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D-optimal chemical balance weighing designs with positively correlated errors: part III

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SUMMARY

In this paper we consider issues related to the determination of chemical balance weighing designs satisfying the criterion of D-optimality. This problem is considered under the assumption that the measurements are equally positively correlated and they have the same variances. We answer the question of how to add four runs to a regular D-optimal chemical balance weighing design in order to obtain a highly D-efficient design. The problem formulated in this way leads to an extension of results contained in a previous paper in *Biometrical Letters*.

Key words: chemical balance weighing design, plan of experiment

1. Introduction

The first stage of conducting an experiment is the preparation of its plan. This should ensure that estimators of unknown objects are obtained that satisfy the specified optimality criteria and that, at the same time, the proposed measurements are feasible. The planning of experiments plays an important role in the field of mathematical statistics. Among various factors that determine the most effective and best experimental plan, it is essential to define the criterion for optimality. Optimality criteria are functions of the information matrix for the design, so that the criterion determines the properties of the estimators. In this paper we formulate conditions for determining the experimental plan of chemical balance weighing designs, which can be used in fields such as agricultural research, economics, and optics.

We consider the design matrix $\mathbf{X} \in \mathbf{\Phi}_{n \times p}\{-1,0,1\}$, 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. Such a matrix is called the matrix of a chemical balance weighing design. The results of n measurement operations of p objects will fit the linear model $\mathbf{y} = \mathbf{X}\mathbf{w} + \mathbf{e}$, where $\mathbf{y}$ is an $n \times 1$ random vector of observations, $\mathbf{w}$ is a $p \times 1$ vector of uknown measurements of objects, and $\mathbf{e}$ 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 $p \times p$ positive definite matrix of the form

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

Assuming just such a form of the variance matrix $\mathbf{G}$, we indicate that the measurement errors are equally positively correlated. For estimation of $\mathbf{w}$ we use the normal equations $\mathbf{X}'\mathbf{G}^{-1}\mathbf{X}\mathbf{w} = \mathbf{X}'\mathbf{G}^{-1}\boldsymbol{y}$. If the matrix $\mathbf{X}$ is of full column rank then the generalized least squares estimator of $\mathbf{w}$ is given by $\widehat{\mathbf{w}} = (\mathbf{X}'\mathbf{G}^{-1}\mathbf{X})^{-1}\mathbf{X}'\mathbf{G}^{-1}\boldsymbol{y}$ and $\mathrm{Var}(\widehat{\mathbf{w}}) = \sigma^2(\mathbf{X}'\mathbf{G}^{-1}\mathbf{X})^{-1}$. The matrix $\mathbf{M} = \mathbf{X}'\mathbf{G}^{-1}\mathbf{X}$ is called the information matrix of the design $\mathbf{X}$.

Several optimality criteria minimizing selected functions of the matrix $\mathbf{M}$ are considered in the literature. One of them is D-optimality. We say that the design $\mathbf{X}_\mathrm{D}$ is D-optimal in the class of all possible design matrices $\mathbf{\Phi}_{n \times p}\{-1,0,1\}$ if $\det\left(\mathbf{X}_\mathrm{D}'\mathbf{G}^{-1}\mathbf{X}_\mathrm{D}\right) = max\left\{\det(\mathbf{M}^{-1}) \colon \mathbf{X} \in \mathbf{\Phi}_{n \times p}\{-1,0,1\}\right\}$.

Assuming that the matrix $\mathbf{G}$ conforms to the structure outlined in equation (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. We mention here the definition and theorem concerning a regular D-optimal design given in the above paper for this particular 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 $\mathbf{X} \in \mathbf{\Phi}_{n \times p}\{-1,0,1\}$.

Definition 1.1. 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), 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, but the inverse may not hold.

Theorem 1.1. 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), 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 $\mathbf{X}'\mathbf{1}_n = \mathbf{0}_p$ states that in each column of $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ the numbers of elements equal to -1 and 1 should be the same. Despite the abundance of theoretical literature offering insights into a wide range of optimal designs, we cannot identify a regular D-optimal design for an arbitrary combination of measurements and objects. Selected issues related to the solution of the problem and some construction methods for D-optimal chemical balance weighing designs appear in the literature; see Ceranka and Graczyk (2014, 2017, 2018), Sathe and Shenoy (1990), and Smaga (2014). Here, we study classes of design matrices that have not previously been considered, and give a new construction method for D-optimal designs.

Our idea is to expand the design matrix of a regular D-optimal chemical balance weighing design and to add $h = 1,2,3,4$ 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. Topics related to the addition of 1, 2 and 3 measurements were presented in Graczyk and Ceranka (2023 a, b). Adding a further measurement will enable us to extend the set of design classes in which we can determine the optimal design.

An important issue here is the efficiency of a chemical balance weighing design with an additional $h = 1,2,3,4$ 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 $\mathrm{D_{eff}}(\mathbf{X}) = \left(\frac{\det\left(\mathbf{X}'\ \mathbf{G}^{-1}\mathbf{X}\ \right)}{\det\left(\mathbf{X}_\mathrm{D}'\mathbf{G}^{-1}\mathbf{X}_\mathrm{D}\right)}\right)^{1/p}$, where $\mathbf{X}_\mathrm{D}$ is a regular D-optimal design.

2. The main result

Following the notation of Graczyk and Ceranka (2023 a, b), let $\mathbf{X}_\gamma \in \mathbf{\Phi}_{(n-\gamma)\times p}\{-1,0,1\}$ be the design matrix of a regular D-optimal chemical balance weighing design with the variance matrix of errors $\sigma^2\mathbf{G}_{\boldsymbol{\gamma}}$, where for $h = 4$, $\mathbf{G}_\gamma = g\left[(1-\rho)\mathbf{I}_{n-\gamma} + \rho\mathbf{1}_{n-\gamma}\mathbf{1}_{n-\gamma}'\right]$, i.e. $\mathbf{X}_\gamma'\mathbf{X}_\gamma = m\mathbf{I}_p$ and $\mathbf{X}_\gamma'\mathbf{1}_{n-\gamma} = \mathbf{0}_p$. The problem is how to add the fourth measurement to the optimal design to obtain a design that fulfils the proposed criteria. With reference to Graczyk and Ceranka (2023 a,b), in order to examine the addition of four measurements, $h = 4$, we consider the design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ in the form

$$\mathbf{X} = \begin{bmatrix} \mathbf{X}_\gamma \\ \boldsymbol{x}_1' \\ \boldsymbol{x}_2' \\ \boldsymbol{x}_3' \\ \boldsymbol{x}_4' \end{bmatrix}, \gamma = 4 \tag{2}$$

where the $\boldsymbol{x}_h$ are any $p \times 1$ vectors of elements equal to $-1, 0$ or 1, $\boldsymbol{x}_h'\boldsymbol{x}_h = t_h$, $\boldsymbol{x}_h'\boldsymbol{x}_{h'} = u_{hh'}, 1 \le t_h \le p, h, h' = 1,2,3,4, h \ne h'$. $\mathbf{X}_\gamma \in \mathbf{\Phi}_{(n-4)\times p}\{-1,0,1\}$ is the design matrix of a regular D-optimal chemical balance weighing design, i.e. $\mathbf{X}_\gamma'\mathbf{X}_\gamma = m\mathbf{I}_p$ and $\mathbf{X}_\gamma'\mathbf{1}_{n-4} = \mathbf{0}_p$.

Theorem 2.1. For any chemical balance weighing design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ in the form (2) with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1),

- If $p \equiv 0(mod4)$ then $\det(\mathbf{M}) \le \left(\frac{m}{1-\rho}\right)^p \left(1 + \frac{p}{m}\right)^3 \left(1 + \beta\frac{p}{m}\right)$
- If $p + 1 \equiv 0(mod4)$ then $\det(\mathbf{M}) \le \left(\frac{m}{1-\rho}\right)^p \left(1 + \frac{p+1}{m}\right)^3 \left(1 + \beta\frac{p-3}{m}\right)$

- If $p+2\equiv 0(mod 4)$ then $\det(\mathbf{M})\le\beta\left(\frac{m}{1-\rho}\right)^p\left[\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^4+8\left(\frac{\rho}{\delta}\right)^2\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)\left(\frac{\rho}{\delta}-\frac{2}{m}\right)+\left(\frac{\rho}{\delta}-\frac{2}{m}\right)^4-2\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^2\right]\left[2\left(\frac{\rho}{\delta}\right)^2\left(\frac{\rho}{\delta}-\frac{2}{m}\right)^2\right]-4\left(\frac{\rho}{\delta}\right)^2\left(\frac{\rho}{\delta}-\frac{2}{m}\right)^2$
- If $p+3\equiv 0(mod 4)$ then $\det(\mathbf{M})\le\left(\frac{m}{1-\rho}\right)^p\left(1+\frac{p-1}{m}\right)^3\left(1+\beta\frac{p+3}{m}\right)$
  where $\delta=1+\rho(n-5),\beta=\frac{\delta}{1+\rho(n-1)}$.

Proof. For $\mathbf{G}$ of the form (1) and $\mathbf{X}_1'\mathbf{1}_{n-4}=\mathbf{0}_p$ we have
$\mathbf{M}=\frac{1}{1-\rho}\left(\mathbf{X}_4'\mathbf{X}_4+[\boldsymbol{x}_1\ \ \boldsymbol{x}_2\ \ \boldsymbol{x}_3\ \ \boldsymbol{x}_4]\left(\mathbf{I}_4-\frac{\rho}{1+\rho(n-1)}\mathbf{1}_4\mathbf{1}_4'\right)\begin{bmatrix}\boldsymbol{x}_1'\\ \boldsymbol{x}_2'\\ \boldsymbol{x}_3'\\ \boldsymbol{x}_4'\end{bmatrix}\right)$. According to Theorem 18.1.1 of Harville (1997) we have $\det(\mathbf{M})=\det(\mathbf{X}_4'\mathbf{X}_4)\det\left(\mathbf{I}_4-\frac{\rho}{1+\rho(n-1)}\mathbf{1}_4\mathbf{1}_4'\right)\det\left(\mathbf{I}_4-\frac{\rho}{1+\rho(n-1)}\mathbf{1}_4\mathbf{1}_4'\right)^{-1}+\begin{bmatrix}\boldsymbol{x}_1'\\ \boldsymbol{x}_2'\\ \boldsymbol{x}_3'\\ \boldsymbol{x}_4'\end{bmatrix}(\mathbf{X}_4'\mathbf{X}_4)^{-1}[\boldsymbol{x}_1\ \ \boldsymbol{x}_2\ \ \boldsymbol{x}_3\ \ \boldsymbol{x}_4]$.
Since $\det\left(\mathbf{I}_4-\frac{\rho}{1+\rho(n-1)}\mathbf{1}_4\mathbf{1}_4'\right)=\beta$ we have $\det(\mathbf{M})=\beta\left(\frac{m}{1-\rho}\right)^p\det(\theta)$, where $\theta=\mathbf{I}_4+\frac{\rho}{\delta}\mathbf{1}_4\mathbf{1}_4'+\frac{1}{m}\begin{bmatrix}\boldsymbol{x}_1'\\ \boldsymbol{x}_2'\\ \boldsymbol{x}_3'\\ \boldsymbol{x}_4'\end{bmatrix}[\boldsymbol{x}_1\ \ \boldsymbol{x}_2\ \ \boldsymbol{x}_3\ \ \boldsymbol{x}_4]$. From the assumption $\boldsymbol{x}_h'\boldsymbol{x}_h=t_h,\ \boldsymbol{x}_h'\boldsymbol{x}_{h'}=u_{hh'},\ h,h'=1,2,3,4,h\neq h'$ it follows that
$\det(\theta)=\mathrm{A}+2\mathrm{B}+\mathrm{C}-\mathrm{D}-2\mathrm{E}$, where
$\mathrm{A}=\prod_{h=1}^{4}\left(1+\frac{\rho}{\delta}+\frac{t_h}{m}\right)$,
$\mathrm{B}=\sum_s\left(1+\frac{\rho}{\delta}+\frac{t_s}{m}\right)\prod_{h,h'}\left(1+\frac{\rho}{\delta}+\frac{u_{hh'}}{m}\right),\ \ h,h'=1,2,3,4,h\neq h'\neq s,$
$\mathrm{C}=\sum_{h,q,\varpi}\left(\frac{\rho}{\delta}+\frac{u_{1h}}{m}\right)^2\left(\frac{\rho}{\delta}+\frac{u_{q\varpi}}{m}\right)^2,\ \ h,q,\varpi=2,3,4,h\neq q\neq\varpi,$
$\mathrm{D}=\sum_s\prod_{h,h',s,t}\left(1+\frac{\rho}{\delta}+\frac{t_h}{m}\right)\left(1+\frac{\rho}{\delta}+\frac{t_{h'}}{m}\right)\left(\frac{\rho}{\lambda}+\frac{u_{st}}{m}\right)^2,\ h,h',q,\varpi=1,2,3,4,h<h'\neq q\neq\varpi,$
$\mathrm{E}=\sum_q\prod_{h,h',\varpi}\left(\frac{\rho}{\delta}+\frac{u_{1h}}{m}\right)\left(\frac{\rho}{\delta}+\frac{u_{1h'}}{m}\right)\left(\frac{\rho}{\delta}+\frac{u_{2q}}{m}\right)\left(\frac{\rho}{\delta}+\frac{u_{\varpi 4}}{m}\right),h=2,h'=3,q=4,\varpi=3$ or $h=2,h'=4,q=\varpi=3$ or $h=3,h'=4,q=3,\varpi=2$.

The maximum of $\det(\mathbf{M})$ is attained if and only if $t_h = p, h = 1,2,3,4$, and consequently $\det(\mathbf{M}) \leq \beta\left(\frac{m}{g(1-\rho)}\right)^p \left[\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^4 + 2\mathrm{H} + \mathrm{C} - \mathrm{D} - 2\mathrm{E}\right]$,
$\mathrm{H} = \left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)\sum_{h,h'}\left(\frac{\rho}{\delta}+\frac{u_{hh'}}{m}\right),\ \ h,h' = 1,2,3,4, h \neq h'$.
Let us note that $u_{hh'}, h, h' = 1,2,3,4, h \neq h'$ are the scalar products of two rows of the matrix having elements $-1, 1, 0$ and $u_{hh'} = 0$ if and only if $p \equiv 0(mod4)$. In this case $\det(\mathbf{M}) \leq \left(\frac{m}{1-\rho}\right)^p \left(1+\frac{p}{m}\right)^4 \left(1+\beta\frac{p}{m}\right)$.
If $p+1 \equiv 0(mod4)$ then the maximum of $\det(\mathbf{M})$ is attained if and only if $u_{hh'} = -1, h, h' = 1,2,3,4, h \neq h'$, then
$\det(\mathbf{M}) \leq \beta\left(\frac{m}{1-\rho}\right)^p \left[\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^4 + 8\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)\left(\frac{\rho}{\delta}-\frac{1}{m}\right)^3 - 6\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^2\left(\frac{\rho}{\delta}-\frac{1}{m}\right)^2 - 3\left(\frac{\rho}{\delta}-\frac{1}{m}\right)^4\right] = \left(\frac{m}{1-\rho}\right)^p\left(1+\beta\frac{p-3}{m}\right)\left(1+\frac{p+1}{m}\right)^3$.
If $p+2 \equiv 0(mod4)$ then the maximum of $\det(\mathbf{M})$ is attained if and only if $u_{hh'} = -2$ or 0 with 0 occurring four times and -2 occurring twice, $h, h' = 1,2,3,4, h \neq h'$. Then
$\det(\mathbf{M}) \leq \beta\left(\frac{m}{1-\rho}\right)^p \left\{\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^4 + 8\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)\left(\frac{\rho}{\delta}-\frac{2}{m}\right)\left(\frac{\rho}{\delta}\right)^2 + \left(\frac{\rho}{\lambda}-\frac{2}{m}\right)^4 - 2\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^2\left[2\left(\frac{\rho}{\delta}\right)^2 + \left(\frac{\rho}{\delta}-\frac{2}{m}\right)^2\right] - 4\left(\frac{\rho}{\delta}\right)^2\left(\frac{\rho}{\delta}-\frac{2}{m}\right)^2\right\}$
If $p+3 \equiv 0(mod4)$ then the maximum of $\det(\mathbf{M})$ is attained if and only if $u_{hh'} = 1, h, h' = 1,2,3,4, h \neq h'$. Then
$\det(\mathbf{M}) \leq \beta\left(\frac{m}{1-\rho}\right)^p \left[\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^4 + 8\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)\left(\frac{\rho}{\delta}+\frac{1}{m}\right)^3 - 6\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^2\left(\frac{\rho}{\delta}+\frac{1}{m}\right)^2 - 3\left(\frac{\rho}{\delta}-\frac{2}{m}\right)^4\right] = \left(\frac{m}{1-\rho}\right)^p\left(1+\frac{p-1}{m}\right)^3\left(1+\beta\frac{p+3}{m}\right)$.
This completes the proof of the theorem.

Definition 2.1. Any chemical balance weighing design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ of the form (2) with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1), is regular D-optimal if and only if

- If $p \equiv 0(mod4)$ then $\det(\mathbf{M}) = \left(\frac{m}{1-\rho}\right)^p\left(1+\frac{p}{m}\right)^3\left(1+\beta\frac{p}{m}\right)$
- If $p+1 \equiv 0(mod4)$ then $\det(\mathbf{M}) = \left(\frac{m}{1-\rho}\right)^p\left(1+\frac{p+1}{m}\right)^3\left(1+\beta\frac{p-3}{m}\right)$

- If $p+2 \equiv 0(mod 4)$ then $\det(\mathbf{M}) = \beta\left(\frac{m}{1-\rho}\right)^p \left\{\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^4 + 8\left(\frac{\rho}{\delta}\right)^2\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)\left(\frac{\rho}{\delta}-\frac{2}{m}\right) + \left(\frac{\rho}{\delta}-\frac{2}{m}\right)^4 - 2\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^2\left[2\left(\frac{\rho}{\delta}\right)^2 + \left(\frac{\rho}{\delta}-\frac{2}{m}\right)^2\right] - 4\left(\frac{\rho}{\delta}\right)^2\left(\frac{\rho}{\delta}-\frac{2}{m}\right)^2\right\}$
- If $p+3 \equiv 0(mod 4)$ then $\det(\mathbf{M}) = \left(\frac{m}{1-\rho}\right)^p \left(1+\frac{p-1}{m}\right)^3 \delta\left(1+\beta\frac{p+3}{m}\right)$

Theorem 2.2. Any chemical balance weighing design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ of the form (2) with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1), is D-optimal if

- $\boldsymbol{x}'_h\boldsymbol{x}_h = p, h = 1,2,3,4$ and
- $u_{hh'} =$

$$\begin{cases} 0 & \text{if } p \equiv 0(mod4) \\ -1 & \text{if } p+1 \equiv 0(mod4) \\ -2 \text{ or } 0, 0 \text{ occurs four times}, -2 \text{ occurs twice} & \text{if } p+2 \equiv 0(mod4) \\ 1 & \text{if } p+3 \equiv 0(mod4) \end{cases}.$$

The D-efficiency of a design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ of the form (2) with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1), equals

$$\mathrm{D_{eff}}(\mathbf{X}) = \frac{\boldsymbol{m}}{\boldsymbol{m+4}} \begin{cases} \sqrt[p]{\left(1+\frac{p}{m}\right)^3\left(1+\beta\frac{p}{m}\right)} & \text{for } p \equiv 0(mod4) \\ \sqrt[p]{\left(1+\frac{p+1}{m}\right)^3\left(1+\beta\frac{p-3}{m}\right)} & \text{for } p+1 \equiv 0(mod4) \\ \sqrt[p]{\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^3+\tau} & \text{for } p+2 \equiv 0(mod4) \\ \sqrt[p]{\left(1+\frac{p+1}{m}\right)^3\left(1+\beta\frac{p-3}{m}\right)} & \text{for } p+3 \equiv 0(mod4) \end{cases},$$

where

$$\tau = 8\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)\left(\frac{\rho}{\delta}-\frac{2}{m}\right)\left(\frac{\rho}{\delta}\right)^2 + \left(\frac{\rho}{\delta}-\frac{2}{m}\right)^4 - 2\left(1+\frac{\rho}{\delta}+\frac{p}{m}\right)^2\left[2\left(\frac{\rho}{\delta}\right)^2 + \left(\frac{\rho}{\delta}-\frac{2}{m}\right)^2\right] - 4\left(\frac{\rho}{\delta}\right)^2\left(\frac{\rho}{\delta}-\frac{2}{m}\right)^2.$$

Theorem 2.3. For any chemical balance weighing design $\mathbf{X} \in \mathbf{\Phi}_{n\times p}\{-1,0,1\}$ of the form (2) with the variance matrix of errors $\sigma^2\mathbf{G}$, where $\mathbf{G}$ is of the form (1), $\mathrm{D}_{\mathrm{eff}}(\mathbf{X})$ is a decreasing function of the variable ρ.

Proof. Let $p \equiv 0(mod4)$ and $\rho_1 < \rho_2$. We consider $\frac{m}{m+4}\sqrt[p]{\left(1+\frac{p}{m}\right)^3\left(1+\beta_1\frac{p}{m}\right)} > \frac{m}{m+4}\sqrt[p]{\left(1+\frac{p}{m}\right)^3\left(1+\beta_2\frac{p}{m}\right)}$. Hence $1+\frac{\beta_1 p}{m} - 1 - \frac{\beta_2 p}{m} = \frac{3p(\rho_2-\rho_1)}{m(1+\rho_1(n-1))(1+\rho_2(n-1))} > 0$. The steps of the proof for the cases $p+1 \equiv 0(mod4), p+2 \equiv 0(mod4), p+3 \equiv 0(mod4)$ are analogous.

3. Examples

Example 3.1: $p \equiv 0(mod4)$

In order to construct $\mathbf{X} \in \mathbf{\Phi}_{19\times 4}\{-1,0,1\}$, according to the theory presented above, we consider the matrix $\mathbf{X}_4 \in \mathbf{\Phi}_{15\times 4}\{-1,0,1\}$ of the D-optimal chemical balance weighing design

$$\mathbf{X}_4' = \begin{bmatrix} 0 & - & - & + & 0 & - & + & - & 0 & + & - & - & + & + & + \\ + & 0 & - & - & - & 0 & - & + & - & 0 & + & - & + & + & + \\ - & + & 0 & - & + & - & 0 & - & - & - & 0 & + & + & + & + \\ + & - & + & 0 & - & + & - & 0 & + & - & - & 0 & + & + & + \end{bmatrix},$$

where "+" denotes "+1" and "−" denotes "−1". Hence

$$\mathbf{X} = \begin{bmatrix} & \mathbf{X}_4 & & \\ + & + & + & - \\ + & + & - & + \\ + & - & + & + \\ - & + & + & + \end{bmatrix} \in$$

$\mathbf{\Phi}_{19\times 4}\{-1,0,1\}$ is a chemical balance weighing design with efficiency $\mathrm{D}_{\mathrm{eff}}(\mathbf{X}) = 0.9701$ for $\rho = 0.1$ and $\mathrm{D}_{\mathrm{eff}}(\mathbf{X}) = 0.9632$ for $\rho = 0.2$.

Example 3.2: $p+1 \equiv 0(mod4)$

To construct $\mathbf{X} \in \mathbf{\Phi}_{16\times 5}\{-1,0,1\}$, we consider the matrix $\mathbf{X}_4 \in \mathbf{\Phi}_{12\times 5}\{-1,0,1\}$ of the D-optimal chemical balance weighing design

$$\mathbf{X}_4' = \begin{bmatrix} + & + & - & 0 & 0 & + & - & - & + & 0 & - & 0 \\ + & + & - & - & 0 & 0 & + & 0 & - & + & 0 & - \\ + & + & + & - & - & 0 & 0 & - & 0 & - & + & 0 \\ + & + & 0 & + & - & - & 0 & 0 & - & 0 & - & + \\ + & + & 0 & 0 & + & - & - & + & 0 & - & 0 & - \end{bmatrix}.$$

Hence $\mathbf{X} = \begin{bmatrix} & & \mathbf{X}_4 & & \\ + & + & + & - & + \\ + & + & - & + & + \\ + & - & + & + & + \\ - & + & + & + & + \end{bmatrix} \in \mathbf{\Phi}_{16\times 5}\{-1,0,1\}$ is a chemical balance weighing design with efficiency $\mathrm{D_{eff}}(\mathbf{X}) = 0.9606$ for $\rho = 0.1$ and $\mathrm{D_{eff}}(\mathbf{X}) = 0.9564$ for $\rho = 0.2$.

Example 3.3: $p + 2 \equiv 0(mod4)$

In order to form $\mathbf{X} \in \mathbf{\Phi}_{16\times 6}\{-1,0,1\}$, we consider the matrix $\mathbf{X}_4 \in \mathbf{\Phi}_{12\times 6}\{-1,0,1\}$ of the D-optimal chemical balance weighing design

$$\mathbf{X}_4' = \begin{bmatrix} + & - & 0 & - & - & 0 & - & + & 0 & + & + & 0 \\ 0 & + & - & 0 & - & - & 0 & - & + & 0 & + & + \\ - & 0 & + & - & 0 & - & + & 0 & - & + & 0 & + \\ - & - & 0 & + & - & 0 & + & + & 0 & - & + & 0 \\ 0 & - & - & 0 & + & - & 0 & + & + & 0 & - & + \\ - & 0 & - & - & 0 & + & + & 0 & + & + & 0 & - \end{bmatrix}.$$

Hence $\mathbf{X} = \begin{bmatrix} & & \mathbf{X}_4 & & & \\ + & + & + & - & + & + \\ + & + & - & + & - & + \\ + & - & + & + & + & - \\ - & + & + & + & - & - \end{bmatrix} \in \mathbf{\Phi}_{16\times 6}\{-1,0,1\}$ is a chemical balance weighing design with efficiency $\mathrm{D_{eff}}(\mathbf{X}) = 0.9572$ for $\rho = 0.1$ and $\mathrm{D_{eff}}(\mathbf{X}) = 0.9459$ for $\rho = 0.2$.

Example 3.4: $p + 3 \equiv 0(mod4)$

In order to form $\mathbf{X} \in \mathbf{\Phi}_{18\times 7}\{-1,0,1\}$, we consider the matrix $\mathbf{X}_4 \in \mathbf{\Phi}_{14\times 7}\{-1,0,1\}$ of the D-optimal chemical balance weighing design

$$\mathbf{X}_4' = \begin{bmatrix} 0 & + & + & + & 0 & - & 0 & + & 0 & - & - & - & 0 & 0 \\ 0 & 0 & + & - & + & 0 & + & - & + & - & 0 & 0 & - & 0 \\ - & 0 & 0 & + & + & + & 0 & 0 & 0 & + & 0 & - & - & - \\ 0 & + & 0 & 0 & - & + & + & 0 & - & - & + & 0 & 0 & - \\ + & 0 & + & 0 & 0 & + & - & 0 & - & 0 & - & + & - & 0 \\ + & - & 0 & + & 0 & 0 & + & - & 0 & 0 & - & 0 & + & - \\ + & + & - & 0 & + & 0 & 0 & - & - & 0 & 0 & - & 0 & + \end{bmatrix}.$$

Hence $\mathbf{X} = \begin{bmatrix} & & & \mathbf{X}_4 & & & \\ + & + & + & - & + & + & + \\ + & + & - & + & + & - & - \\ + & - & + & + & - & - & + \\ - & + & + & + & - & + & - \end{bmatrix} \in \mathbf{\Phi}_{18\times 7}\{-1,0,1\}$ is a chemical balance weighing design with efficiency $\mathrm{D_{eff}}(\mathbf{X}) = 0.8666$ for $\rho = 0.1$ and $\mathrm{D_{eff}}(\mathbf{X}) = 0.8644$ for $\rho = 0.2$.

4. Discussion

In this paper, we have established D-optimal designs within novel categories of chemical balance weighing designs. Since it is impractical to determine the optimal design within every combination of objects and measurements, the exploration of new construction methods remains pertinent and warrants further investigation. Notable among the numerous instances of utilization of such designs are the studies by Harwit and Sloane (1979) and by Katulska (1984). Graczyk (2013) demonstrated the use of weighing design matrices as experimental plans in research concerning plant protection. Potential future applications of these designs include experimental plans founded on factor designs.

REFERENCES

Bulutoglu D.A., Ryan K.J. (2009): D-optimal and near D-optimal 2_k fractional factorial designs of resolution V, Journal of Statistical Planning and Inference 139: 16–22.

Ceranka B., Graczyk M. (2014): Regular D-optimal spring balance weighing designs: construction, Acta Universitatis Lodziensis, Folia Oeconomica 302: 111-125.

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., Ceranka B. (2023a): D-optimal chemical balance weighing designs with positively correlated errors: part I, Biometrical Letters 60(1): 53-63.

Graczyk M., Ceranka B. (2023b): D-optimal chemical balance weighing designs with positively correlated errors: part II, Biometrical Letters 60(2): 177-186.

Graczyk M. (2013): Some applications on weighing designs, Biometrical Letters 50: 15-26.

Harville D.A. (1997): Matrix Algebra from Statistician's Perspective. Springer-Verlag, New York Inc.

Harwit M., Sloane N.J.A. (1979): Hadamard transform optics. New York, Academic Press.

Katulska K. (1984): Zastosowanie teorii układów wagowych do badania upraw paszowych i w geodezji, Czternaste Colloquium Metodologiczne z Agro-Biometrii, PAN: 195-208.

Sathe Y.S., Shenoy R.G. (1990): Construction method for some A- and D-optimal weighing designs when n=3(mod4), Journal of Statistical Planning and Inference 24: 369-375.

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.