Optimization of nitrogen-15 production by isotopic exchange in nitrox system

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Abstract

The main aim of this study was to optimize the operating parameters for ^{15}N producing plant, used at the National Institute for Research and Development of Isotopic and Molecular Technologies from Cluj-Napoca, for high-level enrichment of nitrogen -15 by isotopic exchange reactions between nitrogen oxides (NO, NO_2) and aqueous solution of nitric acid (HNO_3). The optimization was based on the theory of ideal cascades applied for square cascades. The theoretical results were compared with experimental data obtained in total reflux conditions, using the final column of the ^{15}N producing plant. Good agreement was obtained between the predicted optimized operating conditions and the experimental data results.

Theoretical background

Steady state of countercurrent column

The fundamental equation of isotope separation[1]:

$$PN_P = PN + \varepsilon LN(1-N) - \frac{L}{2}\frac{dN}{ds}, \qquad (1)$$

which gives the following integral:

$$\int_0^S ds = \int_{N_0}^{N_s} \frac{LdN}{2(-\varepsilon LN^2 + (\varepsilon L + P)N - PN_P)}, \qquad (2)$$

from which it follows:

$$s = \frac{1}{\varepsilon \boldsymbol{\Delta}(\psi)} \tanh^{-1}\left[\frac{(N_s - N_0)\boldsymbol{\Delta}\psi}{(N_s - 2N_sN_0 + N_0) - (N_s - N_0)\psi}\right], \qquad (3)$$

where

$$\boldsymbol{\Delta}(\psi) = [1 + 2\psi(1 - 2N_P) + \psi^2]^{1/2}, \qquad (4)$$

$$\psi = \frac{P}{\varepsilon L}, \Psi' = \frac{\Psi}{P}. \qquad (5)$$

When $N_s = N_P$ and $s = S$ equation (4) becomes:

$$S = \frac{1}{\varepsilon \boldsymbol{\Delta}(\psi)} \tanh^{-1}\left[\frac{(N_P - N_0)\boldsymbol{\Delta}\psi}{(N_P - 2N_PN_0 + N_0) - (N_P - N_0)\psi}\right]. \qquad (6)$$

The total flow of the square cascade may be determined from:

$$T_F = \sum_{s=0}^{S} L_s = SL_S. \qquad (7)$$

Symbols

P	Product flow [mol/s]
N	Mole fraction of the desired isotope [mol/mol]
ε	Enrichment factor [-]
L	Flow rate [mol/s]
s	Stage (theoretical plates) number [-]
N_P	Mole fraction of the desired isotope in product flow (S[th] stage) [mol/mol]
N_s	Mole fraction of the desired isotope fed into s[th] stage [mol/mol]
N_0	Mole fraction of the desired isotope feed flow [mol/mol]
Ψ	Normalized rate of production [-]

[1] Cohen, K.: The Theory of Isotope Separations, McGraw-Hill, New York, 1951

Experimental results

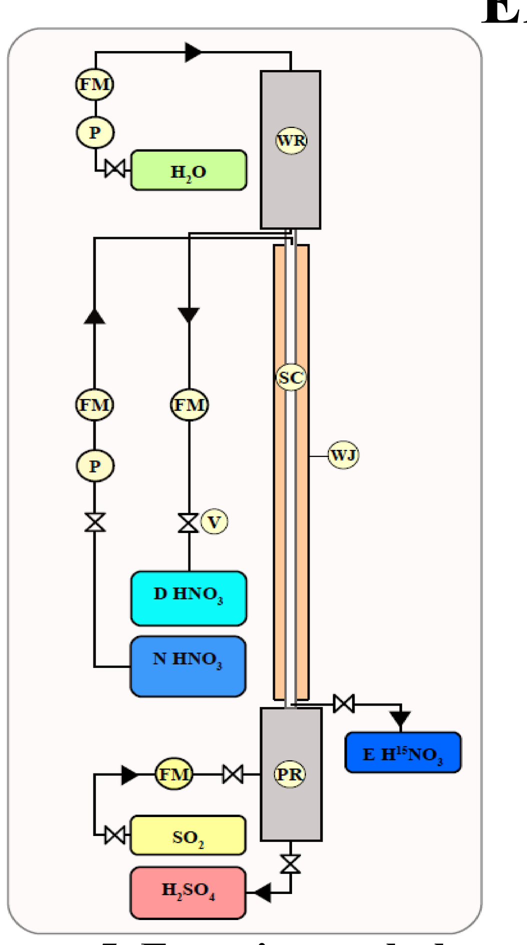
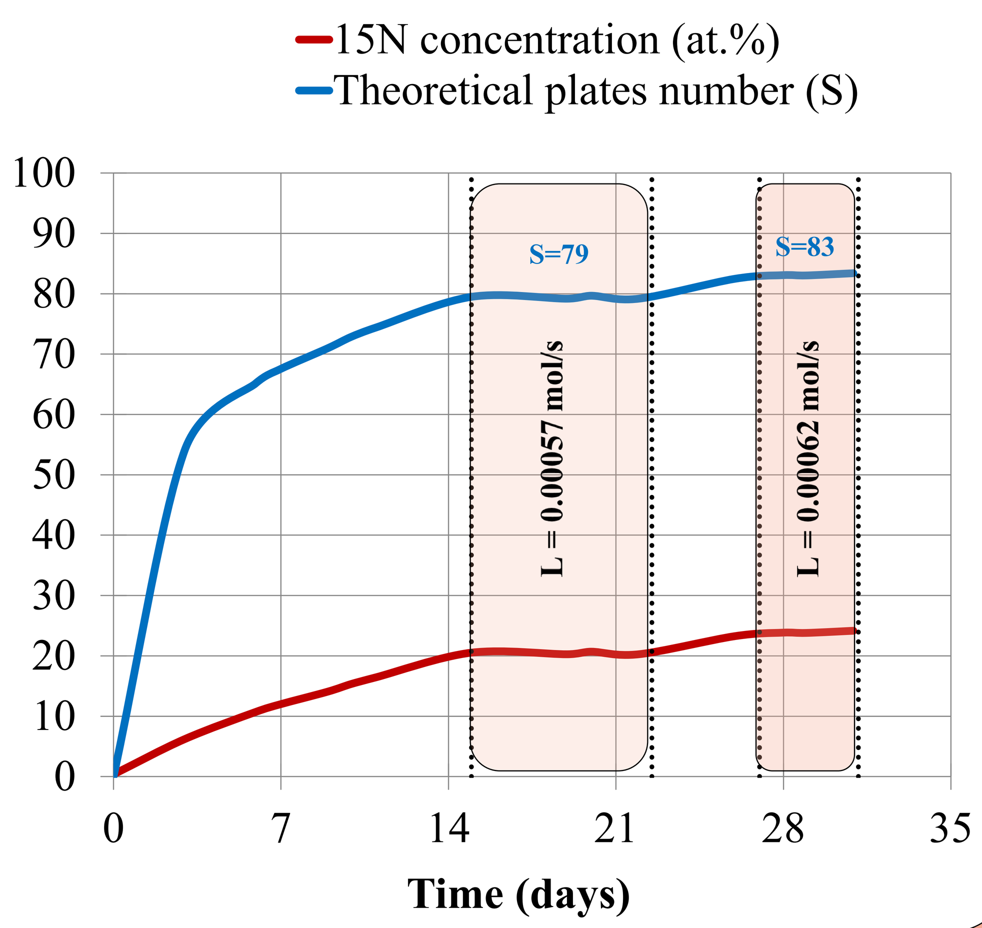


Figure 5. Experimental plant



Acknowledgments

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Theoretical results

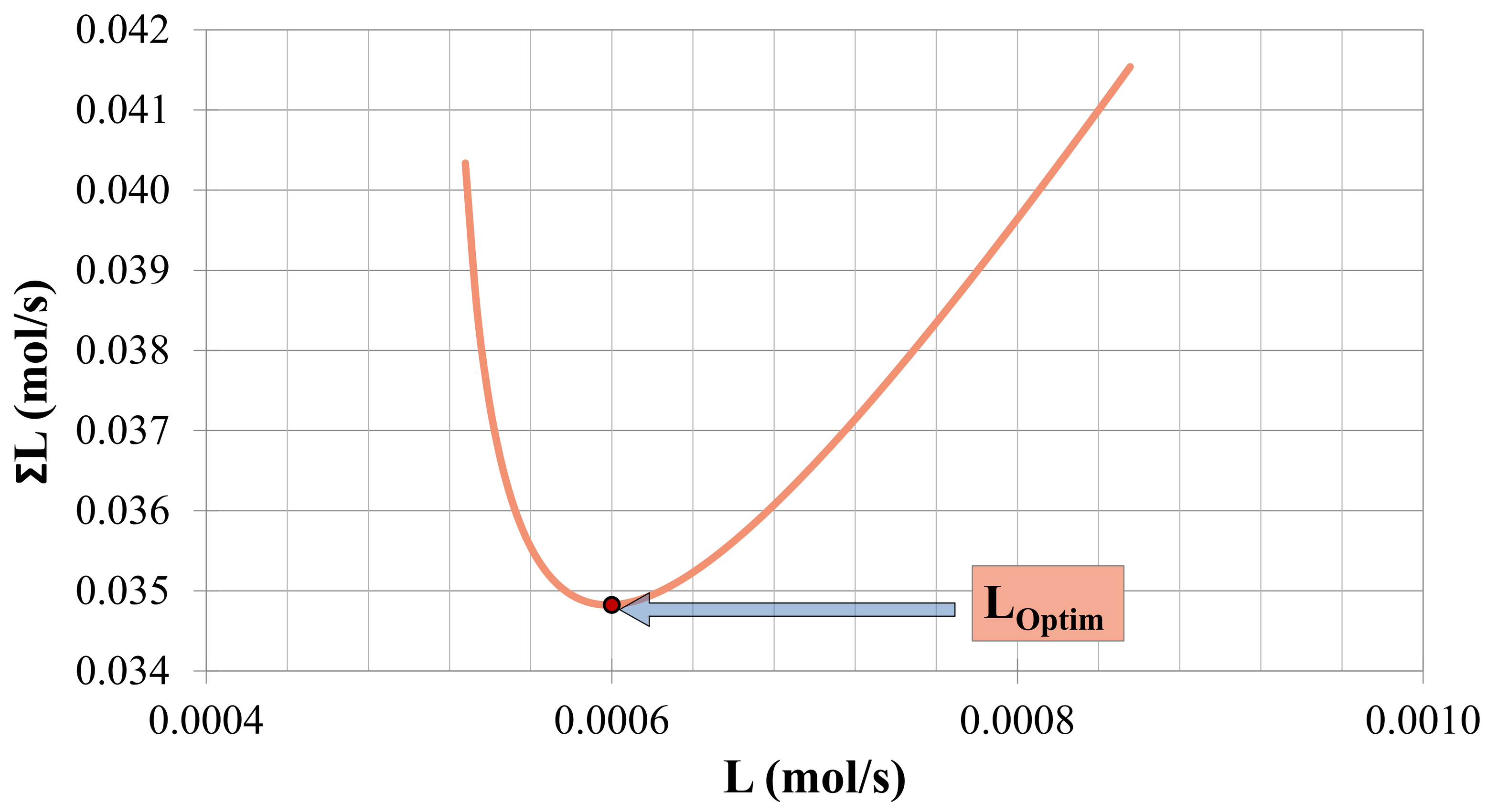

Figure 1. The variation of total flow of separating column vs. flow rate
(P=0.0384 mol^{15}N/day, N_0=0.00365, N_P=0.237)

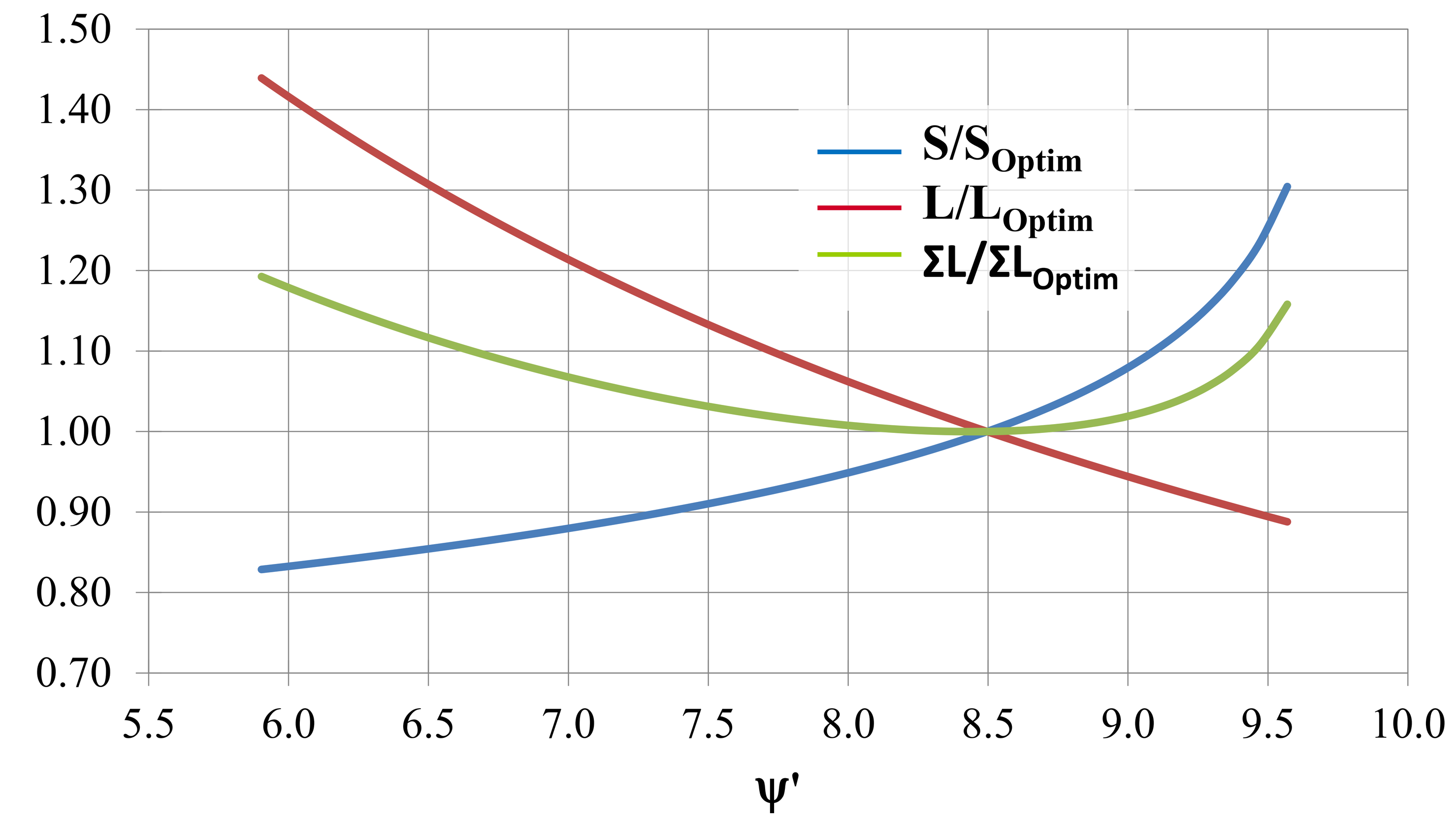

Figure 2. The variation S/S_{optim}, L/L_{optim} and $\Sigma L/\Sigma L_{Optim}$ vs. Ψ′
(P=0.0384 mol^{15}N/day, N_0=0.00365, N_P=0.237)

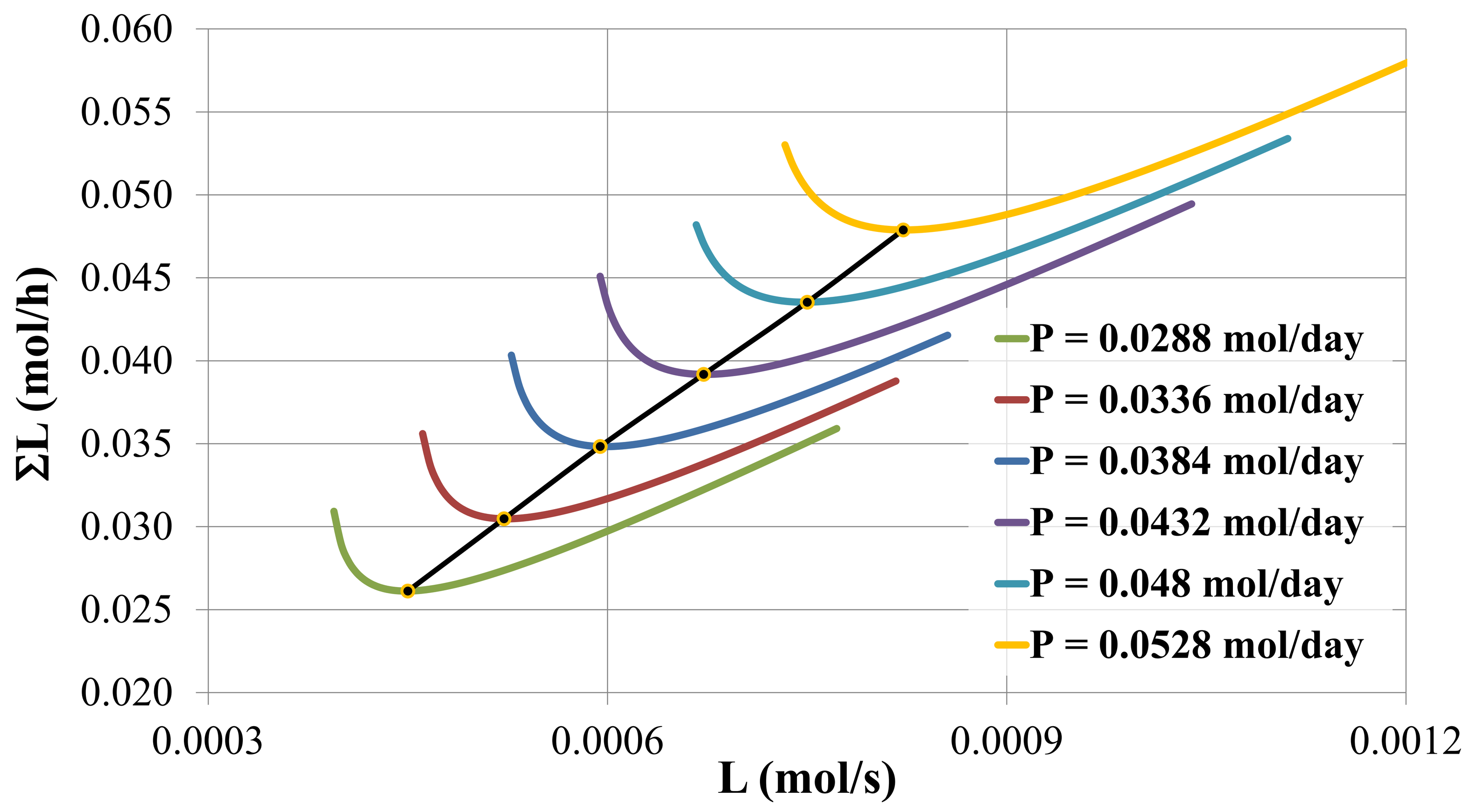

Figure 3. The variation of total flow of separating column vs. flow rate

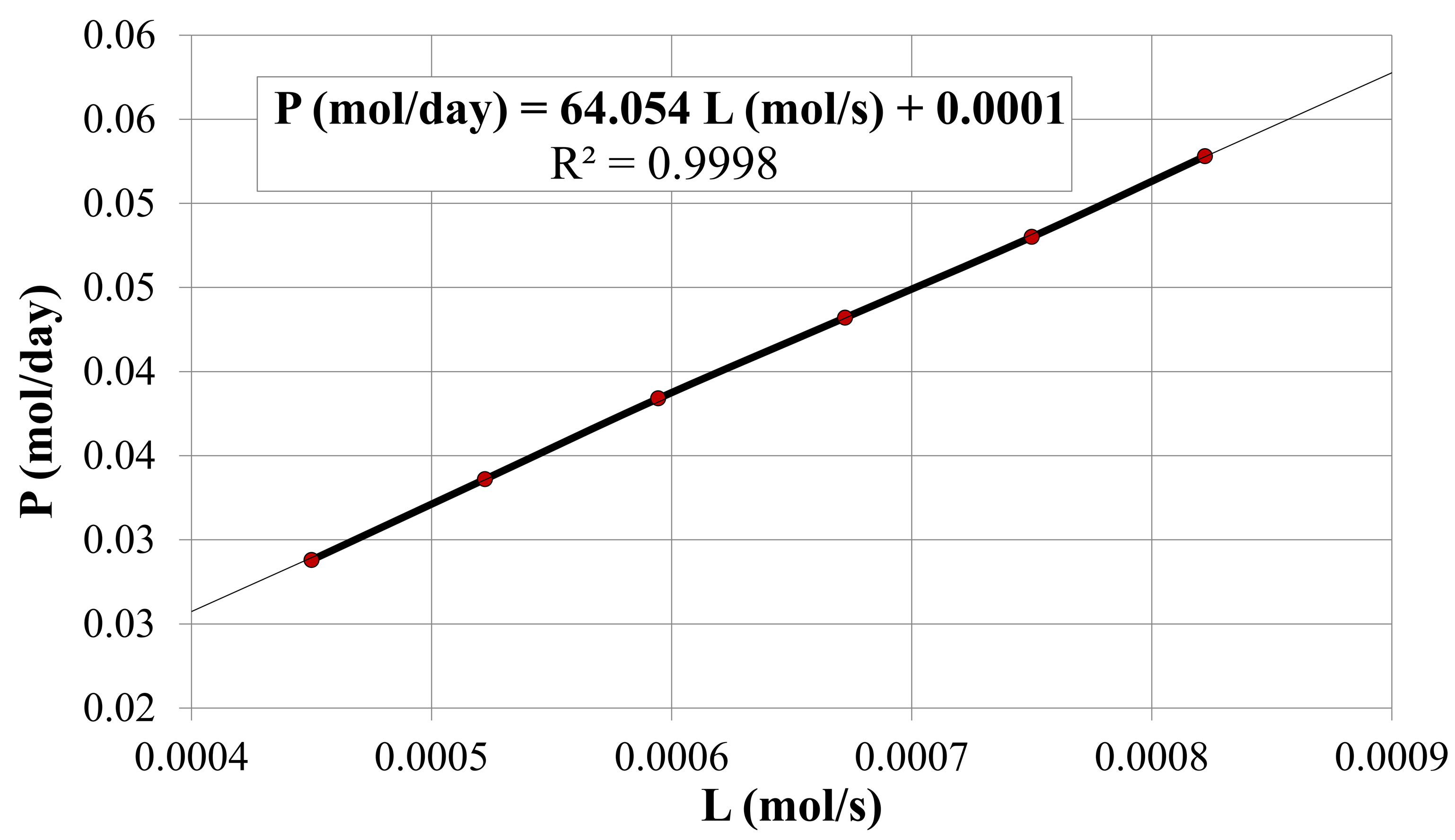

Figure 4. The variation of the product flows. flow rate