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# ON SOME D-OPTIMAL CHEMICAL BALANCE WEIGHING DESIGN WITH $n \equiv 2 \pmod{4}$

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### Summary

In this paper, we consider the chemical balance weighing designs for estimation of individual unknown weights of three objects using D-optimality criterion. We assume that the error components create a first-order autoregressive process AR(1). Then, the covariance matrix of random errors has known form, which does not have to be identity matrix and depends on known parameter  $\rho$ . In this paper, we prove D-optimality of some design from Bora-Senta and Moyssiadis (1999), if  $n \equiv 2 \pmod{4}$ , in the whole class of designs for three objects and some  $\rho \leq 0$ . Under these assumptions, we present the necessary and sufficient conditions such that the weighing design for three objects is D-optimal. These conditions can be used to construct D-optimal designs.

**Keywords and phrases:** D-optimal chemical balance weighing design, first-order autoregressive process AR(1)

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## 1. Introduction

In the paper we consider the chemical balance, where each object can be placed on one of two pans (left and right). A reading represents the total weight of the objects on the pans. We would like to choose a chemical balance weighing design that is optimal with respect to D-optimality criterion, which we define below.

At the beginning, we introduce a model for chemical balance weighing design for three objects. We estimate the true unknown weights  $\omega_1, \omega_2, \omega_3$  of three objects employing *n* measuring operations using a chemical balance. Let  $y_1, y_2, ..., y_n$  denote the observations in these *n* operations, respectively. We assume that the observations follow the linear model  $\mathbf{y} = \mathbf{X}\boldsymbol{\omega} + \boldsymbol{\varepsilon}$ , where  $\mathbf{y} = [y_1, y_2, ..., y_n]'$  is an  $n \times 1$  vector of observations,  $\boldsymbol{\omega} = [\omega_1, \omega_2, \omega_3]'$  is the vector of unknown weights of objects, the  $n \times 3$  matrix  $\mathbf{X} = [x_{ij}]$  is called the design matrix, the vector  $\boldsymbol{\varepsilon} = [\varepsilon_1, \varepsilon_2, ..., \varepsilon_n]'$  is the vector of error components. In the chemical balance weighing design, we suppose that  $x_{ij} = -1$  ( $x_{ij} = 1$ ) if the *j*th object is placed on the left (right) pan during the *i*th weighing operation. We consider the case when the random errors form an AR(1) process which implies that  $E(\boldsymbol{\varepsilon}) = [0, 0, ..., 0]'$  is an  $n \times 1$  nil vector and  $Var(\boldsymbol{\varepsilon}) = 1/(1 - \rho^2)\mathbf{S}$ , where  $\mathbf{S} = (\rho^{|r-d|})_{r,d=1}^n$  and  $-1 < \rho < 1$ . We identify the design with its matrix  $\mathbf{X}$ .

The D-optimal chemical balance weighing design maximizes the determinant of the information matrix  $\mathbf{X}'\mathbf{S}^{-1}\mathbf{X}$ . More precisely, the design  $\widetilde{\mathbf{X}}$  is D-optimal in the class of the designs  $C \subseteq M_{n\times 3}(\pm 1)$ , where the set  $M_{n\times p}(\pm 1)$  consists of all matrices with n rows, p columns and elements 1 or -1, if det $(\widetilde{\mathbf{X}}'\mathbf{S}^{-1}\widetilde{\mathbf{X}}) = \max{\det(\mathbf{X}'\mathbf{S}^{-1}\mathbf{X}) : \mathbf{X} \in C}$ .

The case, when the matrix **S** is the identity matrix ( $\rho = 0$ ), is well known and the D-optimal designs are considered in many papers (see, e.g. Galil and Kiefer (1980), or Jacroux et al. (1983)). For  $\rho \neq 0$ , Bora-Senta and Moyssiadis (1999) gave some conjectures (based on several exhaustive searches) about Doptimal chemical balance weighing designs with matrices  $\mathbf{X} = [\mathbf{1}_n \mid \mathbf{x} \mid \mathbf{y}] \in M_{n \times 3}(\pm 1)$ , where  $\mathbf{1}_n$  is the vector of *n* ones. These conjectures were proved in Li and Yang (2005) and Yeh and Lo Huang (2005)  $n \equiv 0 \pmod{4}, \rho \in (-1,1)$ and  $n \equiv 2 \pmod{4}$ ,  $\rho > 0$ . For for some  $-1 < \rho \le 0$  and  $n \equiv 0 \pmod{4}$ , some construction of D-optimal design in the class of designs such that each column of the design matrix  $\mathbf{X}$  contains at least one 1 and one -1 were considered in Katulska and Smaga (2010) and Katulska and Smaga (accepted).

Some results about D-optimal designs in the classes of designs with matrices  $\mathbf{X} = [\mathbf{x} | \mathbf{y} | \mathbf{z}] \in M_{n \times 3}(\pm 1)$  and  $\mathbf{X} = [\mathbf{1}_n | \mathbf{x} | \mathbf{y} | \mathbf{z}] \in M_{n \times 4}(\pm 1)$  for some  $\rho \ge 0$  are given in Katulska and Smaga (2012) and Katulska and Smaga (2011), respectively.

### 2. D-optimal chemical balance weighing designs

In this section, we present the main results but first we give some definitions and supporting results.

For any vector  $\mathbf{x} = [x_1, x_2, ..., x_n] \in M_{n \times 1}(\pm 1)$ , we define the numbers

$$cs(\mathbf{x}) = \#\{i : x_i = -x_{i+1}, 1 \le i \le n-1\},\$$

 $fcs(\mathbf{x}) = \min\{i : x_i = -x_{i+1}, 1 \le i \le n-1\},\$ 

$$scs(\mathbf{x}) = \min\{i : i > fcs(\mathbf{x}), x_i = -x_{i+1}, 1 \le i \le n-1\}.$$

We obtain the following lemma directly from properties of determinants (see Horn and Johnson, 1985).

**Lemma 2.1.** If  $\mathbf{X} \in M_{n \times p}(\pm 1)$  and  $\mathbf{G}$  is the  $n \times n$  real matrix, then the determinant of the matrix  $\mathbf{X}'\mathbf{G}\mathbf{X}$  does not change if we interchange two columns of the matrix  $\mathbf{X}$  or we multiply any column of this matrix by -1.

Below, we remind well known inequality.

**Lemma 2.2.** (Fischer's inequality). If  $\mathbf{P} = \begin{bmatrix} \mathbf{B} & \mathbf{C} \\ \mathbf{C}^T & \mathbf{D} \end{bmatrix}$  is a positive definite matrix that is partitioned so that  $\mathbf{B}$  and  $\mathbf{D}$  are square and nonempty, then  $\det(\mathbf{P}) \le \det(\mathbf{B}) \det(\mathbf{D})$  and the equality holds if and only if  $\mathbf{C} = \mathbf{0}$ .

**Lemma 2.3.** Suppose that  $n \equiv 2 \pmod{4}$  and  $\lambda = 0, 1, 2, \dots, n-1$ . If  $\Delta = (n-2)(1-\rho)^2 + 2(1-\rho)$ ,  $\rho \neq 0$  and  $\mathbf{x} \in M_{n \times 1}(\pm 1)$ , then  $cs(\mathbf{x}) = \lambda$  if and only if  $\mathbf{x}' \mathbf{A} \mathbf{x} = \Delta + 4\lambda\rho$ , where

$$\mathbf{A} = \begin{bmatrix} 1 & -\rho & 0 & \cdots & 0 & 0 \\ -\rho & 1+\rho^2 & -\rho & \cdots & 0 & 0 \\ 0 & -\rho & 1+\rho^2 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & \cdots & 1+\rho^2 & -\rho \\ 0 & 0 & 0 & \cdots & -\rho & 1 \end{bmatrix}.$$
 (2.1)

Proof. The thesis follows from equality

**x**' **Ax** = (*n*−2)(1+ρ<sup>2</sup>)+2−2ρ(*x*<sub>1</sub>*x*<sub>2</sub> + *x*<sub>2</sub>*x*<sub>3</sub> +···+ *x*<sub>*n*−1</sub>*x*<sub>*n*</sub>). ■

The next lemma follows from proofs in Yeh and Lo Huang (2005) and some direct calculations.

# Lemma 2.4. Let

 $\mathbf{x} = [x_1, x_2, ..., x_n]', \mathbf{y} = [y_1, y_2, ..., y_n]' \in M_{n \times 1}(\pm 1), n \equiv 2 \pmod{4}$  and the matrix **A** is defined by (2.1). (a) If  $cs(\mathbf{x}) = cs(\mathbf{y}) = 1$ ,  $fcs(\mathbf{x}) > fcs(\mathbf{y})$ , then

(a) If  $cs(\mathbf{x}) = cs(\mathbf{y}) = 1$ ,  $fcs(\mathbf{x}) > fcs(\mathbf{y})$ , then

$$\mathbf{x}'\mathbf{A}\mathbf{y} = \begin{cases} (n-2fcs(\mathbf{x})+2fcs(\mathbf{y})-2)(1-\rho)^2+2(1-\rho) & \text{if } x_1 = y_1 \\ -((n-2fcs(\mathbf{x})+2fcs(\mathbf{y})-2)(1-\rho)^2+2(1-\rho)) & \text{if } x_1 \neq y_1 \end{cases}.$$

(b) If cs(x) = 0, cs(y) = 2, then

$$\mathbf{x}'\mathbf{A}\mathbf{y} = \begin{cases} (n+2fcs(\mathbf{y})-2scs(\mathbf{y})-2)(1-\rho)^2 + 2(1-\rho) & \text{if } x_1 = y_1 \\ -((n+2fcs(\mathbf{y})-2scs(\mathbf{y})-2)(1-\rho)^2 + 2(1-\rho)) & \text{if } x_1 \neq y_1 \end{cases}.$$

(c) If cs(x) = 0, cs(y) = 1, then

$$\mathbf{x}' \mathbf{A} \mathbf{y} = \begin{cases} (2 f c s(\mathbf{y}) - n)(1 - \rho)^2 & \text{if } x_1 = y_1 \\ -(2 f c s(\mathbf{y}) - n)(1 - \rho)^2 & \text{if } x_1 \neq y_1 \end{cases}.$$

(d) If  $cs(\mathbf{x}) = 1$ ,  $fcs(\mathbf{x}) = n/2$ ,  $cs(\mathbf{y}) = 2$ ,  $b = fcs(\mathbf{y})$ ,  $c = scs(\mathbf{y})$ , then

$$\mathbf{x}'\mathbf{A}\mathbf{y} = \begin{cases} 2(b+c-n)(1-\rho)^2 & \text{if } x_1 = y_1, b < n/2, c > n/2 \\ -2(b+c-n)(1-\rho)^2 & \text{if } x_1 \neq y_1, b < n/2, c > n/2 \\ (n-4)(1-\rho)^2 + 2(1+\rho^2) & \text{if } (x_1 \neq y_1, b = 1, c = n/2) \text{ or} \\ (x_1 = y_1, b = n/2, c = n-1) & \text{if } (x_1 = y_1, b = 1, c = n/2) \text{ or} \\ (x_1 \neq y_1, b = n/2, c = n-1) & \text{if } (x_1 \neq y_1, b = n/2, c = n-1) \end{cases}$$

Now, we formulate new theorems concerning D-optimal chemical balance weighing designs under the assumption that the random errors form a process AR(1). First, we prove that some design is D-optimal weighing design for three objects and some  $\rho \le 0$ .

**Theorem 2.5.** Let  $n \equiv 2 \pmod{4}$ ,  $n \neq 2$  and  $\rho \in (-1, -1/(n-2)] \cup \{0\}$  if n = 6, 10, ..., 22, and  $\rho \in (-4/(n-8), -1/(n-2)] \cup \{0\}$  if  $n \ge 26$ . Then the design with the matrix

$$\hat{\mathbf{X}} = \begin{bmatrix} 1 & 1 & 1 \\ \vdots & \vdots & \vdots \\ 1 & 1 & 1 \\ 1 & 1 & -1_2 \\ \vdots & \vdots & \vdots \\ 1 & 1 & -1_2 \\ \vdots & \vdots & \vdots \\ 1 & 1 & -1_2 \\ \vdots & \vdots & \vdots \\ 1 & -1 & -1 \\ \vdots & \vdots & \vdots \\ 1 & -1 & -1 \\ 1 & -1 & 1_3 \\ \vdots & \vdots & \vdots \\ 1 & -1 & 1 \end{bmatrix}$$
(2.2)

where elements with indices 1, 2 and 3 are in positions (n/2+1, 2), ((n-2)/4+2, 3), (3(n-2)/4+2, 3), respectively, is D-optimal chemical balance weighing design for three objects.

**Proof.** (Sketch) The inverse of the matrix **S** is equal to  $\mathbf{S}^{-1} = 1/(1-\rho^2)\mathbf{A}$ , where the matrix **A** is given by (2.1). The matrix **A** is positive definite. From definition of D-optimal design and the inverse of the matrix **S** we obtain the D-optimal design in the class of designs  $C \subseteq M_{n \times p}(\pm 1)$  maximizes the determinant of the matrix **X'AX** among all  $\mathbf{X} \in C$ .

From Lemmas 2.3 and 2.4 for the matrix  $\hat{\mathbf{X}}$  of the form (2.2), we have

$$\det(\hat{\mathbf{X}}'\mathbf{A}\hat{\mathbf{X}}) = \det\begin{bmatrix}\Delta & 0 & 2(1-\rho)\\0 & \Delta+4\rho & 0\\2(1-\rho) & 0 & \Delta+8\rho\end{bmatrix} = (\Delta+4\rho)[\Delta(\Delta+8\rho)-4(1-\rho)^2].$$

When  $\rho = 0$ , then the matrix **A** is the identity matrix and

$$\det(\hat{\mathbf{X}}'\mathbf{A}\hat{\mathbf{X}}) = \det(\hat{\mathbf{X}}'\hat{\mathbf{X}}) = \det\begin{bmatrix}n & 0 & 2\\0 & n & 0\\2 & 0 & n\end{bmatrix} = n^3 - 4n.$$

Hence  $\hat{\mathbf{X}}$  is D-optimal from Jacroux et al. (1983). From now on, we assume that  $\rho \neq 0$ . It is easy to see that the matrix  $\mathbf{X'AX}$  is positive definite. By Lemma 2.1, we can suppose  $x_1 = y_1 = z_1 = 1$  and consider only the designs with matrices  $\mathbf{X} = [\mathbf{x} | \mathbf{y} | \mathbf{z}] \in C_1 \cup C_2 \cup C_3$ , where

$$C_{1} = \{ [\boldsymbol{\alpha} \mid \boldsymbol{\beta} \mid \boldsymbol{\gamma}] \in M_{n \times 3}(\pm 1) : cs(\boldsymbol{\alpha}) \ge 1, cs(\boldsymbol{\beta}) \ge 1, cs(\boldsymbol{\gamma}) \ge 2 \},$$

$$C_{2} = \{ [\boldsymbol{\alpha} \mid \boldsymbol{\beta} \mid \boldsymbol{\gamma}] \in M_{n \times 3}(\pm 1) : cs(\boldsymbol{\alpha}) = 0, cs(\boldsymbol{\beta}) \ge 1, cs(\boldsymbol{\gamma}) \ge 1 \},$$

$$C_{3} = \{ [\boldsymbol{\alpha} \mid \boldsymbol{\beta} \mid \boldsymbol{\gamma}] \in M_{n \times 3}(\pm 1) : cs(\boldsymbol{\alpha}) = cs(\boldsymbol{\beta}) = cs(\boldsymbol{\gamma}) = 1 \}.$$

We show that  $\det(\hat{\mathbf{X}}'\mathbf{A}\hat{\mathbf{X}}) \ge \det(\mathbf{X}'\mathbf{A}\mathbf{X})$  for all  $\mathbf{X} \in C_i$ , i = 1, 2, 3. For example, we present the proof if  $\mathbf{X} = [\mathbf{x} | \mathbf{y} | \mathbf{z}] \in C_1$ . Then from Hadamard's inequality, the determinant of the matrix  $\mathbf{X}'\mathbf{A}\mathbf{X}$  is less or equal to the product of the diagonal elements of this matrix, ie  $\det(\mathbf{X}'\mathbf{A}\mathbf{X}) \le (\mathbf{x}'\mathbf{A}\mathbf{x})(\mathbf{y}'\mathbf{A}\mathbf{y})(\mathbf{z}'\mathbf{A}\mathbf{z})$ . From Lemma 2.3, we obtain the inequalities  $\mathbf{x}'\mathbf{A}\mathbf{x} \le \Delta + 4\rho$ ,  $\mathbf{y}'\mathbf{A}\mathbf{y} \le \Delta + 4\rho$ ,  $\mathbf{z}'\mathbf{A}\mathbf{z} \le \Delta + 8\rho$ . Therefore we conclude  $\det(\mathbf{X}'\mathbf{A}\mathbf{X}) \le (\Delta + 4\rho)^2 (\Delta + 8\rho)$  and  $\det(\hat{\mathbf{X}}'\mathbf{A}\hat{\mathbf{X}}) - \det(\mathbf{X}'\mathbf{A}\mathbf{X}) \ge \det(\hat{\mathbf{X}}'\mathbf{A}\hat{\mathbf{X}}) - (\Delta + 4\rho)^2 (\Delta + 8\rho)$ 

= 4(∆+4 $\rho$ )[-(n-2) $\rho^3$  + (2n-11) $\rho^2$  - (n-2) $\rho$ -1] > 0, which completes the proof. ■

From the proof of Theorem 2.5, it follows that the design  $\hat{\mathbf{X}}$  given by (2.2) is D-optimal in some large subclass of the class  $M_{n\times 3}(\pm 1)$  for all  $\rho \in (-1, -1/(n-2)] \cup \{0\}$ , what we describe in the following corollary.

**Corollary 2.6.** If  $\rho \in (-1, -1/(n-2)] \cup \{0\}$  and  $n \equiv 2 \pmod{4}$ ,  $n \neq 2$ , then the design  $\hat{\mathbf{X}}$  given by (2.2) is D-optimal in the class  $\{[\boldsymbol{\alpha} \mid \boldsymbol{\beta} \mid \boldsymbol{\gamma}] \in M_{n\times 3}(\pm 1) : cs(\boldsymbol{\alpha}) \ge 0, cs(\boldsymbol{\beta}) \ge 1, cs(\boldsymbol{\gamma}) \ge 2 \text{ or } cs(\boldsymbol{\alpha}) = cs(\boldsymbol{\beta}) = cs(\boldsymbol{\gamma}) = 1\}.$ 

Now, we prove some necessary and sufficient conditions under which the design for the three objects is the D-optimal.

**Theorem 2.7.** If *n* and  $\rho$  are the same as in Theorem 2.5,  $\mathbf{X}^* = [\mathbf{x}^* | \mathbf{y}^* | \mathbf{z}^*] \in M_{n \times 3}(\pm 1)$ , then the design  $\mathbf{X}^*$  is D-optimal in the class of designs for three objects if and only if

$$\mathbf{X}^*' \mathbf{A} \mathbf{X}^* = \begin{bmatrix} \Delta & 0 & \pm 2(1-\rho) \\ 0 & \Delta + 4\rho & 0 \\ \pm 2(1-\rho) & 0 & \Delta + 8\rho \end{bmatrix}$$
(2.3)

exact to permuting columns of the matrix  $\mathbf{X}^*$ .

**Proof.** We present the proof if  $\rho \neq 0$ . First, we prove the sufficient condition. If the design  $\mathbf{X}^*$  satisfies the equality (2.3), then by Theorem 2.5 we obtain  $\det(\mathbf{X}^* \cdot \mathbf{A}\mathbf{X}^*) = \det(\hat{\mathbf{X}} \cdot \mathbf{A}\hat{\mathbf{X}})$ , so the design  $\mathbf{X}^*$  is D-optimal in  $M_{n\times 3}(\pm 1)$ . Now, we present the necessary condition. Assume that  $\mathbf{X}^*$  is the D-optimal design for three objects. So by Theorem 2.5, we conclude that  $\det(\mathbf{X}^* \cdot \mathbf{A}\mathbf{X}^*) = \det(\hat{\mathbf{X}} \cdot \mathbf{A}\hat{\mathbf{X}}) = (\Delta + 4\rho)[\Delta(\Delta + 8\rho) - 4(1-\rho)^2]$ . From the proof of Theorem 2.5, we obtain  $\det(\mathbf{X}^* \cdot \mathbf{A}\mathbf{X}^*) > \det(\mathbf{X} \cdot \mathbf{A}\mathbf{X})$  for all designs  $\mathbf{X} \in M_{n\times 3}(\pm 1) \setminus B$ , where

$$B = \left\{ [\boldsymbol{\alpha} \mid \boldsymbol{\beta} \mid \boldsymbol{\gamma}] : cs(\boldsymbol{\alpha}) = 0, cs(\boldsymbol{\beta}) = 1, cs(\boldsymbol{\gamma}) = 2, fsc(\boldsymbol{\beta}) = \frac{n}{2}, scs(\boldsymbol{\gamma}) - fcs(\boldsymbol{\gamma}) \neq \frac{n}{2} \right\}.$$

If  $\mathbf{X}^* \in B$ , then from Lemma 2.3 it follows that  $\mathbf{x}^* \mathbf{A} \mathbf{x}^* = \Delta, \mathbf{y}^* \mathbf{A} \mathbf{y}^* = \Delta + 4\rho$  and  $\mathbf{z}^* \mathbf{A} \mathbf{z}^* = \Delta + 8\rho$ . By Lemma 2.1:  $\det(\mathbf{X}^* \mathbf{A} \mathbf{X}^*) = \det([\mathbf{x}^* | \mathbf{z}^* | \mathbf{y}^*] \mathbf{A} [\mathbf{x}^* | \mathbf{z}^* | \mathbf{y}^*]).$  From Fischer's inequality, we obtain the following inequality

$$\det(\mathbf{X}^* \mathbf{A}\mathbf{X}^*) \le (\Delta + 4\rho) [\Delta(\Delta + 8\rho) - (\mathbf{x}^* \mathbf{A}\mathbf{z}^*)^2].$$
(2.4)

The equality in (2.4) holds if and only if  $\mathbf{x}^* \mathbf{A} \mathbf{y}^* = \mathbf{y}^* \mathbf{A} \mathbf{z}^* = 0$ . Moreover, from the fact that  $scs(\mathbf{z}^*) - fcs(\mathbf{z}^*) \neq n/2$  and Lemma 2.4 (b), it follows that  $(\mathbf{x}^* \mathbf{A} \mathbf{z}^*)^2 \geq 4(1-\rho)^2$  and the equality holds if and only if  $\mathbf{x}^* \mathbf{A} \mathbf{z}^* = \pm 2(1-\rho)$ . Therefore, we obtain the following inequality

$$\det(\mathbf{X}^* \mathbf{A}\mathbf{X}^*) \le (\Delta + 4\rho)[\Delta(\Delta + 8\rho) - 4(1-\rho)^2] = \det(\hat{\mathbf{X}}^* \mathbf{A}\hat{\mathbf{X}}). \quad (2.5)$$

But as we noted at the beginning of the proof in the inequality (2.5) there must be equality. So  $\mathbf{x}^* \cdot \mathbf{A} \mathbf{y}^* = \mathbf{y}^* \cdot \mathbf{A} \mathbf{z}^* = 0$ ,  $\mathbf{x}^* \cdot \mathbf{A} \mathbf{z}^* = \pm 2(1-\rho)$  and the matrix  $\mathbf{X}^* \cdot \mathbf{A} \mathbf{X}^*$  has the form (2.3).

**Theorem 2.8.** Let  $n \equiv 2 \pmod{4}, n \neq 2$  and  $\rho \in (-1, -1/(n-2)]$  if  $n = 6, 10, \dots, 22$ , and  $\rho \in (-4/(n-8), -1/(n-2)]$  if  $n \ge 26$ . Then the design  $\mathbf{X}^* = [\mathbf{x}^* | \mathbf{y}^* | \mathbf{z}^*] \in M_{n \times 3}(\pm 1)$  is D-optimal in the class of designs for three objects if and only if  $cs(\mathbf{x}^*) = 0$ ,  $cs(\mathbf{y}^*) = 1$ ,  $cs(\mathbf{z}^*) = 2$  and  $fcs(\mathbf{y}^*) = n/2$ ,  $fcs(\mathbf{z}^*) = (n-2)/4 + 1$ ,  $scs(\mathbf{z}^*) = 3(n-2)/4 + 1$  exact to permuting columns of the matrix  $\mathbf{X}^*$ .

**Proof.** The sufficient condition is easy to see, because from Lemmas 2.3 and 2.4, we conclude that the matrix  $\mathbf{X}^* \cdot \mathbf{A} \mathbf{X}^*$  has the form (2.3) and hence by Theorem 2.7, the design  $\mathbf{X}^*$  is D-optimal design for three objects. Proof of necessary condition is as follows. Let  $\mathbf{X}^*$  be the D-optimal design for three objects. So the matrix  $\mathbf{X}^* \cdot \mathbf{A} \mathbf{X}^*$  has the form (2.3) by Theorem 2.7.

 $\mathbf{x}^* \mathbf{A} \mathbf{x}^* = \Delta \Leftrightarrow cs(\mathbf{x}^*) = 0,$ From Lemma 2.3, it follows that  $\mathbf{v}^* \mathbf{A} \mathbf{v}^* = \Delta + 4\rho \Leftrightarrow cs(\mathbf{v}^*) = 1$  $\mathbf{z}^* \mathbf{A} \mathbf{z}^* = \Delta + 8\rho \Leftrightarrow cs(\mathbf{z}^*) = 2.$ and Moreover, from Lemma 2.4 (c), we have  $\mathbf{x}^* \cdot \mathbf{A} \mathbf{y}^* = \pm (2 f c s(\mathbf{y}^*) - n)(1 - \rho)^2 = 0$ , so  $f c s(\mathbf{y}^*) = n/2$ . From the equality  $x^* A z^* = \pm 2(1-\rho)$ Lemma 2.4 and (b), we obtain  $scs(\mathbf{z}^*) - fcs(\mathbf{z}^*) = n/2 - 1$ . Hence and from the fact that  $\mathbf{y}^* \mathbf{A}\mathbf{z}^* = 0$  we have (by Lemma 2.4 (d))  $fcs(\mathbf{z}^*) < n/2$ ,  $scs(\mathbf{z}^*) > n/2$  and hence  $\mathbf{y}^* \cdot \mathbf{A}\mathbf{z}^* = \pm 2(fcs(\mathbf{z}^*) + scs(\mathbf{z}^*) - n)(1 - \rho)^2 = 0$  which implies  $fcs(\mathbf{z}^*) + scs(\mathbf{z}^*) = n$ .

Therefore  $fcs(\mathbf{z}^*) = (n-2)/4+1$ ,  $scs(\mathbf{z}^*) = 3(n-2)/4+1$ . So the thesis is proved.

Using Theorems 2.7 and 2.8, D-optimal chemical balance weighing designs (other than  $\hat{\mathbf{X}}$ ) for the three objects under the assumption that the random errors form a process AR(1) can be constructed.

#### References

Bora-Senta E., Moyssiadis C. (1999). An algorithm for finding exact D- and A-optimal designs with n observations and k two-level factors in the presence of autocorrelated errors. *J. Combin. Math. Combin. Comput.* 30, 149-170.

Galil Z., Kiefer J. (1980). D-optimum weighing designs. Ann. Statist. 8, 1293-1306.

- Horn R.A., Johnson C.R. (1985). Matrix Analysis. Cambridge University Press, Cambridge.
- Jacroux M., Wong C.S., Masaro J.C. (1983). On the optimality of chemical balance weighing design. *Journal of Statistical Planning and Inference* 8, 231-240.
- Katulska K., Smaga Ł. (2010). On some construction of D-optimal chemical balance weighing designs. Coll. Biom. 40, 155-164.
- Katulska K., Smaga Ł. (2011). D-optimal biased chemical balance weighing designs. Coll. Biom. 41, 143-153.
- Katulska K., Smaga Ł. (2012). D-optimal chemical balance weighing designs with autoregressive errors. *Metrika* DOI: 10.1007/s00184-012-0394-8.
- Katulska K., Smaga Ł., D-optimal chemical balance weighing designs with  $n \equiv 0 \pmod{4}$  and 3 objects. *Communications in Statistics Theory and Methods* (accepted).
- Li C.H., Yang S.Y. (2005). On a conjecture in D-optimal designs with  $n \equiv 0 \pmod{4}$ . *Linear Algebra and its Applications* 400, 279-290.
- Yeh H.G., Lo Huang M.N. (2005). On exact D-optimal designs with 2 two-level factors and n autocorrelated observations. *Metrika* 61, 261-275.