABLE 6.22. Incomplete block

design for model (II) with $s = 4$,

$b = 6$ and $n = 24$.

Block no.	Design points
1	(0, 0, 1) (0, 1, 0) (0.55, 0, 0.45) (0.55, 0.45, 0)
2	(0, 0, 1) (0, 1, 0) (0.55, 0, 0.45) (0.55, 0.45, 0)
3	(0, 0, 1) (1, 0, 0) (0, 0.55, 0.45) (0.45, 0.55, 0)
4	(0, 0, 1) (1, 0, 0) (0, 0.55, 0.45) (0.45, 0.55, 0)
5	(0, 1, 0) (1, 0, 0) (0, 0.45, 0.55) (0.45, 0, 0.55)
6	(0, 1, 0) (1, 0, 0) (0, 0.45, 0.55) (0.45, 0, 0.55)

TABLE 6.23. Incomplete block

design for model (II) with

$s = 5$, $b = 2$ and $n = 10$.

TABLE 6.24. Incomplete block

design for model (II) with $s = 5$,

$b = 3$ and $n = 15$.

Block no.	Design points	Block no.	Design points
1	(0, 0, 1)	1	(0, 0, 1)
	(0, 1, 0)		(0, 1, 0)
	(0, 0.46, 0.54)		(0, 0.5, 0.5)
	(0.54, 0, 0.46)		(0.55, 0, 0.45)
	(0.58, 0.42, 0)		(0.55, 0.45, 0)
2	(0, 0, 1)	2	(0, 0, 1)
	(1, 0, 0)		(1, 0, 0)
	(0, 0.54, 0.46)		(0.5, 0, 0.5)
	(0.46, 0, 0.54)		(0, 0.55, 0.45)
	(0.42, 0.58, 0)		(0.45, 0.55, 0)
		3	(0, 1, 0)
			(1, 0, 0)
			(0.5, 0.5, 0)
			(0, 0.45, 0.55)
			(0.45, 0, 0.55)

TABLE 6.25. Incomplete block design for model (II)

with $s = 5$, $b = 4$ and $n = 20$.

Block no.	Design points
1	(0, 0, 1)
	(0, 1, 0)
	(0, 0.5, 0.5)
	(0.55, 0, 0.45)
	(0.55, 0.45, 0)
2	(0, 0, 1)
	(1, 0, 0)
	(0.5, 0, 0.5)
	(0, 0.55, 0.45)
	(0.45, 0.55, 0)
3	(0, 1, 0)
	(1, 0, 0)
	(0.5, 0.5, 0)
	(0, 0.45, 0.55)
	(0.45, 0, 0.55)
4	(0, 0, 1)
	(0, 1, 0)
	(1, 0, 0)
	(0, 0.5, 0.5)
	(0.5, 0, 0.5)

of $|M^{11}(\xi_n)|^{-1}$ for the step from $s = 2$ to $s = 3$ is much larger than for any other unit increase in block size. The experiments with $s = 2$ are relatively inefficient and are, if possible, avoided.

The design for $s = 2$, $b = 12$ and $n = 24$ in Table 6.11 is a relatively general one for $s = 2$ and the investigator could use the first ℓ blocks for a ℓ -block design. For $b = 6$ and $n = 12$; and $b = 11$ and $n = 22$, the designs with a largest $|M^{11}(\xi_n)|^{-1}$ are subsets of the design for $b = 12$ and $n = 24$. Table 6.26 shows the $|M^{11}(\xi_n)|^{-1}$ when using the first $\frac{1}{2}n$ blocks of Table 6.11, and the ratio of this determinant to the maximum $|M^{11}(\xi_n)|^{-1}$ arrived at by generating a design, for $n = 10, 14, 16$ and 18 . Only for $n = 10$ would the use of the first 5 blocks of Table 6.11 not yield a design with a relatively large $|M^{11}(\xi_n)|^{-1}$.

TABLE 6.26. Values of $|M^{11}(\xi_n)|^{-1}$ obtained by using subsets of the blocks of the design for $s = 2$, $b = 12$ and $n = 24$, and the ratio $|M^{11}(\xi_n)|^{-1}/\max|M^{11}(\xi_n)|^{-1}$, for different numbers, n of observations and b of blocks for $s = 2$.

n	b	$ M^{11}(\xi_n) ^{-1}$	ratio
10	5	0.1265×10^{-9}	.52
14	7	0.3116×10^{-9}	.92
16	8	0.3457×10^{-9}	.90
18	9	0.4029×10^{-9}	.99

Similarly for $s = 3$ the design in Table 6.15 for $n = 18$ and $b = 6$ seem the most useful when compared with the other designs for $s = 3$. For $s = 4$ the basic design seems to be that for $b = 4$ and $n = 16$ in Table 6.21. The basic design for $s = 5$ occurs in Table 6.24 where $b = 3$ and $n = 15$.

Care should, however, be taken when extrapolating these designs to values of $n > 24$ because larger experiments are not necessarily mere replications of the basic blocks. For example the design in Table 6.11 with $s = 2$, $b = 12$ and $n = 24$ is not the design in Table 6.6 for $b = 6$ and $n = 12$ replicated twice.

The cubic model (III)

For the quadratic model only designs with blocks up to size $s = 5$ were considered. The reason for this limitation is the assumption that the smaller block sizes will be more important in practice.

Table 6.27 contains the values of $|M(\xi_n)|$, $|M^{11}(\xi_n)|^{-1}$ and E for a number of balanced, partially balanced and cyclic incomplete block designs using the 10 design points of the D-optimum design for the third order model as 'treatments'. This design is given in Table 4.2. The balanced incomplete block designs were those given by Cochran and Cox (1957), their plan numbers being given; the partially balanced incomplete block designs are from Bose et al (1954), their plan numbers being used; and the cyclic incomplete block designs are a selection of those given by John (1966) and are identified by their generating sets. Unfortunately virtually all the designs require very large numbers of observations. In this case partially balanced and

TABLE 6.27. Values of $|M(\xi_n)|$, $|M^{11}(\xi_n)|^{-1}$ and E for the balanced (BIB), partially balanced (PBIB) and cyclic (CIB) incomplete block designs utilising the D-optimum design for the cubic model (III), for different numbers, n of observations and b of blocks and the corresponding block sizes s. Here $|M_{11}(\xi_n)| = 0.4486 \times 10^{-23}$ throughout.

Plan Type and No.	n	b	s	$ M(\xi_n) $	$ M^{11}(\xi_n) ^{-1}$	E
BIBD 11.14	90	45	2	0.1846×10^{-96}	0.2262×10^{-25}	0.0050
11.15	90	30	3	0.1317×10^{-65}	0.3012×10^{-24}	0.0671
11.16	60	15	4	0.4467×10^{-39}	0.8694×10^{-24}	0.1938
11.17	90	18	5	0.1280×10^{-43}	0.1554×10^{-23}	0.3464
11.18	90	15	6	0.1153×10^{-38}	0.2244×10^{-23}	0.5002
*	90	10	9	0.4012×10^{-31}	0.4012×10^{-23}	0.8942
PBIBD S17	40	10	4	0.6845×10^{-32}	0.6845×10^{-24}	0.1526
S18	40	5	8	0.2772×10^{-25}	0.3465×10^{-23}	0.7725
S19	60	10	6	0.2163×10^{-31}	0.2163×10^{-23}	0.4822
SR16	40	8	5	0.5608×10^{-29}	0.1470×10^{-23}	0.3277
R14	80	20	4	0.3137×10^{-47}	0.8224×10^{-24}	0.1833
T1	20	5	4	0.5476×10^{-26}	0.6845×10^{-24}	0.1526
T6	30	10	3	0.2371×10^{-32}	0.2371×10^{-24}	0.0529
T9	30	6	5	0.9811×10^{-27}	0.1281×10^{-23}	0.3211
T15	30	5	6	0.1731×10^{-25}	0.2163×10^{-23}	0.4823
CIBD (0,1,3)	30	10	3	0.2357×10^{-32}	0.2357×10^{-24}	0.0525
(0,1,4)(0,2,5)	60	20	3	0.1086×10^{-47}	0.2848×10^{-24}	0.0635
(0,1,2,5)	40	10	4	0.8116×10^{-32}	0.8116×10^{-24}	0.1809
(0,1,2,4)(0,1,3,6)	80	20	4	0.3270×10^{-47}	0.8572×10^{-24}	0.1911
(0,1,2,4)(0,1,3,6)	100	25	4	0.6064×10^{-56}	0.8617×10^{-24}	0.1921
(0,1,5,6)	50	10	5	0.1498×10^{-31}	0.1498×10^{-23}	0.3340
(0,1,2,5,7)	50	10	5	0.1530×10^{-31}	0.1530×10^{-23}	0.3410
(0,1,2,3,6)	100	20	5	0.5836×10^{-47}	0.1530×10^{-23}	0.3410
(0,1,2,3,5)(0,1,3,5,7)	100	20	5	0.5893×10^{-47}	0.1549×10^{-23}	0.3444
(0,1,2,3,6)(0,1,3,5,7)	60	10	6	0.2224×10^{-31}	0.2224×10^{-23}	0.4958
(0,1,2,3,5,6)	70	10	7	0.2878×10^{-31}	0.2878×10^{-23}	0.6416
(0,1,2,3,4,5,7)	80	10	8	0.3474×10^{-31}	0.3474×10^{-23}	0.7745

cyclic incomplete block designs with unequal replication would prove to be very useful. Such methods, unfortunately, do not exist as yet.

Table 6.28 contains the values of $|M(\xi_n)|$, $|M^{11}(\xi_n)|^{-1}$ and E and the table in which that design appears for all the viable combinations of $n \leq 24$ and $2 \leq s \leq 5$ used to generate incomplete block mixture designs for the cubic model. Tables 6.29 to 6.42 contain the generated designs. Alternative designs are given in some cases where a design with a slightly smaller $|M^{11}(\xi_n)|^{-1}$ has more homogenous mixtures.

The problem of a local maximum for $|M^{11}(\xi_n)|^{-1}$ becomes very important when using this model. Table 6.43 gives a few examples of local maxima for $|M^{11}(\xi_n)|^{-1}$ that had been reached and the final maxima achieved by making some subjective changes to the design and then using the maximisation routine again. It is also difficult to decide whether the design with the maximum possible $|M^{11}(\xi_n)|^{-1}$ had been reached because the design with a local maximum sometimes has a more homogenous mixture pattern than another with a larger $|M^{11}(\xi_n)|^{-1}$. Compare for example the designs of Tables 6.40(a) and 6.40(b) for $n = 24$, $b = 6$ and $s = 4$ and quoted in Table 6.43.

The designs differ appreciably for different block sizes. There are, however, always some design points near the centre of the design space varying from permutations of (0.2, 0.35, 0.55) to (0.3, 0.3, 0.4). The non-vertex mixtures on the bounding lines of the design space, the triangle, vary from permutations of (0, 0.2, 0.8) to (0, 0.5, 0.5). The mixture proportions tend to be closer to the former and thus to

TABLE 6.28. Values of $|M(\xi_n)|$, $|M^{11}(\xi_n)|^{-1}$ and E and the number of the table in which the design is given for the incomplete block mixture designs generated for the different block sizes s, numbers b of blocks and n of observations for fitting the cubic model (III) to the full simplex.

s	b	n	$ M(\xi_n) $	$ M^{11}(\xi_n) ^{-1}$	E	Table No.
2	9	18	0.1592×10^{-32}	0.7613×10^{-26}	0.0115	6.29
	10	20	0.1140×10^{-33}	0.1140×10^{-25}	0.0070	6.30
	11	22	0.5511×10^{-35}	0.1299×10^{-25}	0.0089	6.31
	12	24	0.2559×10^{-36}	0.1585×10^{-25}	0.0120	6.32
3	5	15	0.1256×10^{-26}	0.1570×10^{-24}	0.0937	6.33
	6	18	0.1701×10^{-27}	0.2205×10^{-24}	0.0997	6.34
	7	21	0.1698×10^{-28}	0.2854×10^{-24}	0.1015	6.35
	8	24	0.1017×10^{-29}	0.2667×10^{-24}	0.0095	6.36
4	3	12	0.1472×10^{-24}	0.4417×10^{-24}	0.2025	6.37a
	3	12	0.1507×10^{-24}	0.4520×10^{-24}	0.1944	6.37b
	4	16	0.1011×10^{-25}	0.6418×10^{-24}	0.3155	6.38
	5	20	0.6283×10^{-26}	0.7854×10^{-24}	0.2170	6.39
	6	24	0.5968×10^{-27}	0.7734×10^{-24}	0.2467	6.40a
	6	24	0.6297×10^{-27}	0.8161×10^{-24}	0.2747	6.40b
5	3	15	0.3259×10^{-24}	0.9778×10^{-24}	0.5452	6.41
	4	20	0.9310×10^{-25}	0.1490×10^{-23}	0.3698	6.42

TABLE 6.29. Incomplete block

design for model (III) with

$s = 2$, $b = 9$ and $n = 18$.

Block no.	Design points
1	(0, 0.2, 0.8) (0, 0.8, 0.2)
2	(0.2, 0, 0.8) (0.8, 0, 0.2)
3	(0.2, 0.8, 0) (0.8, 0.2, 0)
4	(0, 0, 1) (0.225, 0.225, 0.55)
5	(0, 1, 0) (0.225, 0.55, 0.225)
6	(1, 0, 0) (0.55, 0.225, 0.225)
7	(0, 0.5, 0.5) (0.5, 0.25, 0.25)
8	(0.5, 0, 0.5) (0.25, 0.5, 0.25)
9	(0.5, 0.5, 0) (0.25, 0.25, 0.5)

TABLE 6.30. Incomplete block

design for model (III) with

$s = 2$, $b = 10$ and $n = 20$.

Block no.	Design points
1	(0, 0.2, 0.8) (0, 0.8, 0.2)
2	(0.2, 0, 0.8) (0.8, 0, 0.2)
3	(0.2, 0.8, 0) (0.8, 0.2, 0)
4	(0, 0, 1) (0.225, 0.275, 0.5)
5	(0, 1, 0) (0.225, 0.5, 0.275)
6	(0, 0.5, 0.5) (0.5, 0.25, 0.25)
7	(0, 0, 1) (0.4, 0.6, 0)
8	(0, 1, 0) (0.4, 0, 0.6)
9	(1, 0, 0) (0.575, 0.425, 0)
10	(1, 0, 0) (0.575, 0, 0.425)

TABLE 6.31. Incomplete block

design for model (III) with

$s = 2$, $b = 11$ and $n = 22$.

Block no.	Design points
1	(0, 0.2, 0.8) (0, 0.8, 0.2)
2	(0.2, 0, 0.8) (0.8, 0, 0.2)
3	(0.2, 0.8, 0) (0.8, 0.2, 0)
4	(0, 0, 1) (0.24, 0.26, 0.5)
5	(0, 1, 0) (0.26, 0.48, 0.26)
6	(1, 0, 0) (0.5, 0.26, 0.24)
7	(0, 0, 1) (0.46, 0.54, 0)
8	(0, 1, 0) (0.52, 0, 0.48)
9	(1, 0, 0) (0, 0.56, 0.44)
10	(0, 0.44, 0.56) (0.36, 0, 0.64)
11	(0.54, 0.46, 0) (0.62, 0, 0.38)

TABLE 6.32. Incomplete block

design for model (III) with

$s = 2$, $b = 12$ and $n = 24$.

Block no.	Design points
1	(0, 0.2, 0.8) (0, 0.8, 0.2)
2	(0.2, 0, 0.8) (0.8, 0, 0.2)
3	(0.2, 0.8, 0) (0.8, 0.2, 0)
4	(0, 0, 1) (0.24, 0.24, 0.52)
5	(0, 1, 0) (0.24, 0.52, 0.24)
6	(1, 0, 0) (0.52, 0.24, 0.24)
7	(0.24, 0.26, 0.5) (0.58, 0.42, 0)
8	(0.26, 0.5, 0.24) (0.42, 0, 0.58)
9	(0.5, 0.24, 0.26) (0, 0.58, 0.42)
10	(0, 0, 1) (0.38, 0.62, 0)
11	(0, 1, 0) (0.62, 0, 0.38)
12	(1, 0, 0) (0, 0.38, 0.62)

TABLE 6.33. Incomplete block

design for model (III) with

$s = 3$, $b = 5$ and $n = 15$.

Block no.	Design points
1	(0, 0, 1) (0.225, 0.775, 0) (0.8, 0.2, 0)
2	(0, 1, 0) (0.225, 0, 0.775) (0.8, 0, 0.2)
3	(1, 0, 0) (0, 0.225, 0.775) (0, 0.775, 0.225)
4	(0, 0, 1) (0, 0.475, 0.525) (0.45, 0.25, 0.3)
5	(0.5, 0, 0.5) (0.525, 0.475, 0) (0.2, 0.425, 0.375)

TABLE 6.34. Incomplete block

design for model (III) with

$s = 3$, $b = 6$ and $n = 18$.

Block no.	Design points
1	(0, 0, 1) (0.225, 0.775, 0) (0.775, 0.225, 0)
2	(0, 1, 0) (0.225, 0, 0.775) (0.775, 0, 0.225)
3	(1, 0, 0) (0, 0.225, 0.775) (0, 0.775, 0.225)
4	(0, 0, 1) (0.44, 0.24, 0.32) (0, 0.48, 0.52)
5	(0.5, 0, 0.5) (0.24, 0.42, 0.34) (0.7, 0.3, 0)
6	(0, 1, 0) (1, 0, 0) (0.4, 0.6, 0)

TABLE 6.35. Incomplete block

design for model (III) with

$s = 3$, $b = 7$ and $n = 21$.

Block no.	Design points
1	(0, 0.22, 0.78) (0, 0.78, 0.22) (0.32, 0, 0.68)
2	(0.22, 0, 0.78) (0.78, 0, 0.22) (0.68, 0.32, 0)
3	(0.22, 0.78, 0) (0.78, 0.22, 0) (0, 0.68, 0.32)
4	(0, 0, 1) (0.28, 0.24, 0.48) (0.38, 0.62, 0)
5	(0, 1, 0) (0.26, 0.46, 0.28) (0.62, 0, 0.38)
6	(1, 0, 0) (0.48, 0.28, 0.24) (0, 0.38, 0.62)
7	(0, 0, 1) (0, 1, 0) (1, 0, 0)

TABLE 6.36. Incomplete block

design for model (III) with

$s = 3$, $b = 8$ and $n = 24$.

Block no.	Design points
1	(0, 0.22, 0.78) (0, 0.78, 0.22) (0.28, 0, 0.72)
2	(0.22, 0, 0.78) (0.78, 0, 0.22) (0.72, 0.28, 0)
3	(0.22, 0.78, 0) (0.78, 0.22, 0) (0, 0.72, 0.28)
4	(0, 0, 1) (0.275, 0.275, 0.45) (0.46, 0.54, 0)
5	(0, 1, 0) (0.275, 0.45, 0.275) (0.54, 0, 0.46)
6	(1, 0, 0) (0.45, 0.275, 0.275) (0, 0.46, 0.54)
7	(0, 0, 1) (0, 1, 0) (1, 0, 0)
8	(0, 0.28, 0.72) (0.72, 0, 0.28) (0.28, 0.72, 0)

TABLE 6.37a. Incomplete block design for model (III) with $s = 4$, $b = 3$ and $n = 12$.

TABLE 6.37b. Incomplete block design for model (III) with $s = 4$, $b = 3$ and $n = 12$.

Block no.	Design points	Block no.	Design points
1	(0, 0, 1)	1	(0, 0, 1)
	(0.25, 0.25, 0.5)		(0.24, 0.24, 0.52)
	(0.25, 0.75, 0)		(0.26, 0.74, 0)
	(0.75, 0.25, 0)		(0.74, 0.26, 0)
2	(0, 1, 0)	2	(0, 1, 0)
	(0.25, 0.5, 0.25)		(0.24, 0.52, 0.24)
	(0.25, 0, 0.75)		(0.26, 0, 0.74)
	(0.75, 0, 0.25)		(0.74, 0, 0.26)
3	(1, 0, 0)	3	(1, 0, 0)
	(0.5, 0.25, 0.25)		(0.52, 0.24, 0.24)
	(0, 0.25, 0.75)		(0, 0.26, 0.74)
	(0, 0.75, 0.25)		(0, 0.74, 0.26)

TABLE 6.38 . Incomplete block

design for model (III) with

$s = 4, b = 4$ and $n = 16$.

Block no.	Design points
1	(0, 0, 1) (0, 1, 0) (0, 0.58, 0.42) (0.34, 0.66, 0)
2	(0, 0, 1) (1, 0, 0) (0.4, 0, 0.6) (0.66, 0.34, 0)
3	(0, 0.25, 0.75) (0, 0.78, 0.22) (0.46, 0.28, 0.26) (0.24, 0, 0.76)
4	(0.22, 0.78, 0) (0.8, 0.2, 0) (0.24, 0.32, 0.44) (0.74, 0, 0.26)

TABLE 6.39. Incomplete block

design for model (III) with

$s = 4, b = 5$ and $n = 20$.

Block no.	Design points
1	(0, 0, 1) (1, 0, 0) (0, 0.325, 0.675) (0.625, 0, 0.375)
2	(0, 0, 1) (1, 0, 0) (0.65, 0.35, 0) (0.3, 0.325, 0.375)
3	(0, 1, 0) (0.375, 0.325, 0.3) (0, 0.225, 0.775) (0, 0.775, 0.225)
4	(0, 1, 0) (0.325, 0.675, 0) (0.225, 0, 0.775) (0.775, 0, 0.225)
5	(0.3, 0, 0.7) (0, 0.7, 0.3) (0.225, 0.775, 0) (0.775, 0.225, 0)

TABLE 6.40a. Incomplete block

design for model (III) with

$s = 4$, $b = 6$ and $n = 24$.

TABLE 6.40b. Incomplete block

design for model (III) with

$s = 4$, $b = 6$, and $n = 24$.

Block no.	Design points	Block no.	Design points
1	(0, 0, 1) (0.3, 0.26, 0.44) (0.76, 0.24, 0) (0.24, 0.76, 0)	1	(0, 0, 1) (0.28, 0.26, 0.46) (0.78, 0.22, 0) (0.25, 0.75, 0)
2	(0, 1, 0) (0.28, 0.44, 0.28) (0.2, 0, 0.8) (0.76, 0, 0.24)	2	(0, 1, 0) (0.26, 0.46, 0.28) (0.22, 0, 0.78) (0.75, 0, 0.25)
3	(0, 0, 1) (0.44, 0.28, 0.28) (0, 0.44, 0.56) (0.32, 0, 0.68)	3	(1, 0, 0) (0.46, 0.28, 0.26) (0, 0.78, 0.22) (0, 0.25, 0.75)
4	(1, 0, 0) (0, 0.22, 0.78) (0, 0.74, 0.26) (0.32, 0, 0.68)	4	(0, 0, 1) (0.7, 0, 0.3) (0.72, 0.28, 0) (0, 0.66, 0.34)
5	(0, 1, 0) (1, 0, 0) (0.7, 0, 0.3) (0.52, 0.48, 0)	5	(0, 1, 0) (0, 0.3, 0.7) (0.28, 0, 0.72) (0.66, 0.34, 0)
6	(0, 0.74, 0.26) (0, 0.26, 0.74) (0.24, 0.76, 0) (0.76, 0.24, 0)	6	(1, 0, 0) (0.3, 0.7, 0) (0, 0.72, 0.28) (0.34, 0, 0.66)

TABLE 6.41. Incomplete block

design for model (III) with

$s = 5$, $b = 3$ and $n = 15$.

Block no.	Design points
1	(0, 0, 1) (0.2, 0.25, 0.55) (0, 0.75, 0.25) (0.725, 0, 0.275) (0.675, 0.325, 0)
2	(0, 1, 0) (0.25, 0.55, 0.2) (0.75, 0.25, 0) (0, 0.275, 0.725) (0.325, 0, 0.675)
3	(1, 0, 0) (0.55, 0.2, 0.25) (0.25, 0, 0.75) (0.275, 0.725, 0) (0, 0.675, 0.325)

TABLE 6.42. Incomplete block

design for model (III) with

$s = 5$, $b = 4$ and $n = 20$.

Block no.	Design points
1	(0, 0, 1) (0.25, 0.75, 0) (0.775, 0.225, 0) (0, 0.675, 0.325) (0.7, 0, 0.3)
2	(0, 1, 0) (0.25, 0, 0.75) (0.75, 0, 0.25) (0, 0.3, 0.7) (0.35, 0.375, 0.275)
3	(1, 0, 0) (0, 0.25, 0.75) (0, 0.75, 0.25) (0.3, 0, 0.7) (0.3, 0.7, 0)
4	(0, 0, 1) (0, 1, 0) (1, 0, 0) (0.3, 0.3, 0.4) (0.675, 0.325, 0)

that of the D-optimum design namely permutations of (0, 0.28, 0.72). The designs differ so much that no general recommendations can be made.

TABLE 6.43. Local maxima and maxima obtained for $|M^{11}(\xi_n)|^{-1}$ when generating incomplete block designs for fitting the cubic model (III) for some block sizes s , numbers, b of blocks and sizes, n of experiments.

			$ M^{11}(\xi_n) ^{-1}$	
s	b	n	Local max.	Maximum
2	9	18	0.7212×10^{-26}	0.7613×10^{-26}
3	6	18	0.1938×10^{-24}	0.2205×10^{-24}
4	4	16	0.6408×10^{-24}	0.6418×10^{-24}
4	6	24	0.7734×10^{-24}	0.8161×10^{-24}
5	4	20	0.1399×10^{-24}	0.1490×10^{-24}

7. CONCLUSION

The results of Sections 3.3, 4.3 and 5.3 show that when designing mixture experiments for any simplex design space only the full simplex need be considered since the design for a simplex subspace is a suitably scaled version of the design for the full simplex.

Section 4.3 shows that the simplex-lattice designs for the full simplex design space are D-optimum only for linear and quadratic models. The D-optimum designs for fitting linear, quadratic and cubic models for any simplex design space can be obtained by extending the corresponding D-optimum designs for the two-dimensional, 3-component simplex given in Table 4.2.

In Section 3.3 it was shown that the only method so far published for obtaining designs for design spaces constrained to a subspace of the full simplex, namely that of McLean and Anderson (1966), yields D-optimum designs very rarely. If a $(q-1)$ dimensional non-simplex subspace of the full simplex can be transformed to a $(q-1)$ -cube by a linear transformation, the method given by Kiefer (1961a) can be used to obtain D-optimum designs. No generalisations can, however, be made about the nature of the $\text{var} \{ \hat{\eta}(x) \}$ functions and the nature of the D-optimum designs for other non-regular design spaces. The D-optimum design will most probably require observations to be taken in unequal proportions at the relatively small number of distinct design points.

The method developed in Section 6.1 and used in Section 6.3 to generate incomplete block mixture designs did not yield any simple method for blocking mixture experiments. The incomplete block designs

generated for the 3-component mixtures could be extended to higher dimensions by extension and permutation of the mixture proportions but the efficiencies of the resulting designs are unknown.

A comparison of Table 4.2 with Tables 6.4 to 6.25 and 6.29 to 6.42 shows that the incomplete block mixture designs generated in Section 6.3 never consisted solely of points from the D-optimum or simplex-lattice designs. The larger the block size becomes, however, the closer the mixture proportions get to that of the D-optimum design. The usual incomplete block designs proved only slightly less efficient than the corresponding generated designs and could therefore possibly be used in more complex cases where generating a design would be too lengthy. As Tables 6.2 and 6.27 show, the usual incomplete block designs that are available from the literature tend to require a large number of blocks and consequently a large number of observations.

The method of Section 6.1 could perhaps be used to generate split-plot mixture designs by keeping the proportions of one or more of the components fixed in a block. The implications of this application is, however, still to be investigated.

There are several problems connected with the design of mixture experiments that have not yet been solved. A major one is the problem of mixture designs for sequential experimentation. The parameterisation of Cox (1971) could prove useful in this case if the reference mixture is taken common to two successive experiments. The problem of the mixture equivalent of rotatable designs is also still unresolved.

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