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ABOUT THE CHEMICAL BALANCE WEIGHING DESIGNS WITH CORRELATED ERRORS

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Abstract: The paper deals with the problem of estimation of individual weights of objects using the chemical balance weighing design. We assume that not all objects are included in each weighing operation. Addtionally we assume that the number of times in which each object is weighed is less then number of all measurement operations. The errors are correlated and they have equal variances. We give the lower bound of variance of each of the estimators and the sufficient and necessary conditions under which this lower bound is attained. The new construction methods for the optimum chemical balance weighing design are given. They are based on the incidence matrices of the balanced incomplete block designs and the ternary balanced block designs.

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1. Introduction

Let us consider the linear model

$$\mathbf{y} = \mathbf{X}\mathbf{w} + \mathbf{e},$$

which describe how to find unknown measurements of p objects using n weighing

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operations according to the design matrix $\mathbf{X}$. In the above model $\mathbf{y}$ is an $n \times 1$ random column vector of the observed weights, the design matrix $\mathbf{X}$ belongs to the class of $n \times p$ matrices of elements equal to $-1, 0$ or 1 and in which maximum number of elements equal to -1 and 1 in each column is equal to m, i.e. $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$, $\mathbf{w}$ is an $p \times 1$ column vector representing unknown weights of objects and $\mathbf{e}$ is an $n \times 1$ random column vector of errors such that $E(\mathbf{e}) = \mathbf{0}_n$ and $E(\mathbf{ee}') = \sigma^2\mathbf{G}$, where $\mathbf{0}_n$ is an $n \times 1$ column vector of zeros,

$$\mathbf{G} = g\left[(1-\rho)\mathbf{I}_n + \rho\mathbf{1}_n\mathbf{1}_n^{'}\right], \quad g > 0, \quad \frac{-1}{n-1} < \rho < 1. \tag{1}$$

Now, if $\mathbf{X}'\mathbf{G}^{-1}\mathbf{X}$ is nonsingular, the least squares estimator of $\mathbf{w}$ is given by

$$\hat{\mathbf{w}} = (\mathbf{X}'\mathbf{G}^{-1}\mathbf{X})^{-1}\mathbf{X}'\mathbf{G}^{-1}\mathbf{y}$$

and the variance matrix of $\hat{\mathbf{w}}$ is of the form

$$\text{Var}(\hat{\mathbf{w}}) = \sigma^2(\mathbf{X}'\mathbf{G}^{-1}\mathbf{X})^{-1}.$$

Hotelling [3] studied the estimation problem for the case $\mathbf{G} = \mathbf{I}_n$ and under the assumption that in each measurement operation all objects are included. In this paper we generalize the assumption given above. In each measurement operation there is number of objects which are not included. The similar problem was considered in Ceranka et al [2]. They gave the lower bound of variance of each of the estimators and the definition of the optimal design.

Definition 1. In the nonsingular chemical balance weighing design with the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$, with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is given in (1), the variance of $\hat{w}_j$ for a particularly j, such that $j = 1, 2, ..., p$, can not be less then

$$\text{Var}(\hat{w}_j) = \begin{cases} \dfrac{\sigma^2 g(1-\rho)}{m} & \text{if} \quad 0 \le \rho < 1\,, \\ \dfrac{\sigma^2 g(1-\rho)}{m - \frac{\rho}{1+\rho(n-1)}(m-2u)^2} & \text{if} \quad \frac{-1}{n-1} < \rho < 0\,, \end{cases}$$

where $m = \max(m_1, m_2, ..., m_p)$, m_j represents the number of elements equal to -1 and 1 in j-th column of $\mathbf{X}$, $u = \min(u_1, u_2, ..., u_p)$, u_j represents the number of elements equal to -1 in j-th column of the matrix $\mathbf{X}$, $\quad j = 1, 2, ..., p$.

In the same paper they have given the necessary and sufficient conditions under which the chemical balance weighing design with the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$, and with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1), is optimal.

Theorem 1. *Let $0 \leq \rho < 1$. Any nonsingular chemical balance weighing design with the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ and with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is given in (1), is optimal if and only if*

$$\mathbf{X}^{'}\mathbf{X} = m\mathbf{I}_p \qquad \text{and} \qquad \mathbf{X}^{'}\mathbf{1}_n = \mathbf{0}_p. \tag{2}$$

Theorem 2. *Let $\frac{-1}{n-1} < \rho < 0$. Any nonsingular chemical balance weighing design with the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ and with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is given in (1), is optimal if and only if*

$$\mathbf{X}^{'}\mathbf{X} = m\mathbf{I}_p - \frac{\rho(m-2u)^2}{1+\rho(n-1)}(\mathbf{I}_p - \mathbf{1}_p\mathbf{1}_p^{'}),$$

$$u_1 = u_2 = ... = u_p = u \tag{3}$$

and $\mathbf{X}^{'}\mathbf{1}_n = \mathbf{z}_p$, where $\mathbf{z}_p$ is an $p \times 1$ column vector, for which j-th element is equal to $(m-2u)$ or $-(m-2u)$, $j = 1,2,...,p$.

But in Ceranka et al [2] some methods of construction of the design matrix $\mathbf{X}$ were not given. Because of this reason in present paper we give the method of construction of the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$. It is based on the incidence matrices of the balanced incomplete block designs and the ternary balanced block designs.

2. Construction of the Design Matrix

Now, we remind the definitions of the balanced incomplete block designs given in Raghavarao [4] and the ternary balanced block designs given in Billington [1].

A balanced incomplete block design there is an arrangement of v treatments in b blocks, each of size k, in such a way, that each treatment occurs at most ones in each block, occurs in exactly r blocks and every pair of treatments occurs together in exactly λ blocks. The integers v, b, r, k, λ are called the parameters of the balanced incomplete block design. Let $\mathbf{N}$ be the incidence matrix of balanced incomplete block design. It is straightforward to verify that

$$vr = bk\,, \quad \lambda(v-1) = r(k-1), \quad \mathbf{NN}' = (r-\lambda)\mathbf{I}_v + \lambda\mathbf{1}_v\mathbf{1}_v',$$

where $\mathbf{1}_v$ is the $v \times 1$ vector of units.

A ternary balanced block design is defined as the design consisting of b blocks, each of size k, chosen from a set of objects of size v, in such a way that each of the v treatments occurs r times altogether and 0, 1 or 2 times in each block (2 appears at least ones) and each of the distinct pairs appears λ times. Any ternary balanced block design is regular, that is, each treatment occurs alone in ρ_1 blocks and is repeated two times in ρ_2 blocks, where ρ_1 and ρ_2 are constant for the design. Let $\mathbf{N}$ be the incidence matrix of the ternary balanced block design. It is straightforward to verify that

$$vr = bk, \quad r = \rho_1 + 2\rho_2,$$

$$\lambda(v-1) = \rho_1(k-1) + 2\rho_2(k-2) = r(k-1) - 2\rho_2,$$
$$\mathbf{NN'} = (\rho_1 + 4\rho_2 - \lambda)\mathbf{I}_v + \lambda\mathbf{1}_v\mathbf{1}_v' = (r + 2\rho_2 - \lambda)\mathbf{I}_v + \lambda\mathbf{1}_v\mathbf{1}_v'.$$

Let $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ be the design matrix of the chemical balance weighing design given in the form

$$\mathbf{X} = \begin{bmatrix} 2\mathbf{N}_1' - \mathbf{1}_{b_1}\mathbf{1}_v' \\ \mathbf{N}_2' - \mathbf{1}_{b_2}\mathbf{1}_{v'} \end{bmatrix}, \tag{4}$$

where $\mathbf{N}_1$ is the incidence matrix of the balanced incomplete block design with the parameters $v,\ b_1,\ r_1,\ k_1,\ \lambda_1$ and $\mathbf{N}_2$ is the incidence matrix of the ternary balanced block design with the parameters $v,\ b_2,\ r_2,\ k_2,\ \lambda_2,\ \rho_{12},\ \rho_{22}$. In this design each of the $p = v$ objects is weighed $m = b_1 + b_2 - \rho_{12}$ times in $n = b_1 + b_2$ weighing operations.

Lemma 1. *The chemical balance weighing design with the matrix* $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ *given in the form (4) is nonsingular if and only if*

$$2k_1 \neq k_2 \quad \text{or} \quad 2k_1 = k_2 \neq v.$$

Proof. For the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ given by (4) we have

$$\mathbf{X}'\mathbf{X} = \left[4(r_1 - \lambda_1) + r_2 + 2\rho_{22} - \lambda_2\right]\mathbf{I}_v + \eta\mathbf{1}_v\mathbf{1}_v', \tag{5}$$

where $\eta = b_1 - 4(r_1 - \lambda_1) + b_2 + \lambda_2 - 2r_2$ and

$$\det(\mathbf{X}'\mathbf{X}) = \left[4(r_1 - \lambda_1) + r_2 + 2\rho_{22} - \lambda_2\right]^{v-1} \cdot \left[\frac{r_1}{k_1}(v - 2k_1)^2 + \frac{r_2}{k_2}(v - k_2)^2\right].$$

Evidently $4(r_1 - \lambda_1) + r_2 + 2\rho_{22} - \lambda_2 > 0$, then $\det(\mathbf{X}'\mathbf{X}) = 0$ if and only if $v = 2k_1$ and $v = k_2$. So, the lemma is proved. □

The optimality conditions given in Ceranka et al [2] are depended on the parameter ρ which is connected with the matrix $\mathbf{G}$. This implies that the methods of construction of the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ are depended on ρ, either. Hence we have the following theorem.

Theorem 3. *Let $0 \leq \rho < 1$. Any nonsingular chemical balance weighing design with the matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ given by (4) and with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1), is optimal for estimation unknown measurements of objects if and only if*

$$b_1 - 4(r_1 - \lambda_1) + b_2 + \lambda_2 - 2r_2 = 0 \tag{6}$$

and

$$b_1 - 2r_1 + b_2 - r_2 = 0. \tag{7}$$

Proof. For the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ in the form (4) we have (5). Since $0 \leq \rho < 1$ then (5) and (2) imply the conditions (6) and (7). □

If the chemical balance weighing design given by matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ of the form (4) with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1) and $0 \leq \rho < 1$ is optimal then

$$\mathrm{Var}\,(\hat{w}_j) = \frac{\sigma^2 g(1-\rho)}{b_1 + b_2 - \rho_{12}}, \quad j = 1, 2, ..., v.$$

Theorem 4. *Let $\frac{-1}{n-1} < \rho < 0$. Any nonsingular chemical balance weighing design with the matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ given by (4) and with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1), is optimal for estimation unknown measurements of objects if and only if*

$$\rho = \frac{\eta}{(2r_1 - b_1 + r_2 - b_2)^2 - \eta(b_1 + b_2 - 1)} \tag{8}$$

and

$$\eta < 0. \tag{9}$$

Proof. From the Theorem 2 it derivers that the chemical balance weighing design $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ in the form (4) with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1), is optimal if and only if the conditions (3) are true. From the last one of them it follows that $\mathbf{z}_p$ is equal to $(m-2u)\mathbf{1}_p$ or $-(m-2u)\mathbf{1}_p$, where $m - 2u = 2r_1 - b_1 + r_2 - b_2$. Now from the first condition of (3) and from (5) we have $\eta = \frac{\rho(2r_1 - b_1 + r_2 - b_2)^2}{1+\rho(b_1+b_2-1)}$, which complete the proof. □

If the chemical balance weighing design given by matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ of the form (4) with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1) and $\frac{-1}{n-1} < \rho < 0$ is optimal then

$$\mathrm{Var}\,(\hat{w}_j) = \frac{\sigma^2 g(1-\rho)}{b_1 + b_2 - \rho_{12} - \eta}, \quad j = 1, 2, ..., v.$$

Now, we give the parameters of the balanced incomplete block designs and the ternary balanced block designs based on which we form the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ of the optimum chemical balance weighing design in the form (4) with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is given in (1).

Based on the series of the balanced incomplete block designs given in Raghavarao [4] and the ternary balanced block designs given in Billington [1] we have formulated the following theorems.

Theorem 5. *Let $0 \le \rho < 1$. If the parameters of the balanced incomplete block designs and ternary balanced block designs are equal to:*

(i) $v = 4s^2$, $b_1 = 4s^2$, $r_1 = 2s^2+s$, $k_1 = 2s^2+s$, $\lambda_1 = s^2+s$ *and* $v = 4s^2$, $b_2 = 4s^3$, $r_2 = 2s(2s^2-1)$, $k_2 = 2(2s^2-1)$, $\lambda_2 = 4s(s^2-1)$, $\rho_{12} = 4s(s^2-1)$, $\rho_{22} = s$, $s = 2, 3, ...,$

(ii) $v = 16s^2$, $b_1 = 16s^2$, $r_1 = 2s(4s+1)$, $k_1 = 2s(4s+1)$, $\lambda_1 = 2s(2s+1)$ *and* $v = 16s^2$, $b_2 = 16s^3$, $r_2 = 4s(4s^2-1)$, $k_2 = 4(4s^2-1)$, $\lambda_2 = 8s(2s^2-1)$, $\rho_{12} = 16s(s^2-1)$, $\rho_{22} = 6s$, $s = 2, 3, ...,$

(iii) $v = 36s^2$, $b_1 = 36s^2$, $r_1 = 3s(6s+1)$, $k_1 = 3s(6s+1)$, $\lambda_1 = 3s(3s+1)$ *and* $v = 36s^2$, $b_2 = 72s^3$, $r_2 = 6s(12s^2-1)$, $k_2 = 3(12s^2-1)$, $\lambda_2 = 12s(6s^2-1)$, $\rho_{12} = 18s(4s^2-1)$, $\rho_{22} = 6s$, $s = 1, 2, ...,$

then the chemical balance design with the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ given by (4) and with the variance matrix of errors $\sigma^2\mathbf{G}$, where the matrix $\mathbf{G}$ is of the form (1), is optimal.

Theorem 6. *Let $\frac{-1}{n-1} < \rho < 0$. If for a given ρ the parameters of the balanced incomplete block designs and ternary balanced block designs are equal to:*

(i) $\rho = \frac{-1}{16s+u-5}$, $v = 4s$, $b_1 = 2(4s-1)$, $r_1 = 4s-1$, $k_1 = 2s$, $\lambda_1 = 2s-1$ *and* $v = 4s$, $b_2 = 8s+u-2$, $r_2 = 8s+u-2$, $k_2 = 4s$, $\lambda_2 = 8s+u-4$, $\rho_{12} = u$, $\rho_{22} = 4s-1$, $s = 2, 3, ...,$ $u = 1, 2, ...,$

(ii) $\rho = \frac{-1}{16s+u+3}$, $v = 4s+1$, $b_1 = 2(4s+1)$, $r_1 = 4s$, $k_1 = 2s$, $\lambda_1 = 2s-1$ *and* $v = 4s+1$, $b_2 = 8s+u+1$, $r_2 = 8s+u+1$, $k_2 = 4s+1$, $\lambda_2 = 8s+u-1$, $\rho_{12} = u+1$, $\rho_{22} = 4s$, $s, u = 1, 2, ...,$ $4s+1$ *is a prime or a prime power,*

(iii) $\rho = \frac{-3}{36s+3u+7}$, $v = 4s+1$, $b_1 = 2(4s+1)$, $r_1 = 4s$, $k_1 = 2s$, $\lambda_1 = 2s-1$ *and* $v = 4s+1$, $b_2 = 4s+u$, $r_2 = 4s+u$, $k_2 = 4s+1$, $\lambda_2 = 4s+u-1$, $\rho_{12} = u$, $\rho_{22} = 2s$, $s = 1, 2,$ $u = 1, 2, ...,$

(iv) $\rho = \frac{-3}{72s+3u+58}$, $v = 8s+7$, $b_1 = 8s+7$, $r_1 = 4s+3$, $k_1 = 4s+3$, $\lambda_1 = 2s+1$ *and* $v = 8s+7$, $b_2 = 16s+u+13$, $r_2 = 16s+u+13$, $k_2 = 8s+7$, $\lambda_2 = 16s+u+11$, $\rho_{12} = u+1$, $\rho_{22} = 2(4s+3)$, $s, u = 1, 2, ...,$

(v) $\rho = \frac{-1}{8((t+s)^2-s(t-1))+4t+u}$, $v = (2s+1)^2$, $b_1 = 4t(2s+1)$, $r_1 = 4st$, $k_1 = s(2s+1)$, $\lambda_1 = t(2s-1)$ *and* $v = (2s+1)^2$, $b_2 = 8s(s+1)+u+1$, $r_2 = 8s(s+1)+u+1$, $k_2 = (2s+1)^2$, $\lambda_2 = 8s(s+1)+u-1$, $\rho_{12} = u+1$, $\rho_{22} = 4s(s+1)$, $s, u, t = 1, 2, ...,$ $4t \geq 2s+1$

(vi) $\rho = \frac{-3}{36s^2+3u-14}$, $v = 4s^2-1$, $b_1 = 4s^2-1$, $r_1 = 2s^2-1$, $k_1 = 2s^2-1$, $\lambda_1 = s^2-1$ *and* $v = 4s^2-1$, $b_2 = 8s^2+u-3$, $r_2 = 8s^2+u-3$, $k_2 = 4s^2-1$, $\lambda_2 = 8s^2+u-5$, $\rho_{12} = u+1$, $\rho_{22} = 2(2s^2-1)$, $s, u = 1, 2, ...,$

(vii) $\rho = \frac{-1}{2(t+s)^2+6s^2+u-3}$, $v = 4s^2$, $b_1 = 4st$, $r_1 = t(2s-1)$, $k_1 = s(2s-1)$, $\lambda_1 = t(s-1)$ *and* $v = 4s^2$, $b_2 = 8s^2+u-2$, $r_2 = 8s^2+u-2$, $k_2 = 4s^2$, $\lambda_2 = 8s^2+u-4$, $\rho_{12} = u$, $\rho_{22} = 4s^2-1$, $s, t = 2, 3, ...,$ $t \geq s$

(viii) $\rho = \frac{-2}{u(u+8s)+6(8s-1)}$, $v = 12s$, $b_1 = 2(12s-1)$, $r_1 = 12s-1$, $k_1 = 6s$, $\lambda_1 = 6s-1$ *and* $v = 12s$, $b_2 = 4us$, $r_2 = u(4s-1)$, $k_2 = 3(4s-1)$, $\lambda_2 = 2u(2s-1)$, $\rho_{12} = u(4s-3)$, $\rho_{22} = u$, $s = 1, 2, ...,$ $u = 4, 5, ...,$

(ix) $\rho = \frac{-1}{s+114}$, $v = 7$, $b_1 = 21$, $r_1 = 6$, $k_1 = 2$, $\lambda_1 = 1$ *and* $v = 7$, $b_2 = s+13$, $r_2 = s+13$, $k_2 = 7$, $\lambda_2 = s+11$, $\rho_{12} = s+11$, $\rho_{22} = 6$, $s = 1, 2, ...,$

(x) $\rho = \frac{-1}{s^2+15s+65}$, $v = 9$, $b_1 = 18$, $r_1 = 8$, $k_1 = 4$, $\lambda_1 = 3$ *and* $v = 9$, $b_2 = 3(s+4)$, $r_2 = 2(s+4)$, $k_2 = 6$, $\lambda_2 = s+5$, $\rho_{12} = 8$, $\rho_{22} = s$, $s = 1, 2, ...,$

(xi) $\rho = \frac{-1}{4s(4s+3)+u+7}$, $v = 9$, $b_1 = 12s$, $r_1 = 4s$, $k_1 = 3$, $\lambda_1 = s$ and $v = 9$, $b_2 = u+8$, $r_2 = u+8$, $k_2 = 9$, $\lambda_2 = u+7$, $\rho_{12} = u$, $\rho_{22} = 4$, $s, u = 1, 2, ...,$

(xii) $\rho = \frac{-1}{s+175}$, $v = 12$, $b_1 = 33$, $r_1 = 11$, $k_1 = 4$, $\lambda_1 = 3$ and $v = 12$, $b_2 = s + 22$, $r_2 = s + 22$, $k_2 = 12$, $\lambda_2 = s + 20$, $\rho_{12} = s$, $\rho_{22} = 11$, $s = 1, 2, ...,$

(xiii) $\rho = \frac{-1}{s+62}$, $v = 13$, $b_1 = 13$, $r_1 = 4$, $k_1 = 4$, $\lambda_1 = 1$ and $v = 13$, $b_2 = s + 25$, $r_2 = s + 25$, $k_2 = 13$, $\lambda_2 = s + 23$, $\rho_{12} = s + 1$, $\rho_{22} = 12$, $s = 1, 2, ...,$

then the chemical balance design with the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1, 0, 1)$ *given by (4) and with the variance matrix of errors* $\sigma^2\mathbf{G}$*, where the matrix* $\mathbf{G}$ *is of the form (1), is optimal.*

Theorem 7. *Let* $\rho = \frac{-1}{(u+1)(4s^2-1)+u(t-1)(u(t-1)+2)}$. *If the parameters of the balanced incomplete block designs are equal to* $v = 4s^2 - 1$, $b_1 = 4s^2 - 1$, $r_1 = 2s^2 - 1$, $k_1 = 2s^2 - 1$, $\lambda_1 = s^2 - 1$ *and the parameters of the ternary balanced block designs are equal to* $v = 4s^2 - 1$, $b_2 = u(4s^2 - 1)$, $r_2 = u(4s^2 - t)$, $k_2 = 4s^2 - t$, $\lambda_2 = u(4s^2 - 2t + 1)$, $\rho_{12} = u(4s^2 - t^2 + 2(t - 1))$, $\rho_{22} = \frac{1}{2}u(t - 1)(t - 2)$, $s = 2, 3, ...,$ $u = 1, 2, ...,$ $t = 3, 4, 5,$ *except the case* $s = 2$ *and* $t = 5$*, then the chemical balance design with the design matrix* $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1, 0, 1)$ *given by (4) and with the variance matrix of errors* $\sigma^2\mathbf{G}$*, where the matrix* $\mathbf{G}$ *is of the form (1), is optimal.*

Theorem 8. *Let* $\rho = \frac{-1}{(u+1)(4s+3)+u(t+1)(u(t+1)+2)}$. *If the parameters of the balanced incomplete block designs are equal to* $v = 4s+3$, $b_1 = 4s+3$, $r_1 = 2s+1$, $k_1 = 2s+1$, $\lambda_1 = s$ *and the parameters of the ternary balanced block designs are equal to* $v = 4s + 3$, $b_2 = u(4s + 3)$, $r_2 = u(4s + 2 - t)$, $k_2 = 4s + 2 - t$, $\lambda_2 = u(4s - 2t + 1)$, $\rho_{12} = u(4s - t^2 - 2(t - 1))$, $\rho_{22} = \frac{1}{2}ut(t + 1)$, $t = 1, 2, 3,$ $s = t, t + 1, ...,$ $u = 1, 2, ...,$ *except the case* $s = t = 3$*, then the chemical balance design with the design matrix* $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1, 0, 1)$ *given by (4) and with the variance matrix of errors* $\sigma^2\mathbf{G}$*, where the matrix* $\mathbf{G}$ *is of the form (1), is optimal.*

Theorem 9. *Let* $\rho = \frac{-1}{(u+1)(8s+7)+u(t+1)(u(t+1)+2)}$. *If the parameters of the balanced incomplete block designs are equal to* $v = 8s+7$, $b_1 = 8s+7$, $r_1 = 4s+3$, $k_1 = 4s+3$, $\lambda_1 = 2s+1$ *and the parameters of the ternary balanced block designs are equal to* $v = 8s+7$, $b_2 = 8s+7$, $r_2 = u(8s+6-t)$, $k_2 = 8s + 6 - t$, $\lambda_2 = u(8s - 2t + 5)$, $\rho_{12} = u(8s + 7 - (t + 1)^2)$, $\rho_{22} = \frac{1}{2}ut(t + 1)$, $t = 1, 2, 3,$ $u, s = 1, 2, ...,$ *except the case* $s = 1$ *and* $t = 3$*, then the chemical balance design with the design matrix* $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1, 0, 1)$ *given by*

(2) and with the variance matrix of errors $\sigma^2\mathbf{G}$, where the matrix $\mathbf{G}$ is of the form (1), is optimal.

Theorem 10. *Let $\rho = \frac{-1}{14s^2+u-3}$. If the parameters of the balanced incomplete block designs are equal to:*

(i) $v = 4s^2, \quad b_1 = 4s^2, \quad r_1 = 2s^2 + s, \quad k_1 = 2s^2 + s, \quad \lambda_1 = s^2 + s,$

(ii) $v = 4s^2, \quad b_1 = 4s^2, \quad r_1 = 2s^2 - s, \quad k_1 = 2s^2 - s, \quad \lambda_1 = s^2 - s,$

and the parameters of the ternary balanced block designs are equal to $v = 4s^2, \quad b_2 = 8s^2+u-2, \quad r_2 = 8s^2+u-2, \quad k_2 = 4s^2, \quad \lambda_2 = 8s^2+u-4, \quad \rho_{12} = u, \quad \rho_{22} = 4s^2 - 1, \quad s, u = 1, 2, ...,$ *then the chemical balance design with the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1, 0, 1)$ given by (4) and with the variance matrix of errors $\sigma^2\mathbf{G}$, where the matrix $\mathbf{G}$ is of the form (1), is optimal.*

Theorem 11. *Let $\rho = \frac{-1}{(u+2)(4s+1)+u(t+1)(u(t+1)+4)+3}$. If the parameters of the balanced incomplete block designs are equal to $v = 4s + 1, \quad b_1 = 2(4s + 1), \quad r_1 = 4s, \quad k_1 = 2s, \quad \lambda_1 = 2s - 1$ and the parameters of the ternary balanced block designs are equal to $v = 4s + 1, \quad b_2 = u(4s + 1), \quad r_2 = u(4s-t), \quad k_2 = 4s-t, \quad \lambda_2 = u(4s-2t-1), \quad \rho_{12} = u(4s+1-(t+1)^2), \quad \rho_{22} = \frac{1}{2}ut(t + 1), \quad t = 1, 2, 3, \quad u, s = 1, 2, ..., \quad 4s + 1$ is a prime or a prime power, then the chemical balance design with the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1, 0, 1)$ given by (4) and with the variance matrix of errors $\sigma^2\mathbf{G}$, where the matrix $\mathbf{G}$ is of the form (1), is optimal.*

Theorem 12. *Let $\frac{-1}{n-1} < \rho < 0$. If for a given ρ the parameters of the balanced incomplete block designs are equal to: $v = 5, \quad b_1 = 10, \quad r_1 = 4, \quad k_1 = 2, \quad \lambda_1 = 1$ and the parameters of the ternary balanced block designs are equal to:*

(i) $\rho = \frac{-1}{s+19}, \quad v = 5, \quad b_2 = s + 9, \quad r_2 = s + 9, \quad k_2 = 5, \quad \lambda_2 = s + 7, \quad \rho_{12} = s + 1, \quad \rho_{22} = 4,$

(ii) $\rho = \frac{-3}{s^2+21s+51}, \quad v = 5, \quad b_2 = 5(s + 1), \quad r_2 = 4(s + 1), \quad k_2 = 4, \quad \lambda_2 = 3s + 2, \quad \rho_{12} = 4s, \quad \rho_{22} = 2,$

(iii) $\rho = \frac{-1}{4s^2+29s+55}, \quad v = 5, \quad b_2 = 5(s + 2), \quad r_2 = 3(s + 2), \quad k_2 = 3, \quad \lambda_2 = s + 3, \quad \rho_{12} = s + 6, \quad \rho_{22} = s,$

$s = 1, 2, ...,$ then the chemical balance design with the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1, 0, 1)$ given by (4) and with the variance matrix of errors $\sigma^2\mathbf{G}$, where the matrix $\mathbf{G}$ is of the form (1), is optimal.

Theorem 13. *Let* $\frac{-1}{n-1} < \rho < 0$*. If for a given* ρ *the parameters of the balanced incomplete block designs are equal to* $v = 12,\quad b_1 = 22,\quad r_1 = 11,\quad k_1 = 6,\quad \lambda_1 = 5$ *and the parameters of the ternary balanced block designs are equal to:*

(i) $\rho = \frac{-1}{14},\quad v = 12,\quad b_2 = 18,\quad r_2 = 15,\quad k_2 = 10,\quad \lambda_2 = 11,\quad \rho_{12} = 1,\quad \rho_{22} = 7,$

(ii) $\rho = \frac{-1}{4s^2+26s+61},\quad v = 12,\quad b_2 = 3(2s+5),\quad r_2 = 2(2s+5),\quad k_2 = 8,\quad \lambda_2 = 2(s+3),\quad \rho_{12} = 6-2s,\quad \rho_{22} = 3s+2,\quad s = 0,1,2,$

$s = 1,2,...,$ *then the chemical balance design with the design matrix* $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ *given by (4) and with the variance matrix of errors* $\sigma^2\mathbf{G}$*, where the matrix* $\mathbf{G}$ *is of the form (1), is optimal.*

Theorem 14. *Let* $\frac{-1}{n-1} < \rho < 0$*. If for a given* ρ *the parameters of the balanced incomplete block designs are equal to* $v = 4s+3,\quad b_1 = 4s+3,\quad r_1 = 2s+1,\quad k_1 = 2s+1,\quad \lambda_1 = s$ *and the parameters of the ternary balanced block designs are equal to:*

(i) $\rho = \frac{-3}{36s+3u+22},\quad v = 4s+3,\quad b_2 = 8s+u+5,\quad r_2 = 8s+u+5,\quad k_2 = 4s+3,\quad \lambda_2 = 8s+u+3,\quad \rho_{12} = u+1,\quad \rho_{22} = 2(2s+1),\quad s = 1,2,...,$

(ii) $\rho = \frac{-2}{16s+2u+9},\quad v = 4s+3,\quad b_2 = 4s+u+2,\quad r_2 = 4s+u+2,\quad k_2 = 4s+3,\quad \lambda_2 = 4s+u+1,\quad \rho_{12} = u,\quad \rho_{22} = 2s+1,\quad s = 2,3,$

$u = 1,2,...,\quad 4s+3$ *is a prime or a prime power, then the chemical balance design with the design matrix* $\mathbf{X} \in \mathbf{\Phi}_{n\times p,m}(-1,0,1)$ *given by (4) and with the variance matrix of errors* $\sigma^2\mathbf{G}$*, where the matrix* $\mathbf{G}$ *is of the form (1), is optimal.*

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