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QUALITY IMPROVEMENT THROUGH EXPERIMENTS WITH MIXTURES

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1. Introduction

Mixture experiments are typical for chemical, food, metallurgical and other industries. The aim of these experiments is to find optimal component proportions that provide desired values of some product performance characteristics. Let A_i, $i = 1, 2, \ldots, q$ represent the amounts of the components in a mixture. The component proportions can be expressed as follows:

$$X_i = \frac{A_i}{\sum\limits_{i=1}^{q} A_i}. \tag{1}$$

They are subject to following constraints:

$$0 \le X_i \le 1, \qquad i = 1, 2, \ldots, q$$

$$\sum_{i=1}^{q} X_i = 1. \tag{2}$$

Let η is the undisturbed performance characteristic of a product. Usually there are disturbances in the production process and as a result the measured value of the performance characteristic is $y = \eta + \varepsilon$. Sheffé (1958) proposed canonical models for mixture experiments. A second order canonical model is:

$$\eta = \sum_{i=1}^{q} \beta_i X_i + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} \beta_{ij} X_i X_j. \tag{3}$$

First order canonical model can be obtained from (3) if we put $\beta_{ij} = 0$. In this paper we consider only first and second order canonical models, but the results can easily be extended for other canonical models.

In the production process errors occur due to poor measurements or violations of technological prescriptions. Let e_i, $i = 1, 2, \ldots, q$ are the errors in component amounts. The component proportions in a mixture experiment that is subject to errors in the component amounts are:

$$x_i = \frac{A_i + e_i}{\sum\limits_{i=1}^{q} (A_i + e_i)} \tag{4}$$

It is easy to see that for these components constraints (2) also hold.

The errors in component amounts are transmitted to the response and cause variations in the performance characteristics, which exist even if $\varepsilon = 0$. Vuchkov and Boyadjieva (2001) proposed a model-based approach to description of error transmission for regression experiments, which are not subject to constraints (2). In the polynomial models with errors the product or process parameters are expressed as $p_i + e_i$ and the performance characteristic mean and variance models are developed. In mixture experiments this approach cannot be directly used, because the errors occur both in the nominator and denominator of component proportions. Therefore the model is nonlinear with respect to errors and the distribution moments cannot be directly calculated.

Steiner and Hamada (1997) considered the problem of error transmission in mixture experiments. They used simulations to estimate the transmitted variance. To do that they need the relative probability of each simulated point. In this paper we propose an analytical approach that is free of simulations.

2. Mean and variance models for mixture experiments

Vuchkov and Boyadjieva (2001), pages 390-391, used Taylor expansion for nonlinear performance characteristic η. Mean and variance models were obtained for product performance characteristics with factors that are subject to errors e_i with zero expectations and variances $\sigma^2(e_i)$. These models can be used for description of mean and variance in mixture experiments. In this context and if the errors are independent and normally distributed, the models can be written as follows:

Mean value:

$$\tilde{y} = \eta + \sum_{i=1}^{q} h_{ii} \sigma^2 (e_i), \tag{5}$$

Variance:

$$\sigma^2 = \sum_{i=1}^{q} \delta_i^2 \sigma^2(e_i) + 2\sum_{i=1}^{q} h_{ii}^2 \sigma^4(e_i) + 4\sum_{i=1}^{q-1}\sum_{j=i+1}^{q} h_{ij}^2 \sigma^2(e_i)\sigma^2(e_j) +$$

$$+2\sum_{i=1}^{q} \delta_i \sigma^2(e_i)\left(3\sigma^2(e_i)\, t_{ii}^{(i)} + \sum_{j=1, j\neq i}^{q} \sigma^2(e_j)\, t_{jj}^{(i)} + 2\sum_{j=1, j\neq i}^{q} \sigma^2(e_i)\, t_{ij}^{(i)}\right) + \sigma_\varepsilon^2. \tag{6}$$

Following notations are used in these formulae:

$$\delta_i = \frac{\partial \eta}{\partial A_i}, \tag{7}$$

$$h_{ii} = \frac{1}{2}\frac{\partial^2 \eta}{\partial A_i^2}, \quad h_{ij} = \frac{1}{2}\frac{\partial^2 \eta}{\partial A_i \partial A_j}, \tag{8}$$

$$t_{ii}^{(i)} = \frac{1}{3!}\frac{\partial^3 \eta}{\partial A_i^3}, \quad t_{jj}^{(i)} = \frac{1}{3!}\frac{\partial^3 \eta}{\partial A_i \partial A_j^2}, \quad t_{ij}^{(i)} = \frac{1}{3!}\frac{\partial^3 \eta}{\partial A_i^2 \partial A_j} \tag{9}$$

The derivatives for the second order mixture model are:

$$\delta_i = \frac{1}{\sum\limits_{i=1}^{q} A_i}\Delta_i, \tag{10}$$

$$h_{ii} = \frac{1}{\left(\sum\limits_{i=1}^{q} A_i\right)^2} H_{ii}, \quad h_{ij} = \frac{1}{\left(\sum\limits_{i=1}^{q} A_i\right)^2} H_{ij}, \tag{11}$$

$$t_{ii}^{(i)} = \frac{1}{\left(\sum\limits_{i=1}^{q} A_i\right)^3} T_{ii}^{(i)}, \quad t_{ii}^{(j)} = \frac{1}{\left(\sum\limits_{i=1}^{q} A_i\right)^3} T_{ii}^{(j)}, \quad t_{ij}^{(j)} = \frac{1}{\left(\sum\limits_{i=1}^{q} A_i\right)^3} T_{ij}^{(j)}, \tag{12}$$

where the values of Δ_i, H_{ii}, H_{ij}, $T_{ii}^{(i)}$, $T_{ii}^{(j)}$, $T_{ij}^{(j)}$ can be written through the undisturbed component proportions as follows:

$$\Delta_i = \beta_i (1 + X_i) + \sum_{i\neq j} (\beta_j + \beta_{ij}) X_j - 2\eta, \tag{13}$$

$$H_{ii} = \eta + 2\sum_{u=1}^{q-1}\sum_{v=u+1}^{q} \beta_{uv} X_u X_v - 2\sum_{u\neq i} \beta_{iu} X_u - \beta_i, \tag{14}$$

$$H_{ij}{=}\eta{+}2\sum_{u=1}^{q-1}\sum_{v=u+1}^{q} \beta_{uv} X_u X_v {-} \beta_{ij}(X_i{+}X_j) {-} \sum_{u\neq i}(\beta_{iu}{+}\beta_{ju})X_u {+} (\beta_{ij} - \beta_i - \beta_j)/2,$$

$$T_{ii}^{(i)} = \beta_i + 3(1 - X_i)\sum_{u\neq i} \beta_{iu} X_u - 3\sum_{u\neq i}\sum_{v\neq i,u} \beta_{uv} X_u X_v - \eta, \tag{15}$$

$$T_{ii}^{(j)}{=}T_{ij}^{(i)}{=}\frac{1}{3}(2\beta_i{+}\beta_j){+}\frac{1}{3}\beta_{ij}(3X_i{+}6X_j{-}9X_iX_j{-}2){+}(2{-}3X_i)\sum_{u\neq i,j} \beta_{iu} X_u +$$

$$+ (1 - 3X_j)\sum_{u\neq i,j} \beta_{ju} X_u - 3\sum_{u\neq i,j}\sum_{v\neq i,j,u} \beta_{uv} X_u X_v - \eta, \tag{16}$$

$$T_{jj}^{(i)} = T_{ij}^{(j)} \tag{17}$$

The proportion of the error in a mixture component is $e_i/\sum A_i$. Its variance is equal to

$$\sigma_i^2 = \frac{\sigma^2(e_i)}{\left(\sum A_i\right)^2}.$$

Therefore the proportion's standard deviation is

$$\sigma(e_i) = \sigma_i\left(\sum A_i\right). \tag{18}$$

Putting (10)-(12) and (18) in (5) and (6) we obtain mean and variance models expressed only through component proportions, and not the component amounts in explicit form. These models are as follows:

Mean value:

$$\tilde{y} = \eta + \sum_{i=1}^{q} H_{ii}\sigma_i^2, \tag{19}$$

Variance:

$$\sigma^2 = \sum_{i=1}^{q} \Delta_i^2\sigma_i^2 + 2\sum_{i=1}^{q} H_{ii}^2\sigma_i^4 + 4\sum_{i=1}^{q-1}\sum_{j=i+1}^{q} H_{ij}^2\sigma_i^2\sigma_j^2 +$$

Table 1. Second order simplex design for rubber mixture

No.	X_1	X_2	X_3	y
1	1	0	0	100
2	0	1	0	69.5
3	0	0	1	65.5
4	0.5	0.5	0	77.5
5	0.5	0	0.5	71.5
6	0	0.5	0.5	82

$$+2\sum_{i=1}^{q}\Delta_i\sigma_i^2\left(3\sigma_i^2T_{ii}^{(i)}+\sum_{j=1,j\neq i}^{q}\sigma_j^2T_{jj}^{(i)}+2\sum_{j=1,j\neq i}^{q}\sigma_j^2T_{ij}^{(i)}\right)+\sigma_\varepsilon^2. \tag{20}$$

These models make it possible to calculate the mean and variance values of the performance characteristic in any point of the simplex and this way to improve the product quality.

Minimization of the transmitted error variance

The quality improvement problem can be defined as minimization of variance (20) under the condition that the mean value of the performance characteristic is on target. Other method is to minimize the Taguchi's loss function, which for our problem can be written as follows (Taguchi, 1996):

$$L=k\left[(\tilde{y}-\tau)^2+\sigma^2\right] \tag{21}$$

In this formula $\tau$is target value for the performance characteristic, while $\tilde{y}$ and σ^2 can be calculated by use of formulae (19) and (20).

Example

Consider a formulation for protector of truck tires. The performance characteristic of interest y is modulus of 300 % elongation of the mixture formulation. It is studied as function of three types of synthetic rubber: X_1 (Bulex 1500), X_2 (Bulex M-27) and X_3(SKD). A second order simplex lattice is used and the results of experiments are given in Table 1.

Following canonical model was found and proved to be adequate:

$$\hat{y}=100X_1+69.5X_2+65.6X_3-29X_1X_2-45X_1X_3+58X_2X_3. \tag{22}$$

In order to study the transmitted variation we take into account that there are not errors when some of the component proportions equals 0. That is why all calculations were carried out for points without zero proportions. We considered a smaller simplex defined by the following vertices: V_1 (0.1;0.8;0.1), V_2 (0.1;0.1;0.8) and V_3 (0.8;0.1;0.1). The values of biased response in the vertices and centers of the edges were calculated by use of formula (19) and the biased response was approximated by use of second order canonical model. We obtained the following model for the biased response:

$$\tilde{y} = 102.13X_1 + 68.65X_2 + 64.5X_3 - 31.61X_1X_2 - 49.05X_1X_3 + \\ + 63.22X_2X_3 \tag{23}$$

There is some difference in the coefficients of (23) as compared with (22), which is due to the errors in the component proportions. Contours of the predicted and biased responses based on (22) and (23) are on Fig. 1 and Fig. 2.

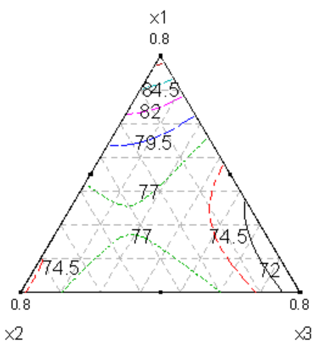

Fig.1. Predicted response

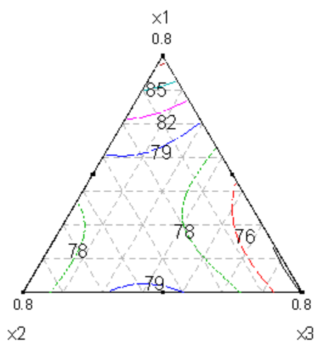

Fig. 2. Biased response

Variance was also calculated in the vertices and edge centers of the restricted simplex area by formula (20). The response variance was neglected in this model. The following canonical model approximated it:

$$\sigma^2 = 97.3X_1 + 40.5X_2 + 56.5X_3 - 251.7X_1X_2 - 252.6X_1X_3 - \\ - 133.7X_2X_3 + 190.2X_1X_2X_3$$

Fig. 3 shows the contours of variance transmitted from factors to the response. It is interesting to note that the minimum of variance can be obtained for component proportions corresponding to the saddle point of the response surface, where the first derivatives of the response are equal to zero. An explanation of this property for second order polynomial models of the response is given in Vuchkov and Boyadjieva (2001), pages 294-299. This example shows that this property holds also for second order canonical models too.

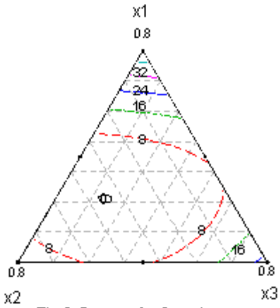

Fig. 3. Contour plot for variance

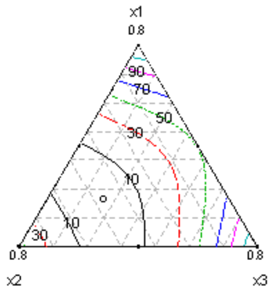

Fig. 4. Contour plot for L

The optimal component proportions depend on the target definition. For example if the modulus of 300 % elongation is required to be no more than 80, then from Fig. 2 and Fig. 3 we see that the optimal proportions that make the variance equal to 0 are approximately $X_1 = 0.31, X_2 = 0.45, X_3 = 0.24$. Fig. 4 shows other solution based on Taguchi's loss function with target $\tau = 80$. In this case the optimal proportions are: $X_1 = 0.26, X_2 = 0.48, X_3 = 0.26$. These two solutions are not too different.

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