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Analysis of ^{235}U Enrichment by Chemical Exchange in U(IV)-U(VI) System on Anionite

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Abstract: A theoretical study about ^{235}U enrichment by the chemical exchange method in U(IV)-U(VI) system on anion-exchange resins is presented. The ^{235}U isotope concentration profiles along the band were numerically calculated using an accurate mathematical model and simulations were carried out for the situation of product and waste withdrawal and feed supply. By means of numerical simulation, an estimation of the migration time, necessary for a desired enrichment degree, was obtained. The required migration distance, the production of uranium 3 at.% ^{235}U per year and the plant configuration are calculated for different operating conditions. An analysis of the process scale for various experimental conditions is also presented.

Keywords: ^{235}U enrichment, U(VI)/U(IV) chemical exchange, anion exchange resin, numerical simulation

INTRODUCTION

Research studies on uranium isotope separation by a chemical exchange method were done since the late 1940's and the first successful semi-commercial

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plant achieved an enrichment of 3 at.% ^{235}U starting from the natural abundance, in 1982 (1).

Theoretical approaches of isotope separation by ion exchange chromatography date from the 1950's, when Spedding (2) published experimental and theoretical results on nitrogen isotope separation. A fundamental equation of isotope separation by displacement chromatography was formulated by Fujii (3), based on the concept of the relative movement of isotopes in the enrichment and diffusion processes. Aoki (4), derived a set of equations to describe the enrichment profile including dynamic behavior, and proposed a numerical method to solve them. The simulations were made for the case of band operation and used Spedding's experimental data on nitrogen isotopes separation. A theoretical study using an appropriate mathematical analysis model of uranium isotopes separation by chemical exchange, in a reverse breakthrough operation, was developed by the authors of this paper (5). The model parameters were identified from experimental data and simulations were carried out for different experimental conditions.

A simulation study of the enrichment process in the case of feed supply, product, and waste withdrawal was developed recently (6). By means of numerical simulation, a few parameters, useful for cascade analysis, such as the amount of chemicals and migration time have been determined. An important study on plant scale, providing some basic relations for computing the necessary migration length and the width of the enriched zone, was done by Fujii (3). The research in this direction was further developed by Ohwaki (7) who proposed a method for the calculation of the volume of resin necessary to obtain a desired ^{15}N enrichment in a nitrogen isotope separation plant.

This paper presents a theoretical approach of uranium isotope enrichment by a chemical exchange method. The plant scale, to obtain a certain enrichment of ^{235}U, was estimated based on some numerical simulation results and analyzed for various operating conditions.

EXPERIMENTAL

The chemical method of uranium isotopes separation is based on the electronic exchange reaction between U(IV) and U(VI), represented by the relation:

$$^{238}\mathrm{U(VI)}_{(\mathrm{R})} + {}^{235}\mathrm{U(IV)}_{(\mathrm{S})} \underset{k_b}{\overset{k_f}{\rightleftarrows}} {}^{235}\mathrm{U(VI)}_{(\mathrm{R})} + {}^{238}\mathrm{U(IV)}_{(\mathrm{S})} \tag{1}$$

where the subscripts (R) and (S) refer to the resin and solution phases. The single stage separation factor α, equal to the equilibrium constant $K = k_f/k_b$, was determined in previous studies (8, 9) to be 1.0014 ± 0.003.

The process is considered to take place in a series of columns, packed with anion exchange resin, as shown in Fig. 1. A uranium adsorption band is realized in the upper part of the resin bed of the first column and then it is eluted down with a reducing agent.

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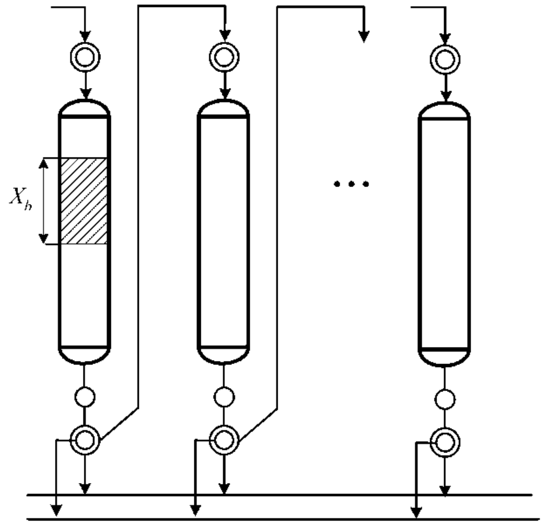

Figure 1. Schematic diagram of the process.

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The isotope exchange reaction (1) occurs repeatedly between adsorbed U(VI) ions and solute U(IV), as the uranium band descends through the column. The concentration of ^{235}U increases near the rear boundary of the adsorption band and decreases gradually, towards the front edge of the band.

MATHEMATICAL MODEL

A mathematical model of the process, proposed by Aoki et al. (4), consists of a system of two nonlinear partial differential equations. It describes the variations of ^{235}U mole fraction in solution and resin phases, due to the movement of the eluent, the internal concentration gradient, and the exchange reaction, and is given by:

$$\begin{aligned}\frac{\partial N_S(x,t)}{\partial t} = {} & -V_S\frac{\partial N_S(x,t)}{\partial x} + D\frac{\partial^2 N_S(x,t)}{\partial t^2} - \\ & -(1-\gamma)k'(N_S(x,t) - N_R(x,t) \\ & + \varepsilon N_S(x,t)(1 - N_R(x,t)))\end{aligned} \tag{2}$$

$$\begin{aligned}\frac{\partial N_R(x,t)}{\partial t} = {} & \gamma k'(N_S(x,t) - N_R(x,t) \\ & + \varepsilon N_S(x,t)(1 - N_R(x,t)))\end{aligned} \tag{3}$$

where:

t – the time, [h],

x – the distance measured downwards from the rear edge of the band, [cm]

$N_S(x,t)$ and $N_R(x,t)$ - the ^{235}U mole fractions in solution and resin phase respectively,[-]

V_S - the solution velocity, [cm/sec]

D - the diffusion coefficient of ^{235}U in solution, [cm^2/h]

ε - the enrichment coefficient, [-]

γ - the ratio between the amount of uranium in solution and adsorbed on resin. If the band velocity is denoted V_b, then γ can be written as (4):

$$\gamma = \frac{V_b}{V_S} \tag{4}$$

k'- a coefficient (4) given by the relation:

$$k' = \frac{k_b}{f(1-f)} C_T \tag{5}$$

where k_b is the backward reaction rate constant (1), f is the column void fraction, and C_T is defined as the 'overall' concentration of uranium in solution and resin (4).

From the assumption that the uranium isotopes are confined in the band and diffusion occurs only inside the adsorption band, the imposed boundary conditions are Danckwerts-type (10).

At the rear edge, where $x = x_R(t)$, the boundary condition is written as:

$$D\frac{\partial N_S(x,t)}{\partial x} = V_S(N_S(x,t) - N^*) \tag{6}$$

where N^* is the mole fraction of ^{235}U in the entering stream.

At the front edge, where $x = x_F(t)$, the following relation applies:

$$\frac{\partial N_S(x,t)}{\partial x} = 0 \tag{7}$$

NUMERICAL PROCEDURE

The equations (2) and (3) are valid only in the uranium adsorption band. Since the band moves, the spatial domain also moves with constant velocity V_b. Previous studies (4, 6) used an explicit finite differences (Total Variation Diminishing) scheme on a rectangular mesh, that proceeds one mesh size (Δx) at each time step (Δt) along with the band, where $\Delta x = V_b \Delta t$.

The numerical method used in this paper is based on a coordinate transformation. The variable x, measured from the rear edge of the uranium adsorption band, is time dependent, as shown in Fig. 2a.

The physical variable $x(t)$, can take values between two moving limits $x_R(t)$ and $x_F(t)$, where the subscripts R and F stand for rear (R) and front (F) edges of the band, as described by the relation:

$$x_R(t) \le x(t) \le x_F(t) \tag{8}$$

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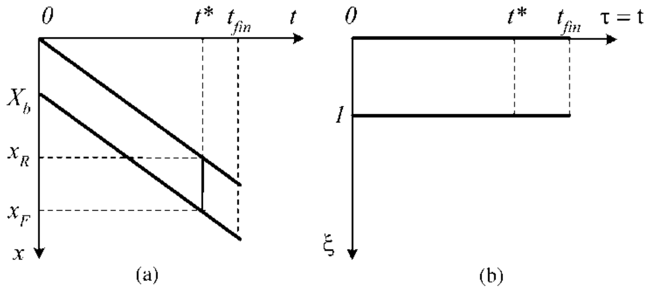

(a) (b)

Figure 2. Transformation of coordinate system.

From Fig. 2a, the band edges vary in time according to the relations:

$$x_R(t) = V_b t, \quad x_F(t) = X_b + V_b t \tag{9}$$

where X_b is the band length, [cm].

A coordinate transformation $(x, t) \rightarrow (\xi, \tau)$, with $\tau = t$, is introduced in order to map the space-time physical domain into a rectangular one, as shown in Fig. 2b. In the new set of variables x was replaced with the computational variable $\xi \in [0,1]$, according to the relation:

$$\xi = \frac{x - x_F(t)}{x_R(t) - x_F(t)} \tag{10}$$

Relations (9) can be introduced into (10) and the new variable ξ is calculated as:

$$\xi = \frac{x - V_b t}{X_b + V_b t - V_b t} = \frac{x - V_b t}{X_b} \tag{11}$$

The inverse transformation $x \rightarrow \xi$ is then given by:

$$x = \xi X_b + V_b \tau \tag{12}$$

In the new coordinate system the partial derivatives from (2) and (3) are calculated using the chain rule of differentiation and the new system of partial differential equations (written in variables ξ and t, since $t = \tau$) is:

$$\begin{aligned}\frac{\partial N_S(\xi, t)}{\partial t} = {} & \frac{V_b - V_S}{X_b} \cdot \frac{\partial N_S(\xi, t)}{\partial \xi} + \frac{D}{X_b^2} \frac{\partial^2 N_S(\xi, t)}{\partial t^2} \\ & - (1 - \gamma) k'(N_S(\xi, t) - N_R(\xi, t) \\ & + \varepsilon N_S(\xi, t)(1 - N_R(\xi, t)))\end{aligned} \tag{13}$$

$$\begin{aligned}\frac{\partial N_R(\xi, t)}{\partial t} = {} & \frac{V_b}{X_b} \cdot \frac{\partial N_R(\xi, t)}{\partial \xi} + \gamma k'(N_S(\xi, t) \\ & - N_R(\xi, t) + \varepsilon N_S(\xi, t)(1 - N_R(\xi, t)))\end{aligned} \tag{14}$$

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The rear edge boundary condition is transformed in the same manner and re-written as:

$$\frac{D}{X_b}\frac{\partial N_S(\xi, t)}{\partial \xi} = V_S(N_S(\xi, t) - N^*), \quad \text{for} \quad \xi = 1 \tag{15}$$

For the front edge of the band the boundary condition is:

$$\frac{\partial N_S(\xi, t)}{\partial \xi} = 0, \quad \text{for} \quad \xi = 0 \tag{16}$$

The initial conditions are set equal to the mole fraction of natural isotope abundance uranium, $N_0 = 0.007203$.

The system of partial differential equations (13, 14) was solved using the numerical method of lines (11). All spatial derivatives, represented by differentiation operators with respect to the computational variable ξ, were replaced by finite difference approximations and the resulting system of ordinary differential equations (ODE) was solved. Due to the stiff character of the system, the ODE solver used was a variable order one, based on the numerical differentiation formulas.

Given the constants: V_b, V_S, X_b, k', D, ε, and the initial condition N_0, the equations (13, 14) can be solved and the values of the ^{235}U mole fraction in solution and resin, along the band, can be determined at any time. The simulation study may be used to obtain the time for which the system reaches the steady state. A variation of the above mentioned constants, followed by a numerical simulation, could give information about the concentration profile along the band at any time.

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SIMULATION RESULTS

In order to determine the transient behavior of the system and the time necessary to obtain a desired enrichment, the system of nonlinear partial differential equations was solved using the above mentioned procedure. The numerical calculations were performed until a required ^{235}U mole fraction in solution was obtained. When the values of the parameters occurring in the model are the ones presented in Table 1, the profile of ^{235}U mole fraction in solution resulted as showed in Fig. 3. The time when the simulation stopped, i.e. the ^{235}U mole fraction reached the required value, was determined to be 28 days.

The solution velocity (V_S) can be calculated for a real-life operating process as the ratio between eluent flow-rate and the column cross-sectional area. The parameter k' was determined in previous studies (4, 5) by fitting experimental data with the numerical solution of the mathematical model. In this theoretical study, k' was considered to be of the same order as the ratio $V_S/HETP$ (4).

Table 1. Simulation data

Band velocity, V_b	100 [cm/h]
Solution velocity, V_S	333.3 [cm/h]
Uranium diffusion coefficient in solution phase (12)	$5 \times 10^{-6} \times 3600$ [cm^2/h]
Height equivalent to a theoretical plate, HETP (13)	0.05 [cm]
Band length, X_b	300 [cm]
Enrichment coefficient, ε	0.0013 [-]
Mole fraction of natural abundance uranium, N_0	0.007203 [-]
Required ^{235}U mole fraction, N_L	0.03 [-]

The concentration profile was obtained as an S-shaped curve (Fig. 3), from which the maximum enrichment and the mole fraction of the depleted uranium, at the rear and the front edge respectively of the uranium adsorption band can be accurately determined.

SIMULATION OF THE CONTINUOUS OPERATION

In order to obtain data for the plant scale estimation, the simulation was performed also in the case of continuous operation, when waste and product are withdrawn from the front and respectively, the rear edge of the band,

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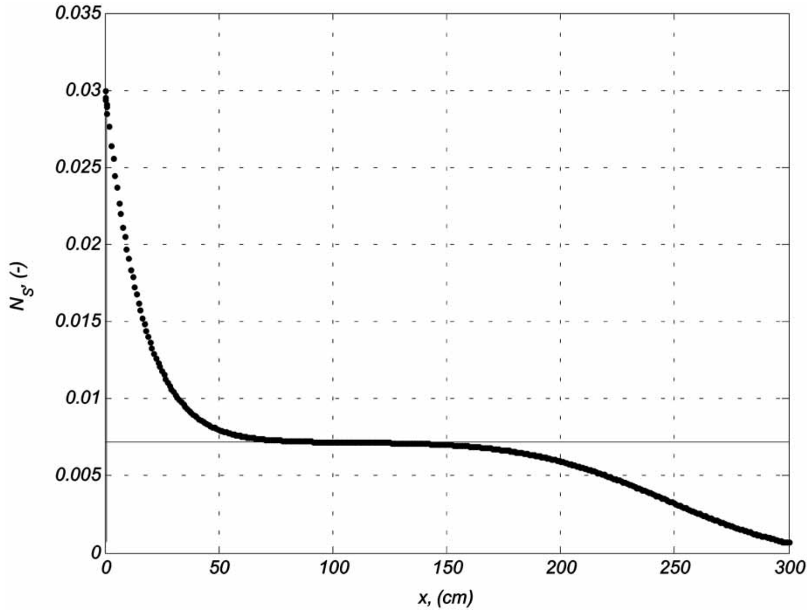

Figure 3. The ^{235}U mole fraction in solution along the band after 28 days of operation.

and natural abundance uranium is supplied as feed. This multiple-stage process proceeds as follows:

- In the first stage of the process the uranium band travels through a series of columns, the uranium isotope profile is gradually developed until it reaches the desired enrichment when the product and isotopic waste are collected from opposite edges of the band. Let t_{eq} be the time interval of this first operation.
- When ^{235}U depleted uranium is withdrawn at the front edge, the length of the band decreases with X_W [cm].
- Uranium solution of natural abundance is supplied as feed in the region of the band where natural isotope abundance uranium is detected. During this operation, the length of the band increases with X_F [cm].
- The product is extracted from the last X_P [cm] at the rear end of the band. In order to maintain a band of constant length (X_b), when X_W and X_P are fixed, the feed must be supplied so that $X_F = X_W + X_P$.
- A new stage of the continuous operation starts now, with another enrichment step, until the desired ^{235}U mole fraction is obtained at the rear edge of the band. The time necessary to complete this operation is denoted t_{stage}.

In this simulation study, we considered that waste is withdrawn from a distance $X_W = 5$ [cm] and product from $X_P = 1$ [cm]. The variation in time of ^{235}U mole fraction in solution, at the two edges of the band, is presented in Fig. 4.

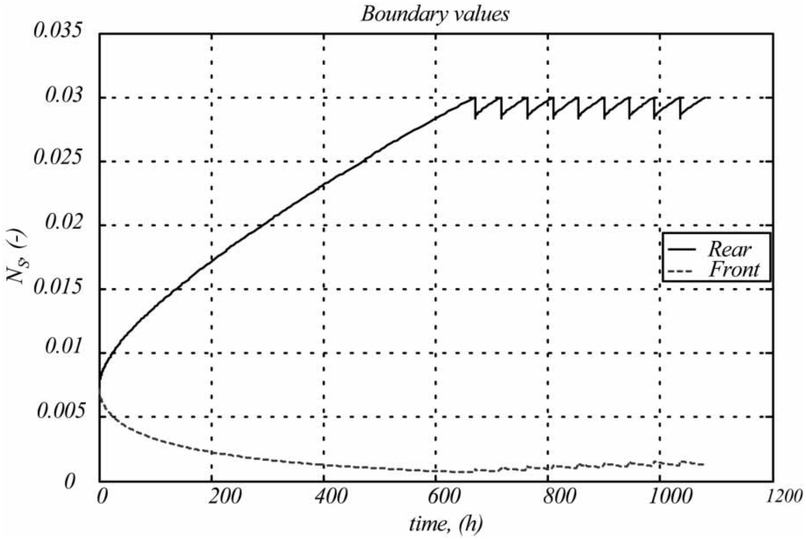

Figure 4. The time variation of ^{235}U mole fraction in solution at the rear and front edges of the band.

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According to the data given in Table 1, the time necessary for X_W [cm] band migration is $t_{waste} = X_W/V_b = 0.5$ [h], while the rear edge of the band is descending the same distance. The ^{235}U mole fraction was considered unchanged during the waste withdrawal, because the time needed for this operation (t_{waste}) is much smaller than the time of a significant enrichment in the rear edge of the band. The time necessary for the product withdrawal, equal to $X_P/V_b = 0.01$ [h], was small enough so as to neglect, during this period, any depletion at the front edge of the band.

The same assumption was made for the case of feed supply, when the rear edge of the band is at rest, but the front edge proceeds down the column $(X_W + X_P)$ cm.

The simulation was also carried out in the next stage of enrichment until the ^{235}U mole fraction in solution became equal to the desired value $N_L = 0.03$. The time of this operation is equal to 45 hours and the mole fraction of the product varied between 0.029 and 0.03.

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PRODUCTION

In order to estimate the amount of ^{235}U with the mole fraction of 0.03, which can be produced in a year, the column and resin characteristics have to be set. The size of one column, the adsorption capacity and the density of three anion-exchange resins, determined in a previous study (14), for the temperature of 70°C, are presented in Table 2.

During the operation of product withdrawal, the uranium contained in a volume V_r of resin is eluted from the rear edge of the band. The mass of that resin, denoted m_r, can be calculated from:

$$m_r = V_r \cdot \rho = \frac{\pi \cdot d^2}{4} \cdot X_P \cdot f \cdot \rho \tag{17}$$

Table 2. Column size and the characteristics of anion-exchange resins (14)

Column	Column length, L	300 [cm]
	Column diameter, d	100 [cm]
	Void fraction, f	0.6 [-]
Resin		
Dowex Marathon	Adsorption capacity, q	238.63 [mg U/g resin]
	Density, ρ	1.08 [g / cm^3]
Dowex 1-X8	Adsorption capacity, q	235.24 [mg U/g resin]
	Density, ρ	1.04 [g / cm^3]
CEPU - M5	Adsorption capacity, q	182.47 [mg U/g resin]
	Density, ρ	1.173 [g / cm^3]

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and then, the amount of uranium (m_U), eluted from the resin (m_r), having the ^{235}U mole fraction equal to 0.03, result as:

$$m_U = m_r \cdot q \tag{18}$$

This amount of enriched uranium can be extracted only after a time t_{stage}, which can be calculated by means of a numerical simulation, according to the procedure described before. Taking into account the time interval of the first stage of enrichment t_{eq}, the production of enriched uranium in one year, P [kg/year] is calculated as:

$$P = (365 \cdot 24 - t_{eq}) \cdot \frac{1}{t_{stage}} \cdot m_U \tag{19}$$

The migration distance of every new enrichment stage, L_{stage}, can be calculated from:

$$L_{stage} = V_b \cdot t_{stage} \tag{20}$$

The results of the study of three cases are given in Table 3. The only variation of the theoretical data is the length of the rear edge of the column, from which the product is withdrawn. Because it is desired to extract uranium solution having the ^{235}U mole fraction as close as possible to 0.03 in this case, small distances X_P are considered: 1 cm, 0,5 cm and 0,3 cm. The values of the model parameters are the ones presented in Table 1, the column and resin characteristics are given in Table 2.

As expected, the numerical simulation indicated that for smaller distances X_P, the time interval (t_{stage}) necessary for the ^{235}U mole fraction to reach again 0.03, is smaller.

Table 3. Calculated values of uranium (3% at ^{235}U) production per one year

No.	Theoretical data and time values resulted from simulation	Resin type	Amount of uranium withdrawn in one stage, m_U, [kg]	Production, P, [kg/year]	Migration distance of one stage, L_{stage}, [cm]
1	X_P=1 [cm]	Dowex Marathon	1.21	218.28	4500
	$t_{eq} = 28 \times 24$ [h]	Dowex 1-X8	1.15	207.21	
	$t_{stage} = 45$ [h]	CEPU - M5	1.008	181.28	
2	$X_P = 0.5$ [cm]	Dowex Marathon	0.60	327.42	1500
	$t_{eq} = 28 \times 24$ [h]	Dowex 1-X8	0.57	310.81	
	$t_{stage} = 15$ [h]	CEPU - M5	0.50	271.92	
3	$X_P = 0.3$ [cm]	Dowex Marathon	0.36	368.35	800
	$t_{eq} = 28 \times 24$ [h]	Dowex 1-X8	0.34	349.66	
	t_{stage}=8 [h]	CEPU - M5	0.30	305.91	

Although the amount of uranium solution, withdrawn in one stage, decreases as the distance X_P decreases, the yearly production is larger because the product withdrawal is performed more frequently.

If the product is extracted from the last 0.5 cm of the band (case 2, Table 3) instead of the last 1 cm (case 1), the yearly production increases with about 50%, for all resin types. If X_P is 0.3 cm, the production increases with 70% by comparison with case 1.

Separation Plant Configuration

Using the previously obtained results, we are able to propose an appropriate configuration of the separation plant. It is desired to withdraw the product at the bottom of one column and to design the plant configuration so that the amount of resin used is as small as possible.

Taking the size of one column: 1 m diameter and 3 m length, the band length $X_b = 3$ m, and the band velocity, $V_b = 100$ cm/h = 24 m/day in all three cases presented in Table 3, the results of the calculation are presented in Table 4.

In case 1, a plant consisting of a series of three identical columns is proposed. During the first enrichment stage, which lasts for $t_{eq} = 28$ days, the band has to cover continuously the series of columns, travelling with 24 m/day. If the total distance which has to be covered in one day is 9 m, the frequency of resin bed use per day is 24/9 = 2,66. The total distance to be covered for the whole first stage is equal to $V_b t_{eq} = 24 \times 28$ m = 672 m. If the band travels in the series of three columns for 75 times, it will migrate on a distance of 75×9 m = 675 m which is very close to the necessary distance of 672 m. Moreover, the necessary migration distance at

Table 4. Plant configuration

No.	Theoretical data and time values resulted from simulation	Migration distance of one stage, L_{stage}, [m]	Number of columns	Frequency of resin use, [1/day]	Withdrawal point
1	$X_P = 1$ [cm] $t_{eq} = 28$[days] $t_{stage} = 45$ [h]	45	3	2.66	Bottom edge of the last column
2	$X_P = 0{,}5$ [cm] $t_{eq} = 28$ [days] $t_{stage} = 15$ [h]	15	3	2.66	Bottom edge of the each column
3	$X_P = 0{,}3$ [cm] $t_{eq} = 28$ [days] $t_{stage}=8$ [h]	8	4	2	Bottom edge of the last column

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each of the next stages is 45 m, i.e. all the three columns have to be covered for 5 times. In this case it is convenient to consider as product withdrawal point the bottom of the last column.

The same configuration may be designed for case 2, with the only difference that starting from the second stage of enrichment, the product has to be withdrawn after every 15 m of migration, or 5 column lengths covered. In this case, withdrawal has to be possible at the bottom edge of every column.

In case 3, a four-column plant might be appropriate. The frequency of resin bed use per day is $24/12 = 2$. The total migration distance of 672 m can be covered if the band travels 56 times through the columns. Only one withdrawal point would be necessary for continuous operation, located at the bottom of the last column.

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CONCLUSION

Although there are several studies available which deal with the calculation of the parameters to be used in the design of an enrichment plant, none of them give a complete description of how to determine all the data necessary. There is a large number of degrees of freedom in choosing the parameters of a plant. Many of them cannot be calculated, but they can be determined either from the application of the experimental technique, or from numerical simulation and numerical parameter identification, in case there are at least a few experimental data available.

This study combines numerical simulation of the isotope separation process with a set of *a priori* established data in order to estimate the plant scale for a desired enrichment.

For a given mole fraction of ^{235}U, band velocity and eluent flow-rate, the time necessary to obtain the desired concentration profile and the time of every new enrichment stage were calculated.

The amount of uranium 3 at.% ^{235}U produced in a year was evaluated in different cases of resin type and product withdrawal conditions.

The plant scale was calculated for the same case and an appropriate configuration was proposed.

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