SOME DESIGNS AND MODELS FOR MIXTURE EXPERIMENTS FOR THE SEQUENTIAL EXPLORATION OF RESPONSE SURFACES

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1. INTRODUCTION

Scheffe' (1958, 1963) introduced simplex-lattices and simplex-centroid designs for experiments with mixtures. The symmetric-simplex designs of Murty and Das (1968) are generalised form of these designs and are capable of uniform exploration of the simplex.

Box and Hunter (1957), in their pioneer work on exploration of response surfaces, emphasised that a response surface design should be such that it forms a nucleus from which a satisfactory design of order $(d+1)$ can be built in case the assumed polynomial of degree d proves inadequate. Mixture designs, so far available in literature, have the limitation of not being suitable for fitting into a sequential programme of exploring the response surfaces unless proper choice of mixtures is made. The present work is an effort in this direction. The case when certain process variables are present, has also been dealt with. Varying order models and designs involving mixture and process variables have been suggested which can be used for sequential exploration of response surfaces of both the types of variables.

2. MIXTURE DESIGNS FOR SEQUENTIAL EXPLORATION OF RESPONSE SURFACES

For an n-component mixture experiment Scheffe' (1958, 1963) suggested the fitting of following linear, quadratic and cubic models :

$$Y = \sum_{1 \leqslant i < n} \beta_i x_i \qquad \ldots(2\cdot1)$$

$$Y = \sum_{1 \leqslant i \leqslant n} \beta_i x_i + \sum_{1 \leqslant i < j \leqslant n} \beta_{ij} x_i x_j \qquad \ldots(2\cdot2)$$

and

$$Y = \sum_{1 \leqslant i \leqslant n} \beta_i x_i + \sum_{1 \leqslant i < j \leqslant n} \beta_{ij} x_i x_j + \sum_{1 \leqslant i < j < k \leqslant n} \beta_{ijk} x_i x_j x_k \qquad \ldots(2\cdot3)$$

where Y is ihe response, x_i is the ith mixture component and β's are the regression coefficients.

Let us now consider the following designs:

$$D_L \;.\quad n \text{ points of type } \left[\frac{1}{p}\ \frac{p-1}{(n-1)p}\ \frac{p-1}{(n-1)p}\cdots\frac{p-1}{(n-1)p}\right]$$

$$D_Q \;:\quad \binom{n}{2} \text{ points of type } \left[\frac{1}{p}\ \frac{1}{p}\ \frac{p-2}{(n-2)p}\ \frac{p-2}{(n-2)p}\cdots\frac{p-2}{(n-2)p}\right]$$

$$D_C \;:\quad \binom{n}{3} \text{ points of type } \left[\frac{1}{p}\ \frac{1}{p}\ \frac{1}{p}\ \frac{p-3}{(n-3)p}\ \frac{p-3}{(n-3)p}\cdots\frac{p-3}{(n-3)p}\right]$$

where p can take positive integral values depending upon the region of immediate interest.

If a linear model is desired to be fitted the design D_L may be more appropriate. Of these, some or all the points are to be repeated so as to obtain the error component. For providing d.f. for lack of fit component, one may also add either the total mixture $(1/n, 1/n\ldots\ldots 1/n)$ or n mixtures of the type $(1/n-1,\ 1/n-1\ldots\ldots,$ $(1/n-1,\ 0)$.

If the linear model is found unsatisfactory, the quadratic model (2·2) may be tried through mixture points of the initial design and the set of points D_Q. Similarly, for fitting the cubic model (2·3) one has to add further the design points D_Q.

It may be remarked here that all the design sets D_L, D_Q and D_C are special forms of the symmetric-simplex designs of Murty and Das (1968). We must emphasize that our choice is intended to make existing designs suitable for sequential exploration.

3. Varying Order Models in Presence of Process Variables

When certain process variables are present Murty and Das (1968) propose that a combined quadratic model involving quadratic terms both in mixture and process variables should be taken for approximating the response surface in the region pf interest. The estimation procedure for such a model is somewhat cumbersome.

Besides this difficulty in estimation, it would be difficult to say that mixture and process variable would exhibit a similar pattern in respect of their response surfaces For example, it is just possible that the mixture variables may regress in a quadratic fashion while the process variables may not—the process variables may have a linear or cubic surface. The experimental conditions may, also some times, restrict the number of levels for process variables to two, and thereby make the design unsuitable for a quadratic or higher order model in process variables.

Thus, it is observed that the model should be such that it allows the mixture variables to have any order say, d_1 and the process variables to have the order d_2 irrespective of each other. When $d_1=d_2=2$, the combined model is the same as suggested by Murty and Das (1958).

Consider now the model with $d_1=d_2=1$:

$$Y=\beta'_0+\sum_i \beta_i' x_i+\sum_j \beta_j' z_j+\sum_{i,j} \beta_{ij}' x_i z_j \qquad \ldots(3\cdot1)$$

which reduces to

$$Y=\sum_i \beta_i x_i+\sum_{i,j} \beta_{ij} x_i z_j \qquad \ldots(3\cdot2)$$

by virtue of the substitutions

$$\beta_i=\beta_0'+\beta_i' \text{ and } \beta_{ij}=\beta'_{ij}+\beta'_j \qquad \ldots(3\cdot3)$$

where z_j is the j-th process variable ; $j=1, 2, \ldots, p$.

The term $\Sigma\ \beta_{ij}' x_{iu} z_{ju}$ in (3·1) has been included with a special motive of examining the behaviour of the mixtures over different level combinations of process variables. It may be observed from (3·2) and (3·3) that the main effects of the process variables are not estimable as such, but they are confounded with the interaction between process and mixture variables.

The estimation of the parameters of the model (3·2) and the models which follow can be easily done on the lines suggested by Murty and Das (1968). It can be seen that for the designs that we suggest in section 4, the parameters β_i and β_{ij} of the model (3·2) are orthogonal to each other.

The sequential programme starts with the fitting of the model (3·2). If the test for lack of fit indicates inadequate exploration of the response surface, the following models (3·4) and (3·5) with other values of d_1 and d_2 may be tried one by one. Since β_i and β_{ij} are orthogonal to each other, the regression *S.S.* can be partitioned into these two components. In case of signifiance of β_i, the model (3·5) may be tried first. One can pass on to the model of Murty and Das (1968) with $d_1=d_2=2$, if these models prove to be inadequate, (*i*) $d_1=1, d_2=2$

$$Y= \sum_{i}\beta_i x_i + \sum \beta_{ij} z_j^2+\sum_{j<j'}\beta_{jj'} z_j z_j'+ \sum_{i,j}\beta_{ij} x_{j'} z_j \quad ...(3\cdot 4)$$

(*ii*) $d_1=2, d_2=1$

$$Y= \sum_{i} \beta_i x_i +\sum_{i<i'} \beta_{ii}' x_i x_i' + \sum_{i,j}\beta_{ij} x_i z_j \quad ...(3\cdot 5)$$

4. Design for Fitting the Models

For the models (3·2) and (3·5), the mixture variables may be chosen in accordance with the arguments formulated in sections 2·1 and 2·3. For process variables, a 2^p factorial design with level as -1 and 1 is adopted. It may be observed that these designs are again special forms of symmetric-simplex x Factorial designs of Murty and Das (1968).

For other models, the process variable part of the design is augmented by adding $(2p+1)$ process variable points of the type $(0, 0, ..., 0)$, $(\pm\alpha, 0, ..., 0)$ $(0, \pm\alpha, 0, ... 0)$, ..., $(0, 0, ...\pm\alpha)$ to make it a central composite design of Box and Wilson (1951). The value of α can be chosen to make the regression coefficients orthogonal to one another or to minimise the bias that arises if the true form of the process variable part of the response surface is not quadratic. The value of α can be also so chosen that the central composite design becomes a second order rotatable design of Box and Hunter (1957).

It is interesting to note that the choice of a central composite or a second order rotatable design for process variables has some distinct advantages over the design of Scheffe' (1963), and Murty and Das (1968) where an s^p, $(s \geqslant 3)$ factorial design is taken for process variables. If a central composite design is chosen, the estimation problems are much simplified because all the sums of powers or products of powers of z_{ju} and all the sums of products of powers of x_{iu} and z_{ju} in which at least one power of z_{ju} is odd are zero. Further, if the central composite design is a second order rotatable design of Box and Hunter (1957), then it also satisfies the condition $\Sigma z_j^4 = 3 \Sigma z_j^2 z_j'^2$. Another advantage with the choice of central composite design is that the number of design points will always be considerably less compared to the number in a complete or fractional 3^p factorial design. Further, the coefficients β_{jj} of the squared terms are estimated with relatively low precision from a 3^p factorial as pointed out by Box and Wilson (1951).

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