



MODELING AND SIMULATION ISOTOPIC SEPARATION CASCADE FOR ^{15}N

Mihaela Ligia UNGUREȘAN^a, L. JÄNTSCHI^a

^a Technical University of Cluj-Napoca, Faculty of Science and Engineering of Materials, Chemistry Department, Muncii Street, 101-103, Cluj, 3400, ROMANIA

SUMMARY

To continue the papers [1- 5], here are presented some introductory aspects for modeling and simulation of a separation cascade for ^{15}N . We operate with second-order partial differential equation using a local-iterative linearization procedure (LLI). [6, 7]

The first stage presents the parameters of experimental separation cascade for ^{15}N . In the next stage, we present the modeling and simulation of the first and second separation columns, based on graphical interpretation.

Keywords: modeling and simulation, separation column, partial differential equation

INTRODUCTION

A separation method for ^{15}N is the isotopic exchange $^{14}\text{N}/^{15}\text{N}$ on a cascade with nitric acid 10M and nitric oxide circulating in countercurrent (Figure 1).

The general equations that describe the isotopic exchange in countercurrent, based on isotopic balance for the gas and liquid phase are [3]:

$$\begin{cases} h \cdot \frac{\partial n}{\partial t} - l \cdot \frac{\partial n}{\partial z} = -T \\ H \cdot \frac{\partial N}{\partial t} - L \cdot \frac{\partial N}{\partial z} = +T \end{cases} \quad (1)$$

where: z – the height at which takes place the isotopic exchange (m); N , (n) – the isotopic concentration for the isotope in study, considered in liquid (gas) phase (at%); L , (l) – the liquid (gas) flow expressed in number of atoms per time units (at N/sec.); H , (h) – the liquid (gas) phase retention on the separation column, expressed in number of atoms per volume unit (at N/cm³); T – the transport rate of the isotopic species (at N/cm³).

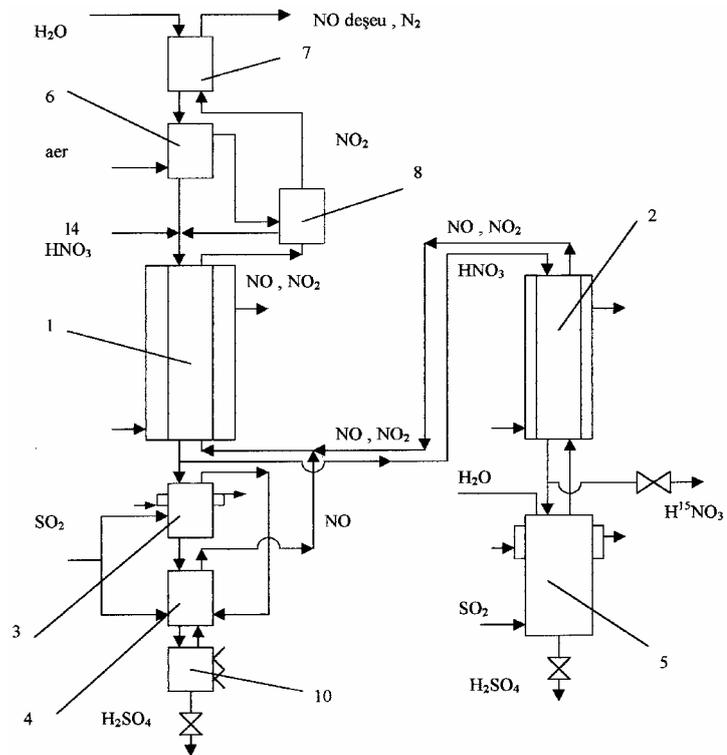


Figure 1. The installation for isotopic separation ¹⁵N through isotopic exchange NO, NO₂ – HNO₃

The final equation is:

$$(H+h) \cdot \frac{\partial N}{\partial t} = \frac{L \cdot l}{K} \cdot \frac{\partial^2 N}{\partial z^2} - \frac{\partial}{\partial z} [L \cdot (\alpha - 1) \cdot N \cdot (1 - N) + P \cdot N] \quad (2)$$

where: $P = L - 1$, represents the production of the isotopic separation column (at N/sec).

The hypotheses imposed retentions in liquid and gas phase are constant on the whole column's length; L, l – the liquid and gas flows are constant in every column's point; α – the elementary separation factor is constant; K – the transport constant is equal on the whole column's length.

The analogical model (2) of a separation column for ¹⁵N can be written as:

$$(P_{100} + P_{101} \cdot y) \cdot \frac{\partial y}{\partial a} + P_{01} \cdot \frac{\partial y}{\partial b} + P_{20} \cdot \frac{\partial^2 y}{\partial a^2} = 0 \quad (3)$$

where: $a = z$ – the unit's height (m); $b = t$ – time (h); $y = N$ – the ¹⁵N concentration (%); and: $P_{100} = L \cdot (\alpha - 1) + P$; $P_{101} = 2 \cdot L \cdot (1 - \alpha) + P$;

$$P_{01} = H + h; P_{20} = -\frac{L \cdot l}{K} = -\frac{L^2}{K} \quad (\text{for } P = 0) \quad (4)$$

MATERIALS AND METHODS

Separation is realized with the help of two columns tight up in series, where the first column makes the separation until 8.3% ¹⁵N, and the second one until 99.9 %. The first column is characterized by the following parameters:

$\alpha = 1.055$; $H_1 = 430$ at. N/m³; $h_1 = 2.8$ at. N/m³; $l_1 = L_1 = 1.6 \cdot 10^{-2}$ at. N/s = 1382.4 at. N/day; $K = 4.7 \cdot 10^{-4}$ at. N/s·m³ = 4080.8 at N/day·m³. The height of the column is 7 meters, and the separation reaches the stationary stage after a period of 14 days.

If we calculate the concentration at the end of the column $z_f = 7$ m in different moments we obtain the following experimental data:

Table I. Experimental data, first column

t (days)	0	1	2	3	4	5	6
N (%)	0.365	1.4	2.3	3.3	4.1	4.9	5.5
T (days)	7	8	9	12	14	15	16
N (%)	6.1	6.5	6.8	8.0	8.2	8.25	8.3

that lead us to the experimental graphic in Figure 2.

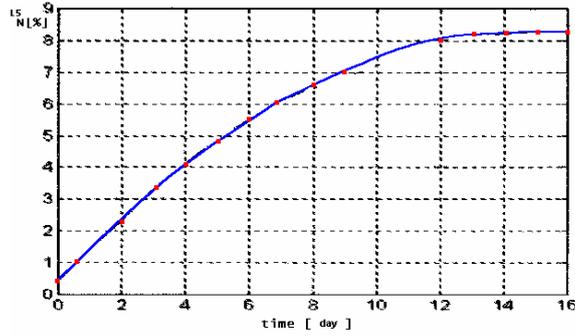


Figure 2. The variation of the isotopic concentration as a function of time obtained from experiments in the first separation column

The second column is characterized by the following parameters:

$\alpha = 1.055$; $H_2 = 0.6$ at. N/m^3 ; $h_2 = 4 \cdot 10^{-4}$ at. N/m^3 ; $l_2 = L_2 = 5.3 \cdot 10^{-4}$ at. $N/s = 46.08$ at. N/day ; $K = 4.7 \cdot 10^{-4}$ at. $N/s \cdot m^3 = 4080.8$ at $N/day \cdot m^3$;

The height of the column is 7 meters, and the separation reaches the stationary stage in a period of 16 days.

If we measure the concentration at the end of the column $z_f = 7$ m at different periods of time we obtain the following experimental data if the second column is being fed with the product of the first column in equilibrium (8,3% ¹⁵N):

Table II. Experimental data, second column

t (days)	0	1	2	3	4	5	6
N (%)	8,3	9	13	19	27	35	42
T (days)	7	8	9	12	14	15	16
N (%)	50	62	70	90	96	98	99

that lead us to the experimental graphic in Figure 3.

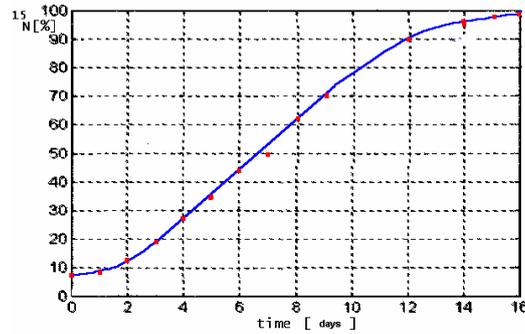


Figure 3: The variation of the isotopic concentration as a function of time obtained from experiments in the second separation column

RESULTS AND DISCUSSION

Because the nonlinear differential equation that describes a column of isotopic separation has infinity of solutions, we chose for modeling a function that corresponds to the experimental conditions and which is its solution.

Because the analytical solution cannot be calculated we chose the following expression:

$$N_1 = C_0 + (C_1 \cdot e^{\alpha_1 t} + C_2 \cdot e^{\alpha_2 t} + C_3 \cdot e^{\alpha_3 t} + C_4 \cdot e^{\alpha_4 t}) \cdot z \quad (4)$$

where: N_1 – the concentration of the isotope ^{15}N (%); z – the height in the column (m); t – the time (days).

We pick: $\alpha_1 = -0.01$; $\alpha_2 = -0.02$; $\alpha_3 = -0.01$; $\alpha_4 = -0.02$.

The coefficients C_0, \dots, C_4 are calculated from the following conditions:

- For $z = 0$, at the entrance of the separation column, the concentration of the isotope ^{15}N is constant 0.365 %. That means that $C_0 = 0.365$;
- At the beginning of the separation process, $t = 0$ days, at the end of the column, $z = 7$ m, the concentration is 0.365 %;
- At the end of the separation process, $t = 14$ days, at the end of the column, $z = 7$ m, the concentration is 8.3 %;
- Concentration is stable after 14 days with a slope of 0.1;

e) The concentration's variation depending on the height in a certain moment presents a point of inflexion at the middle of the column $z = 3.5$ m:

$$\left. \frac{\partial N}{\partial t} \right|_{t=14, z=7} = 0.1, \quad \left. \frac{\partial^2 N}{\partial t^2} \right|_{z=3.5} = 0 \quad (5)$$

From the conditions presented above we have:

$$C_0 = 0.365; C_1 = 90.513; C_2 = -53.002; C_3 = -79.124; C_4 = 41.776. \quad (6)$$

For the second column, fed with the product obtained from the first column, the concentration is:

$$N_2 = N_1 + (C_1 \cdot e^{\alpha_1 t} + C_2 \cdot e^{\alpha_2 t} + C_3 \cdot e^{\alpha_3 z} + C_4 \cdot e^{\alpha_4 z} + C_5 \cdot e^{\alpha_5 z}) \cdot z \quad (7)$$

where: N_1 – is the exit from the first column $N(t, z = 7)$; $N_1 = -262.209 \cdot 633.592 \cdot e^{(-0.1 \cdot t)} - 371.017 \cdot e^{(-0.2 \cdot t)}$; z – the height in the column (m); t – the time (days);

We chose: $\alpha_1 = -0.01$; $\alpha_2 = -0.02$; $\alpha_3 = -0.04$; $\alpha_4 = -0.08$; $\alpha_5 = -0.16$;

The coefficients C_1, \dots, C_5 are obtain from the following conditions:

- a) At the end of the column, after 16 days, we have an isotopic concentration of 99%;
- b) At the beginning of the separation process, $t = 0$, in the entire column and the concentration is 8.3%;
- c) From the experimental results obtain on concentration, the variations depending on time at the end of the column at $t = 16$ days, are $(99-98)/1 = 1$;
- d) For $t = 0$ we have a variation of 1.635;
- e) The experimental graphic of the concentration depending on time presents an inflexion at $t = 7$ days (measured at $z = 7$):

$$\left. \frac{\partial N_2}{\partial t} \right|_{t=16, z=7} = 1, \quad \left. \frac{\partial N_2}{\partial t} \right|_{t=0, z=7} = 1.635, \quad \left. \frac{\partial N_2}{\partial t} \right|_{t=7, z=7} = 0 \quad (8)$$

From the conditions presented above we have:

$$C_1 = -2267.939; C_2 = 4528.087; C_3 = -2894.313; C_4 = -670.687; C_5 = -36.521 \quad (9)$$

Using programs of numerical modeling and simulation by the method LLI realized in MATHLab for the integration as function of time and the height in the column we have obtained the simulation in Figure 4 and 5.

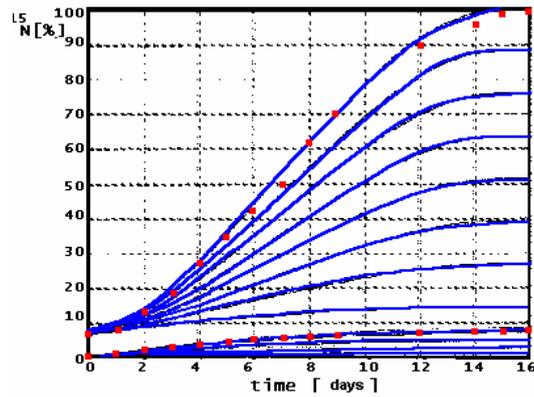


Figure 4. The concentration of ^{15}N as a function of time for different height in the two columns (0, 1, ..., 7m)

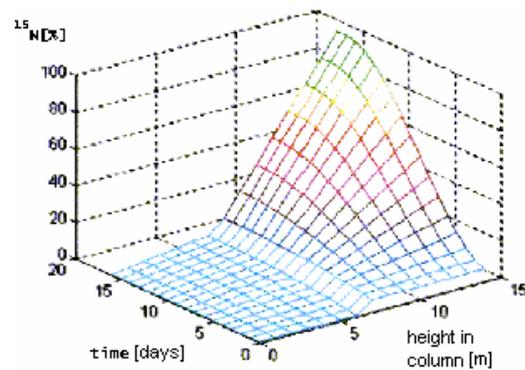


Figure 5. The concentration of ^{15}N as a function of time ($t = 0, \dots, 16$ days) and the height in the two columns ($z = 0, 1, \dots, 7\text{m}; z = 7, \dots, 14\text{m}$)

CONCLUSION

The chosen variant of approximation satisfies well enough the differential equation that modeled mathematically the two columns of the separation cascade and corresponds to the experimental values that were measured (the curves with experimental points) (•).[8]

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