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ULTRACENTRIFUGE SEPARATIVE POWER MODELING WITH MULTIVARIATE REGRESSION USING COVARIANCE MATRIX

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Abstract: In this work, the least-squares methodology with covariance matrix is applied to determine a data curve fitting in order to obtain a performance function for the separative power δU of an ultracentrifuge as a function of variables that are experimentally controlled. The experimental data refer to 460 experiments on the ultracentrifugation process for uranium isotope separation. The process control variables, which significantly influence the δU values, are chosen in order to give information on the ultracentrifuge behaviour when submitted to several levels of feed flow F and cut θ and product line pressure Pp. The response curves are made relating the separative power with the control variables F, θ and Pp, to compare the fitted model with the experimental data and finally to calculate their optimized values.

Keywords: ultracentrifuge; uranium hexafluoride; isotopic separation; covariance matrix; least-squares method.

Resumo: Neste trabalho, método dos quadrados mínimos com matriz de covariância é aplicada para determinar um formato de curva dos dados a fim obter uma função de desempenho para a energia de separação δU de uma ultracentrífuga em função de variáveis que são controladas experimentalmente. Os dados experimentais correspondem a 460 experiências no processo de ultracentrifugação para a separação do isótopo de urânio. As variáveis de controle do processo, que influenciam significativamente os valores de δU, são escolhidas a fim de dar a informação sobre o comportamento da ultracentrífuga quando submetidas a diversos níveis de fluxo F, da variável corte θ e de pressão da linha Pp. As curvas de resposta são feitas relacionando a energia de separação com as variáveis de controle F, θ e Pp, para comparar o modelo com os dados experimentais e para calcular finalmente seus valores otimizados.

Palavras-chave: ultracentrífuga; uranium hexafluorido; separação isotópica; matriz de covariância; método dos quadrados mínimos.

I. INTRODUCTION

A gas ultracentrifuge, as schematized in Fig. 1 is composed of a long, thin vertical cylinder (rotor), rotating around its axis at a high velocity inside a case under vacuum. The process gas, assumed to be a binary isotopic mixture with $^{235}UF_6$ and $^{238}UF_6$, inside the cylinder is subjected to a centrifuge force that establishes a pressure gradient in the radial direction, increasing from the center to the rotor wall (Jordan, 1980). That

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pressure distribution, proportional to mass, is slightly dissimilar for the different isotopes. This results in a partial separation of the feed F, into two fractions: an enriched one (product) and another depleted (waste) in the desired isotope ($^{235}UF_6$). The ultracentrifuge performance and production capacity evaluation is usually done by means of the required work to isotope separation, which is proportional to the amount of processed material and to the obtained separation degree. The dependent variable that best defines the separative efficiency of any isotope separation unit, is the separative power or capacity δU, given by the following expression:

$$\delta U = P*\frac{R_p - 1}{R_p + 1}*\ln R_p + W*\frac{R_w - 1}{R_w + 1}*\ln R_w - F*\frac{R_f - 1}{R_f + 1}*\ln Rf \qquad (1)$$

where the operational variables F, P and W are the streams of feed, product and waste; x, y
and z are the isotope desired compositions, respectively; and the response variables are the
abundance rations of product R_p=y/(1-y) and waste R_w=x/(1-x).

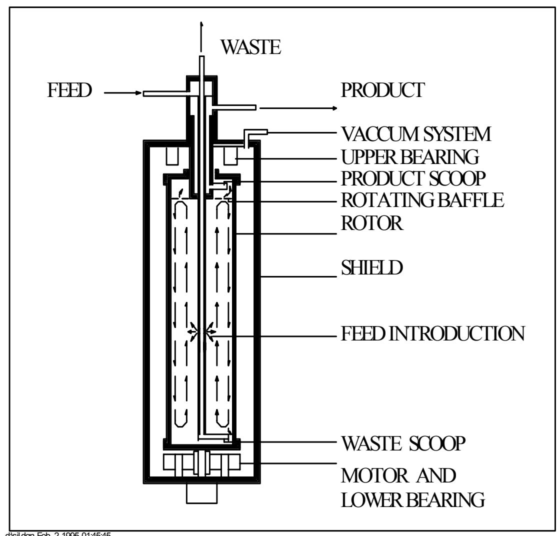

Figure 1: Countercurrent ultracentrifuge design

II. EXPERIMENTS

An isotopic separation test consists in the operation of an ultracentrifuge in a bench plant shown in Fig. 2. The ultracentrifuge receives an injection of a binary isotopic mixture with $^{235}UF_6$ and $^{238}UF_6$ as feed flow F and permits the extractions of the product flow P and waste flow W. Samples are collected for verification of the separation obtained by the measures of the abundance ratio of the enriched and depleted streams, R_p and R_w, respectively, allowing to calculate the separative power δU, given by Eq. (1). Defining the cut θ as the relation between the product and feed flow and fixing the product pressure line p_p, several groups of data are generated with the variation of the cut θ and the feed flow F. Each of them is denominated a separation experiment, resulting in an ultracentrifuge performance function like δU (F, θ, Pp).

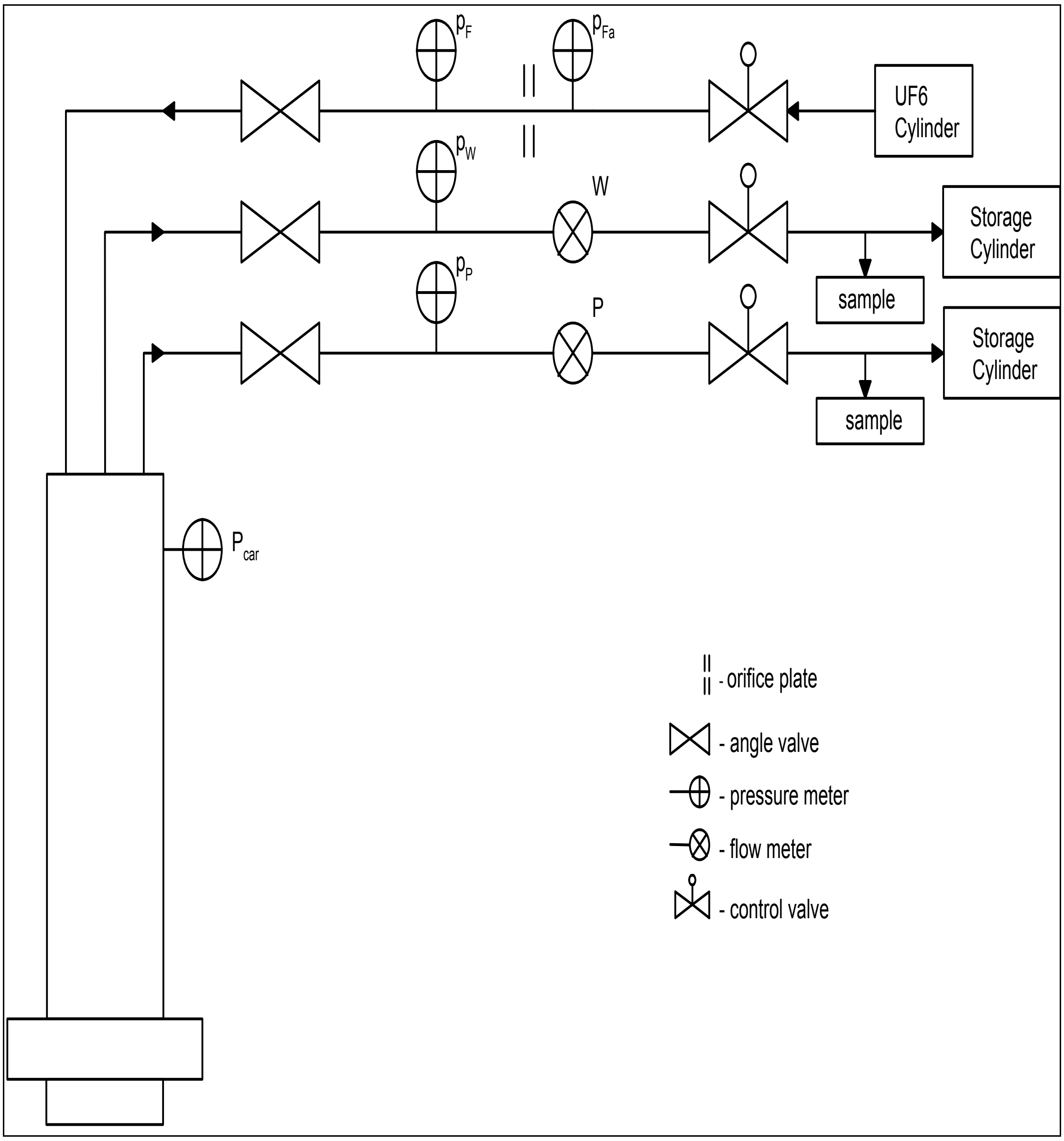

Figure 2: Experimental bench plant design.

III. STATISTICAL THEORY

The measurements of R_P, R_W, P and W, involved in the separative power determination δU, provide correlated uncertainties and define a covariance between them. These statistical uncertainties are propagated in Eq. (1) in order to obtain a final δU uncertainty by the expression (Cowan,1998):

$$(\sigma_{\delta U})^2 \approx \sum_{i=1}^{n} \left(\frac{\partial \delta U}{\partial x_i} \right)^2 \sigma_i^2 \tag{2}$$

where x_i are the independent variables R_P, R_W, P and W, σ_i express their respective variances. The R_P and R_W variances are directly given by mass spectrometry analysis while the P and W variances are calculated from mass flowmeters calibration curves. Each δU experimental data covariance matrix is calculated by the expression:

$$\left(V_{\delta U}\right)_{ij} = \sum_{l=1}^{L} \rho_{ijl} e_{il} e_{jl} \quad \left(i, j = 1, n\right) \tag{3}$$

where e_{il}, e_{jl} are the partial uncertainty magnitude of any independent variable R_P, R_W, P and W; ρ_{ijl} represents the microcorrelations between these variable measurements due to each attribute *l*. The process analysis permits to determine these microcorrelations values with safety. The δU experimental data fitting through a performance function of the kind δU (F, θ, internal variables) is obtained due to δU and (F,θ, Pp) relation that may be written as a second order polynomial given by:

$$Y = \beta_0 + \sum \beta_i x_i + \sum \beta_{ii} x_i^2 + \sum \beta_{ij} x_i x_j + \sum \beta_{iij} x_i^2 x_j + \sum \beta_{iijj} x_i^2 x_j^2 \qquad i \neq j \tag{4}$$

where Y is the response (δU), β_i are the equation coefficients and x_i , x_j are the controlled variables (F, θ, Pp). This equation is used to evaluate the linear, quadratic and interaction effects between these variables providing the project matrix A that contains all the fitted model explained variables. The Eq. (4) is a linear function in the β_i parameters and although we can perform the least-squares method to any function, in this case the chi-square and estimators resulting values have desired properties: the estimators and their variances can be analytically obtained, they will be unbiased with minimum variance no matter the number of experiments or the experimental data distribution function. According to the least-square method with covariance matrix, the best possible solution is the one which minimizes the chi-square χ^2. The χ^2 value for this particular problem is given by (Smith, 1981 and 1993):

$$\chi^2 = \left(\delta U_{\exp} - \delta U_{calc}\right)^t * V_{\delta U}^{-1} * \left(\delta U_{\exp} - \delta U_{calc}\right) \tag{5}$$

where $\delta U_{calc} \approx A\beta$, and β is the coefficients estimates vector of the fitted equation. Under the following conditions: (i) the δU experimental data is distributed according to a normal with a known covariance matrix, which permits to use the chi-square statistic, (ii) the fitted function, Eq.(4) is linear in the coefficients β_i, allowing to obtain an analytical solution for Eq.(5) and (iii) the functional form of the fitted function, Eq.(4), is corrected, , i. e., it is possible to obtain the minimum deviation between the experimental and predicted values, so the quadratic form χ^2 should be distributed in conformity with the chi-square tables, allowing to evaluate the model goodness-of-fit (Cowan, 1998).

The desired least-square solution is given by:

$$\beta = V_\beta \, A^t \, V_{\delta U}^{-1} \, \delta U_{\exp} \tag{6}$$

where the covariance matrix for the solution β is given by:

$$V_\beta = \left(A^t \, V_{\delta U}^{-1} A\right)^{-1} \tag{7}$$

that gives the coefficients estimates variances and covariances of the experimental data fitted curves. In this case, a FORTRAN program (Migliavacca, 2004) is used.

IV. RESULTS AND DISCUSSION

The experimental data performed with only one ultracentrifuge covered the whole domain of interest, consisting of eight levels of feed flow F, seven levels of cut θ and five levels of the product pressure line Pp, resulting in a group of 460 experiments. Due to secret character inherent to the process development, the sensitive data were codified, with all variables related to arbitrary units. The isotopic abundance ratios R_F, R_P and R_W , and the flow values F, P and W, with their respective uncertainties; the separative power δU and cut θ experimental values are presented in Tab. (1). In Tab. (2), are presented the coefficients estimates of the fitted equation, the determination coefficient, the chi-square and normalized chi-square and in Tab. (3) are presented their variances and covariances in the upper triangle and their correlations in the lower triangle.

Table 1: Codified variables and their uncertainties values

Exp	R_F ($x10^{-04}$)	R_p ($x10^{-03}$)	R_w ($x10^{-04}$)	σ_{Rf} ($x10^{-06}$)	σ_{Rp} ($x10^{-07}$)	σ_{Rw} ($x10^{-07}$)	P	W	σ_P	σ_W	θ	Pp	δU
1	9.8086	1.1982	8.9588	1.1900	4.2337	4.11	67.478	75.853	0.124	0.734	0.4631	1.29	1.02
2	9.8114	1.1622	9.0964	2.3143	12.1834	0.79	98.048	102.216	0.112	0.675	0.4995	1.29	1.01
3	9.8114	1.1645	9.0994	2.3143	15.8560	8.67	97.547	102.216	0.112	0.675	0.4945	1.29	1.03
4	9.8114	1.1619	9.1356	2.3143	6.6741	28.51	98.048	102.216	0.112	0.675	0.4922	1.29	0.98
5	9.8014	1.1784	9.0483	2.0429	18.4626	1.36	86.021	90.278	0.116	0.700	0.4790	1.29	1.04
6	9.8014	1.1779	9.0272	2.0429	6.7377	12.43	85.018	91.273	0.117	0.698	0.4837	1.29	1.05
7	9.8014	1.1729	9.0604	2.0429	19.3306	5.87	86.021	90.278	0.116	0.700	0.4863	1.29	0.99
...	...	...	...	...	...	...	...	...	...	...	...	...	...
460	9.9400	1.0760	8.5600	2.4900	6.6000	6.20	105.56	47.003	0.098	0.810	0.8170	0.72	0.57

Table 2: Model coefficients estimates and model goodness-of-fit parameters

β_0	β_1	β_2	β_{11}
0.1711	2.8038	0.3830	-3.368
β_{22}	β_{223}	β_{1133}	β_{2233}
-0.4533	3,517E-03	3.986E-06	-9.5467E-06
R^2	χ^2	χ^2_{red}	
0,9268	418.76	0.94	

Table 3: Covariance and correlation matrices

β_0	β_1	β_2	β_{11}	β_{22}	β_{223}	β_{1133}	β_{2233}
0,002	-0,003	0,001	-0,001	0,001	8,7E-9	-1,2E-9	1,8E-8
-0,614	**0,011**	0,000	1,0E-4	-0,005	-5,43E-8	9,5E-9	-2,5E-6
0,631	0,042	**4,0E-04**	-7,6E-4	0,000	-1,68E-9	2,5E-9	-8,5E-7
-0,705	0,038	-0,897	**0,002**	-6,8E-4	-3,18E-10	4,1E-10	-9,0E-8
0,382	-0,548	-0,054	-0,019	**0,006**	-2,93E-8	3,8E-9	-1,2E-6
0,173	-0,465	-0,083	-0,006	-0,339	**1,21E-12**	-1,7E-13	4,3E-11
-0,065	0,228	0,346	0,022	0,121	-0,386	**1,6E-13**	-4,5E-11
0,003	-0,205	-0,390	-0,017	-0,133	0,337	-0,970	**1,42E-8**

As an additional verifying of possible residuals serial correlation, the Fig. 3 presents the model residuals distribution in positive and negative values around zero, characterizing a random scattering. In Figs. 4 - 5 are presented the residuals graphs against the controlled variables, which permit to evaluate the regression model residuals heteroscedasticity degree (Vasconcellos and Portella, 2001).

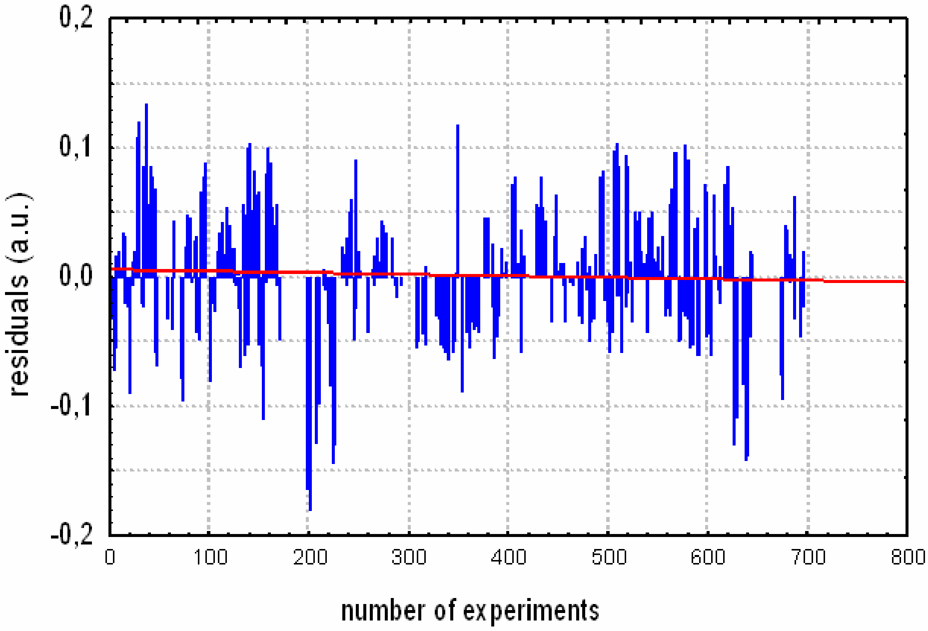

Figure 3: Residuals against number of experiments

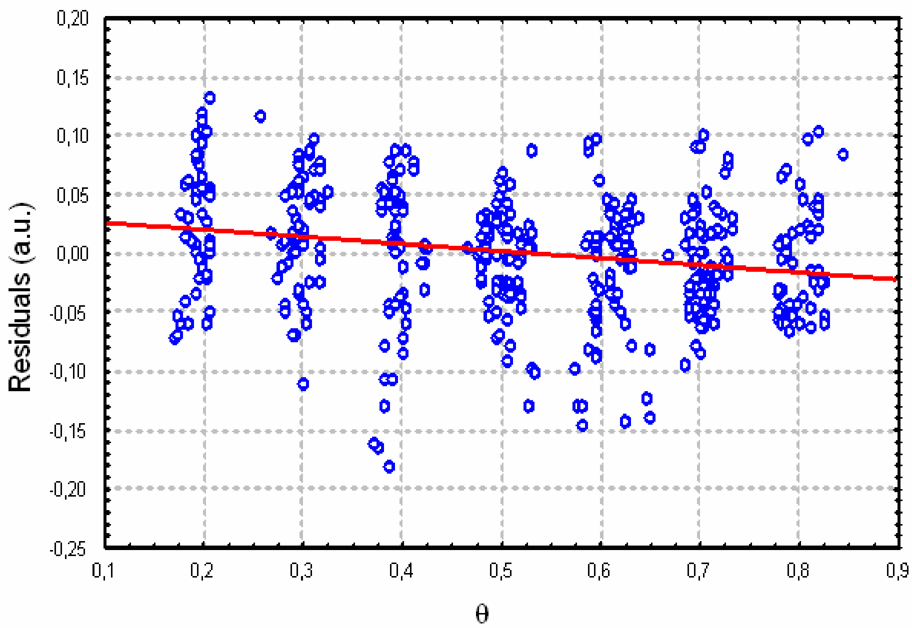

Figure 4: Residuals against θ.

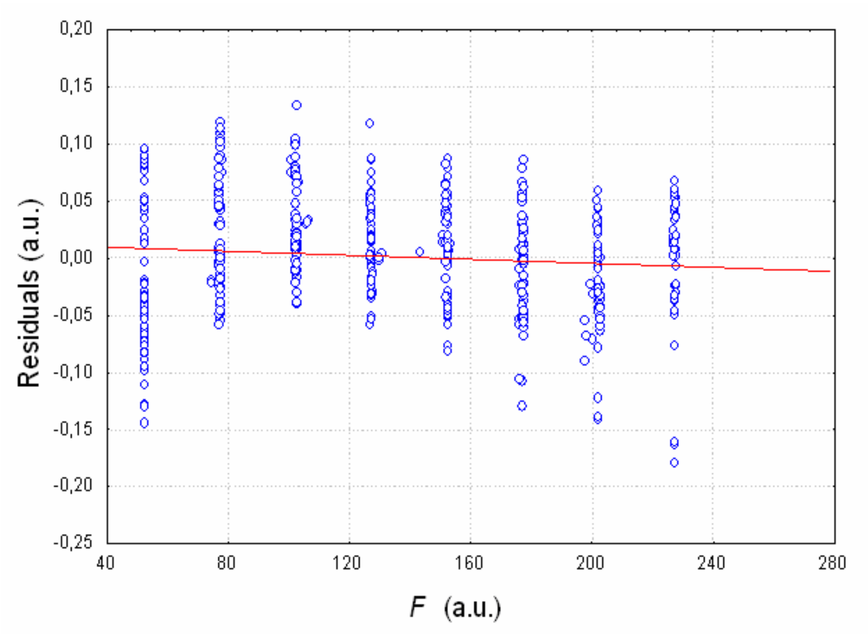

Figure 5: Residuals against F.

Through Figs. 6 - 7 it is possible to verify how satisfactorily the theoretic curve fits the experimental data and finally in Figs. 8 - 9 are presented the response surface of the separative power δU, against F and θ, and, Pp θ, that allows visualizing the δU behavior in the ultracentrifugation process to find the optimum values of the operational controlled variables.

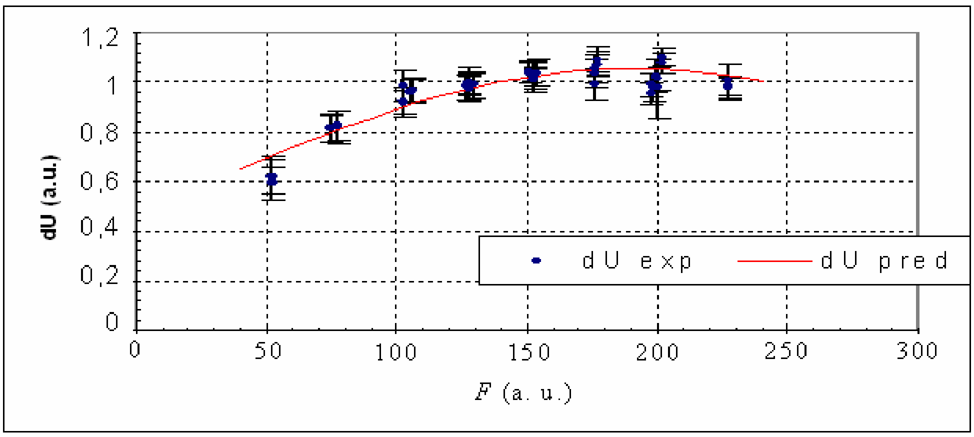

Figure 6: δU against F.

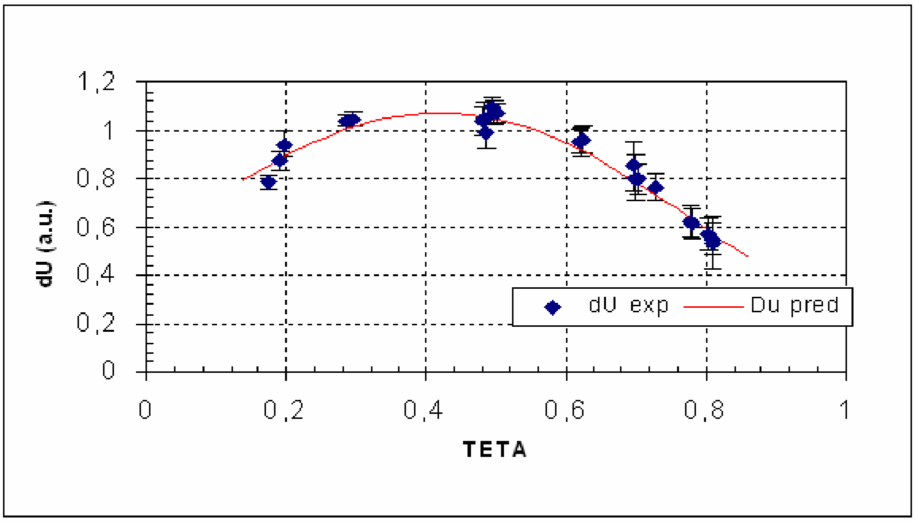

Figure 7: δU against θ.

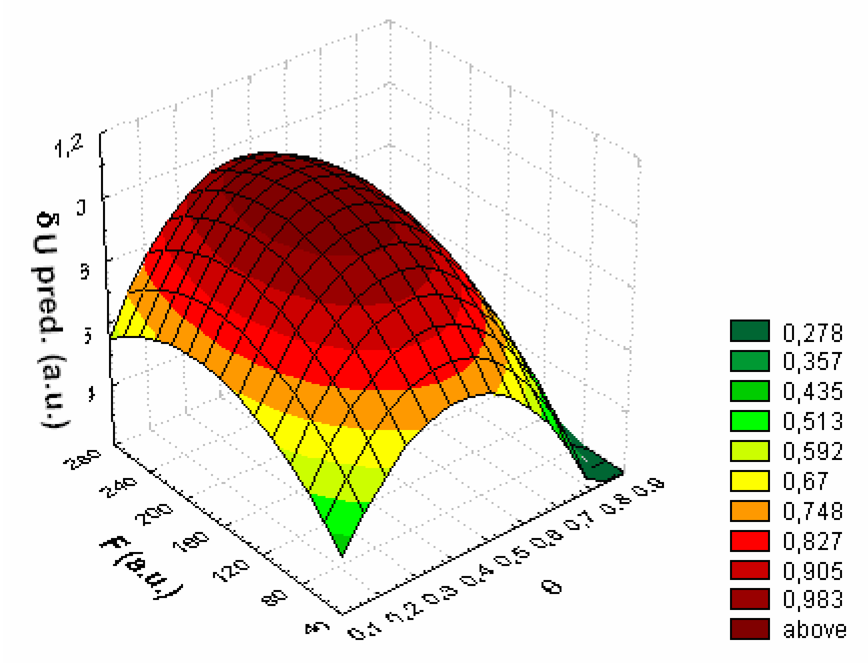

Figure 8: δU against F and θ.

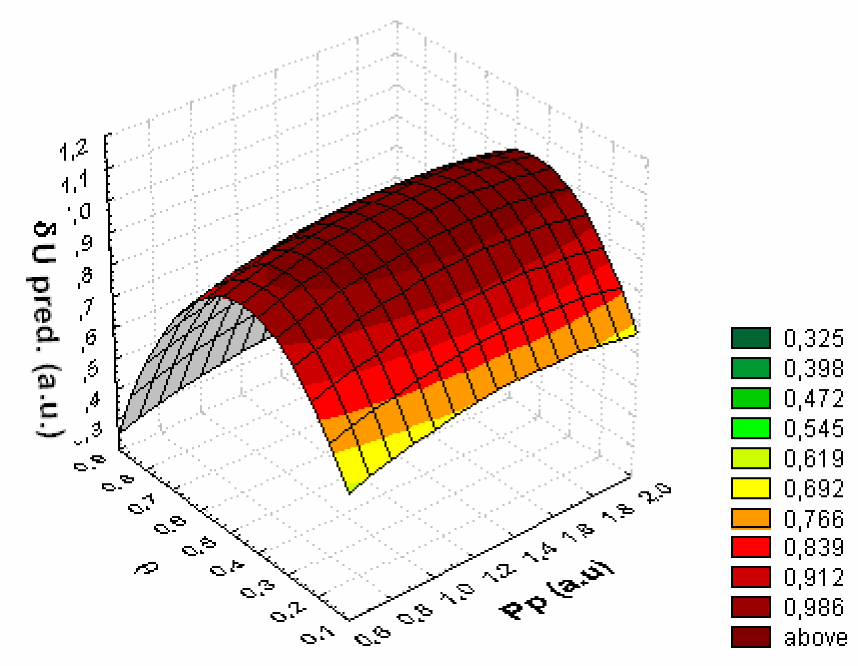

Figure 9: *δU* against *Pp* and *θ.*

V. CONCLUSIONS

The least-squares method with covariance matrix was successfully applied to determine the ultracentrifuge separative power δU fitting curve against experimentally controlled variables. The normalized chi-square obtained showed a very reasonable agreement between the experimental δU data dispersion and the uncertainties estimated through their covariance matrix. The fitted model was able to explain the experimental data due to the determination coefficient (R^2= 0,9268). In Figs. 4 - 5, it is possible to verify that there is no visible pattern between the residuals and the control variables and finally through the response curve graphs, Figs. 6 - 7, the theoretical model is showed to be reasonably fitted to the experimental data.

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