DOI: 10.2478/bile-2025-0012

Biometrical Letters
Vol. 62 (2025), No. 2, 215-225

New classes of D-optimal chemical balance weighing designs with correlated errors

Małgorzata Graczyk[1], Bronisław Ceranka[1], Teresa Oliveira[2]

[1]Department of Mathematical and Statistical Methods, Poznań University of Life Sciences, Wojska Polskiego 28, 60-638 Poznań, Poland, e-mails: malgorzata.graczyk@up.poznan.pl, bronislaw.ceranka@up.poznan.pl
[2]Department of Mathematical and Statistical Methods, Universidade Aberta, Rua da Escola Politécnica 147, 1269-001 Lisboa, Portugal, e-mail: teresa.oliveira@uab.pt

SUMMARY

This study presents several results concerning the determination of chemical balance weighing designs that satisfy the criterion of D-optimality. The designs are examined under the assumption that all measurements exhibit equal positive correlation and identical variances. We introduce a method for augmenting a regular D-optimal chemical balance weighing design with additional measurements in order to obtain a highly D-efficient chemical balance weighing design.

Key words: chemical balance weighing, D-optimality, design of experiments

1. Introduction

Let us consider the linear model $\mathbf{y} = \mathbf{Xw} + \mathbf{e}$, where $\mathbf{y}$ is an $n \times 1$ random vector of observations, the matrix of a chemical balance weighing design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$, where $\mathbf{\Phi}_{n\times p}\{-1,0,1\}$ denotes the class of $n \times p$ matrices $\mathbf{X} = (x_{ij})$, $i = 1,2,\dots,n$, $j = 1,2,\dots,p$, of known elements equal to –1, 0 or 1. Moreover, $\mathbf{w}$ is a $p \times 1$ vector of unknown measurements of objects, $\mathbf{e}$ is an $n \times 1$ random vector of errors with E($\mathbf{e}$)=$\mathbf{0}_n$ and Var($\mathbf{e}$)=$\sigma^2\mathbf{G}$, where $\mathbf{0}_n$ is the $n \times 1$ vector with zero elements everywhere, σ^2 is a constant, and $\mathbf{G}$ is a known $n \times$ n positive definite matrix of the form

$$\mathbf{G} = (1-\rho)\mathbf{I}_n + \rho\mathbf{1}_n\mathbf{1}_n', \quad 0 < \rho < 1. \tag{1.1}$$

Such a form of the matrix $\mathbf{G}$ is used when the measurement errors are equally positively correlated. In order to estimate $\mathbf{w}$ we use the normal equations $\mathbf{X}'\mathbf{G}^{-1}\mathbf{X}\mathbf{w} = \mathbf{X}'\mathbf{G}^{-1}\boldsymbol{y}$, and if the matrix $\mathbf{X}$ is of full column rank then the generalized least squares estimator of $\mathbf{w}$ is given by $\hat{\mathbf{w}} = (\mathbf{X}'\mathbf{G}^{-1}\mathbf{X})^{-1}\mathbf{X}'\mathbf{G}^{-1}\boldsymbol{y}$ and $\mathrm{Var}(\widehat{\mathbf{w}}) = \sigma^2(\mathbf{X}'\mathbf{G}^{-1}\mathbf{X})^{-1}$. Many theoretical studies offer insights that inform the choice of optimal designs (see, for example, Ceranka and Graczyk 2017, 2018). However, for various combinations of a number of objects and a number of measurements, existing construction techniques described in the literature—such as those of Ceranka and Graczyk (2014, 2015)—do not allow us to obtain a regular D-optimal design. Here, we introduce a new construction method to address this limitation. Assuming that the matrix G conforms to the structure outlined in equation (1.1), Ceranka and Graczyk (2014) provide criteria for establishing a regular D-optimal chemical balance weighing design. They also present examples illustrating how these criteria are applied, utilizing the incidence matrices of balanced incomplete block designs. In Graczyk and Ceranka (2023) some issues regarding the optimality of chemical balance weighing designs are presented for another form of the matrix $\sigma^2\mathbf{G}$. In the present paper, we study D-optimal designs; the design $\mathbf{X}_{\mathrm{D}}$ is D-optimal in the class of all possible design matrices $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ if

$$\det\left(\mathbf{X}_{\mathrm{D}}'\mathbf{G}^{-1}\mathbf{X}_{\mathrm{D}}\right) = max\left\{\det(\mathbf{M}^{-1}) : \mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}\right\}, \tag{1.2}$$

where $\mathbf{M} = \mathbf{X}'\mathbf{G}^{-1}\mathbf{X}$. Next, we present the definition and theorem regarding regular D-optimal designs given in the above paper for this case. Let $m = max\{m_1, m_2, \dots, m_p\}$, where m_j, $j = 1,2,\dots,p$, represents the number of elements equal to –1 or 1 in the j^{th} column of $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$.

It should be noted that according to the definition given in Ceranka and Graczyk (2014), any chemical balance weighing design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1.1), is regular D-optimal if and only if $\det(\mathbf{M}) = \left(\frac{m}{1-\rho}\right)^p$. Let us note that a regular D-optimal design is D-optimal, whereas the converse statement may not be true. Moreover, any chemical balance weighing design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1.1), is regular D-optimal if and only if $\mathbf{X}'\mathbf{X} = m\mathbf{I}_p$ and $\mathbf{X}'\mathbf{1}_n = \mathbf{0}_p$ if $0 < \rho < 1$.

The condition $\mathbf{X}'\mathbf{1}_n = \mathbf{0}_p$ indicates that in each column of $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ the numbers of ones and negative ones in each column are the same. It is not possible to determine a regular D-optimal design for an arbitrary combination of numbers of measurements n and objects p. Proposed solutions of this problem and some methods of constructing D-optimal chemical balance weighing designs are given in the literature; see Ceranka and Graczyk (2014, 2015, 2017, 2018). In this work, we examine classes of design matrices that have not been previously addressed, and propose a new method for constructing D-optimal designs. The idea is to expand the design matrix of a regular D-optimal chemical balance weighing design and add $s = 1,2,3$ measurements in order to obtain the design matrix of a D-optimal chemical balance weighing design in the class $\mathbf{\Phi}_{n\times p}\{-1,0,1\}$, a class in which a regular D-optimal chemical balance weighing design does not exist.

An important issue here is the efficiency of a chemical balance weighing design with $s = 1,2,3$ additional measurements. Following the definition of Bulutoglu and Ryan (2009), the D-efficiency of a design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ is

$$\mathrm{D_{eff}}(\mathbf{X}) = \left(\frac{\det\left(\mathbf{X}'\ \mathbf{G}^{-1}\mathbf{X}\ \right)}{\det\left(\mathbf{X}_\mathrm{D}'\mathbf{G}^{-1}\mathbf{X}_\mathrm{D}\right)}\right)^{1/p}, \tag{1.3}$$

where $\mathbf{X}_\mathrm{D}$ is a regular D-optimal design. Some results relating to the efficiency of weighing designs were given by Smaga (2014).

2. The main result

Let us consider the design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ in the form

$$\mathbf{X} = \begin{bmatrix} \mathbf{X}_s \\ \boldsymbol{x}_1' \\ \boldsymbol{x}_2' \\ \boldsymbol{x}_3' \end{bmatrix} \tag{2.1}$$

where

(i) $\mathbf{X}_s \in \mathbf{\Phi}_{(n-s)\times p}\{-1,0,1\}$ is the design matrix of a regular D-optimal chemical balance weighing design,

(ii) the variance matrix of errors $\sigma^2\mathbf{G}$ is of the form

$$\mathbf{G} = \begin{bmatrix} \mathbf{G}_s & \mathbf{0}_{n-s} \\ \mathbf{0}'_{n-s} & \mathbf{I}_s \end{bmatrix}, \text{ where } \mathbf{G}_s = (1-\rho)\mathbf{I}_{n-s} + \rho\mathbf{1}_{n-s}\mathbf{1}'_{n-s}, 0 < \rho < 1, \quad (2.2)$$

(iii) $\mathbf{G}^{-1} = \frac{1}{1-\rho}\begin{bmatrix} \mathbf{I}_{n-s} - \frac{\rho}{1+\rho(n-s-1)}\mathbf{1}_{n-s}\mathbf{1}'_{n-s} & \mathbf{0}_{n-s}\mathbf{0}'_s \\ \mathbf{0}_s\mathbf{0}'_{n-s} & (1-\rho)\mathbf{I}_s \end{bmatrix}$

(iv) $\boldsymbol{x}_h$ are any $p \times 1$ vectors of elements equal to -1, 0 or 1, $\boldsymbol{x}'_h\boldsymbol{x}_h = t_h$, $\boldsymbol{x}'_h\boldsymbol{x}_{h'} = \boldsymbol{x}_{h'}\boldsymbol{x}'_h = u_{hh'}$,

(v) $s = 1,2,3,\ h, h' = 1,2,\dots,s,\ h \neq h'$, $1 \leq t_h \leq p$,

(vi) $\mathbf{M} = \frac{1}{1-\rho}\left(\mathbf{X}'_s\mathbf{X}_s + [\mathbf{1}'_s \otimes \boldsymbol{x}_h]\left(\mathbf{I}_2 - \frac{\rho}{1+\rho(n-1)}\mathbf{1}_2\mathbf{1}'_2\right)[\mathbf{1}_s \otimes \boldsymbol{x}'_h]\right)$.

Let us recall a theorem given by Harville (1997), which will be used later in the paper.

Lemma 2.1. (Theorem 18.1.1 in Harville, 1997) Let **R** represent an $n \times n$ matrix, **S** an $n \times m$ matrix, **T** an $m \times m$ matrix, and **U** an $m \times m$ matrix. If **R** and **T** are nonsingular, then

$$|\mathbf{R} + \mathbf{STU}| = |\mathbf{R}||\mathbf{T}||\mathbf{T}^{-1} + \mathbf{U}\mathbf{R}^{-1}\mathbf{S}|.$$

Theorem 2.1. Let $s = 1$. For any chemical balance weighing design in the form (2.1) $\mathbf{X} = \begin{bmatrix} \mathbf{X}_s \\ \boldsymbol{x}'_1 \end{bmatrix} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where **G** is of the form (2.2),

$$\det(\mathbf{M}) \leq \left(\frac{m}{1-\rho}\right)^p \left(1 + \frac{p}{m}\cdot(1-\rho)\right). \quad (2.3)$$

Proof. For the variance matrix of errors given in the form (2.2) we have

$$\mathbf{G}^{-1} = \frac{1}{1-\rho}\begin{bmatrix} \mathbf{I}_{n-1} - \frac{\rho}{1+\rho(n-2)}\mathbf{1}_{n-1}\mathbf{1}'_{n-1} & \mathbf{0}_{n-1} \\ \mathbf{0}'_{n-1} & 1-\rho \end{bmatrix}.$$

For $s = 1$, the design matrix $\mathbf{X}_s$ is regular D-optimal and $\mathbf{X}_s'\mathbf{X}_s = m\mathbf{I}_p$. Since $\mathbf{X}_1'\mathbf{1}_{n-1} = \mathbf{0}_p$ then $\mathbf{M} = \frac{1}{1-\rho}(\mathbf{X}_1'\mathbf{X}_1 + (1-\rho)\boldsymbol{x}_1\boldsymbol{x}_1')$ and hence $\det(\mathbf{M}) = \left(\frac{1}{1-\rho}\right)^p \det(\mathbf{X}_s'\mathbf{X}_s + (1-\rho)\boldsymbol{x}_1'\boldsymbol{x}_1)$.

According to Theorem 18.1.1 in Harville (1997) we have

$$\det\left(\mathbf{X}_s'\mathbf{X}_s + \frac{1+\rho(n-2)}{1+\rho(n-1)}\boldsymbol{x}_1'\boldsymbol{x}_1\right) = m^p\left(1 + \frac{(1-\rho)t_1}{m}\right).$$

Consequently $\det(\mathbf{M}) = \left(\frac{m}{1-\rho}\right)^p\left(1 + \frac{(1-\rho)t_1}{m}\right)$. We maximize $\det(\mathbf{M})$ under a given matrix $\mathbf{G}$. Because $\det(\mathbf{M})$ is a function of t_1, $\det(\mathbf{M})$ is maximal if and only if $t_1 = p$. Hence the theorem.

Theorem 2.2. Let $s = 1$. Any chemical balance weighing design of the form (2.1) $\mathbf{X} = \begin{bmatrix}\mathbf{X}_s \\ \boldsymbol{x}_1'\end{bmatrix} \in \boldsymbol{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (2.2), is D-optimal if

(i) $\mathbf{X}_s'\mathbf{X}_s = m\mathbf{I}_p$

(ii) $\mathbf{X}_s'\mathbf{1}_{n-1} = \mathbf{0}_p$

(iii) $\boldsymbol{x}_1'\boldsymbol{x}_1 = p$.

Proof. It is easy to see that $\det(\mathbf{M})$ as given by (2.3) attains its upper bound if conditions (i)–(iii) are fulfilled.

The D-efficiency of a design of the form (1.2) $\mathbf{X} = \begin{bmatrix}\mathbf{X}_s \\ \boldsymbol{x}_1'\end{bmatrix} \in \boldsymbol{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (2.2), equals $\mathrm{D_{eff}}(\mathbf{X}) = \frac{m}{m+1}\sqrt[p]{1 + \frac{p}{m}\cdot(1-\rho)}$.

Theorem 2.3. Let $s = 2$. For any chemical balance weighing design in the form (2.1) $\mathbf{X} = \begin{bmatrix}\mathbf{X}_s \\ \boldsymbol{x}_1' \\ \boldsymbol{x}_2'\end{bmatrix} \in \boldsymbol{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (2.2),

$$\det(\mathbf{M}) \le \left(\frac{m}{1-\rho}\right)^p \cdot \begin{cases} \left(1+\frac{(1-\rho)p}{m}\right)^2 & \text{for even } p \\ \left(1+\frac{(1-\rho)(p-1)}{m}\right)\left(1+\frac{(1-\rho)(p+1)}{m}\right) & \text{for odd } p \end{cases}. (2.4)$$

Proof. For the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ of the form (2.4) and with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (2.2), we have

$$\det(\mathbf{M}) = \left(\frac{1}{1-\rho}\right)^p \det(\mathbf{X}_s'\mathbf{X}_s + (1-\rho)\textstyle\sum_{h=1}^{s} \boldsymbol{x}_h\boldsymbol{x}_h').$$

According to Theorem 18.1.1 in Harville (1997) we obtain

$$\det(\mathbf{M}) = \left(\frac{1}{1-\rho}\right)^p m^p(1-\rho)^2 \det\left(\frac{1}{1-\rho}\mathbf{I}_2 + \frac{1}{m}\begin{bmatrix}\boldsymbol{x}_1 & \boldsymbol{x}_2\end{bmatrix}\begin{bmatrix}\boldsymbol{x}_1' \\ \boldsymbol{x}_2'\end{bmatrix}\right).$$

From the assumptions $\boldsymbol{x}_h'\boldsymbol{x}_h = t_h$, $\boldsymbol{x}_h'\boldsymbol{x}_{h'} = \boldsymbol{x}_{h'}\boldsymbol{x}_h' = u_{hh'}$ it follows that $\det(\mathbf{M}) = \frac{m^p}{(1-\rho)^{p-s}}\left(\left(\frac{1}{1-\rho}+\frac{t_1}{m}\right)\left(\frac{1}{1-\rho}+\frac{t_2}{m}\right) - \left(\frac{u_{12}}{m}\right)^2\right)$.

The maximum of $\det(\mathbf{M})$ is attained if and only if the maximum of $\frac{1}{1-\rho}+\frac{t_h}{m}, h = 1,2,\dots,s$ is attained and simultaneously the minimum of $\left(\frac{u_{12}}{m}\right)^2$ is attained. $\det(\mathbf{M})$ as a function of t_h attains a maximal value if and only if $t_h = p$ and consequently $\det(\mathbf{M}) = \frac{m^p}{(1-\rho)^{p-s}}\left(\left(\frac{1}{1-\rho}+\frac{p}{m}\right)^2 - \left(\frac{u_{12}}{m}\right)^2\right)$. Let us note that u_{12} is the scalar product of two rows of the matrix having elements -1 and 1; $u_{12} = 0$ if and only if p is even. In this case $\det(\mathbf{M}) \le \left(\frac{m}{1-\rho}\right)^p\left(1+\frac{(1-\rho)p}{m}\right)^2$. If p is odd, the condition $u_{12} = 0$ is never fulfilled. Therefore, in such a case the maximum of $\det(\mathbf{M})$ is attained if and only if $u_{12} = \pm 1$. Then $\det(\mathbf{M}) \le \left(\frac{m}{1-\rho}\right)^p\left(1+\frac{(1-\rho)(p-1)}{m}\right)\left(1+\frac{(1-\rho)(p+1)}{m}\right)$. Hence the theorem.

Theorem 2.4. Let $s = 2$. Any chemical balance weighing design of the form (2.1) $\mathbf{X} = \begin{bmatrix}\mathbf{X}_s \\ \boldsymbol{x}_1' \\ \boldsymbol{x}_2'\end{bmatrix} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (2.2), is D-optimal if

(i) $\mathbf{X}_s'\mathbf{X}_s = m\mathbf{I}_p$

(ii) $\mathbf{X}_s'\mathbf{1}_{n-s} = \mathbf{0}_p$

(iii) $\boldsymbol{x}_h'\boldsymbol{x}_h = p, h = 1,2$

(iv) $u_{12} = \begin{cases} 0 & \text{for even } p \\ -1 & \text{for odd } p \end{cases}.$

Proof. It is easy to see that $\det(\mathbf{M})$ as given by (2.4) attains its upper bound if $\mathbf{X}_s'\mathbf{X}_s = m\mathbf{I}_p$, $\mathbf{X}_s'\mathbf{1}_{n-s} = \mathbf{0}_p$ and $\boldsymbol{x}_h'\boldsymbol{x}_h = p$. Moreover, for an even number of objects if $\boldsymbol{x}_{h'}\boldsymbol{x}_h' = p$ and for an odd number of objects if $\boldsymbol{x}_{h'}\boldsymbol{x}_h' = \pm 1, h, h' = 1,2, \dots, s,\ h \neq h'$.

The D-efficiency of a design $\mathbf{X} \in \boldsymbol{\Phi}_{n\times p}\{-1,0,1\}$ of the form (2.1) $\mathbf{X} = \begin{bmatrix} \mathbf{X}_s \\ \boldsymbol{x}_1' \\ \boldsymbol{x}_2' \end{bmatrix} \in \boldsymbol{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (2.2), equals

$$\mathrm{D_{eff}}(\mathbf{X}) = \frac{m}{m+s}\begin{cases} \sqrt[p]{\left(1+\frac{(1-\rho)p}{m}\right)^2} & \text{if } p \equiv 0\ (mod\ 2) \\ \sqrt[p]{\left(1+\frac{(1-\rho)(p-1)}{m}\right)\left(1+\frac{(1-\rho)(p+1)}{m}\right)} & \text{if } p+1 \equiv 0\ (mod\ 2) \end{cases}.$$

Theorem 2.5. Let $s = 3$. For any chemical balance weighing design in the form (2.1) $\mathbf{X} = \begin{bmatrix} \mathbf{X}_s \\ \boldsymbol{x}_1' \\ \boldsymbol{x}_2' \\ \boldsymbol{x}_3' \end{bmatrix} \in \boldsymbol{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (2.2),

(i) If $p \equiv 0\ (mod\ 4)$ then $\det(\mathbf{M}) \leq \left(\frac{m}{1-\rho}\right)^p \left(1+\frac{(1-\rho)p}{m}\right)^3$

(ii) If $p+2 \equiv 0\ (mod\ 4)$then

$$\det(\mathbf{M}) \leq \left(\frac{m}{1-\rho}\right)^p \left(1+\frac{(1-\rho)p}{m}\right)\left(1+\frac{(1-\rho)(p-2)}{m}\right)\left(1+\frac{(1-\rho)(p+2)}{m}\right)$$

(iii) If $p+1 \equiv 0\ (mod\ 4)$then

$$\det(\mathbf{M}) \leq \left(\frac{m}{1-\rho}\right)^p \left(\left(1+\frac{(1-\rho)p}{m}\right)^3 - 2\left(\frac{1-\rho}{m}\right)^3 - 3\frac{1-\rho}{m^2}\left(1+\frac{(1-\rho)p)}{m}\right)\right)$$

(iv) If $p+3 \equiv 0\ (mod\ 4)$then

$$\det(\mathbf{M}) \leq \left(\frac{m}{1-\rho}\right)^p \left(\left(1+\frac{(1-\rho)p}{m}\right)^3 + 2\left(\frac{1-\rho}{m}\right)^3 - 3\frac{1-\rho}{m^2}\left(1+\frac{(1-\rho)p)}{m}\right)\right).$$

Proof. Let $s = 3$. For the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ of the form (2.4) and with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (2.2), we have $\det(\mathbf{M}) = \left(\frac{1}{1-\rho}\right)^p \det(\mathbf{X}_s'\mathbf{X}_s + (1-\rho)\sum_{h=1}^{s}\boldsymbol{x}_h\boldsymbol{x}_h')$. According to Theorem 18.1.1 in Harville (1997) we obtain

$$\det(\mathbf{M}) = \left(\frac{1}{1-\rho}\right)^p m^p(1-\rho)^3 \det\left(\frac{1}{1-\rho}\mathbf{I}_3 + \frac{1}{m}\begin{bmatrix}\boldsymbol{x}_1 & \boldsymbol{x}_2 & \boldsymbol{x}_3\end{bmatrix}\begin{bmatrix}\boldsymbol{x}_1' \\ \boldsymbol{x}_2' \\ \boldsymbol{x}_3'\end{bmatrix}\right).$$

From the assumptions $\boldsymbol{x}_h'\boldsymbol{x}_h = t_h,\ \boldsymbol{x}_h'\boldsymbol{x}_{h'} = \boldsymbol{x}_{h'}\boldsymbol{x}_h' = u_{hh'}$ it follows that $\det(\mathbf{M}) = \frac{m^p}{(1-\rho)^{p-s}}\left(\gamma_1\gamma_2\gamma_3 + 2\frac{u_{12}u_{13}u_{23}}{m^3} - \gamma_1\left(\frac{u_{23}}{m}\right)^2 - \gamma_2\left(\frac{u_{13}}{m}\right)^2 - \gamma_3\left(\frac{u_{12}}{m}\right)^2\right)$, where $\gamma_1 = \frac{1}{1-\rho} + \frac{t_1}{m}$, $\gamma_2 = \frac{1}{1-\rho} + \frac{t_2}{m}, \gamma_3 = \frac{1}{1-\rho} + \frac{t_3}{m}$. We are interested in determining the maximum of $\det(\mathbf{M})$ in the class $\mathbf{\Phi}_{n\times p}\{-1,0,1\}$. It is clear that $t_h = p, h = 1,2,\dots,s$. Consequently,

$$\det(\mathbf{M}) \leq \frac{m^p}{(1-\rho)^{p-3}}\left(\left(\frac{1}{1-\rho} + \frac{p}{m}\right)^2 + 2\frac{u_{12}u_{13}u_{23}}{m^3} - \left(\frac{1}{1-\rho} + \frac{p}{m}\right)\frac{u_{12}^2 + u_{13}^2 + u_{23}^2}{m^2}\right)$$

and the equality holds if and only if $t_h = p, h = 1,2,\dots,s$. Let us note that each of u_{12}, u_{13}, u_{23} is the scalar product of two rows of the matrix having elements -1 and 1, and $u_{12} = u_{13} = u_{23} = 0$ if and only if $p \equiv 0\ (mod\ 4)$. In this case $\det(\mathbf{M}) \leq \left(\frac{m}{1-\rho}\right)^p\left(1 + \frac{(1-\rho)p}{m}\right)^3$. If $p + 2 \equiv 0\ (mod\ 4)$ then the maximum of $\det(\mathbf{M})$ is attained if and only if $u_{hh'} = \pm 2$ or 0 with 0 occurring twice and ± 2 occurring once. Then

$$\det(\mathbf{M}) \leq \left(\frac{m}{1-\rho}\right)^p\left(1 + \frac{(1-\rho)p}{m}\right)\left(1 + \frac{(1-\rho)(p-2)}{m}\right)\left(1 + \frac{(1-\rho)(p+2)}{m}\right).$$

If $p + 1 \equiv 0\ (mod\ 4)$ then the maximum of $\det(\mathbf{M})$ is attained if and only if $u_{hh'} = -1$ or $u_{hh'} = \pm 2$ or 0 with $+1$ occurring twice and -1 occurring once. Then $\det(\mathbf{M}) \leq \left(\frac{m}{1-\rho}\right)^p\left(\left(1 + \frac{(1-\rho)p}{m}\right)^3 - 2\left(\frac{1-\rho}{m}\right)^3 - 3\frac{1-\rho}{m^2}\left(1 + \frac{(1-\rho)p)}{m}\right)\right)$.

If $p + 3 \equiv 0\ (mod\ 4)$ then the maximum of $\det(\mathbf{M})$ is attained if and only if $u_{hh'} = +1$ or $u_{hh'} = \pm 2$ or 0 with -1 occurring twice and $+1$ occurring once.

Then $\det(\mathbf{M}) \le \left(\frac{m}{1-\rho}\right)^p \left(\left(1+\frac{(1-\rho)p}{m}\right)^3 + 2\left(\frac{1-\rho}{m}\right)^3 - 3\frac{1-\rho}{m^2}\left(1+\frac{(1-\rho)p)}{m}\right)\right)$.

Hence the theorem.

Theorem 2.6. Let $s = 3$. Any chemical balance weighing design of the form (2.1) $\mathbf{X} = \begin{bmatrix} \mathbf{X}_s \\ \boldsymbol{x}_1' \\ \boldsymbol{x}_2' \\ \boldsymbol{x}_3' \end{bmatrix} \in \boldsymbol{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (2.2), is D-optimal if

(i) $\mathbf{X}_s'\mathbf{X}_s = m\mathbf{I}_p$

(ii) $\mathbf{X}_s'\mathbf{1}_{n-s} = \mathbf{0}_p$

(iii) $\boldsymbol{x}_h'\boldsymbol{x}_h = p, h = 1,2$

and

a. If $p \equiv 0\ (mod\ 4)$ then $u_{hh'} = 0$,
b. If $p + 2 \equiv 0\ (mod\ 4)$ then $u_{hh'} = \pm 2$ or 0 with 0 occurring twice and ± 2 occurring once,
c. If $p + 1 \equiv 0\ (mod\ 4)$ then $u_{hh'} = -1$ or $u_{hh'} = \pm 2$ or 0 with $+1$ occurring twice and -1 occurring once,
d. If $p + 3 \equiv 0\ (mod\ 4)$ then $u_{hh'} = +1$ or $u_{hh'} = \pm 2$ or 0 with -1 occurring twice and $+1$ occurring once.

Proof. It is easy to see that $\det(\mathbf{M})$ as given by (2.4) attains its upper bound if the above conditions are satisfied. The D-efficiency of a design $\mathbf{X} \in \boldsymbol{\Phi}_{n\times p}\{-1,0,1\}$ of the form (2.1) $\mathbf{X} = \begin{bmatrix} \mathbf{X}_s \\ \boldsymbol{x}_1' \\ \boldsymbol{x}_2' \\ \boldsymbol{x}_3' \end{bmatrix} \in \boldsymbol{\Phi}_{n\times p}\{-1,0,1\}$ with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (2.2) for $s = 3$, equals

$$\mathrm{D_{eff}}(\mathbf{X}) = \frac{m}{m+s}\begin{cases} \sqrt[p]{\left(1+\frac{(1-\rho)p}{m}\right)^{3}}, & p \equiv 0 \ (mod\ 4) \\ \sqrt[p]{\left(1+\frac{(1-\rho)p}{m}\right)\left(1+\frac{(1-\rho)(p-2)}{m}\right)\left(1+\frac{(1-\rho)(p+2)}{m}\right)}, & p+2 \equiv 0 (mod\ 4) \\ \sqrt[p]{\left(1+\frac{(1-\rho)p}{m}\right)^{3}-2\left(\frac{1-\rho}{m}\right)^{3}-3\frac{1-\rho}{m^2}\left(1+\frac{(1-\rho)p)}{m}\right)}, & p+1 \equiv 0 \ (mod\ 4) \\ \sqrt[p]{\left(1+\frac{(1-\rho)p}{m}\right)^{3}+2\left(\frac{1-\rho}{m}\right)^{3}-3\frac{1-\rho}{m^2}\left(1+\frac{(1-\rho)p)}{m}\right)}, & p+3 \equiv 0 \ (mod\ 4) \end{cases}$$

3. Discussion

This paper has identified several classes of chemical balance weighing designs that achieve D-optimality or exhibit high D-efficiency. However, for certain combinations of a number of objects and a number of measurements, it is not possible to derive a design possessing the properties described here. Nonetheless, additional avenues for constructing such designs should be explored, particularly those that build on existing block design matrices. It is worth noting that numerous applications of weighing designs exist; see for example the work of Banerjee (1975), which discusses their use in fields such as medicine, economics, and operational research, and papers by Sloane and Harwit (1976) and Koukouvinos and Seberry (1997) concerning the planning of experiments in optics. The possibility of using the weighing design matrix as an experimental plan in plant protection research is discussed by Graczyk (2013). In turn, Graczyk (2014) describes an experimental plan used in a comparison of the impact of NPK and various types of organic matter applied under the fore crop on the health of the stem base of the successor plant, spring barley. Additional applications of the designs examined here may arise in any context where the experimental scheme is derived from a factorial design.

References

Banerjee K.S. (1975): Weighing Designs For Chemistry, Medicine, Economics, Operations Research, Statistics. Marcel Dekker Inc., New York.

Bulutoglu D.A., Ryan K.J. (2009): D-optimal and near D-optimal 2k fractional factorial designs of resolution V. Journal of Statistical Planning and Inference 139: 16–22.
Ceranka B., Graczyk M. (2014): The problem of D-optimality in some experimental design. International Journal of Mathematics and Computer Applications Research, 4: 11–18.
Ceranka B., Graczyk M. (2015): Relations between ternary designs and D-optimal weighing designs with non-negative correlated errors. Colloquium Biometricum 145: 23–34.
Ceranka B., Graczyk M. (2017): Highly D-efficient weighing design and its construction. Acta Universitatis Lodziensis, Folia Oeconomica 331: 143–151.
Ceranka B., Graczyk M. (2018): Highly D-efficient weighing designs for an even number of objects. Revstat Statistical Journal 16(4): 475–486.
Graczyk M. (2013): Some applications on weighing designs. Biometrical Letters 50(1): 15–26.
Graczyk M. (2014): Relations between randomized block designs and weighing designs in examples. Colloquium Biometricum 44: 97–107.
Graczyk M., Ceranka B. (2023): D-optimal chemical balance weighing designs with positively correlated errors: part I. Biometrical Letters 60(1): 53–63.
Harville, D.A. (1997): Matrix Algebra from a Statistician's Perspective. Springer, New York. https://doi.org/10.1007/b98818
Koukouvinos Ch., Seberry J. (1997): Weighing matrices and their applications. Journal of Statistical Planning and Inference 6:, 91–101.
Sloane N.J.A., Harwit M. (1976): Masks for Hadamard transform optics, and weighing designs. Applied Optics 5: 107–114.
Smaga Ł. (2014): Necessary and sufficient conditions in the problem of D-optimal weighing designs with autocorrelated errors. Statistics and Probability Letters 92: 12–16.