Lecture Notes in Statistics 1028

B. K. Sinha
N. K. Mandal
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Optimal Mixture Experiments

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We dedicate this monograph to all the researchers in this fascinating topic whose valuable contributions have paved the way of our thought process in writing this monograph. All of them have been with us in spirit and action. We fondly hope we have not disappointed them

Foreword

It gives me much pleasure to welcome this comprehensive work on the optimality aspects of Mixture Designs. Kiefer formulated various optimality criteria and laid foundations for the study of optimal designs—both exact and approximate. Scheffé initiated the study of mixture designs and introduced a model for these designs. The current monograph brings these together in a cohesive way. It deals with Scheffé's model and some other models. Optimality aspects studied include: (a) optimal designs for the estimation of parameters in mixture models and (b) optimal designs for optimal mixtures under mixture models. This is followed by applications of mixture experiments in various fields such as agriculture and pharmaceuticals. The monograph concludes with a study of several variants and extensions. It also gives directions for further research in this area of study which has just opened up. The authors have taken a significant step in promoting research in this area of study by putting together all known work in this area and by indicating directions for further work. They deserve our gratitude for this important contribution.

Waterloo, Canada, February 2014 Kirti R. Shah

Preface

Two of us had necessary research experience in the broad area of optimal designs. One of us had necessary exposition in the area of response surface-related optimal designs. The remaining one had adequate experience in handling factorial designs. Two of us ventured into this emerging area of optimal mixture designs several years back. The other two joined hands and strengthened the research collaboration to the extent that all of us together could see the emergence of a research-level monograph within a reasonable time frame.

We have tried to present and explain, in our own way, the techniques needed for handling the optimal designing problems related to the general area of mixture models and specific areas of applications of such models.

We will consider our efforts rewarded if the readers find the presentations in order and derive enough creative interest in pursuing the research topics further.

Professor Kirti R. Shah has showered unbounded research opportunity on two of us for over 20 long years or so. He has obliged us by very kindly agreeing to write the Foreword for this monograph.

We have a very special point to make. The whole exercise was academically challenging to one of us who had no exposure to this area of research. But with sheer interest, enthusiasm, and dedication, this special collaborator exceeded all our expectations and rightfully deserved a position on the front page of the publication.

Kolkata, February 2014

B. K. Sinha
N. K. Mandal
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P. Das

Acknowledgments

This monograph is the end product of a research project entitled *Optimum Regression Designs* that was undertaken by us in the Department of Statistics, University of Calcutta, and was supported by the UPE (University with Potential for Excellence) Grant received from the University Grants Commission, India. We are thankful to the department and our colleagues for providing a congenial and supportive atmosphere during the preparation of the monograph. We also thank Prof. S. P. Mukherjee, our revered teacher and Centenary Professor of Statistics in the department [now retired], for his continued interest and encouragement.

At different stages, three workshops on this theme were conducted by us at (i) Kalyani University's Department of Statistics, (ii) Indian Agricultural Statistics Research Institute, New Delhi, and (iii) Calcutta University's Department of Statistics. We thank the participants for their interest and questions/queries which helped us shape the presentation material for this monograph.

We feel highly indebted to Mr. Aparesh Chatterjee for giving time and energy to create the latex file of the entire manuscript. We have been corresponding with him over the last one year in all permutations and combinations—at times even from overseas—with frequent and quick changes in the draft. We are truly happy to note that he has favored us by not departing in the middle of the chaos! He indeed deserves a special mention and our thanks for bearing with our demands with all smiles and extreme courtesy at all stages!!

Finally, with heartfelt thanks, we express our gratitude to our family members for their love and unlimited support, and our appreciation to Ms. Sagarika Ghosh of Springer for her valued help at various stages.

Kolkata, February 2014

Bikas Kumar Sinha
Nripes Kumar Mandal
Manisha Pal
Premadhis Das

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Chapter 1
Mixture Models and Mixture Designs: Scope of the Monograph

Abstract We introduce standard mixture models and standard mixture designs as are well known in the literature (vide Cornell 2002). Some of the less known models are also introduced briefly. Next we explain the frameworks of exact and approximate [or, continuous] mixture designs. We mention about known applications of mixture experiments in agriculture, food processing, and pharmaceutical studies. We also provide a chapter-wise brief summary of the contents covered in the monograph.

Keywords Scheffé's homogeneous mixture models · Becker's mixture model · Draper–St. John's mixture model · Simplex lattice designs · Simplex centroid designs · Axial designs · Exact designs · Approximate designs · Applications · Agriculture · Pharmacy · Food processing

1.1 Introduction

This monograph features state-of-the-art research findings on various aspects of mixture experiments, mainly from the point of view of *optimality*. We have freely consulted available books and journals on mixture experiments and optimal experimental designs. There is no denying the fact that a considerable number of research articles has been published in this specific area dealing with mixture model specifications and related data analyses; however, emphasis on finding optimal mixture experiments has been relatively less pronounced. With a thorough understanding of the tools and techniques in the study of optimal designs [in discrete and continuous design settings], we ventured into this relatively new area of research a few years back and we were fascinated by the niceties of the elegant results—already known in the literature and further researched out by our team. We are happy to work on this monograph and bring it to the attention of optimal design theorists in a most comprehensive manner—covering basic and advanced results in the area of optimal mixture experiments.

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1.2 Mixture Models

Let $\boldsymbol{x} = (x_1, x_2, \ldots, x_q)$ denote the vector of proportions of q mixing components and $\eta(\boldsymbol{x})$ be the corresponding mean response. The factor space is a simplex, given by

$$\mathcal{X} = \{\boldsymbol{x} = (x_1, \ldots, x_q) : x_i \geq 0, \quad i = 1, 2, \ldots, q; \ \sum_{i=1}^{q} x_i = 1\}. \tag{1.2.1}$$

Scheffé (1958) introduced the following models in *canonical* forms of different degrees to represent the mean response function $\eta(\boldsymbol{x})$:

$$\text{Linear: } \eta(\boldsymbol{x}) = \sum_i \beta_i x_i \tag{1.2.2}$$

$$\text{Quadratic: } \eta(\boldsymbol{x}) = \sum_i \beta_i x_i + \sum_{i<j} \beta_{ij} x_i x_j \tag{1.2.3}$$

$$\text{Cubic: } \eta(\boldsymbol{x}) = \sum_i \beta_i x_i + \sum_{i<j} \beta_{ij} x_i x_j + \sum_{i<j<k} \beta_{ijk} x_i x_j x_k \tag{1.2.4}$$

$$\text{Special Cubic: } \eta(\boldsymbol{x}) = \sum_i \beta_i x_i + \sum_{i<j} \beta_{ij} x_i x_j + \sum_{i<j<k} \beta_{ijk} x_i x_j x_k + \sum_{i<j} \beta_{ij} x_i x_j (x_i - x_j). \tag{1.2.5}$$

In the above, we have used generic notations for the model parameters in different versions of mixture models. Using the identity $\Sigma x_i = 1$, model (1.2.3) can be converted to a canonical *homogeneous* quadratic model:

$$\eta(\boldsymbol{x}) = \sum_i \beta_{ii} x_i^2 + \sum_{i<j} \beta_{ij} x_i x_j. \tag{1.2.6}$$

In the present study, we shall be concerned with the canonical models (1.2.2) and (1.2.3) or (1.2.6) or some other equivalent versions of it. There are other types of mixture models introduced in the literature. In order to differentiate them, we may refer to the above models as 'standard mixture models'. It is to be noted that the use of the identity $\Sigma x_i = 1$ may lead to remodeling very deceptively. For example, the quadratic model (1.2.3) may be modified to the cubic model, thus inviting more parameters [with intriguing parametric relations] and more design points for estimation. Also proper interpretation of the parameters may be a bit confusing and complicated. Note that unlike in the usual regression models, the constant term β_0 has been dropped from all mixture models, as otherwise, the β-parameters become non-estimable. Further to this, in the mixture model setup, β_0 does not have any obvious interpretation like the intercept!

As was mentioned above, there are some other 'non-standard' mixture models introduced and studied in the literature. We will deal with symmetrized versions

of two such models viz. Becker's homogeneous model of degree one (1968) and Draper–St. John's model (1977).
Becker's model is given by

$$\eta(\boldsymbol{x}) = \beta_1 x_1 + \beta_2 x_2 + \cdots + \beta_q x_q + \beta_{12}\frac{x_1 x_2}{x_1 + x_2} + \cdots + \beta_{q-1q}\frac{x_{q-1}x_q}{x_{q-1} + x_q}$$
$$0 \le x_i \le 1, \forall i;\; x_i + x_j > 0,\; \forall i < j. \tag{1.2.7}$$

As a matter of fact, Becker introduced a general representation of (1.2.7) as is given below:

$$\eta(\boldsymbol{x}) = \sum_i \beta_i x_i + \sum_{i<j} \beta_{ij}\frac{x_i x_j}{x_i + x_j} + \cdots + \sum_{i<j<k} \beta_{ijk}\frac{x_i x_j x_k}{(x_i + x_j + x_k)^2} + \cdots$$
$$0 \le x_i \le 1, \forall\, i; x_i + x_j > 0,\; \forall\, i < j. \tag{1.2.8}$$

Besides the above, there are two other homogeneous models of degree one suggested by Becker. For the sake of completeness, these are also displayed below:

$$\eta(\boldsymbol{x}) = \sum_i \beta_i x_i + \sum_{i<j} \beta_{ij}\min(x_i, x_j) + \sum_{i<j<k} \beta_{ijk}\min(x_i, x_j, x_k) + \cdots$$
$$0 < x_i < 1, \forall\, i. \tag{1.2.9}$$

$$\eta(\boldsymbol{x}) = \sum_i \beta_i x_i + \sum_{i<j} \beta_{ij}(x_i x_j)^{1/2} + \cdots + \sum_{i<j<k} \beta_{ijk}(x_i x_j x_k)^{1/3} + \cdots$$
$$0 < x_i < 1, \forall\, i. \tag{1.2.10}$$

Draper–St. John's model is given by

$$\eta(\boldsymbol{x}) = \beta_1 x_1 + \cdots + \beta_q x_q + \frac{\alpha_1}{x_1} + \cdots + \frac{\alpha_q}{x_q}, \quad 0 < x_i < 1,\; \forall\, i. \tag{1.2.11}$$

Two other entirely different models will be introduced in later chapters.

1.3 Mixture Designs

Mixture designs are essentially design layouts with the descriptions of the distinct design points or vectors of the type $\boldsymbol{x}$ of mixing proportions inside the simplex, along with specification of their corresponding masses. In a sense, a mixture design is full of arbitrariness, in terms of the design points and their mass distribution. Generally, we first consider a collection of design points and then attribute a mass distribution to them. Loosely speaking, a collection of design points is also referred to as a mixture design. [The underlying mass distribution is tacitly understood to be defined at a subsequent stage, with a positive mass attributed to each design point in the collection.] In this sense, following three are the most commonly used standard mixture designs introduced by Scheffé (1958, 1963).

1.3.1 Simplex Lattice Designs

This class of designs consists of all feasible combinations of the mixing proportions wherein each proportion comprises of the values $(0, 1/m, 2/m, \ldots, m/m = 1)$ for a given integer parameter $m > 1$. Though there are $(m+1)^q$ possible combinations, only those combinations $(x_1, x_2, \ldots, x_q)$ are feasible which satisfy $x_1 + x_2 + \cdots + x_q = 1$. In a lattice design with q components and a given integer parameter m, the support set of design points is called a (q, m) simplex lattice. For a (q, m) simplex lattice design, there are $C(q + m - 1, m)$ design points where $C(a, b)$ stands for the usual binomial coefficient involving positive integers $a \geq b > 0$. .

For example, for a (4, 3) simplex lattice, the design points are given by [(1, 0, 0, 0) and its 3 variations, (1/3, 1/3, 1/3, 0) and its 3 variations, (2/3, 1/3, 0, 0) and its 11 variations]—with a total of 20 design points.

1.3.2 Simplex Centroid Designs

The centroid of a set of q nonzero coordinates in a q-dimensional coordinate system is the unique point $(1/q, 1/q, \ldots, 1/q)$. On the other hand, centroid of a set of t nonzero coordinates in a q-dimensional coordinate system is not unique. There are $C(q, t)$ centroid points of the form $[(1/t, 1/t, \ldots, 1/t, 0, 0, \ldots, 0); (1/t, 1/t, \ldots, 0, 1/t, 0, 0, \ldots, 0); \ldots\ldots (0, 0, \ldots, 0, 1/t, 1/t, \ldots, 1/t)]$.

A simplex centroid design deals exclusively with the centroids of the coordinate system, starting with exactly one nonzero component in the mixture (having q centroid points) and extending up to q nonzero components (having unique centroid point displayed above). Thus a simplex centroid design in the q-dimensional coordinate system contains $2^q - 1$ points.
As an example, for $q = 4$, there is a total of 15 points in the simplex centroid design:

[(1/4, 1/4, 1/4, 1/4); (1/3, 1/3, 1/3, 0); (1/3, 1/3, 0, 1/3); (1/3, 0, 1/3, 1/3); (0, 1/3, 1/3, 1/3); (1/2, 1/2, 0, 0); (1/2, 0, 1/2, 0); (1/2, 0, 0, 1/2); (0, 1/2, 1/2, 0); (0, 1/2, 0, 1/2); (0, 0, 1/2, 1/2); (1, 0, 0, 0); (0, 1, 0, 0); (0, 0, 1, 0); (0, 0, 0, 1)].

1.3.3 Axial Designs

It is to be noted that both simplex lattice and simplex centroid designs contain boundary points, i.e., points on the vertices, edges, and faces, *except* the centroid point $(1/q, 1/q, \ldots, 1/q)$ which lies inside the simplex. On the other hand, designs with interior points on the axis joining the points $x_i = 0, x_j = 1/(q-1),\ \forall\, j (\neq i)$ and $x_i = 1, x_j = 0,\ \forall\, j (\neq i)$ are called axial designs. Thus axial designs contain points of the form $[\{1+(q-1)\Delta\}/q, (1-\Delta)/q, \ldots, (1-\Delta)/q]$ and their permutations, $-1/(q-1) < \Delta < 1$. These designs are essentially 'interior point' designs.

For example, taking $q = 4$ and $\Delta = 0.20$, we can form an axial design with the points [(0.4, 0.2, 0.2, 0.2); (0.2, 0.4, 0.2, 0.2); (0.2, 0.2, 0.4, 0.2); (0.2, 0.2, 0.2, 0.4)].

Note that for $q = 4$, we have a choice of Δ such as $-0.33 < \Delta < 1.00$, and an axial design in general terms will be formed out of a few choices for Δ in the stated range.

In this monograph, we have dealt with such well-known mixture models and address the questions of optimal/efficient estimation of the model parameters and their meaningful functions. It turns out that the above three types of standard mixture designs occupy central stage during the investigation on optimal mixture experiments. These basic standard mixture designs will be discussed again in Chap. 3.

1.4 Exact Versus Approximate or Continuous Designs

An exact design deals with integer number of replications of the design points, thereby resulting into a design with a totality of an exact integer number of observations. For example, [(0.4, 0.3, 0.2, 0.1), (3); (0.2, 0.4, 0.1, 0.3), (4); (0.1, 0.1, 0.4, 0.4), (2); (0.1, 0.2, 0.2, 0.5), (4) produces an exact design with 4 distinct design points, with repeat numbers 3, 4, 2, 4, respectively, so that altogether 13 observations are produced upon its application. On the other hand, an example of a continuous design is given by [(0.4, 0.3, 0.2, 0.1), (0.3); (0.2, 0.4, 0.1, 0.3), (0.4); (0.1, 0.1, 0.4, 0.4), (0.2); (0.1, 0.2, 0.2, 0.5), (0.1). Here, again we have four distinct design points with respective 'mass' distribution given by 0.3, 0.4, 0.2, 0.1. In applications, for a given total number of observations, say $N = 30$ observations, the respective repeat numbers for the above design points are given by 9, 12, 6, 3. For N not a multiple of 10, we make nearest integer approximations as usual. The exact-design version of a continuous design has the above interpretation.

In optimality theory for 'regression models', almost exclusively, continuous design frameworks have been used. In this monograph as well, we will deal exclusively with this framework.

1.5 Applications of Mixture Methodology

Mixture experiments are commonly encountered in industrial product formulations, such as in food processing, chemical formulations, textile fibers, and pharmaceutical drugs. Some examples follow.

1. A large number of these experiments are also carried out in agriculture, where a fixed quantity of inputs such as fertilizer, irrigation water, insecticide, or pesticide

is applied as a mixture of two or more components to a crop. This makes the yield a function of the ingredient proportions.

2. In pharmaceutical drug preparation, polymers and diluents play important roles in the preparation of an inert matrix tablet, and a study of optimum mixture designs is necessary for the estimation of the parameters of the model defining the relationship between the mean response and the proportions of polymers and diluents used.
3. Intercropping is an important feature of dryland farming and has proved very useful for survival of small and marginal farmers in tropical and subtropical regions. In replacement series agricultural experiments, the component crop is introduced by replacing a part of the main crop. For fixed area under each experimental unit, the mean response is found to depend only on the proportions of the area allotted to the crops.
4. Experiments are conducted in food/horticulture technology to identify the best blending of fruit juice/pulp of lime, aonla, grape, pineapple, and mango that maximizes the responses (viz. hedonic scores on color, aroma, taste, and all taken together) among some specified mixing proportions. This presupposes establishing a relationship between the mean responses and the blending proportions, which helps to estimate the optimum blending.

1.6 Chapter-Wise Coverage of Topics

Not to obscure the flow of the chapters and the sustained interest of the readers, in Chap. 2, we make a concerted attempt to review the vast literature on optimal regression designs—admittedly much to the discontent of a serious reader—only to 'relate' to what is required for an understanding of the nature of optimal mixture experiments and the underlying optimality criteria—as discussed in this monograph! This is justified once we recognize that mixture models are effectively special types of regression models. Standard concepts of exact and approximate (or continuous) designs are well known; for the sake of completeness, these are introduced here. Next, in Chap. 3, we have introduced some of the commonly encountered mixture models and, thereafter, discussed about estimation of the underlying model parameters at length. This is done with reference to linear and quadratic homogeneous mixture models only. Some other models are deferred to latter chapters. Specific mixture designs with appealing features are also introduced in this chapter with the aim of setting the tone for the kind of optimal mixture experiments generally expected to be encountered in such studies.

Chapter 4 onwards, we deal exclusively with optimality studies in the context of mixture experiments and associated model parameters, or parametric functions with meaningful interpretations. The results are vast and varied and spread out in many directions. We progress in a manner that seemed most appealing to us in terms of the thought process of the researchers. We mention in passing that only the two standard optimality criteria [viz., A- and D-optimality] have been mostly dealt with in this monograph. In Chap. 4, we treat problems related to optimal estimation of natural parameters in mixture models due to Scheffé with reference to the unconstrained factor space [a simplex]; and in Chap. 5, we deal separately with some naturally arising constraints involving the factor space. Chapter 6 is meant for discussions on natural parameters in other mixture models. In the next four chapters (Chaps. 7–10), we discuss at length the problems associated with optimal estimation of some nonlinear functions of the model parameters. These functions arise naturally while one tries to maximize the ‘expected output’ as per the model specifications. Scheffé model, Darroch–Waller and Log-contrast models are taken up in these chapters. Lastly, we discuss some applications in Chap. 11 and a few miscellaneous diverse topics in Chap. 12. The topics are: robust mixture designs, optimality in Scheffé’s and Darroch–Waller models with random regression coefficients, optimality in mixture–amount model, multi-response mixture models, and mixture designs in blocks.

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Chapter 2
Optimal Regression Designs

Abstract In this chapter, we review the theory of optimum regression designs. Concept of continuous design and different optimality criteria are introduced. The role of *de la Garza phenomenon* and *Loewner order domination* are discussed. Equivalence theorems for different optimality criteria, which play an important role in checking the optimality of a given otherwise prospective design, are presented. These results are repeatedly used in later chapters in the search for optimal mixture designs. We present standard optimality results for single variable polynomial regression model and multivariate linear and quadratic regression model. Kronecker product representation of the model(s) and related optimality results are also discussed.

Keywords Continuous design · Optimality criteria · de la Garza phenomenon · Loewner order domination · Polynomial regression models · Equivalence theorem · Optimum regression designs

2.1 Introduction

In this chapter, we will discuss optimality aspects of regression designs in an *approximate* (or, *continuous*) design setting defined below.

Let y be the observed response at a point $(x_1, x_2, \ldots, x_k) = \boldsymbol{x}$ varying in some k-dimensional experimental domain $\mathcal{X}$ following the general linear model

$$y(\boldsymbol{x}) = \eta(\boldsymbol{x}, \boldsymbol{\beta}) + e(\boldsymbol{x}), \tag{2.1.1}$$

with usual assumptions on error component $e(\boldsymbol{x})$, viz. mean zero and uncorrelated homoscedastic variance σ^2; $\eta(\boldsymbol{x}, \boldsymbol{\beta})$ is the mean response function involving k or more unknown parameters. Once for all, we mention that $\boldsymbol{x}$ will represent a combination of the mixing components, the number of such components will be understood

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