SEPARATION PROCESS OPTIMIZATION OF THE PASTE FROM THE ALUMINUM CATHODE, IN THE PROCESS OF RECOVERY OF COBALT FROM USED BATTERIES

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The paper presents the results of the mathematical modeling of the process of recovering the paste with Cobalt content from the aluminum foil-the cathode of the used Li-Ion batteries. Using an active research program - a second order orthogonal program (PO2), the optimal conditions for the process have been established which will lead to a maximum recovery of the active paste.

Keywords: mathematical modeling, factorial program, recycle Li-Ion batteries, $LiCoO_2$ compound.

1. Introduction

Li-ion batteries being rechargeable batteries, have been designed to withstand numerous charging / discharging cycles. Thus, after 500 to 10,000 cycles of loading / unloading (depending on the application for which they were designed), they will stop working. In 2018, around 97,000 tons of recycled batteries in China and 18,000 tons in South Korea were recycled, countries that manufacture a large amount of such batteries [1].

Currently, most used batteries come from electronic products, and are mainly LCO type with cobalt content of about 17%. In 2018 over 14,000 tons of cobalt was recovered by recycling (about 10 of the metal extracted from primary sources / ore). Although in the future, as cobalt is replaced in these batteries with other metals (Ni, Ti) and its share will decrease, today the recovery of Cobalt from spent Li-ion batteries is very topical (Fig. 1). Considering also that most of

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the used batteries do not reach recycling (about 50%), the batteries being stored or hoarded [1].

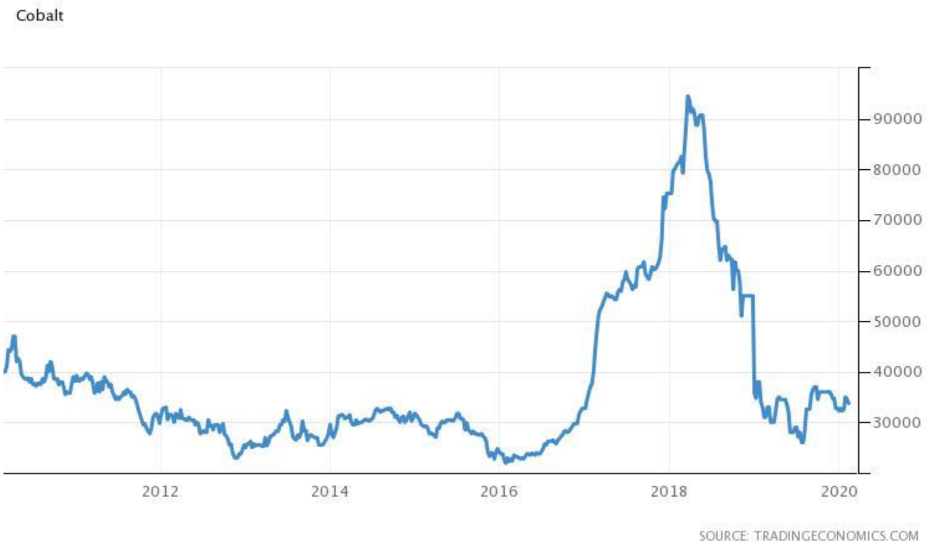

Fig. 1. Prices of Cobalt in the last 10 years [1]

From the point of view of the recycling technologies, the greatest difficulties encountered appear at the disassembly / extrication and the separation by components. Today many technologies of recycling of Li-ion batteries (pyro-metallurgical, hydro-metallurgical or combined technologies) are known and applied but the research in the field is far from finished [2, 3, 4, and 5].

In the Hydrometallurgical Laboratory of the Department of Engineering and Management of Metallic Materials of the Faculty of Materials Science and Engineering, from the University Politehnica of Bucharest, attempts were made to separate/recover the active paste with Cobalt content by means of the ultrasound treatment applied in acidic environment using non-polluting organic acids [6] - citric acid [7, 8, 9], acetic acid [10] lactic acid [11, 12]. The results obtained were encouraging; the next step was to optimize these technologies. For this, the problem proposed to be solved must be well defined and the influencing factors must be significant. These influencing factors will be assigned two levels of variation (upper and lower) equal far from the base level, around which the mathematical modeling will be performed [13, 14, 15]. The complete factorial experiences with k factors of influence, with two levels of variation are symbolized by $EFC\ 2^k$ [16].

2. Materials and methods

The ultrasonic cleaning machine (Emmi12-HC) used has the following technical specifications: housing – stainless steel, cleaning frequency = 45 kHz;

cleaning time = 1- 60 min; volume = 1.2 l; heating temperature = 20 - 80 ºC; bath dimension 200x100x65 mm; ultrasonic power= 50/75/100W [7, 8].

The operation of separating the active paste with Co content from the aluminum foil constituting the cathode of the battery was performed in acidic environment (lactic acid) at a constant pH of 1.7M. The ultrasound duration was kept constant for 4 minutes. The influence of the temperature and the power of the ultrasonic bath on the separation efficiency of the paste from the cathode foil was studied [11, 12]. The starting temperature T = 75ºC and the power output of 65W were chosen as starting point. The basic levels, the ranges of variation, the upper and lower levels for the two factors taken into account are presented in Table 1.

Table 1

The basic levels and the ranges of variation of the factors

Factor	X_1 (power source, W)	X_2 (temperature, ºC)
Code	X_1	X_2
The basic level, z_{i0}	65	75
Variation range, Δz_i	15	7
Upper level, (+1)	80	82
Lower level, (-1)	50	68

It starts from the linear determination of the form:

$$\tilde{y}_1 = b_o + b_1 x_1 + b_2 x_2$$

Mathematical model that will be calculated using the factorial matrix 2^2 presented in Table 2, which includes the experimental results.

Table 2

The matrix of the programmed experiment together with the obtained experimental results

Exp. no.	X_0	X_1	X_2	$X_1 X_2$	$x_1' = x_1^2 - \frac{2}{3}$	$x_2' = x_2^2 - \frac{2}{3}$	y
1	+1	-1	-1	+1	+1/3	+1/3	33.59
2	+1	-1	+1	-1	+1/3	+1/3	60.23
3	+1	+1	+1	+1	+1/3	+1/3	91.85
4	+1	+1	-1	-1	+1/3	+1/3	48.56
5	+1	+1	0	0	+1/3	-2/3	68.24
6	+1	-1	0	0	+1/3	-2/3	33.47
7	+1	0	+1	0	-2/3	+1/3	76.33
8	+1	0	-1	0	-2/3	+1/3	42.91
9	+1	0	0	0	-2/3	-2/3	54.08

a) Calculation of the linear model coefficients

$$b_o = \frac{1}{N}\sum_{u=1}^{N=4} y_u \quad b_i = \frac{\sum_{u=1}^{N} x_{iu} y_u}{\sum_{u=1}^{N} (x_{iu})^2} \quad b_{ij} = \frac{\sum_{u=1}^{N} (x_{iu} x_{ju}) y_u}{\sum_{u=1}^{N} (x_{iu} x_{ju})^2}$$

$b_0 = 58.58$ $\quad$ $b_1 = 11.65$ $\quad$ $b_2 = 17.48$

b) Statistical verification of values

The verification is done on condition that the absolute value of coefficient b_i is higher than the confidence interval of this coefficient Δb_i:

$|b_i| \geq |\Delta b_i|$

$|\Delta b_i| = t_{\alpha;N} \cdot |s_{b_i}|$, where:

$t_{\alpha;N}$ - the Student criterion for the significance threshold α and the number N of degrees of freedom;

s_{b_i} - the mean squared deviation wherewith the coefficient b_i is calculated

$s_{b_i} = \pm\sqrt{s_{b_i}^2}$;

$s_{b_i}^2$ - the dispersion wherewith the coefficient b_i is calculated.

c) Calculation of dispersion of reproducibility (s_o^2)

Three experiments will be performed at the basic level (x1 = x2 = 0); the results are presented in Table 3.

Table 3

Calculation of dispersion of the reproducibility of the experiments (the experimental error)

Exp. no.	y_u	$\bar{y}$	$(y_u - \bar{y})$	$(y_u - \bar{y})^2$	ν_2 = n-1
1	54.08	53.82	1.26	1.58	2
2	55.27		1.45	2.10	
3	52.13		-1.69	2.85	
$s_o^2 = 3.26$				6.53	

Statistical verification of the linear model coefficients

Mean squared deviation in case of coefficients:

$s_{b_0}^2 = s_{b_1}^2 = s_{b_2}^2 = \frac{s_o^2}{N} = \frac{3.26}{4} = 0.815$; $s_{b_o} = 0.9$

$t_{\alpha;N} = t_{0.05;4} = 2.786$

Confidence intervals: $|\boldsymbol{\Delta b_i}| = t_{\alpha;N} \cdot |s_{b_i}| = t_{0.05;4} \cdot |s_{b_o}| = 2.786 \cdot 0.9 = \mathbf{2.49}$

It is found that all coefficients are statistically different from zero, the condition being fulfilled

$|b_i| \geq |\Delta b_i|$

It turns out that the linear model, without taking into account the interaction will be:

$\tilde{y} = 84.36 + 11.64x_1 + 17.48x_2$

that is, the regression equation, without considering the interaction.

The calculation of the dispersion produced by the linear regression equation of the $s^2_{concordance}$ is found in Table 4.

Table 4

Calculation of dispersion s^2_{conc}

Exp. no.	$y_{u_{exp}}$	$\widetilde{y_u}$	$\widetilde{y_u} - y_{u_{exp}}$	$(\widetilde{y_u} - y_{u_{exp}})^2$	ν_1 = N-k'
1	33.59	29.43	- 4.16	17.30	4-3=1 4 = nr.de experiente 3 = coeficientii ec de regresie
2	60.23	64.39	4.16	17.30	
3	91.85	87.67	4.18	17.47	
4	48.56	52.71	4.15	17.22	
$s^2_{conc}=\frac{\sum_{u=1}^{N}(\widetilde{y_u}-y_{u_{exp}})^2}{N-k'} = \frac{69.29}{3-2} = 35.25$				69.29	

$\widetilde{y_u}$ respective $y_{u_{exp}}$ - are the values calculated using the regression equation, respectively obtained experimentally under the conditions of the experience u;

N – k' - the number of degrees of freedom, representing the difference between the number N of experiences and the number *k'* of coefficients in the regression equation (including b_0) [16].

The calculated model is consistent when:

$F_c < F_{0.05;\, \nu;\, \nu}$

$$F_c = \frac{s^2_{conc}}{s^2_o} = \frac{35.25}{3.38} = 33.57 > F_{0.05\,;\,1\,;\,2} = 18.51$$

It result that the linear model does not agree with the analyzed process; we proceed to the determination of a nonlinear model, completing the matrix with the experiments 5-9 required for the central PCCO orthogonal compositional programming of the second order ($\alpha = 1;\ n_0 = 1$).

d) Nonlinear model

The nonlinear equation is a polynomial of the second degree of form [15]:

$$\tilde{y} = b_o + \sum_{i=1}^{k} b_i x_i + \sum_{i=1; j=1 i\neq j}^{k} b_{ij} x_i x_j + \sum_{i=1}^{k} b_{ii} x_i^2$$

In these conditions, the total number of experiences is:

$N = 2^k + 2k + n_0 = 4 + 4 + 1 = 9$

Calculation of non-linear model coefficients:

$$\tilde{y} = \sum_{i=1}^{n} b_i x_i + \sum_{i=1}^{n} b_{ij} x_i x_j + \sum_{i=1}^{n} b_{ii}\, x_i^2 \qquad b'_0 = \sum_{u=1}^{N} \frac{y_u}{N}$$

$$b_i = \frac{\sum_{u=1}^{N} x_{iu} y_u}{\sum_{u=1}^{N} x_{iu}^2}\ ; b_{ij} = \frac{\sum_{u=1}^{N}(x_{iu} x_{ju}) y_u}{\sum_{u=1}^{N}(x_{iu} x_{ju})^2}; b_{ii} = \frac{\sum_{u=1}^{N} x'_{iu} y_u}{\sum_{u=1}^{N}(x'_{iu})^2}\, x'_{iu} = x_{iu}^2 - \overline{x}_i^2$$

$b_0 = 56.58$ $b_{12} = 4.16$
$b_1 = 13.56$ $b_{11} = - 1.78$
$b_2 = 17.22$ $b_{22} = 6.98$

Calculation of the dispersion of the coefficients b_i, b_{ij}, b_{ii}

$$s_{b_1}^2 = s_{b_2}^2 = \frac{s_0^2}{\sum_{u=1}^{N} x_{i_u}^2} = 0.54 \qquad s_{b_i} = \pm 0.73$$

$$s_{b_{11}}^2 = s_{b_{22}}^2 = \frac{s_0^2}{\sum_{u=1}^{N} x'^2_{i_u}} = 1.63 \qquad s_{b_{ii}} = \pm 1.27$$

$$s_{b_{ij}}^2 = s_{b_{12}}^2 = \frac{s_0^2}{\sum_{u=1}^{N} (x_{i_u} \cdot x_{j_u})^2} = 0.815 \qquad s_{b_{ij}} = \pm 0.9$$

$$s_{b_0'}^2 = \frac{s_0^2}{N} = \frac{3.26}{9} = 0.36$$

e) Statistical verification of non-linear model coefficients

The tabulated value of the Student Criterion $\boldsymbol{t_T = t_{0.05;\, \nu 0} = t_{0.05;\, 9} = 2.26}$

ν_0 – the number of degrees of freedom of dispersion of reproducibility;
$s_{b_i}^2$; $s_{b_{ii}}^2$; $s_{b_{ij}}^2$; $s_{b_o'}^2$ – dispersions of the regression coefficients;
t_{b_i}; $t_{b_{ij}}$; $t_{b_{ii}}$ – the calculated values of the Student criterion;
The condition of significance of the coefficients: t_{b_i} ; $t_{b_{ij}}$; $t_{ii} > t_T$

$$t_{b_i} = \frac{|b_i|}{s_{b_i}} ;\ t_{b_1} = \frac{|b_1|}{s_{b_1}} = \frac{|13.56|}{0.73} = 18.57 > t_T;$$
$$t_{b_2} = \frac{|b_2|}{s_{b_2}} = \frac{|17.22|}{0.73} = 23.58 > t_T;$$
$$t_{b_{ij}} = \frac{|b_{ij}|}{s_{b_{ij}}} = t_{b_{12}} = \frac{|b_{12}|}{s_{b_{12}}} = \frac{|4.16|}{0.9} = 4.62 > t_T;$$
$$t_{b\,ii} = t_{b\,11} = \frac{|b_{11}|}{s_{b_{11}}} = \frac{|-1.74|}{1.27} = \frac{1.74}{1.27} = 1.37 < t_T$$
$$t_{b\,ii} = t_{b\,22} = \frac{|b_{22}|}{s_{b_{22}}} = \frac{|3.12|}{0.72} = \frac{6.98}{1.27} = 5.49 > t_T$$

Statistical verification of coefficient b_0

$$s_{b_o}^2 = s_{b_o'}^2 + (\bar{x}_2^2)^2 s_{b_{22}}^2 = 0.9$$
$$s_{b_o} = \pm 0.94$$

$$t_{b_0} = \frac{b_0}{s_{b_0}} = \frac{56.58}{0.94} = 60.19 > t_T$$

The following non-linear model results from the statistical verification of the coefficients:

$$\tilde{y} = 51.98 + 13.56x_1 + 17.22x_2 + 4.16x_1x_2 + 6.98x_2^2$$

f) Verification of model concordance hypothesis

The dispersion caused by the regression equation is calculated.

Table 5

Calculation of dispersion of the nonlinear model

Crt. no.	$y_{u_{exp}}$	$\tilde{y_u}$	$\tilde{y_u} - y_{u_{exp}}$	$(\tilde{y_u} - y_{u_{exp}})^2$	**ν_1 =N-l**
1	33.59	32.34	1.25	1.56	ν_1 =9-5=4
2	60.23	58.46	1.77	3.13	
3	91.85	93.9	-2.05	4	
4	48.56	51.14	-2.58	6.65	
5	68.24	65.54	2.7	7.29	
6	33.47	38.42	-4.95	24.5	
7	76.33	76.18	0.15	0.02	
8	42.91	41.74	1.17	1.36	
9	54.08	51.98	2.10	4.41	
$s_{conc}^2 = 13.23$				52.92	

l - the number of terms in the regression equation, including the free term.

Fischer criterion calculated:

$$F_{calc} = \frac{s_{conc}^2}{s_o^2} = \frac{13.23}{3.26} = 4 < F_{0.05;4;2} < 19.25$$

The Fischer criterion for $\alpha = 0.05$ and $\nu_1 = 4$; $\nu_2 = 2$, is $F_{0.05;\,4;\,2} = 19.25$

It turns out that the calculated nonlinear model:

$$\tilde{y} = 51.98 + 13.56x_1 + 17.22x_2 + 4.16x_1x_2 + 6.98x_2^2$$

is appropriate, expressing with good approximation the actual process analyzed.

Following the modeling, a second degree equation was obtained:

$$\tilde{y} = \sum_{i=1}^{n} b_i x_i + \sum_{i=1}^{n} b_{ij} x_i x_j + \sum_{i=1}^{n} b_{ii} x_i^2$$

whose concordance has been statistically verified, the verification of which results that the mathematical model satisfactorily describes the optimal field. Next we determine the conditions for achieving the optimum and calculating its values:

$$\tilde{Y} - Y_s = B_{11}X_1^2 + B_{22}X_2^2 + \cdots B_{ii}X_k^2$$

where:

$\tilde{Y}$ - the value of the parameter to be optimized in the old coordinate system;
Y_s - the value of the parameter to be optimized in the new axis system;
$X_1, X_2, \dots X_k$ - the factor values in the new axis system;
$B_{11}, B_{22}, \dots B_{kk}$ - coefficients of the regression equation in standard format.

After the translation of the origin of the coordinate system the equation becomes:

$$\tilde{Y} = Y_s + \sum_{\substack{i=1 \\ j=1 \\ i \neq j}}^{n} b_{ij} x_i x_j + \sum_{i=1}^{n} b_{ii} x_i^2$$

$$\tilde{y} = 51.98 + 13.56x_1 + 17.22x_2 + 4.16x_1x_2 + 6.98x_2^2$$

Obtained following a modeling through the programmed experiment by the second order.

Differentiating this equation according to x_1 and x_2 and equaling the partial derivatives with zero, we obtain:

$x_{1c} = 6.8$ $\quad$ $x_{2c} = -3.26$

By replacing these values in the initial equation, we obtain the value of the parameter to be optimized in the new coordinate center $Y_s = 70$.

For the calculation of the canonical coefficients, the characteristic determinant is formed and is canceled:

$$\begin{vmatrix} (b_{11} - B) & \frac{1}{2} b_{12} \\ \frac{1}{2} b_{12} & (b_{22} - B) \end{vmatrix} = \begin{vmatrix} -B & \frac{1}{2} 4.16 \\ \frac{1}{2} 4.16 & (6.98 - B) \end{vmatrix} = 0$$

$B^2 - 6.98B - 4.3264 = 0$

$B_{11} = 7.55$ $\quad$ $B_{22} = -0.57$ $\quad$ $B_{11} > 0$ $\quad$ $B_{22} < 0$, the surface has a minimal.

The number of roots is equal to the number of factors; verifying the correctness of the calculations by means of the relation:

$$\sum_{i=1}^{n} B_{ii} = \sum_{i=1}^{n} b_{ii}$$

$7.552 + (-0.572) = 6.98$

The equation in canonical form will be:

$$\tilde{Y} - 70 = 7.55X_1^2 - 0,57X_2^2$$

Based on the signs of the canonical coefficients, the shape of the response surface is established:

- Elliptical paraboloid, if all the canonical coefficients have the same sign (the plus sign corresponds to a minimum in the center of the surface, and the minus sign corresponds to a maximum);
- Hyperbolic paraboloid, when the coefficients have different signs.

g) Determining the optimal regime - The ridge analysis method based on the Lagrange multipliers method

In order to choose the optimal regimes, the system of equations is formed:

$$\begin{cases}(b_{11}-\lambda)x_1+0{,}5b_{12}x_2+0{,}5b_1=0\\ 0{,}5b_{21}x_1+(b_{22}-\lambda)x_2+0{,}5b_2=0\end{cases}$$

In which λ represents the Lagrange multiplier, and x_1 and x_2 are the coordinates of the searched point.

Solving the system is possible when the values of λ are known, values that depend on the type of the problem. To specify the optimal conditions for the process, the Lagrange multiplier method is applied. In this sense, the Hoerl parameter is calculated with the relation:

$\lambda' = 2\left(a_{\substack{max\\min}} - b_{kk}\right)$;

in which λ' is the Hoerl parameter;

$a_{\substack{max\\min}}$ – the canonical coefficient of maximum or minimum value (depending on the type of problem);

In our case:

$a_{\substack{max\\min}} = -0.57$; $b_{kk} = 6.98$; $a_{min} = -0.57$

Next some values of λ are chosen in the considered interval, the system of equations is solved for each value and the obtained regimes are subjected to an experimental verification.

In case n = 2 the system has the solution:

$$x_1=\frac{0{,}25b_{12}b_2-0{,}5b_1(b_{22}-\lambda)}{(b_{11}-\lambda)(b_{22}-\lambda)-0{,}25b_{12}^2}\ ;\ x_2=\frac{0{,}25b_{12}b_1-0{,}5b_2(b_{11}-\lambda)}{(b_{11}-\lambda)(b_{22}-\lambda)-0{,}25b_{12}^2}$$

Thus:

$\lambda = 1$; $x_1 = -1.447$; $x_2 = -1.417$; $\tilde{y} = 51.98 + 13.56x_1 + 17.22x_2 + 4.16x_1x_2 + 6.98x_2^2 = 30\%$

$\lambda = 2$; $x_1 = 1.11$; $x_2 = -2.19$; $\tilde{y} = 51.98 + 13.56x_1 + 17.22x_2 + 4.16x_1x_2 + 6.98x_2^2 = 52.63\%$

$\lambda = 4$; $x_1 = 0.14$; $x_2 = -2.98$; $\tilde{y} = 51.98 + 13.56x_1 + 17.22x_2 + 4.16x_1x_2 + 6.98x_2^2 = 62.80\%$

$\lambda = 5$; $x_1 = -0.315$; $x_2 = -4$; $\tilde{y} = 51.98 + 13.56x_1 + 17.22x_2 + 4.16x_1x_2 + 6.98x_2^2 = 95.75\%$

$\lambda = 5.2$; $x_1 = -0.43$; $x_2 = -4.34$; $\tilde{y} = 51.98 + 13.56x_1 + 17.22x_2 + 4.16x_1x_2 + 6.98x_2^2 = 110\%$

Results:

For $\lambda = 5.051$; $x_1 = -0.3427$; $x_2 = -4.093$

$\tilde{y} = 51.98 + 13.56x_1 + 17.22x_2 + 4.16x_1x_2 + 6.98x_2^2 = 100\%$

h) Determining the calculation error of the optimization parameter ($\delta_{\tilde{y}}$)

In the present case, the case of a second order program, because the covariate matrix is not diagonal, the calculation relation of the dispersion ($\delta\tilde{y}$) is more complicated, because it has to take into account both the dispersion of the coefficients and the covariance:

$$\delta_{\tilde{y}}^2 = s_{b0}^2 + s_{b_i}^2 \sum_{i=1}^{n} x_i^2 + s_{b_{ii}}^2 \sum_{i=1}^{n} x_i^4 + s_{b_{ij}}^2 \sum_{j>i=1}^{n} x_i^2 x_j^2 + 2cov(b_0 b_{ii}) \sum_{i=1}^{n} x_i^2 + 2cov(b_0 b_{ij}) \sum_{j>i=1}^{n} x_i^2 x_j^2$$

in which:

$e = N_c + 2\alpha^2$, $f = N_c + 2\alpha^4$

$D = 2\alpha^4 H^{-1}[f + (n-1)N_c]$

$E = -2H^{-1}e\alpha^4$

$F = H^{-1}[Nf + (n-2)NN_c - (n-1)e^2]$

$G = H^{-1}(e^2 - NN_c)$

$H = 2\alpha^4[Nf + (n-1)NN_c - ne^2]$

G changes depending on the number of parallel determinations in the center of the program and on the value of parameter α (called "star arm").

$e^2 - NN_c = 0$

Since $N_c = 2^n$ and $N = N_c + 2n + N_0$, the equation becomes:

$\alpha^4 + 2^n\alpha^2 - 2^{n-1}(n + 1/2N_0) = 0$

$\alpha^4 + 2^2\alpha^2 - 2(2 + 1/2) = 0$

$\alpha^4 + 2^2\alpha^2 - 5 = 0$; $\alpha^4 = 1$; $\alpha^2 = 1$; $\alpha = 1$.

In these conditions:

N = number of determinations = 9

$N_c = 2^n = 4$

N_0 = number of determinations in center – 1

$e = N_c + 2\alpha^2 = 6$, $f = N_c + 2\alpha^4 = 6$

$H = 2\alpha^4[Nf + (n-1)NN_c - ne^2] = 2 \cdot 1[9 \cdot 6 + (2-1) \cdot 9 \cdot 4 \cdot (-2) \cdot 36] = 36$

$$D = 2\alpha^4 H^{-1}[f + (n-1)N_c] = \frac{20}{36} = 0.55$$

$$E = -2H^{-1}e\alpha^4 = -\frac{1}{3} = -0.33$$

$$F = H^{-1}[Nf + (n-2)NN_c - (n-1)e^2] = 0.5$$

$$G = H^{-1} = 0$$

$cov(b_0 b_{ii}) = Es_0^2 = -0.33 \cdot 3.26 = -1.07$

$cov(b_0 b_{ij}) = Gs_0^2 = 0$

$s_{b_o}^2 = 0.9$
$s_{b_i}^2 = 0.54$
$s_{b_{ii}}^2 = 1.63$
$s_{b_{ij}}^2 = 0.815$

$\boldsymbol{\delta_{\tilde{y}}^2}$ = 0.9 + 0.54·16 + 1.63·256 + 0.815·1.92 + 2(-1.07)·16 = 0.9 + 8.64 + 417.28 + 1.56 – 34.24 = 394.14

$\boldsymbol{\delta_{\tilde{y}}}$ = 19.85

It turns out that the lower limit of the efficiency has the value
100 – 19.85 = 80.15

In conclusion, the optimal conditions for carrying out the process are the following:

$\widehat{z_1} = z_1^0 + \widehat{x_1} \cdot \Delta z_1$ = 65 + (- 0.3427) · 15 = 65 - 5.14 = 59.86°C;
$\widehat{z_2} = z_2^0 + \widehat{x_2} \cdot \Delta z_2$ = 75 + (- 4.093) · 7 = 75 – 28.65 = 46.35 W;

3. Conclusions

The studies and rough experimental research have emphasized two parameters with significant influence over the electrolysis performances: temperature (z_1), [^{0}C], power of the bath (z_2), [W]. Other parameters, like cleaning time (6 minute), frequency (50 kHz), initial concentration of lactic acid in solution (1,7M), and additions of H_2O_2 (3%), were kept constant, considering having no significant influence. Also, the position of cathode foils in the basket of ultrasonic bath was maintained at 15 mm from the ultrasonic generator.

To determine the optimal conditions for the separation and recovery of the active paste, experiments were made according to an active program, an order two orthogonal program (PO2). The mathematical model obtained with this program was statistically analysed and, with Fisher criterion, the concordance between the model and the experimental data was ascertained. Since the mathematical model was found as adequate, in order to specify the optimal conditions for the process, the Lagrange multiplier method was applied.

The optimal values found are: $\widehat{z_1} = z_1^0 + \widehat{x_1} \cdot \Delta z_1 =$ **59.86**; $\widehat{z_2} = z_2^0 + \widehat{x_2} \cdot \Delta z_2 =$ **46.35, W,** and, taking into consideration the error of calculation of the process performance $\delta_{\tilde{y}}$ the lower limit of the efficiency has the value **80.15**.

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