# BLOCKING OF MIXTURE EXPERIMENTS 

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#### Abstract

A mixture experiment involves mixing various proportions of two or more components to make different compositions of an end product. The methods of analysis of experiments with mixtures seem to be relevant and useful in many areas of agricultural experiment and industrial experiment. The purpose of present study is to analyze the mixture experiments in the presence of block effects by considering linear and quadratic response models.


KEYWORDS: Block Effects, Linear Model, Mixture Experiment, Orthogonal Blocking, Quadratic Model

## Article History

Received: 05 Mar 2018 | Revised: 04 May $2018 \mid$ Accepted: 12 May 2018

## INTRODUCTION

In experiments dealing with mixtures, the characteristics (response) studied depends on the relative proportion of ingredients forming mixture, and the proportions of components will be considered as mixture variables and will be denoted by $\mathrm{X}_{\mathrm{i}}, \mathrm{i}=1,2, \ldots \ldots, \mathrm{k}$.

These proportions ( $X_{i}$ 's) satisfy the restrictions $X_{i} \geq 0$, s for $i=1,2, \ldots . . k$ and $\sum_{i=1}^{k} X_{i}=1$.
The design of mixture experiments has been extensively used in agricultural and industrial experiments. Some of the situations in which these designs could be advantageously used are: split application of fertilizers, intercropping experiments where the interest of the experimenter is to find best crop mixtures, sensory evaluation experiments for making the agricultural and animal products, preparation of fertilizers, insecticides/pesticides mixtures for optimum response, feeding trials in the animal nutritional experiment.

Scheffe $(1958,1963)$ was the first to introduce the concept of mixture experiments and their analysis. The work of Scheffe was extended by Gorman and Hinman (1962), Draper and Lawrance $(1965,1965)$ and Becker $(1970)$. Lambrakis $(1968,1969)$ constructed designs in which all the components of a mixture were present. Murty and Das (1968) considered symmetric simplex design in respect of mixture experiments. According to Box and Hunter (1957), one of the requirements of any response surface is that it should lend itself to blocking. Nigam (1970, 1976) and Murthy and Murty (1992) considered the blocking of designs for mixture experiments. The objective of the present paper is to review the works of Murty (1966), Nigam (1970,1976) and Murthy and Murty (1992) on blocking in mixture experiments with a view
to assess and extend existing knowledge on blocking conditions. For this purpose, analysis of mixture experiments in the presence of block effect is performed considering linear and quadratic response models.

## ANALYSIS OF MIXTURE EXPERIMENTS IN PRESENCE OF BLOCK EFFECTS

## Linear Model

Let us suppose that there are $n$ mixtures distributed in $b$ blocks having $n_{w}(w=1,2, \ldots . b)$ mixtures in the $w^{\text {th }}$ block such that $\sum_{\mathrm{w}=1}^{\mathrm{b}} \mathrm{n}_{\mathrm{w}}=\mathrm{n}$, and a linear model,

$$
\begin{equation*}
Y_{u}=\beta_{1} X_{1 u}+\beta_{2} X_{2 u}+\ldots \ldots . .+\beta_{\mathrm{k}} X_{k u}+\Gamma_{1} Z_{1 u}+\Gamma_{2} Z_{2 u}+\ldots+\Gamma_{\mathrm{b}} Z_{b u}+e_{u} \tag{1.1}
\end{equation*}
$$

Is used to approximate the response Y where $\mathrm{u}=1,2,3$, $\qquad$ n and Yu is the observed response at the $\mathrm{u}^{\text {th }}$ point, $\mathrm{X}_{\mathrm{i}}(\mathrm{i}=1,2, \ldots \mathrm{k})$ is the $\mathrm{i}^{\text {th }}$ mixture variable, $\beta_{\mathrm{i}}$ is the regression coefficient of Y on $\mathrm{X}_{\mathrm{i}}, \Gamma_{w}$ is the effect of the $\mathrm{w}^{\text {th }}$ block.

$$
\begin{aligned}
z_{w u} & =1, \text { if } \mathrm{u}^{\mathrm{th}} \text { point belongs } \mathrm{w}^{\mathrm{th}} \text { block } \\
& =0, \text { otherwise } .
\end{aligned}
$$

$e_{u}$ is the experimental error with mean zero and variance $\sigma^{2}$ (unknown)

Following is the set of $n$ observational equation


$$
Y_{\mathrm{n}}=\beta_{1} X_{1 \mathrm{n}}+\beta_{2} X_{2 \mathrm{n}}+\ldots \ldots \ldots+\beta_{\mathrm{k}} X_{\mathrm{kn}}+\Gamma_{1} Z_{1 \mathrm{n}}+\Gamma_{2} Z_{2 \mathrm{n}}+\ldots+\Gamma_{\mathrm{b}} Z_{\mathrm{bn}}+\mathrm{e}_{\mathrm{u}}
$$

and its corresponding matrix notation is

$$
\begin{equation*}
Y=X \beta+Z \Gamma+e \tag{1.3}
\end{equation*}
$$

Where

$$
\mathrm{Y}_{\mathrm{n} \times 1}=\left[\begin{array}{l}
\mathrm{Y}_{1} \\
\mathrm{Y}_{2} \\
\cdot \\
\cdot \\
\cdot \\
\mathrm{Y}_{\mathrm{n}}
\end{array}\right] \quad \mathrm{e}_{\mathrm{n} \times 1}=\left[\begin{array}{l}
\mathrm{e}_{1} \\
\mathrm{e}_{2} \\
\cdot \\
\cdot \\
\cdot \\
\mathrm{e}_{\mathrm{n}}
\end{array}\right] \quad \quad \mathrm{Y}_{\mathrm{n} \times 1}=\left[\begin{array}{l}
\Gamma_{1} \\
\Gamma_{2} \\
\cdot \\
\cdot \\
\cdot \\
\Gamma_{\mathrm{n}}
\end{array}\right]
$$



The corresponding normal equation for estimating the parameters are obtained from

$$
\begin{align*}
& \mathrm{X}_{1}\left[\begin{array}{ll}
\mathrm{X}_{1}^{\prime} \mathrm{X}_{1} & \mathrm{X}_{1}^{\prime} \mathrm{Z} \\
\mathrm{Z}^{\prime} \mathrm{X}_{1} & \mathrm{Z}^{\prime} \mathrm{Z}
\end{array}\right]\left[\begin{array}{l}
\hat{\beta} \\
\hat{\Gamma}
\end{array}\right]=\left[\begin{array}{l}
\mathrm{X}_{1}^{\prime} \mathrm{Y} \\
\mathrm{Z}^{\prime} \mathrm{Y}
\end{array}\right] \\
& \text { Or } \quad \mathrm{X}_{1}^{\prime} \mathrm{X}_{1} \hat{\beta}+\mathrm{X}_{1}^{\prime} \mathrm{Z} \hat{\Gamma}=\mathrm{X}_{1}^{\prime} \mathrm{Y}  \tag{1.4}\\
&  \tag{1.5}\\
& Z^{\prime} X_{1} \hat{\beta}+Z^{\prime} Z \hat{\Gamma}=Z^{\prime} Y
\end{align*}
$$

If $X_{1}{ }^{\prime} Z \hat{\Gamma}=0$, the regression parameters can be estimated by ignoring the block effects. In this sense, it may be regarded as an orthogonal blocking. Now we consider the various situations under which $X_{1}^{\prime} Z \hat{\Gamma}=0$

1. When $\underset{\mathrm{k} \times \mathrm{b}}{\mathrm{X}_{1}^{\prime} \mathrm{Z}}=\underset{\mathrm{k} \times \mathrm{b}}{0}$

Then $\mathrm{Z}_{\mathrm{b} \times \mathrm{k}}^{\prime} \mathrm{X}_{1}$ is a null matrix of order $\mathrm{b} \times \mathrm{k}$ and under this situation

$$
\begin{equation*}
\hat{\beta}=\left(X_{1}^{\prime} X_{1}\right)^{-1} X_{1}^{\prime} Y \tag{1.6}
\end{equation*}
$$

$$
\begin{equation*}
\hat{\Gamma}=\left(Z^{\prime} Z\right)^{-1} Z^{\prime} Y \tag{1.7}
\end{equation*}
$$

i.e., both the set of parameters are estimable by ignoring the other set of parameters and covariance between them is also zero i.e., $\operatorname{Cov}(\hat{\beta}, \hat{\Gamma})=0$. Thus orthogonality can be achieved. But in mixture experiments $X_{i u} \geq 0$ and hence $X_{1} Z \neq 0$ and orthogonal blocking such that

$$
\begin{aligned}
& \hat{\beta}=\left(X_{1}^{\prime} X_{1}\right)^{-1} X_{1}^{\prime} Y \\
& \hat{\Gamma}=\left(Z^{\prime} Z\right)^{-1} Z^{\prime} Y
\end{aligned}
$$

and $\operatorname{Cov}(\hat{\beta}, \hat{\Gamma})=0$
Cannot be achieved. Further, we consider the possibility of estimating one set of parameters ignoring the other and $\operatorname{Cov}(\hat{\beta}, \hat{\Gamma})=0$
i.e. $\quad \hat{\beta}=\left(X_{1}^{\prime} X_{1}\right)^{-1} X_{1}^{\prime} Y$, and $\operatorname{Cov}(\hat{\beta}, \hat{\Gamma})=0$
2. When $X_{1}^{\prime} Z=a E$

Where $\mathrm{E}_{\mathrm{k} \times \mathrm{b}}$ is a kxb matrix each of whose elements is unity and 'a' is a constant, and $\mathrm{E}_{1 \times \mathrm{b}} \hat{\Gamma}=0$ i.e. $\sum_{w=1}^{b} \hat{\Gamma}_{w}=0$ , then

$$
\begin{align*}
& \hat{\beta}=\left(X_{1}^{\prime} X_{1}\right)^{-1} X_{1}^{\prime} Y  \tag{1.8}\\
& \hat{\Gamma}=\left(Z^{\prime} Z\right)^{-1}\left[Z^{\prime} Y-Z^{\prime} X_{1}\left(X_{1}^{\prime} X_{1}\right)^{-1} X_{1}^{\prime} Y\right]  \tag{1.9}\\
& \text { and } \quad \operatorname{Cov}(\hat{\beta}, \hat{\Gamma})=0
\end{align*}
$$

Blocking designs satisfying the blocking condition

$$
\begin{aligned}
& X_{1}^{\prime} Z=\mathrm{aE}_{\mathrm{k} \times \mathrm{b}} \\
& \text { i.e., } \sum_{\mathrm{u} \in \mathrm{w}} X_{\mathrm{iu}}=\mathrm{a}
\end{aligned} \quad \text { for all } \mathrm{i}=1,2, \ldots \ldots \ldots . \mathrm{k} \text { and } \mathrm{w}=1,2, \ldots \ldots \ldots . . \mathrm{b} .
$$

Have been constructed by Nigum (1970) and Murthy and Murty (1992) for the case $n_{1}=n_{2}=\ldots .=n_{b}$ i.e., for the block of equal size. It is obvious that the blocking condition $\sum_{u \in w} X_{i u}=a$ for any $i$ and $w$ cannot be satisfied by blocks of unequal size. In this case, we can only expect that $\sum_{u \in w} X_{i u}=a_{w}$
3. When

We can write

$$
\begin{array}{ll} 
& \sum_{u \in \mathrm{w}} \mathrm{X}_{1 \mathrm{u}}=\mathrm{a}_{\mathrm{w}}, \sum_{\mathrm{u} \in \mathrm{w}} \mathrm{X}_{2 \mathrm{u}}=\mathrm{a}_{\mathrm{w}}, \ldots \ldots \ldots \ldots . . . \sum_{\mathrm{u} \in \mathrm{w}} \mathrm{X}_{\mathrm{ku}}=\mathrm{a}_{\mathrm{w}} \\
\text { or, } & \sum_{\mathrm{u} \in \mathrm{w}}\left(\mathrm{X}_{1 \mathrm{u}}+\mathrm{X}_{2 \mathrm{u}}+\ldots \ldots .+\mathrm{X}_{\mathrm{ku}}\right)=\mathrm{ka} \mathrm{w}_{\mathrm{w}} \\
\text { or } \quad & \sum_{\mathrm{u} \in \mathrm{w}} 1=\mathrm{ka} \mathrm{w}_{\mathrm{w}} \\
\text { or } \quad & \mathrm{ka}_{\mathrm{w}}=\mathrm{n}_{\mathrm{w}}
\end{array}
$$

Thus,

Under the restriction $\sum_{\mathrm{w}=1}^{\mathrm{b}} \mathrm{n}_{\mathrm{w}} \Gamma_{\mathrm{w}}=0$ and blocking condition $\sum_{\mathrm{u} \in \mathrm{w}} \mathrm{X}_{\mathrm{lu}}=\mathrm{a}_{\mathrm{w}}=\frac{\mathrm{n}_{\mathrm{w}}}{\mathrm{k}}$ $\hat{\beta}=\left(X_{1}^{\prime} X_{1}\right)^{-1} X_{1}{ }^{\prime} Y$
$\hat{\Gamma}=\left(Z^{\prime} Z\right)^{-1}\left[Z^{\prime} Y-Z^{\prime} X_{1}\left(X_{1}^{\prime} X_{1}\right)^{-1} X_{1}^{\prime} Y\right]$
and $\operatorname{Cov}(\hat{\beta}, \hat{\Gamma})=0$
Designs satisfying the above blocking condition are considered by Nigam (1976)
4. When


The following k restriction is required to have an orthogonal blocking

$$
\sum_{\mathrm{w}=1}^{\mathrm{b}} \mathrm{a}_{\mathrm{wi}} \Gamma_{\mathrm{w}}=0, \quad \mathrm{i}=1,2, \ldots \ldots \ldots \ldots, \mathrm{k}
$$

But this is not justified, because of the rank of the matrix

$$
\left[\begin{array}{ll}
\mathrm{X}_{1}^{\prime} \mathrm{X}_{1} & \mathrm{X}_{1}^{\prime} \mathrm{Z} \\
\mathrm{Z}^{\prime} \mathrm{X}_{1} & \mathrm{Z}^{\prime} \mathrm{Z}
\end{array}\right] \text { is (k+b-1) as has been discussed below. Therefore, orthogonal blocking is not possible in this }
$$ case.

Finally, we arrived at the conclusion that for the blocking of mixture experiments with a linear model, one blocking condition (restriction on the mixture variables) $\sum \mathrm{X}_{\mathrm{iu}}=\mathrm{a}_{\mathrm{w}}$ and one blocking restriction on the parameters $\sum_{\mathrm{w}=1}^{\mathrm{b}} \mathrm{a}_{\mathrm{w}} \Gamma_{\mathrm{w}}=0$ is essential. This result holds for all mixture designs not only for symmetric simplex design.

Blocking condition

$$
\sum_{u \in w} X_{l u}=a_{w}=\frac{n_{w}}{k}
$$

Or, $\quad \sum_{u \in w} \frac{X_{i u}}{n_{w}}=\frac{1}{k} \quad$ can be achieved by a suitable arrangement of design points within blocks. Finally, it remains to decide whether we are justified in assuming $\sum \mathrm{n}_{\mathrm{w}} \Gamma_{\mathrm{w}}=0$ or not. This can be answered by finding out the rank of the matrix
$\left[\begin{array}{ll}X^{\prime} X & X^{\prime} Z \\ Z^{\prime} X & Z^{\prime} Z\end{array}\right]$

If its rank is full i.e., $(\mathrm{k}+\mathrm{b})$, then no restriction is a restriction on the parameters. Because we cannot have a solution which satisfies (1.4), (1.5) and the imposed restriction simultaneously. On the other hand, if this rank is ( $\mathrm{k}+\mathrm{b}-\mathrm{h}$ ) then h restrictions are required to have a unique solution and we are quite justified in imposing the h restrictions but more than $h$ restrictions. In the present case, the sum of the last ' $b$ ' rows is equal to the sum of the first ' $k$ ' rows and therefore the matrix is singular and its rank is less than $(k+b)$. The first ' $k$ ' rows are independent and the late ' $b$ ' rows also form an independent set. So, by virtue of the fact that the sum of the two sets of rows is equal. The rank of the matrix is $(k+b-1)$. Therefore, only one restriction on the parameters is required to obtain their estimates.

## Quadratic Model

Let us approximate the response Y by the quadratic model

$$
\begin{equation*}
Y_{u}=\sum_{i=k}^{k} \beta_{i} X_{i}+\sum_{i<j=1}^{k} \beta_{i j} X_{i} X_{j}+\sum_{w=1}^{b} Z_{w u} \Gamma_{w}+e_{u} \quad(u=1,2, \ldots \ldots \ldots \ldots ., n) \tag{2.1}
\end{equation*}
$$

Where the symbols are interpreted as in equation (2.1) of section 2.

The set of n observational equation can be written in matrix form as

$$
\begin{equation*}
\mathrm{Y}=\mathrm{X}_{1} \beta+\mathrm{X}_{2} \delta+\mathrm{Z} \Gamma+\mathrm{e} \tag{2.2}
\end{equation*}
$$

Where $\mathrm{Y}, \mathrm{X}_{\mathrm{i}}, \beta, \mathrm{Z}, \Gamma$ and e are the same as defined previously and $\mathrm{X}_{2}$ is a matrix of order $\mathrm{n} \times \mathrm{k} . \mathrm{C}_{2}$ Having the elements of the type $\left(\mathrm{X}_{\mathrm{iu}} \mathrm{X}_{\mathrm{ju}}\right)$ i.e.


$$
\delta_{\mathrm{k}_{2} \times 1}=\left[\begin{array}{l}
\beta_{12} \\
\beta_{13} \\
\cdot \\
\cdot \\
\beta_{\mathrm{K}-1 \mathrm{~K}}
\end{array}\right]
$$

With this labelling, the normal equations are

$$
\left[\begin{array}{lll}
X_{1}^{\prime} X_{1} & X_{1}^{\prime} X_{2} & X_{1}^{\prime} Z \\
X_{2}^{\prime} X_{1} & X_{2}^{\prime} X_{2} & X_{2}^{\prime} Z \\
Z^{\prime} X_{1} & Z^{\prime} X_{2} & Z^{\prime} Z
\end{array}\right]\left[\begin{array}{l}
\hat{\beta} \\
\hat{\delta} \\
\hat{\Gamma}
\end{array}\right]=\left[\begin{array}{l}
X_{1}^{\prime} Y \\
X_{2}^{\prime} Y \\
Z^{\prime} Y
\end{array}\right]
$$

or,

$$
\begin{align*}
& X_{1}^{\prime} X_{1} \hat{\beta}+X_{1}^{\prime} X_{2} \hat{\delta}+X_{1}^{\prime} Z \hat{\Gamma}=X_{1}^{\prime} Y  \tag{2.3}\\
& X_{2}^{\prime} X_{1} \hat{\beta}+X_{2}^{\prime} X_{2} \hat{\delta}+X_{2}^{\prime} Z \hat{\Gamma}=X_{1}^{\prime} Y  \tag{2.4}\\
& Z^{\prime} X_{1} \hat{\beta}+Z^{\prime} X_{2} \hat{\delta}+Z^{\prime} Z \Gamma \hat{\Gamma}=X_{1}^{\prime} Y \tag{2.5}
\end{align*}
$$

If

$$
\begin{equation*}
X_{1}^{\prime} Z \hat{\Gamma}=0 \tag{2.6}
\end{equation*}
$$

and $\quad X_{2}^{\prime} Z \hat{\Gamma}=0$
Then

$$
\begin{align*}
& {\left[\begin{array}{ll}
\mathrm{X}_{1}^{\prime} \mathrm{X}_{1} & \mathrm{X}_{1}^{\prime} \mathrm{X}_{2} \\
\mathrm{X}_{2}^{\prime} \mathrm{X}_{1} & \mathrm{X}_{2}^{\prime} \mathrm{X}_{2}
\end{array}\right]\left[\begin{array}{l}
\hat{\beta} \\
\hat{\delta}
\end{array}\right]=\left[\begin{array}{l}
\mathrm{X}_{1}^{\prime} \mathrm{Y} \\
\mathrm{X}_{2}^{\prime} \mathrm{Y}
\end{array}\right]} \\
& \text { Or }\left[\begin{array}{l}
\hat{\beta} \\
\hat{\delta}
\end{array}\right]=\left[\begin{array}{ll}
\mathrm{X}_{1}^{\prime} \mathrm{X}_{1} & \mathrm{X}_{1}^{\prime} \mathrm{X}_{2} \\
\mathrm{X}_{2}^{\prime} \mathrm{X}_{1} & \mathrm{X}_{2}^{\prime} \mathrm{X}_{2}
\end{array}\right]^{-1}\left[\begin{array}{l}
\mathrm{X}_{1}^{\prime} \mathrm{Y} \\
\mathrm{X}_{2}^{\prime} \mathrm{Y}
\end{array}\right] \tag{2.8}
\end{align*}
$$

$X_{1}^{\prime} Z \hat{\Gamma}=0$, provide $s$ set of $k$ restrictions on the block parameters and $X_{2}^{\prime} Z \hat{\Gamma}=0$, provides an additional set of ${ }^{\mathrm{k}} \mathrm{C}_{2}$ restrictions. By a suitable arrangement of design points (mixtures) into blocks, the mixture variables can be made to satisfy certain blocking conditions which reduce the k restrictions into one and the other ${ }^{\mathrm{k}} \mathrm{C}_{2}$ restrictions into a single restriction. Various such possibilities are ;

1. When $\mathrm{X}_{1} \mathrm{Z}=\mathrm{a}_{1} \mathrm{E}_{\mathrm{k} \times \mathrm{b}}$
and $X_{2}^{\prime} Z=a_{2} E_{\mathrm{k}_{C_{2} \times \mathrm{b}}}$
then equation (2.6) and (2.7) reduces to

$$
\begin{equation*}
\mathrm{a}_{1} \mathrm{E}_{\mathrm{k} \times \mathrm{b}} \hat{\Gamma}=0 \text { or, } \mathrm{a}_{1} \sum_{w=1}^{b} \hat{\Gamma}_{w}=0 \tag{2.11}
\end{equation*}
$$

and $\quad \mathrm{a}_{2} \mathrm{E}_{\mathrm{k}_{C_{2} \times \mathrm{b}}} \hat{\Gamma}=0$ or, $\mathrm{a}_{2} \sum_{w=1}^{b} \hat{\Gamma}_{w}=0$

Under the blocking conditions (3.8) and (3.9) and assumption $\sum \hat{\Gamma}=0$, the regression parameters are estimable independently of the block parameters of the set of equation (3.7). Designs satisfying the above conditions are obtained by Nigam (1970) and Murty (1992). Equations (3.8) and 3.9 are known as the blocking conditions of Nigam (1970).
2. $X_{1}^{\prime} \mathrm{Z}=\left[\begin{array}{l}\mathrm{a}_{1}{ }^{(1)} \mathrm{a}_{2}{ }^{(1)} \ldots \ldots \ldots . \mathrm{a}_{\mathrm{b}}{ }^{(1)} \\ \mathrm{a}_{1}{ }^{(1)} \mathrm{a}_{2}{ }^{(1)} \ldots \ldots \ldots . \mathrm{a}_{\mathrm{b}}{ }^{(1)} \\ \ldots \ldots \ldots \ldots \ldots \ldots \ldots . . \\ \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots . \\ \mathrm{a}_{1}{ }^{(1)} \mathrm{a}_{2}{ }^{(1)} \ldots \ldots \ldots . \mathrm{a}_{\mathrm{b}}{ }^{(1)}\end{array}\right]$ of order $\mathrm{k} \times \mathrm{b}$

Here it is noticed that $a_{w}{ }^{(1)}=\frac{n_{w}}{k}$

Then equations (3.5) and (3.6) take the form

$$
\begin{align*}
& \sum \mathrm{a}_{\mathrm{w}}{ }^{(1)} \Gamma_{w}=0  \tag{2.15}\\
& \sum \mathrm{a}_{\mathrm{w}}{ }^{(2)} \Gamma_{w}=0 \tag{2.16}
\end{align*}
$$

Where $\mathrm{a}_{\mathrm{w}}{ }^{(1)} \neq \mathrm{a}_{\mathrm{w}}{ }^{(2)}, \mathrm{a}_{\mathrm{w}}{ }^{(1)} \neq \mathrm{a}_{\mathrm{w}^{\prime}}{ }^{(1)}$ and $\mathrm{a}_{\mathrm{w}}{ }^{(2)} \neq \mathrm{a}_{\mathrm{w}^{\prime}}{ }^{(2)}$, for some $\mathrm{w} \neq \mathrm{w}^{\prime}$ equations (2.13) and (2.14) are the modified blocking conditions of Nigam (1976).

Under the above conditions in equations (2.13) and (2.14) and assumptions in equations (2.15) and (2.16), equations (2.3) and (2.4) becomes free from block parameters and therefore the regression parameters can be estimated independently of the block effects. But the rank of $\left(\mathrm{k}+{ }^{\mathrm{k}} \mathrm{C}_{2}+\mathrm{b}\right) \times\left(\mathrm{k}+{ }^{\mathrm{k}} \mathrm{C}_{2}+\mathrm{b}\right)$ matrix

$$
\left[\begin{array}{lll}
X_{1}^{\prime} X_{1} & X_{1}^{\prime} X_{2} & X_{1}^{\prime} Z \\
X_{2}^{\prime} X_{1} & X_{2}^{\prime} X_{2} & X_{2}^{\prime} Z \\
Z^{\prime} X_{1} & Z^{\prime} X_{2} & Z^{\prime} Z
\end{array}\right] \text { is }\left(k+{ }^{k} C_{2}+b-1\right) \text { which reveals that only one restriction can be imposed. }
$$

Thus, the suggestion of Murthy and Murty (1992) of imposing two restrictions on the block parameters is not justified. As a matter of fact, under the modified blocking condition of Nigam (1976), the assumption $\sum \mathrm{a}_{\mathrm{w}}{ }^{(1)} \Gamma_{w}=0$ (which is equivalent to $\sum \mathrm{n}_{\mathrm{w}} \Gamma_{w}=0$ ) is practicable. Under these conditions orthogonal blocking is not possible for any mixture design, including the symmetric simplex design. The reality is that in this situation non-orthogonal blocking is possible and the regression parameters will be estimated by eliminating the block effects. The utility of blocking conditions is that after eliminating the block affects with the help of equation (2.5) from equations (2.3) and (2.4) the pattern of

$$
\left[\begin{array}{ll}
\left(\mathrm{X}_{1}^{\prime} \mathrm{X}_{1}\right)^{*} & \left(\mathrm{X}_{1}^{\prime} \mathrm{X}_{2}\right)^{*} \\
\left(\mathrm{X}_{2}^{\prime} \mathrm{X}_{1}\right)^{*} & \left(\mathrm{X}_{2}^{\prime} \mathrm{X}_{2}\right)^{*}
\end{array}\right]
$$

The adjusted coefficient matrix of regression parameters remains the same as that of the matrix

$$
\left[\begin{array}{ll}
\left(\mathrm{X}_{1}^{\prime} \mathrm{X}_{1}\right) & \left(\mathrm{X}_{1}^{\prime} \mathrm{X}_{2}\right) \\
\left(\mathrm{X}_{2}^{\prime} \mathrm{X}_{1}\right) & \left(\mathrm{X}_{2}^{\prime} \mathrm{X}_{2}\right)
\end{array}\right]
$$

and hence the estimation procedure remains the same. Once again we conclude that orthogonal blocking is not possible here as is suggested by Murthy and Murty (1992).
3. When

and $\quad X_{2}^{\prime} Z=$ Constant $\left[\begin{array}{l}n_{1} n_{2} \ldots \ldots \ldots \ldots \ldots \ldots n_{b} \\ n_{1} n_{2} \ldots \ldots \ldots \ldots \ldots \ldots n_{b} \\ \ldots \ldots \ldots \ldots \ldots \ldots \ldots . . \\ \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots . . \\ n_{1} n_{2} \ldots \ldots \ldots \ldots \ldots . n_{b}\end{array}\right]$ of order ${ }^{k} C_{2} \times b$
or, $\quad \sum_{u \in w} X_{i u}=\frac{n_{w}}{k}$, for $i=1.2 \ldots \ldots . . . k$ and $w=1,2, \ldots \ldots . b$
or, $\quad \sum_{u \in w} X_{i u} X_{j u}=n_{w}$.Constant , for $\mathrm{i}<j=1,2 \ldots \ldots \ldots .$. k and $w=1,2, \ldots \ldots .$. b
Then assumption in equations (3.5) and (3.6) reduces into one assumption as shown below:

$$
\begin{aligned}
& \frac{1}{\mathrm{k}} \sum_{\mathrm{w}=1}^{\mathrm{b}} \mathrm{n}_{\mathrm{w}} \Gamma_{\mathrm{w}}=0 \quad \text { or, } \sum_{\mathrm{w}=1}^{\mathrm{b}} \mathrm{n}_{\mathrm{w}} \Gamma_{\mathrm{w}}=0 \text { and } \\
& \text { Constant } \sum_{\mathrm{w}=1}^{\mathrm{b}} \mathrm{n}_{\mathrm{w}} \Gamma_{\mathrm{w}}=0 \quad \text { or, } \sum_{\mathrm{w}=1}^{\mathrm{b}} \mathrm{n}_{\mathrm{w}} \Gamma_{\mathrm{w}}=0
\end{aligned}
$$

Obviously under blocking conditions in equations (2.17) and (2.18) and the assumption $\sum \mathrm{n}_{\mathrm{w}} \Gamma_{\mathrm{w}}=0$, the orthogonality blocking is possible in the same sense that regression parameters are estimable by ignoring the block and the covariance between the regression parameters and the block parameters is zero. Design satisfying equations (2.17) and (2.18) can be constructed with the use of the theorem (5.1) of Murthy and Murty (1992). The final conclusion is that for orthogonal blocking with the quadratic model the modified blocking conditions of Nigam (1976) have to be modified to get equations (2.17) and (3.12) orthogonal blocking is possible and not under equations (2.13) and (2.14) as is stated by

Murthy and Murty (1992). We also emphasize that the blocking conditions obtained by Nigam (1976) are applicable in general and not only in the case of symmetric simplex designs as mentioned by Murthy and Murty (1992).. The number of blocking conditions required to simplifying the analysis depends on the model used to approximate the response, but the assumptions regarding the block parameters is the only one for any model.

## COMMENTS AND CONCLUSIONS

I. It has been shown that under blocking condition of Nigam (1970) orthogonal blocking can be achieved, provided the blocks are of equal size and the restriction of the form $\quad \sum \Gamma_{\mathrm{w}}=0$ is imposed on the parameters.
II. Under the assumptions, $\sum \mathrm{n}_{\mathrm{w}} \Gamma_{\mathrm{w}}=0$, orthogonal blocking can also be achieved when modified conditions of Nigam (1976) are further modified i.e.,

When $\quad \sum_{u \in w} \frac{\mathrm{X}_{\mathrm{m}}}{\mathrm{n}_{\mathrm{w}}}=\frac{1}{\mathrm{k}}, \quad \quad$ for $\mathrm{i}=1,2,3, \ldots \ldots ., \mathrm{k}$
and $\sum_{u \in w} \frac{X_{i u} X_{j u}}{n_{w}}=$ Constant., for $\mathrm{i}<j=1,2, \ldots \ldots . . . ., k$
III. Blocking conditions are unable to remove the singularity of the adjusted coefficient matrix of the regression parameters. Under these condition the patterns of the adjusted matrix remain the same as that of the matrix obtained by ignoring the block effects. Therefore, the procedure for estimating the regression parameters does not change and no new complexity arises in the analysis due to blocking.

As simplicity and efficiency of the design go hand in hand, the blocking conditions make the design efficient in comparison to the arbitrary blocking.
IV. The blocking conditions are workable for any mixture design not only for symmetric simplex design.
V. It is, in general, true that any parameter, be it treatment effects in binary design or regression effect in mixture experiment, cannot be uniquely estimated in presence of block effects. In order to have unique, some restrictions on the parameters is essential. The implication of this assumption is that, the blocking effect $\Gamma_{\mathrm{w}}$ 's and regression parameters of the type $\beta_{\mathrm{i}}$ 's cannot be estimated uniquely, but their contrast can be estimated. Regression parameters of the type $\beta_{\mathrm{ij}}$ are uniquely estimated.
VI. It is, however, of interest to find that the estimate of $Y$ based on the prediction equation

$$
\mathrm{Y}=\sum_{i} \beta_{\mathrm{i}} \mathrm{X}_{\mathrm{i}}+\sum_{\mathrm{i}<\mathrm{j}} \mathrm{XiXj}_{\mathrm{j}}
$$

Depends upon the assumption on the block parameters and should be taken as an estimate of Y for the circumstances under which the assumption is appropriate. For example, if it is desired to have an estimate under the condition of experiment represented by an average block effect, the reasonable assumption as $\sum n_{w} \Gamma_{w}=0$
and, if the estimate is required for the experimental condition of the $w^{\text {th }}$ block, the assumption should be $\sum \Gamma_{w}=0$.
VII. One use of mixture experiments is to find a suitable optimum combination of the factor component. For this, the regression relation which has been estimated through a mixture experiment has to be differentiated with respect to the component parameter subject to the condition $\sum \mathrm{X}_{\mathrm{i}}=1$ and equated to zero or some suitable constant. These solutions are functions of contrasts among $\beta_{\mathrm{i}}$ 's and $\beta_{\mathrm{ij}}$ 's as such. The estimates of such optimum combination are not subject to the uncertainty in estimates of the regression parameters caused due to the blocking.
VIII. The regression sum of squares and the error sum of squares are unique.

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