# EXPERIMENTS WITH MIXTURES

## AN EXTENTION OF THE MODIFICATION TO THE SIMPLEX - CEDROID DESIGN

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SUMMARY

Scheffè (1963) introduced the Simplex-Centroid Design. Lambrakis (1966) introduced the idea of a modification to this design in which the mixtures of one component, pure mixtures, are replaced by mixtures of q-1 components with equal proportions, (q-1) - nary mixtures. He produced results for the quadratic and cubic cases. In this paper the modification is extended to the quartic case.

### 1. INTRODUCTION

Scheffè (1963) introduced and developed the Simplex-Centroid Design. The main features of this design, in a q-component mixture in which the proportions $x_1, x_2, \ldots, x_q$ of the coponents are in the simplex

$$\sum_{i=1}^{q} x_i = 1, \qquad x_j \geqq 0, \tag{1.1}$$

are the centroid polynomial

$$\eta = \sum_{i=1}^{q} a_i x_i + \sum_{1 \leqq i < j \leqq q} a_{ij} x_i x_j + \sum_{1 \leqq i < j < k \leqq q} a_{ijk} x_i x_j x_k + \cdots + \sum_{1 \leqq i_1 < i_2 < \cdots < i_n \leqq q} a_{i_1 i_2 \ldots i_n} x_{i_1} x_{i_2} \ldots x_{i_n} \tag{1.2}$$

as a regressiou function, and the mixtures of one, two, three, . . . , q components with equal proportions. These mixtures are to be used to estimate the coefficients in the polynomial (1.2). The purpose of this design is the empirical prediction of the response to any mixture of the components when the response depens donly on the proportions of the components present but not on the total amount of the mixture.

Lambrakis (1966) introduced the idea of a modification to this design. In this modification the mixtures of one component, pure mixtures, are replaced by mixtures of q-1 components of equal proportions, (q-1) - nary mixtures. He gave results for the quadratic and cubic cases. The quadratic case was published in 1969 in the J. R. Statist. Soc., series B, as a part of his paper entitled «An alternative to the simplex-lattice design for experiments with mixtures». The cubic case remains unpublished in his Ph. D. Thesis.

In this paper the above mentioned modification is extended to the quartic case and the results of the estimation are included.

## 2. THE QUARTIC GASE

the polynomial is

$$\eta = \sum_{i=1}^{q} a_i x_i + \sum_{1 \leq i < j \leq q} a_{ij} x_i x_j + \sum_{1 \leq i < j < k \leq q} a_{ijk} x_i x_j x_k + \sum_{1 \leq i < j < k < l \leq q} a_{ijkl} x_i x_j x_k x_l \tag{2.1}$$

The mixtures to be used to estimate the coefficients in (2.1) are:

q $(q-1)$—nary $g_i$ $(1 \leq i \leq q)$ with proportions

$$x_1 = x_2 = \ldots = x_{i-1} = x_{i+1} = \ldots = x_q = 1/(q-1)$$

$\binom{q}{2}$ binary $u_{ij}$ $(1 \leq i < j \leq q)$ with proportions $x_i = x_j = 1/2$,

$\binom{q}{3}$ ternary $w_{ijk}$ $(1 \leq i < j < k \leq q)$ with proportions $x_i = x_j = x_k = 1/3$,

$\binom{q}{4}$ quartenary $f_{ijkl}$ $(1 \leq i < j < k < l \leq q)$ with proportions $x_i = x_j = x_k = x_j = 1/4$.

Taking $r_{ij}$, $r_{ijk}$, $r_{ijkl}$, $r_i$ observations for $u_{ij}$, $w_{ijk}$, $f_{ijkl}$, $g_i$ respectively and substituting their observed means $\hat{u}_{ij}$, $\hat{w}_{ijk}$, $\hat{f}_{ijkl}$, $\hat{g}_i$ into (2.1) we have the normal equatios

$$\hat{u}_{ij} = \frac{1}{2}(\hat{a}_i + \hat{a}_j) + \frac{1}{4}\hat{a}_{ij} \tag{2.2}$$

$$\hat{w}_{ijk} = \frac{1}{3}(\hat{a}_i + \hat{a}_j + \hat{a}_k) + \frac{1}{9}(\hat{a}_{ij} + \hat{a}_{ik} + \hat{a}_{jk}) + \frac{1}{27}\hat{a}_{ijk} \tag{2.3}$$

$$\hat{f}_{ijkl} = \frac{1}{4}(\hat{a}_i + \hat{a}_j + \hat{a}_k + \hat{a}_l) + \frac{1}{16}(\hat{a}_{ij} + \hat{a}_{ik} + \hat{a}_{il} + \hat{a}_{jk} + \hat{a}_{jl} + \hat{a}_{kl})$$

$$+ \frac{1}{64}(\hat{a}_{ijk} + \hat{a}_{ijl} + \hat{a}_{ikl} + \hat{a}_{jkl}) + \frac{1}{256}\hat{a}_{ijkl}, \qquad (2.4)$$

$$\hat{g}_i = \frac{1}{q-1} \sum_{\substack{1 \leq r \leq q \\ r \neq i}} \hat{a}_r + \frac{1}{(q-1)^2} \sum_{\substack{1 \leq r < s \leq q \\ r,s \neq i}} \hat{a}_{rs} + \frac{1}{(q-1)^3} \sum_{\substack{1 \leq r < s < t \leq q \\ r,s,t \neq i}} \hat{a}_{rst}$$

$$+ \frac{1}{(q-1)^4} \sum_{\substack{1 \leq r < s < t < v \leq q \\ r,s,t,v \neq i}} \hat{a}_{rstv} \qquad (2.5)$$

Solving the above system of normal equations, we obtain the least - squares estimates of the coefficients

$$\hat{a}_i = \frac{48}{(q-1)(q-3)}[(q-2)\sum\hat{u} - (q-1)\sum\hat{u}_{\bar{i}}]$$

$$- \frac{486}{(q-1)(q-3)(q-4)}[(q-3)\sum\hat{u} - (q-1)\sum\hat{u}_{\bar{i}}]$$

$$+ \frac{1536}{(q-1)(q-3)(q-4)(q-5)}[(q-4)\sum\hat{f} - (q-1)\sum\hat{f}_{\bar{i}}]$$

$$- \frac{6(q-1)^3}{(p-3)(q-4)(q-5)}[\sum\hat{g} - (q-1)\hat{g}], \qquad (2.6)$$

$$\hat{a}_{ij} = 4\hat{u}_{ij} - \frac{96}{(q-1)(q-3)}[2(q-2)\sum\hat{u} - (q-1)(\sum\hat{u}_{\bar{i}} + \sum\hat{u}_{\bar{j}})]$$

$$+ \frac{972}{(q-1)(q-3)(q-4)}[2(q-3)\sum\hat{w} - (q-1)(\sum\hat{w}_{\bar{i}} + \hat{w}_{\bar{j}})]$$

$$- \frac{3072}{(q-1)(q-3)(q-4)(q-5)}[2(q-4)\sum\hat{f} - (q-1)(\sum\hat{f}_{\bar{i}} + \sum\hat{f}_{\bar{j}})]$$

$$+ \frac{12(q-1)^3}{(q-3)(q-4)(q-5)}[2\sum\hat{g} - (q-1)(\hat{g}_i + \hat{g}_j)] \qquad (2.7)$$

$$\hat{a}_{ijk} = 27\hat{w}_{ijk} - 12(\hat{u}_{ij} + \hat{u}_{ik} + \hat{u}_{jk})$$

$$+ \frac{144}{(q-1)(q-3)}[3(q-2)\sum\hat{u} - (q-1)(\sum\hat{u}_{\bar{i}} + \sum\hat{u}_{\bar{j}} + \sum\hat{u}_{\bar{k}})]$$

$$- \frac{1458}{(q-1)(q-3)(q-4)}[3(q-3)\sum\hat{w} - (q-1)(\sum\hat{w}_{\bar{i}} + \sum\hat{w}_{\bar{j}} + \sum\hat{w}_{\bar{k}})]$$

$$+ \frac{4708}{(q-1)(q-3)(q-4)(q-5)}[3(q-4)\sum\hat{f} - (q-1)(\sum\hat{f}_{\bar{i}} + \sum\hat{f}_{\bar{j}} + \sum\hat{f}_{\bar{k}})]$$

$$- \frac{18(q-1)^3}{(q-3)(q-4)(q-5)}[3\sum\hat{g} - (q-1)(\hat{g}_i + \hat{g}_j + \hat{g}_k)], \qquad (2.8)$$

$$\hat{a}_{ijkl} = 256\, \hat{f}_{ijkl} - 108\, (\hat{w}_{ijk} + \hat{w}_{ijl} + \hat{w}_{ikl} + \hat{w}_{jkl})$$
$$+ 32\, (\hat{u}_{ij} + \hat{u}_{ik} + \hat{u}_{il} + \hat{u}_{jk} + \hat{u}_{jl} + \hat{u}_{kl})$$
$$- \frac{192}{(q-1)(q-3)} \left[4(q-2) \sum \hat{u} - (q-1)\left(\sum \hat{u}_{\bar{i}} + \sum \hat{u}_{\bar{j}} + \sum \hat{u}_{\bar{k}} + \sum \hat{u}_{\bar{l}}\right)\right]$$
$$+ \frac{1944}{(q-1)(q-3)(q-4)} \left[4(q-3) \sum \hat{w} - (q-1)\left(\sum \hat{w}_{\bar{i}} + \sum \hat{w}_{\bar{j}} + \sum \hat{w}_{\bar{k}} + \sum \hat{w}_{\bar{l}}\right)\right]$$
$$- \frac{6144}{(q-1)(q-3)(q-4)(q-5)} \left[4(q-4) \sum \hat{f} - (q-1)\left(\sum \hat{f}_{\bar{i}} + \sum f_{\bar{j}} + \sum \hat{f}_{\bar{k}} + \sum \hat{f}_{\bar{l}}\right)\right]$$
$$+ \frac{24(q-1)^3}{(q-3)(q-4)(q-5)} \left[4 \sum \hat{g} - (q-1)(\hat{g}_i + \hat{g}_j + \hat{g}_k + \hat{g}_l)\right] \qquad (2.9)$$

Where

$$\sum \hat{g} = \sum_{i=1}^{q} \hat{g}_i,$$

$$\sum \hat{u} = \sum_{1 \leq i < j \leq q} \hat{u}_{ij}, \quad \sum \hat{u}_{\bar{\nu}} = \sum_{\substack{1 \leq r < s \leq q \\ r,s \neq \nu}} \hat{u}_{rs} \; (\nu = i, j, k, l)$$

$$\sum \hat{w} = \sum_{1 \leq j < j < k \leq q} \hat{w}_{ijk}, \quad \sum \hat{w}_{\bar{\nu}} = \sum_{\substack{1 \leq r < s < t \leq q \\ r,s,t \neq \nu}} \hat{w}_{rst} \; (\nu = i, j, k, l)$$

$$\sum \hat{f} = \sum_{1 \leq i < j < k < l \leq q} \hat{f}_{ijkl}, \quad \sum \hat{f}_{\bar{\nu}} = \sum_{\substack{1 \leq r < s < t < v \leq q \\ r,s,t,v \neq \nu}} \hat{f}_{rstv} \; (\nu = i, j, k, l).$$

Substituting the above least-squares estimates into (2.1) and collecting terms, we obtain the estimated regression function

$$\tilde{\eta} = \sum_{i=1}^{q} \mathbf{a}_i \hat{g}_i + \sum_{1 \leq i < j \leq q} \mathbf{a}_{ij} \hat{u}_{ij} + \sum_{1 \leq i < j < k \leq q} \mathbf{a}_{ijk} \hat{w}_{ijk} + \sum_{1 \leq i < j < k < l \leq q} \mathbf{a}_{ijkl} \hat{f}_{ijkl} \qquad (2.10)$$

where

$$\mathbf{a}_i = - \frac{(q-1)^4}{(q-3)(q-4)(q-5)} \left[\left(1 - 3 \sum x^2 + 8 \sum x^3\right) x_i - 6\left(1 - 2 \sum x^2\right) x_i^2 + 6 x_i^3 - 24 x_i^4\right]$$
$$+ \frac{(q-1)^3}{(q-3)(q-4)(q-5)} \left[1 - 9 \sum x^2 + 14 \sum x^3 - 24 \sum x^4 + 12 \left(\sum x^2\right)^2\right],$$

$$\mathbf{a}_{ij} = 4 x_i x_j \left[2 - 5(x_i + x_j) + 4(x_i^2 + x_j^2) + 4(x_i + x_j)^2 - 4 \sum x^2\right]$$
$$- \frac{8}{q-3} \sum_{r = i,j} \left[\left(1 - 3 \sum x^2 + 8 \sum x^3\right) x_r - 6\left(1 - 2 \sum x^2\right) x_r^2 + 6 x_r^3 - 24 x_r^4\right]$$
$$+ \frac{8}{(q-1)(q-3)} \left[\left(1 - 9 \sum x^2 + 14 \sum x^3 - 24 \sum x^4 + 12 \left(\sum x^2\right)^2\right]$$

$$a_{ijk} = -27\, x_i\, x_j\, x_k \left[3 - 4\,(x_i + x_j + x_k)\right]$$

$$+ \frac{81}{(q-3)(q-4)} \sum_{r=i,j,k} \left[(1 - 3 \sum x^2 + 8 \sum x)\; x_r - 6\,(1 - 2 \sum x^2) x_r^2 + 6 x_r^3 - 24 x_r^4\right]$$

$$- \frac{162}{(q-1)(q-3)(q-4)} \left[1 - q \sum x^2 + 14 \sum x^3 - 24 \sum x^4 + 12\; (\sum x^2)^2\right],$$

$$a_{ijkl} = 256\; x_i x_j x_k x_l$$

$$- \frac{256}{(q-3)(q-4)(q-5)} \sum_{r=i,j,k,l} \left[(1 - 3 \sum x^2 + 8 \sum x^3)\; x_r - 6\,(1 - 2 \sum x^2)\, x_r^2 + 6\, x_r^3 - 24\, x_r^4\right]$$

$$+ \frac{768}{(q-1)(q-3)(q-4)(q-5)} \left[1 - q \sum x^2 + 14 \sum x^3 - 24 \sum x^4 + 12\,(\sum x^2)^2\right]$$

Note that $\sum x^\nu$ stads for $\sum_{i=1}^{q} x_r^\nu$ where $\nu = 2, 3, 4.$

The above results can be applied for any $q \geqq 6$.

A way of checking the above results is to substitute into (2.10) the proportions of the mixtures used and see whether or not expression (2.10) predicts the mean responses of the mixtures. Substituting into (2.10) the proportions of the mixtures used, we verify that

| $a_i$ | $a_{ij}$ | $a_{ijk}$ | $a_{ijkl}$ | $\tilde{\eta}$ |
|---|---|---|---|---|
| 1 | 0 | 0 | 0 | $\hat{g}_i$ |
| 0 | 1 | 0 | 0 | $\hat{u}_{ij}$ |
| 0 | 0 | 1 | 0 | $\hat{w}_{ijk}$ |
| 0 | 0 | 0 | 1 | $\hat{f}_{ijkl}$ |

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