COMPARATIVE ANALYSIS OF DIFFERENT APPROACHES FOR THEORETICAL SIMULATION OF ULTRACENTRIFUGES ISOTOPIC SEPARATION CASCADES

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ABSTRACT

In order to simulate a real cascade performance in terms of the external and internal flowrates and isotopic compositions, it is necessary to solve a system of equations composed of the internal mass balances for the element (U) and for the desired isotopes. Considering the separation of a binary isotopic mixture, for a cascade with n stages, we have a system of $4n$ independent equations with $6n$ unknowns.

This kind of system has infinite solutions, unless we introduce practical or theoretical new equations describing the centrifuge separation performance and/or use approximations in terms of restrictions to the stages behavior.

Depending on the equations and/or restrictions we use, the simulation results can be quite different.

In this paper, six different combinations between theoretical equations and stage restrictions will be analyzed and compared using experimental results, in order to establish the best mathematical model to be used in the theoretical simulation of a real cascade performance.

Key Words: Isotopic Separation, Ultracentrifugation, Cascade Simulation.

I. INTRODUCTION

An ultracentrifuge is a separation device that, when fed with a stream composed of an isotopic mixture, produces two other streams: the product, enriched in the isotopes with lower molecular weight, and the tails, enriched in the isotopes with higher molecular weight.

The main characteristic of the ultracentrifugation process is a high level of separation in only one stage, but with low throughput.

In the design of a plant to produce enriched uranium, the amount of material to be produced and the isotopic compositions to be achieved are specified. In order to obtain material with the specified isotopic compositions, ultracentrifuges are connected in series arrangements, composing separation stages. In order to produce the amount of material specified, ultracentrifuges are connected, in each stage, in parallel arrangements. This complete arrangement is known as an isotopic separation cascade. The separation stages are interconnected in such a way that the feed stream of a generic stage i is composed of the product stream of stage $i-1$ mixed with the tails stream of stage $i+1$.

Normally, the design of an uranium enrichment cascade to perform a given separation task, in terms of product composition and quantity, is made minimizing the operational costs. Considering the material to be processed as a binary mixture, this results in a cascade profile as similar as possible to the ideal one [1], with an established tails composition.

The simulation of the cascade real behavior depends on knowing the ultracentrifuge characteristic curves relating the separative parameters and the flow and pressure variables. Using these curves and the criteria of maximizing the installed separative capacity for the given product and tails compositions, we can obtain, depending on how much the cascade profile is similar to the ideal one, a flow distribution in which some ultracentrifuges can operate very close to the optimal point, but some others can operate far from this same point.

However, using theoretical relations and/or restrictions to the stages behavior, we can achieve results similar to these ones without knowing the real curves cited above. This level of information is sufficient for the starting phase of a cascade design.

In this paper, six different kinds of approximation possible to be used in the theoretical calculation of the internal flow and composition distributions will be compared with experimental results, in order to establish the best method for theoretical cascade simulation.

II. EQUATIONS AND RESTRICTIONS

To establish the flowrate and isotopic composition of all the internal streams of a given cascade, we have to solve a system composed of the following fundamental equations:

1 - material balance for the U compound in each stage (*n* equations):

$$F_i = P_i + W_i \tag{1}$$

where F_i, P_i and W_i are, respectively, the feed, product and tails flowrates;

2 - material balance for the desired isotope (^{235}U) in each stage (*n* equations):

$$F_i z_i = P_i y_i + W_i x_i \tag{2}$$

where z_i, y_i and x_i are, respectively, the feed, product and tails ^{235}U weight percentage;

3 - material balance for the U compound in the streams mixing points (*n* equations):

$$\begin{aligned} &F_1 = W_2 \\ &F_i = P_{i-1} + W_{i+1} + \delta_{i,f} F_c, \quad \text{for } i = 2,..., n-1 \\ &F_n = P_{n-1} \end{aligned} \tag{3}$$

where F_c is the cascade feed flowrate and $\delta_{i,f}$ is equal to 1 for the feed stage, and 0 for the others;

4 - material balance for the desired isotope in the streams mixing points (*n* equations):

$$\begin{aligned} &F_1 z_1 = W_2 x_2 \\ &F_i z_i = P_{i-1} y_{i-1} + W_{i+1} x_{i+1} + \delta_{i,f} F_c z_f, \text{ for } i = 2,..., n-1 \\ &F_n z_n = P_{n-1} y_{n-1} \end{aligned} \tag{4}$$

where z_f is the ^{235}U weight percentage in the feed material.

Introducing in these equations the concepts of cut (θ_i), heads separation factor (β_i) and tails separation factor (γ_i):

$$\theta_i = P_i/F_i \tag{5}$$

$$\beta_i = R_{pi}/R_{fi} = y_i(1-z_i)/[z_i(1-y_i)] \tag{6}$$

$$\gamma_i = R_{fi}/R_{wi} = z_i(1-x_i)/[x_i(1-z_i)] \tag{7}$$

where R_{fi}, R_{pi} and R_{wi} are, respectively, the feed, product and tails ^{235}U abundance ratios, they can be transformed into the following relations:

$$P_i = \theta_i F_i, \quad i=1,..., n \tag{8}$$

$$W_i = (1-\theta_i)F_i, \quad i=1,..., n \tag{9}$$

$$\theta_i = \frac{(\gamma_i - 1)[1 + z_i(\beta_i - 1)]}{\beta_i \gamma_i - 1}, \quad i=1,..., n \tag{10}$$

$$\begin{aligned} &F_1 - (1-\theta_2)F_2 = 0 \\ &-\theta_{i-1}F_{i-1} + F_i - (1-\theta_{i+1})F_{i+1} = \delta_{i,f} F_c, \quad i=2,...,n-1 \\ &-\theta_{n-1}F_{n-1} + F_n = 0 \end{aligned} \tag{11}$$

$$\begin{aligned} &F_1 z_1 - (1-\theta_2)F_2 x_2 = 0 \\ &-\theta_{i-1}F_{i-1}y_{i-1} + F_i z_i - (1-\theta_{i+1})F_{i+1}x_{i+1} - \delta_{i,f} F_c z_f = 0, \quad i=2,...,n-1 \\ &-\theta_{n-1}F_{n-1}y_{n-1} + F_n z_n = 0 \end{aligned} \tag{12}$$

$$y_i = \frac{\beta_i z_i}{1 + z_i(\beta_i - 1)}, \quad i=1,..., n \tag{13}$$

$$x_i = \frac{z_i}{\gamma_i - z_i(\gamma_i - 1)}, \quad i=1,..., n \tag{14}$$

The ideal cascade [1] is defined as the cascade arrangement that minimizes the total internal flowrate and, consequently, the power consumption. The ratio between the calculated flowrate per stage and the optimal feed flow rate of one ultracentrifuge (G) gives the number of centrifuges in each stage. In this case, all the ultracentrifuges operate in the same optimal flow and separation conditions, what means the same feed flowrate, cut (symmetric), heads and tails separation factors ($\beta=\gamma$), that maximize the ultracentrifuge separative power, defined as:

$$\delta U = [\theta \frac{R_p - 1}{R_p + 1} \ln R_p + (1-\theta)\frac{R_w - 1}{R_w + 1} \ln R_w - \frac{R_f - 1}{R_f + 1} \ln R_f] * \\ * G * \frac{238}{352} \tag{15}$$

In the real case, however, it is not possible to maintain all those variables in the optimal point, because we have to round the calculated numbers of centrifuges per stage to integer values and, at the same time, respect the mass balance equations described above.

In order to simulate the real cascade behavior, we can add to this system of equations, based on the ideal cascade behavior, the following restrictions:

- constant cut for all the stages;
- symmetric separative behavior for all the stages ($\beta=\gamma$);
- constant separation factor ($\alpha = \beta*\gamma$) for all the stages;
- constant separative power for all the stages.

The symmetric separative behavior of one ultracentrifuge can be established using theoretical relations to calculate the heads and tails separation factors. The solution of the diffusion-convection equation in the internal centrifuge field gives, for β and γ, the following equations [2]:

$$\beta = \frac{C_1 + \theta G}{\theta G + C_1 \exp[-(C_1 + \theta G) z_e / C_5} \tag{16}$$

$$\gamma = \frac{C_1 \exp\{[C_1 - (1-\theta)G] z_s / C_5\} - (1-\theta)G}{C_1 - (1-\theta)G} \tag{17}$$

where z_e and z_s are related to the feed flowrate introduction position, and C_1 and C_5 are theoretical parameters that can be written as functions of the centrifuge efficiency (e) and its components [2] e_C, e_F and e_I:

$$C_1 = \frac{\sqrt{2}\Delta M}{2RT} \pi \rho D \omega^2 a^3 \sqrt{\frac{e_F e_C}{1-e_C}} = b \sqrt{\frac{e / e_I}{1-e_C}} \tag{18}$$

$$C_5 = \pi a^2 \rho D(1+m^2) = C_5' \frac{1}{1-e_C} \tag{19}$$

The three efficiency components can be calculated assuming a theoretical internal flow profile [2]. An alternative procedure to estimate these components without the assumption of a theoretical internal flow profile is to consider that, in the optimal separation conditions (G_{ot}, α_{ot}, θ_{ot}, δU_{ot} or e_{ot}), the component e_I reaches its maximum value. So, in this point, for which the centrifuge operates in symmetric process [1], the following equation is valid:

$$\beta_{ot} = \frac{b\sqrt{\frac{e/e_I}{1-e_C}} + \theta_{ot} G_{ot}}{\theta_{ot} G_{ot} + b\sqrt{\frac{e/e_I}{1-e_C}} \exp\left[-\left(b\sqrt{\frac{e/e_I}{1-e_C}} + \theta_{ot} G_{ot}\right) \frac{z_e(1-e_C)}{C_5'}\right]} \tag{20}$$

$$\gamma_{ot} = \frac{b\sqrt{\frac{e/e_I}{1-e_C}} \exp\left[\left(b\sqrt{\frac{e/e_I}{1-e_C}} - (1-\theta_{ot})G_{ot}\right) \frac{z_s(1-e_C)}{C_5'}\right] - (1-\theta_{ot})G_{ot}}{\left(b\sqrt{\frac{e/e_I}{1-e_C}} - (1-\theta_{ot})G_{ot}\right)} \tag{21}$$

$$\alpha_{ot} = \beta_{ot} * \gamma_{ot} \tag{22}$$

Using this relation, we can find the value of e_C for which e_I reaches its maximum value. Obtaining e_I and e_C, we can calculate C_1 and C_5 for the optimal point. For the cascade stages, this calculation procedure can be converted into a theoretical restriction:

- constant values of C_1 and C_5 for all the stages.

III. THEORETICAL MODELS FOR CASCADE SIMULATION

Combining the 7*n* mass balance relations and 2*n* of the restrictions cited above, we can construct the different mathematical models described below.

1 - Cascade with constant cut and constant separation factor

In this model, it is assumed that all the stages work with the optimal cut θ_{ot} and the optimal separation factor α_{ot}.

2 - Cascade with constant cut and constant C_1 and C_5 theoretical parameters

In this model, the cut is assumed to be θ_{ot} for all the stages and the heads or the tails separation factors are calculated using the theoretical relations.

3 - Cascade with symmetric stages and constant separation factor

In this model we assume $\beta_{ot} = \gamma_{ot} = \sqrt{\alpha_{ot}}$ for all the stages.

4 - Cascade with symmetric stages and constant C_1 and C_5 parameters

In this model, we assume $\beta = \gamma$ for all the stages, being β or γ calculated using the theoretical relations.

5 - Cascade without restrictions of cut or symmetric behavior and constant C_1 and C_5 parameters

In this model, we assume that β and γ obey the theoretical relations for all the stages.

6 - Cascade with symmetric stages and constant centrifuge efficiency

In this model, we assume that all the centrifuges operate with the maximum separative power δU_{ot} (efficiency e_{ot}) and in the symmetric process ($\beta = \gamma$).

IV. COMPARISON BETWEEN CALCULATED VALUES AND EXPERIMENTAL RESULTS

Experimental results were obtained using four different cascade configurations, with different number of stages and different numbers of centrifuges per stage, operating in their nominal point.

The internal and external flowrates and compositions were calculated, for each cascade configuration, using the six mathematical models described above.

The percentual relative deviation between the calculated values using each model and the experimental results obtained in each cascade for the external variables are registered in Table 1.

As external variables, the product and tails abundance ratios (R_p and R_w), the ratio between the product and feed flowrates (P/F) and the separative capacity (ΔU) are compared.

TABLE 1 - Percentual relative deviation between calculated values and experimental results.

Mathematical Model	Cascade		dR_p (%)	dR_w (%)	dP/F (%)	$d\Delta U$ (%)
1	1		1.5228	-1.1666	-1.7003	5.6400
	2		3.2483	-1.7838	-1.5481	7.0963
	3		-0.2841	0.4127	-0.5487	-1.9951
	4		-11.7699	2.7833	8.0844	-11.5752
		average	-1.8207	0.0614	1.0718	-0.2085
		st. deviation	6.7878	2.0367	4.7029	8.5627
		confidence interval	-12.6200 ≤d≤ 8.9786	-3.1791 ≤d≤ 3.3019	-6.4104 ≤d≤ 8.5540	-13.8318 ≤d≤ 13.4148
2	1		0.8549	0.1296	-1.7003	0.7669
	2		1.1774	0.1049	-1.5481	0.3535
	3		-0.1997	0.4127	-0.5487	-1.8450
	4		-6.0826	-0.3976	8.0844	-1.0285
		average	-1.0625	0.0624	1.0718	-0.4383
		st. deviation	3.3980	0.3370	4.7029	1.2120
		confidence interval	-6.4687 ≤d≤ 4.3438	-0.4737 ≤d≤ 0.5985	-6.4104 ≤d≤ 8.5541	-2.3665 ≤d≤ 1.4900
3	1		1.5228	-1.1666	-1.7003	5.6035
	2		3.1300	-1.7838	-1.3546	7.0073
	3		-1.1280	0.4127	0.6859	-2.0806
	4		-12.7361	2.5845	9.6661	-11.6479
		average	-2.3028	0.0117	1.8243	-0.2794
		st. deviation	7.1737	1.9487	5.3328	8.5672
		confidence interval	-13.7161 ≤d≤ 9.1105	-3.0887 ≤d≤ 3.1121	-6.6603 ≤d≤ 10.3089	-13.9098 ≤d≤ 13.3510
4	1		0.5878	-0.1944	-1.1109	0.6834
	2		0.0533	-0.3148	0.4838	0.2114
	3		-1.1842	0.2162	0.9602	-1.8458
	4		-1.0980	0.1988	0.8787	-0.9620
		average	-0.4103	-0.0236	0.3030	-0.4783
		st. deviation	0.8723	0.2714	0.9652	1.1444
		confidence interval	-1.7981 ≤d≤ 0.9776	-0.4553 ≤d≤ 0.4082	-1.2327 ≤d≤ 1.8387	-2.2991 ≤d≤ 1.3425
5	1		2.7251	1.7498	-6.1891	1.3991
	2		6.6209	1.7838	-10.7886	0.9555
	3		16.5087	2.3777	-20.0274	-1.5960
	4		18.4453	2.3857	-20.7381	-1.4074
		average	11.0750	2.0743	-14.4358	-0.1622
		st. deviation	7.6028	0.3553	7.1250	1.5592
		confidence interval	-1.0210 ≤d≤ 23.1710	1.5091 ≤d≤ 2.6395	-25.7717 ≤d≤ -3.0999	-2.6429 ≤d≤ 2.3185
6	1		0.7213	-0.1944	-1.2242	1.6180
	2		0.6449	-0.5247	0.0968	1.4796
	3		-0.3685	0.0197	0.4152	-0.9801
	4		1.2297	-0.1988	-0.8787	0.9681
		average	0.5569	-0.2246	-0.3987	0.7714
		st. deviation	0.6693	0.2246	0.7776	1.2007
		confidence interval	-0.5079 ≤d≤ 1.6217	-0.5819 ≤d≤ 0.1327	-1.6358 ≤d≤ 0.8384	-1.1388 ≤d≤ 2.6816

With these deviation values, is calculated, for each external variable in each different model, the confidence interval for the real average deviation with 95% of significance level [3].

As internal variables, the separation factors of all stages were experimentally determined for one of the four cascade arrangements (cascade 3). The obtained percentual relative deviations as a function of the relative position of the stages in the enriching and stripping sections of the cascade are shown in Fig. 1.

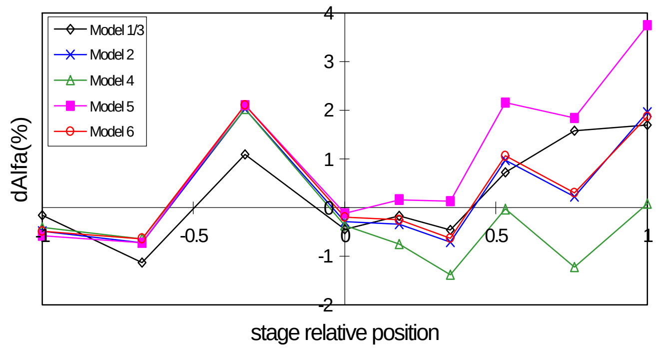

Figure 1. Internal Separation Factors Comparison

As it can be seen in the figure, for five methods (models 1, 2, 3, 4 and 6) we have obtained very good results, with a small percentual relative deviations.

Treating the separation factor deviation of all the stages as independent variables, we can calculate the average value and the 95% confidence interval for the average deviation considering the six different models. The obtained values are shown in Table 2.

TABLE 2 - Average Values for Percentual Relative Deviations of the Internal Separation Factors.

Model	Average Deviation	Standard Deviation	Confidence Interval
1	0.3973	1.0604	-0.0854 ≤d≤ 0.8800
2	0.2187	0.9574	-0.2171 ≤d≤ 0.6545
3	0.3973	1.0604	-0.0854 ≤d≤ 0.8800
4	0.2543	0.9218	-0.1653 ≤d≤ 0.6739
5	1.2100	1.4135	0.5666 ≤d≤ 1.8534
6	0.3467	0.9284	-0.0759 ≤d≤ 0.7693

In order to assure that the internal separation factors can really be treated as independent variables, an analysis of residuals [3] was executed and the obtained residuals (local value - average value) can be seen in Fig. 2.

This figure shows that there is no tendency in the behavior of the analyzed variable in the cascade stages, that permits to consider the stage separation factors as independent variables.

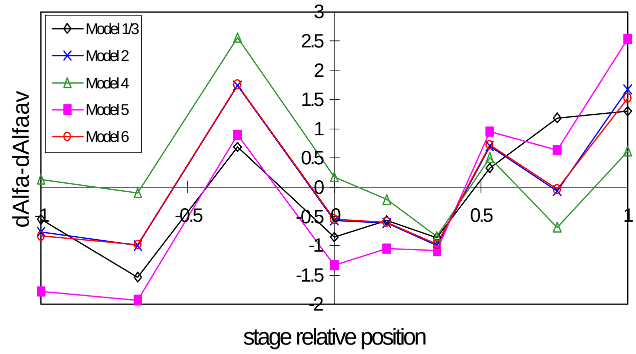

Figure 2. Residuals Analysis

V. CONCLUSIONS

As it can be seen in the tables and figures shown, the best method for the theoretical simulation of a real cascade behavior is the one for which all the stages were considered as operating in symmetric process and with the maximum separative power (model 6), followed by model 4. For this first model, we have obtained the smallest percentual relative deviation between the calculated and measured values for almost all the variables compared. This approximation introduces small errors to the model because of the shape of the curve δU x G in a centrifuge. There is a relatively large feed flowrate interval for which δU is almost constant. This fact permits to fit, for each cascade, an internal feed flowrate profile with different values for each stage without being very far from the real separative power value.

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