Optimal Control of the Carbon Isotopes Cryogenic Separation Process

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The most important demand in an efficient carbon isotope separation is to keep the cryogenic distillation column parameters at their required set points, despite disturbances or uncertainties. One of the key parameters is the column temperature that has a direct influence on the carbon separation coefficient. The nonlinear model of the separation process considers the top and bottom column temperature evolution and their influence on the isotope separation rate. The optimal controller designed in this paper effectively maintains the column temperature at constant levels, rejecting disturbances, for a significant range of model uncertainties.

Key words:
Cryogenic isotope separation, linear quadratic regulator, nonlinear model, uncertainties

Introduction

Carbon represents the fourth most abundant chemical element in the universe, having two stable and one radioactive isotope. Among the stable isotopes, the least abundant, ^{13}C, with a natural amount fraction of approximately 1.1 %, has the most uses in a wide variety of industrial fields and medicine. The major drawback with ^{13}C comes from the difficulty of obtaining it and raising its natural fraction. Carbon isotopes can nowadays be separated using many methods, one of them being the cryogenic distillation of carbon monoxide. Very few aspects regarding the construction and operating conditions of such cryogenic plants are known today, and even less information is available as far as the separation process modeling and control are concerned. However, the efficient control of the carbon monoxide distillation process represents a necessity for large-scale ^{13}C production.

Due to its resemblance to a classic distillation process, several models for carbon isotope separation have been proposed, some based on mass, component and energy balance equations,[1,2] some on the nonlinear wave theory[2] or the Cohen equations.[2,3] However, even though these models capture the essence of the isotope separation process, they can generally be used to determine in each moment and column-coordinate the concentration of the useful isotope, thus describing and modeling the isotope fraction distribution as a function of time and as a function of the column dimension,[2] but not for control purposes.

The purpose of the paper is to design a nonlinear model for ^{13}C cryogenic isotope separation, a model that is specific to the column built at the National Institute for Research and Development of Isotopic and Molecular Technologies Cluj-Napoca (NIRDIMT). The nonlinear model, designed for control purposes, would consist of differential equations describing the evolution of the column temperature and of the isotope transfer rate. The central idea is that, in order to achieve a certain isotope transfer rate, the isotope separation coefficient has to be kept constant. Such a requirement can be transposed in a control demand since the isotope separation coefficient depends directly on the column temperature and its evolution is proportional to the column temperature evolution as a second power function. Thus, by keeping the column temperature constant, the isotope separation process can be controlled efficiently by manipulating some input signals. The designed control strategy consists of a simple linear quadratic regulator[4,5] that can effectively reject temperature disturbances that might occur during plant operation. To test the robustness of the designed controller, simulations were performed considering different ranges for the nonlinear model parameters.

Cryogenic isotope separation column principle of operation

The ^{13}C isotope separation column built at the National Institute for Research and Development of Isotopic and Molecular Technologies Cluj-Napoca (NIRDIMT) consists of a distillation column; the schematic representation is given in Fig. 1. The main components are: distillation column (C), condenser (C1) at the top of the column cooled with liquid nitrogen, boiler (B) at the bottom of the column, and vacuum jacket (M) used for thermal isola-

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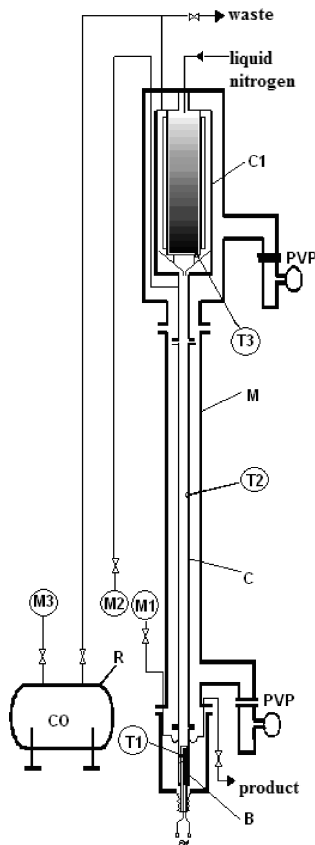e-mail: Cristina.Pop@aut.utcluj.ro

F i g . 1 − *Schematic representation of the* [13]*C cryogenic iso-
tope separation column*

tion, since the column operates at very low temperatures.

Thermal isolation is ensured by vacuum pumps (denoted PVP in Fig. 1), while carbon monoxide is fed as a gas through a feeding system, at an intermediary point in the column, kept initially in a special reservoir (R). Manometers (M1, M2, M3) and temperature transducers (T1, T2, T3) are installed at several points of the distillation column.

The carbon separation process is based on the distillation of carbon monoxide, which has a counter current flow through the column- in an ascending gaseous phase and a descending liquid form. The reflux at the bottom of the column is ensured by boiling the liquid carbon monoxide, while the reflux at the top of the column is ensured by condensing the carbon monoxide vapours. At the bottom and top of the column, probes of carbon monoxide are extracted for analysis.[6]

The principle of carbon isotopes separation relies on the different boiling temperatures of the two carbon isotopes, the predominant ^{12}C and the scarce ^{13}C, the separation coefficient between the two isotopes being defined as the ratio of the vapour pressure of [12]CO and [13]CO:

$$\alpha = \frac{P_{^{12}CO}}{P_{^{13}CO}} \qquad (1)$$

Since the vapour pressure depends on the temperature, so is the separation coefficient. At very low temperatures – around 81 K – the [12]CO vapour pressure is greater than that of [13]CO and thus the carbon isotopes can be separated through distillation, with the 12 carbon isotope accumulating in the gaseous phase and the 13 carbon isotope being predominant in the liquid phase.

The typical values of the separation factor (α) as a function of column temperature are given in Table 1.[2]

T a b l e 1 − *Typical values of the separation factor (α)*

Temperature, T/K	81.85 K	110.05 K	68.15 K
(α)	1.0069	1.0052	1.012

Nonlinear model of the cryogenic carbon isotopes separation column

Considering the equation for the isotope separation coefficient given in (1), and the mentioned aspects relating the latter to column temperature, this paper develops a nonlinear model for the cryogenic isotope separation column presented that considers some simplified differential equations in which the isotope transfer rate depends on the inner column temperatures – that of the condenser area and that of the bottom boiler area. The objective is to design a temperature control loop, by manipulating the level of liquid nitrogen in condenser (h_c) and the electric power at the boiler (P_{el}), so that the closed loop nonlinear system is not affected by external disturbances.

The aim is to develop a nonlinear model of the isotope separation process in terms of differential nonlinear equations, in which the isotope fraction and the two temperatures – bottom and top temperatures – represent the state variables of the nonlinear system, while the condenser liquid nitrogen level and the electric power supplied to the boiler represent the control signals. The output – intended to be controlled – is the final isotope fraction, indirectly controlled by maintaining the condenser and boiler temperatures at their prescribed set points. If we model the isotope fraction as a first state variable, x_1, the temperature in the boiler zone as a second state variable, x_2, and the temperature in the condenser zone as the third variable, x_3, then the nonlinear open loop system can be described as:

$$
\begin{pmatrix} \dot{x}_1 \\ \dot{x}_2 \\ \dot{x}_3 \end{pmatrix} = \begin{pmatrix} -\dfrac{1}{T_I}x_1 + k_{11}(x_2 + x_3) - k_{12}(x_2 - x_3)^2 \\ -\dfrac{1}{T_B}x_2 \\ -\dfrac{1}{T_C}x_3 \end{pmatrix} +
$$

$$
+ \begin{pmatrix} 0 & 0 \\ k_{21} & 0 \\ 0 & k_{31} \end{pmatrix} \begin{pmatrix} P_{el} \\ h_c \end{pmatrix}
\tag{2}
$$

with T_B, T_C and T_I being the boiler, condenser and isotope transfer rate time-constants and k_{11}, k_{12}, k_{21} and k_{31} are specific coefficients. The control vector, as mentioned above, is $u = [P_{el} \quad h_c]^T$. The temperature evolutions are modeled simply as first order transfer functions, while the isotope transfer rate – dependant on the column temperatures as it has been shown – is modeled first of all as a linear equation, $\dot{x}_1 = -\dfrac{1}{T_I}x_1 + k_{11}(x_2 + x_3)$. Due to the fact that the vapor pressure difference – and hence the separation factor (α) and the isotope transfer rate – is inversely proportional to the square of the absolute temperature,[7] a nonlinear term modeling the dependency of the isotope transfer rate upon the square of the absolute temperature is added, thus resulting in the final equation for the isotope transfer rate: $\dot{x}_1 = -\dfrac{1}{T_I}x_1 + k_{11}(x_2 + x_3) - k_{12}(x_2 - x_3)^2$. The linearised model of the isotope separation process is given in the following equation, with the equilibrium point chosen as $x_0 = [2.87 \quad 79 \quad 77]^T$:

$$
\begin{pmatrix} \dot{x}_1 \\ \dot{x}_2 \\ \dot{x}_3 \end{pmatrix} = \begin{pmatrix} -\dfrac{1}{T_I} & k_{11} - 4k_{12} & k_{11} + 4k_{12} \\ 0 & -\dfrac{1}{T_B} & 0 \\ 0 & 0 & -\dfrac{1}{T_C} \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix} +
$$

$$
+ \begin{pmatrix} 0 & 0 \\ k_{21} & 0 \\ 0 & k_{31} \end{pmatrix} \begin{pmatrix} P_{el} \\ h_c \end{pmatrix}
\tag{3}
$$

The nonlinear model parameters were estimated using the experimental data – such as a 25 W electrical power, 20 cm liquid nitrogen level, a 77 K temperature at the condenser, respectively 79 K temperature at the boiler – provided by a dedicated monitoring system implemented on the cryogenic distillation plant at NIRDTIM.[8] Fig. 2 shows a
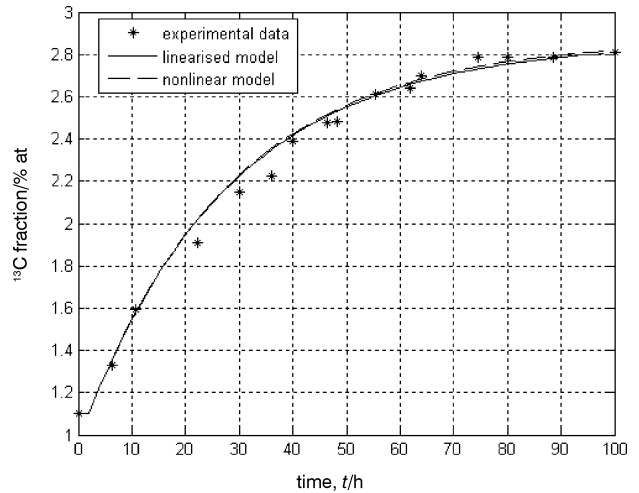

F i g . 2 – *Comparison of the nonlinear and linearised model against experimental data*

comparison between the nonlinear, linearised model and the experimental data. Under the same operating conditions, the nonlinear and linearised models behave similar and also verify the experimental data gathered in exactly the same circumstances.

Optimal controller of the carbon isotope separation process

Controller design

The controller designed for the isotope separation process, in this paper, is a linear quadratic regulator, chosen due to its simplicity and efficiency. Given a discrete linear system:

$$
\mathbf{x}_{k+1} = \mathbf{A} \cdot \mathbf{x}_k + \mathbf{B} \cdot \mathbf{u}_k
\tag{4}
$$

the linear quadratic regulator (LQR) problem is to find a state- variable feedback control:

$$
\mathbf{u}_k = -\mathbf{K} \cdot \mathbf{x}_k
\tag{5}
$$

that minimizes the discrete time performance index:

$$
J = \frac{1}{2} \sum_{i=K}^{\infty} \mathbf{x}_i^T \cdot \mathbf{Q} \cdot \mathbf{x}_i + \mathbf{u}_i^T \cdot \mathbf{R} \cdot \mathbf{u}_i
\tag{6}
$$

with **Q** and **R** being the state (**x**) and input (**u**) weighting matrices.

The solution to the discrete LQR problem consists of solving the Riccati equation:[4]

$$
\mathbf{H} = (\mathbf{A} - \mathbf{B} \cdot \mathbf{K})^T \cdot \mathbf{H} \cdot (\mathbf{A} - \mathbf{B} \cdot \mathbf{K}) \cdot \mathbf{A} + \\ + \mathbf{K}^T \cdot \mathbf{R} \cdot \mathbf{K} + \mathbf{Q}
\tag{7}
$$

with the resulting controller given by:

$$
\mathbf{K} = (\mathbf{R} + \mathbf{B}^T \cdot \mathbf{H} \cdot \mathbf{B})^{-1} \cdot \mathbf{B}^T \cdot \mathbf{H} \cdot \mathbf{A}
\tag{8}
$$

However, since in this case the process differs from a classical disturbance rejection problem, the cost function used in this paper would be written as a tracking problem,[9] meaning that the purpose of the controller would be to not only reject external disturbance inputs, but it should also be able to track some desired values for the controlled outputs.[4] The cost function would then be:[10]

$$J = \sum_{i=k}^{\infty} [(\mathbf{x}_i - \mathbf{x}_d)^T \cdot \mathbf{Q} \cdot (\mathbf{x}_i - \mathbf{x}_d) + $$
$$+ (\mathbf{u}_i - \mathbf{u}_d)^T \cdot \mathbf{R} \cdot (\mathbf{u}_i - \mathbf{u}_d)] \qquad (9)$$

in which $\mathbf{x}_d$ is a vector containing the desired values for the isotope transfer rate, boiler and condenser temperatures, $\mathbf{u}_d$ is a vector containing the steady state values for the control inputs, required to keep the states at their desired values, $\mathbf{x}_d$, and $\mathbf{Q}$ and $\mathbf{R}$ are the weighting matrices for the states and the input vector, respectively. The controller was firstly designed considering the linearised model of the system given in eq. (3), which was then applied to the nonlinear open loop system given in eq. (2).[11,12]

As mentioned before, the control strategy is to keep the column temperature constant, thus the separation factor will also be kept constant and ultimately a properly designed controller would ensure the overall requirement of constant ^{13}C isotope transfer rate, despite output disturbances. However, a few control constraints need to be foreseen when designing the optimal controller.

Two constraints refer to the two control inputs, the liquid nitrogen level and the electrical power. First, the liquid nitrogen level in the condenser should not drop below a critical value of 10 cm. Such a situation would lead to the impossibility of efficiently condensing the vapour upstream, thus compromising the entire separation process. Moreover, the liquid nitrogen level cannot exceed the construction dimensions of the condenser, and thus the upper limit would be around 62 cm. Secondly, for efficient separation and to avoid column flooding, the power supplied to the boiler cannot exceed an upper limit of 45 W. A large variation would affect the separation by modifying the upward gaseous stream.

Finally, a third constraint refers to the vacuum pressure, which from a control engineering point of view, represents an output disturbance and should not drop significantly. Variations in vacuum pressure bring about the loss of the efficient thermal isolation and cause the increment of the inner column temperature.

Robustness of the optimal temperature controller

The linear quadratic regulator was designed by a series of trial and error. Since the states need to be more constrained to follow a desired value and the control inputs have a wider variation range, the major guideline for choosing the state and input weighting matrices was to penalize more strictly the evolution of the system states as compared to the evolution of the input signals. In order to maintain a constant separation coefficient and obtain a desired isotope fraction, the temperatures in the boiler and condenser area need to be kept around 79 K, and 77 K, respectively. Thus, the weighting matrix for the states ($\mathbf{Q}$) was chosen significantly larger than the weighting matrix for the input vector ($\mathbf{R}$),[13] since such a choice would imply a more heavily penalized evolution of the column temperatures as compared to the electrical power and the liquid nitrogen level:

$$\mathbf{Q} = \begin{pmatrix} 9 & 0 & 0 \\ 0 & 18000 & 0 \\ 0 & 0 & 11000 \end{pmatrix} \qquad (10)$$

$$\mathbf{R} = \begin{pmatrix} 2.7 & 0 \\ 0 & 1 \end{pmatrix} \qquad (11)$$

The weights in $\mathbf{R}$ add a higher constraint to the electrical power than the liquid nitrogen level, since the latter can spread over a wider range. The condenser and boiler temperature weights are significantly larger than the weight imposed for the isotope fraction, in $\mathbf{Q}$, since by restraining the evolution of the temperatures, the isotope fraction is indirectly kept at the required value as well.

Desired values for the states, upon which the column operates the most efficiently are: $\mathbf{x}_d = [2.87 \ \ 79 \ \ 77]^T$. The control inputs that ensure the desired isotope fraction and column temperatures, under nominal conditions – without any disturbances acting on the column – and under an accurate identification of the nonlinear system parameters are: $\mathbf{u}_d = [25 \ \ 20]^T$.

The resulting discrete linear quadratic regulator is:

$$\mathbf{K} = \begin{bmatrix} 2.005756 \cdot 10^{-4} & 1.145561 \cdot 10^{2} & 8.004724 \cdot 10^{-9} \\ 0.0011319 & 6.222665 \cdot 10^{-9} & 92.26122 \end{bmatrix} \qquad (12)$$

The control loop, consisting of the optimal controller given in eq. (12) and the nonlinear system in eq. (1) was implemented in Matlab Simulink, with the block scheme represented in Fig. 3.
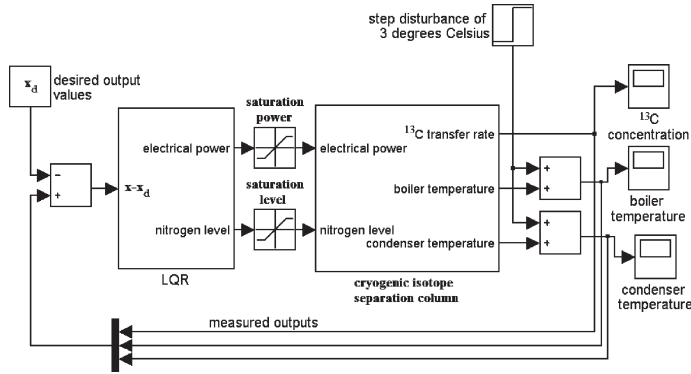
F i g . 3 – *Block scheme of the nonlinear closed loop system controlled with a linear quadratic regulator*

To test the ability of the designed optimal controller, a failure of the vacuum is considered, acting as a disturbance and modelled as a 3-degrees increment in the boiler and condenser temperatures, after 70 h of column normal operation. Such an increment, in an open loop case – without the optimal controller considered – implies a modification of the separation coefficient, and thus would cause significant changes in the isotope transfer rate. Figs. 4–8 present the closed loop simulations for the linear and nonlinear system, considering the disturbance mentioned previously. To test the robustness of the designed controller, uncertainties in the parameters and time constants were considered, such as for T_B, T_C, k_{21} and k_{31} a variation of $\pm 15\ \%$ from nominal value, for k_{12} a $\pm 10\ \%$ variation from nominal value and for T_I and k_{11} only slight variations from nominal value. The simulations include the following case scenarios:

1. T_B, T_C, k_{21}, k_{31} varying between $\pm 15\ \%$ from nominal value, k_{12} varying between $\pm 10\ \%$ from nominal value, T_I, k_{11} varying between $\pm 1\ \%$ (simulations being marked with a black line (–))

2. T_B, T_C, k_{21}, k_{31} varying between $\pm 15\ \%$ from nominal value, k_{12} varying between $\pm 10\ \%$ from nominal value, T_I deviated -5% from nominal value, k_{11} deviated $+ 5\ \%$ from nominal value (simulations being marked with a black line (–))

3. T_B, T_C, k_{21}, k_{31} varying between $\pm 15\ \%$ from nominal value, k_{12} varying between $\pm 10\ \%$ from nominal value, T_I, k_{11} deviated more than $\pm 5\ \%$ from nominal value (simulations being marked with a grey line (–))

Fig. 4 presents the evolution of the boiler temperature, while Fig. 5 presents the evolution of the condenser temperature, considering all case scenarios presented previously. The simulations show that the designed linear quadratic regulator ensures a temperature of approximately 79 K at the boiler area and 77 K at the condenser area, the tempera-
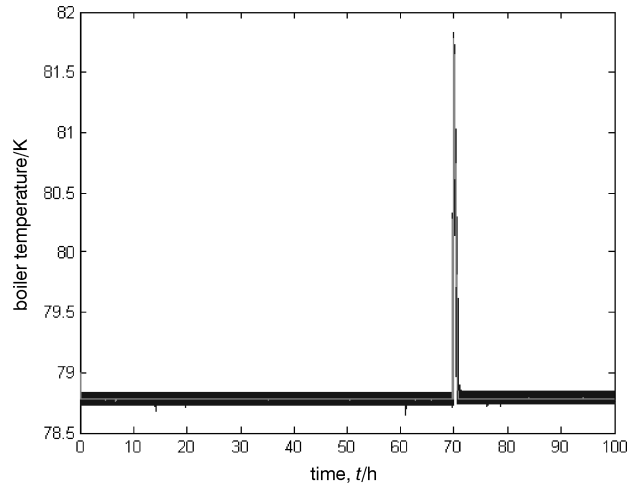

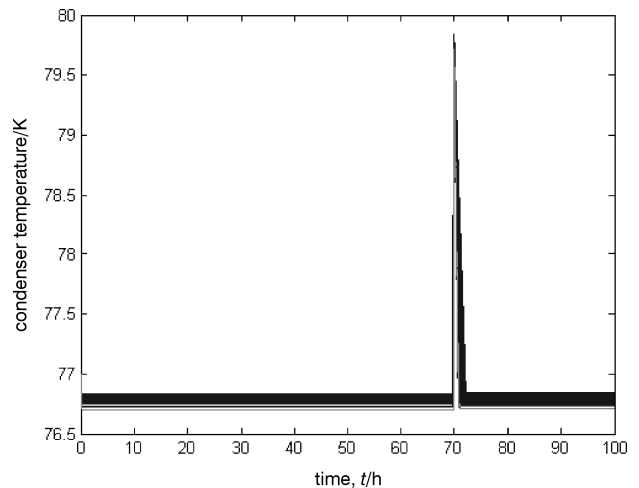F i g . 4 – *Evolution of the boiler temperature, considering a vacuum failure*

F i g . 5 – *Evolution of the condenser temperature, considering a vacuum failure*
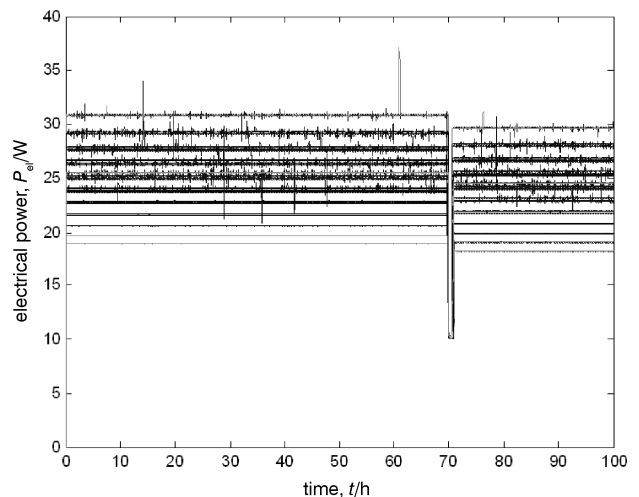

F i g . 6 – *Evolution of the electrical power supplied to the boiler, considering a vacuum failure*
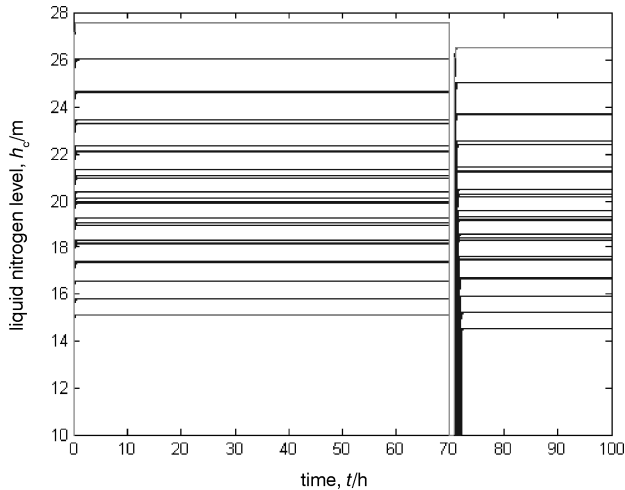
F i g . 7 – *Evolution of the liquid nitrogen level in condenser, considering a vacuum failure*
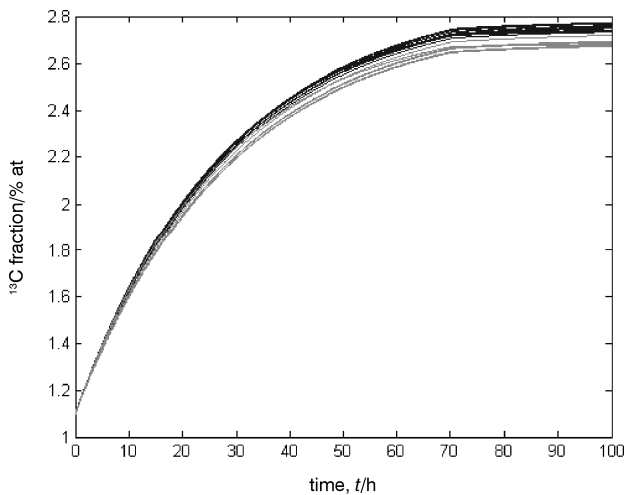

F i g . 8 – *Evolution of the ^{13}C isotope fraction, considering a vacuum failure*

ture disturbance considered of 3-degrees Kelvin being rejected completely.

The input control signal evolutions are given in Fig. 6 and Fig. 7. The electrical power and the liquid nitrogen level respect the control constraints, remaining in the bounded region mentioned in the previous paragraph, despite the modelled disturbance and the uncertainties considered.

Fig. 8 presents the ^{13}C isotope fraction evolution in time. The maximum ^{13}C isotope fraction that can be achieved using the technology available at NIRDIMT and during 100 h of column operation under nominal conditions – 79 K at boiler, 77 K at condenser – and without disturbances is 2.87 % at. The designed controller – under the possible range of uncertainties in the modeling parameters mentioned previously and considering temperature disturbances – can effectively maintain the tempera-

tures around their prescribed values of 79 K, and 77 K, respectively. Consequently, the final isotope fraction achieved is around 2.8 % at. Nevertheless, the simulations show that the closed loop system is highly sensitive to uncertainties in T_I and k_{11}. The designed controller can achieve robust stability, despite large variations of these two parameters, however the performances are no longer met, the final value of the isotope fraction being less than 2.8 % at – the corresponding simulations being marked with a grey line (–).

Conclusions

The simulations prove that the linear quadratic regulator given in (12) can actively reject temperature disturbances as it can be seen from Figs. 4 and 5, in the previous section. Consequently, the isotope fraction is only slightly affected, as shown in Fig. 8. Moreover, the control inputs satisfy the design requirements specified in the previous section as constraints regarding the design of the controller.

The main advantage with the designed controller is its simplicity and effectiveness, the final value of the isotope fraction that can be achieved being approximately 2.8 % at, under temperature disturbances, a value that is close to the maximum achievable isotope fraction using the existing technologies implemented on the actual cryogenic isotope separation column, under no external disturbances. Considering the previously mentioned parameter uncertainties, Figs. 4–8 show that the designed optimal controller is also robust with respect to a ± 15 % variation of T_B, T_C, k_{21} and k_{31}, as well as with respect to a ± 10 % variation from nominal value in terms of the k_{12} parameter. As far as T_I and k_{11} parameters are concerned, the simulations show that the closed loop uncertain systems maintain the robust stability specification, however the steady state value for the isotope fraction is no longer achieved.

L i s t   o f   s y m b o l s

x_1 – ^{13}C isotope fraction, % at

x_2 – boiler temperature, K

x_3 – condenser temperature, K

T_I – time constant of the isotope fraction transfer rate, s

T_B – time constant of the boiler, s

T_C – time constant of the condenser, s

P_{el} − electrical power, W

h_C − liquid nitrogen level, m

T − temperature, K

k_{11}, k_{12}, k_{21}, k_{31} − specific coefficients

x   − state variables vector

x$_d$ − desired values of the state variables vector

u$_d$ − steady state values of the control inputs

u   − control signals vector

K   − linear quadratic regulator

t − time, h

J − optimal cost function

Q   − state variables vector weight matrix

R   − control signals vector weight matrix

H   − solution of the Riccati equation

T1, T2, T3 − temperature transducers

T   − matrix transpose

C   − distillation column

C1   − column condenser

B   − column boiler

M   − vacuum jacket

PVP   − vacuum pumps

R   − carbon monoxide reservoir

M1, M2, M3 − manometers

References

1. *Radoi, A.*, Rev. Chim. (Bucharest, Rom.) **50** (1999).
2. *Gligan, M., Dulf, E. H., Unguresan, M.-L., Festila, C.*, Proc. IEEE Conference on Automation, Quality and Testing, Robotics, Cluj-Napoca, Romania, 2006.
3. *Cohen, K.*, The Theory of Isotope Separation as Applied to the Large-Scale Production of U235, McGraw-Hill Book Company, Inc., New York, 1951.
4. *Lewis, F. L., Syrmos, V. L.*, Optimal Control (Second Edition), John Wiley & Sons, Inc., New York, 1995.
5. *Kwakernaak, H., Sivan, R.*, Linear Optimal Control Systems, John Wiley & Sons, Inc., New York, 1972.
6. *Pop, C. I., Dulf, E. H., Festila, C.*, Proc. IEEE International Conference on Automation, Quality and Testing, Robotics, Cluj-Napoca, Romania, 2008.
7. *Vasaru, Gh.*, Stable Isotopes, (in Romanian), Editura Tehnica, Bucuresti, 1968.
8. *Dulf, E., Stegaru, A., Buzdugan, T., Gligan, M.*, Isotopic and Molecular Processes Conference, Cluj Napoca, 2007.
9. *Budiyono, A., Wibowo, S. S.*, Journal of Bionic Engineering **4** (2007) 271.
10. *Chew, M., Jones, W. E., Wilson, J. A.*, 8th Distillation and Absorption Conference, London, 2006.
11. *Vincent, T. L., Grantham, W. J.*, Nonlinear and Optimal Control Systems, John Wiley & Sons, Inc., New York, 1997.
12. *Naidu, D. S.*, Optimal Control Systems, CRC Press, Florida, 2003.
13. *Zhang, G., Fang, Z., Shu, L.*, Proc. International Conference on Intelligent Systems and Knowledge Engineering, Atlantis Press, Chengdu, 2007.