Control and Simulation of A ¹³C Isotope Separation Process

Roxana M. Motorga, Vlad Muresan, and Mihail Abrudean

Abstract—This paper is presenting a control solution for the concentration of the ¹³C isotope, both at the output and inside of the separation column; the proposed solutions are a relay and a PID controller. The ¹³C isotope results after a distributed parameter process, specifically the chemical exchange process of the carbon dioxide and carbamate. The analytical solution that was determined in order to model the described process is simulated. The evolution of the proposed solution depends on two parameters: the position in the column height and the time. The reference model is simulated in order to reach to the desired concentration using an acceptable flow of solution. The tuning method for the PID controller is Ziegler-Nichols, based on the bipositional relay. The advantage obtained during this control process consists of the possibility of reaching the desired concentration of the ¹³C isotope at any point of height inside the separation column.

Index Terms—¹³C isotope, concentration, control structure, PID, bipositional relay, separation column, simulation, Simulink.

I. INTRODUCTION

There are a few papers in the literature that treats the problem of the ¹³C isotope separation process. Reference [1] is proposing a solution to improving the process of separation using simple control strategies, that focuses mainly on the concentration of the isotope at the bottom of the separation column. The system acquires the desired ¹³C concentration from the separation process using an on-off controller that implies undesired fluctuations in the plant and a lower control accuracy than expected.

The isotope separation process for 13 C requires a specific plant that can be observed in Fig. 1. It consists of five system elements (absorber A, stripper S, heater H, separation column SC and reactor R) that are connected through pipes that circulates the compounds either by their phase or engaged by a pump. The 13 C isotope results by separating the carbon dioxide (CO₂) using ethanolamine.

It can be observed in Fig. 1 that the stripper, the absorber and the separation columns are hachured - this means that those containers consist of steel pack of Helipack type, which have a significant contribution in the separation process of the 13 C isotope [4].

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There are two regimes to obtain this isotope. The production one refers to supplying the plant with CO₂ having a concentration of 99.98% approximately, through pipe 6; being in a gaseous phase it reaches the absorber A (by pipe 5) which is also supplied by the pump through pipe 1 with ethanolamine. These two elements circulate in counter current; the chemical interaction of the absorption of CO₂ in the ethanolamine results in the development of carbamate [2], [3], which settles in the bottom part of the absorber. The so resulted gaseous compound is evacuated using the pipe 4 from the plant, after it reaches the upper part of A - the concentration of the CO₂ contained in the gas is lower than 0.1%. Pipe 3 supplies the separation column with the resulted carbamate; this column is also supplied with the CO₂ obtained after the decomposition of the carbamate in the reactor. The SC is responsible for the chemical exchange that takes place between the two reactants that are circulating in the countercurrent. The process of chemical exchange is responsible to the enrichment of the 13C isotope - can be found in the liquid phase in the separation column's lower part [4]. The quantity of gas from the separation column reaches the absorber through pipe 5. Pipe 2 includes at the output of the separation column a transducer of type mass spectrometer T that measure concentration of the 13C isotope, the most important parameter of the process that is desired to be monitored and controlled. Moreover, the reactor receives the carbamate through pipe 2 and performs the thermal decomposition of it whilst returning the CO2 resulted in this process back to the separation column through pipe 7 - having a higher 13C isotope concentration than the initial conditions or extracts it by pipe 8. In real life processes the two pipes used to supply the CO₂, pipe 6 and 8 are connected together. The stripper is responsible of the result of pure ethanolamine, by completely removing the CO_2 . The heater is responsible for reheating the ethanolamine in order to be recirculated again in the circuit using the pipe 1 by engaging the pump P.

II. PROCESS MODELING

The considered process of separation is considered a distributed parameter process [1] having the imposed output y the concentration of the ¹³C which depends on the position it reaches in the separation column regarding its height and the variable of time (t), which is independent. The process model does not consider the variation of the concentration regarding to the position inside the separation column's transversal section because it is considered insignificant [6].

The separation column is actually a cylinder having the height h = 300 cm; because of its form, making a transversal section would result in a circle of diameter 2 = 2.5 cm [4]. The position in the column's height is noted with H, and

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along with the origin 0 describes the 0H axis - defining the center of the separation column transversal section, in the upper part [6] - presented in Fig. 2.



Fig. 2. The 0H axis.

The process modeling of this system was acquired in [6]; this paper aims to implement the model developed by the authors. The concentration of the ¹³C isotope, y(t,H), depends on both independent parameters; it is impetuous to add the CO₂ using pipe 8 in the separation column in order to have a valid procedure of modeling.

The input flow of ethanolamine influents directly the HETP (the height equivalent to a theoretical plate); Figure 3 presents the Simulink implementations for all the equations in process modeling from [6]. The variables of the system, namely the input flow of ethanolamine and the height of the separation column are implemented using step type signals. All the subsystems referred in the following paragraphs explain the computations. Presented in Fig. 3.

It can be easily remarked that the HEPT(t) depends on its steady-state value, a proportionality constant K_p (connecting the HETP with the flow of ethanolamine), the constant input flow of ethanolamine (F_{in0}) and its instantaneous value ($F_{in}(t)$):

$$HETP(t) = HETP_0 + K_p \cdot (F_{in}(t) - F_{in0}), (1)$$

The HETP_subsystem from Fig. 3 is responsible of the implementation of (1). It can be easily noticed that the HETP depends on the proportionality constant K_p - making a connection between the HETP and the input flow of the ethanolamine.

$$K_p = \frac{HETP_{st1} - HETP_0}{F_{in1} - F_{in0}}.$$
 (2)

Experimentally was determined the value of $K_p =$

 $0.0085(\text{cm}\cdot\text{h})/\text{ml}$ in [6]. Moreover, it can be remarked that the number of theoretical plates depends on the HETP and the height of the separation column:

$$n(t) = \frac{h}{HETP(t)}.$$
(3)

The S(t)_subsystem computes the isotope separation based on knowing the elementary separation factor for the ¹³C isotope regarding the procedure of chemical exchange α = 1.01 and the number of theoretical plates.

$$S(t) = \alpha^{n(t)}.$$
 (4)

An important remark is that the input flow of the ethanolamine affects directly the concentration - a lower flow means a longer period of time required in the separation column to CO_2 and carbamate to interact.

The u(t)_subsystem computes the final value of the input signal of the process as a relation between the natural abundance of the isotope ¹³C $y_0 = 1.108\%$ and the isotope separation:

$$u(t) = y_0 \cdot (S(t) - 1).$$
(5)

Because the isotope separation is a first order process, there is just one time constant which was defined experimentally in [6]. It was remarked that the time constant is depending on the height in the separation column:

$$T = T_{H0} + (T_{Hf-T_{H0}}) \cdot \frac{H}{H_f}.$$
 (6)

The Ft(t)_subsystem implements the first order differential equation:

$$\frac{dF_t(t)}{dt} = -\frac{1}{T(H)} \cdot F_t(t) + \frac{1}{T(H)} \cdot u(t), \quad (7)$$

in which is described the relation defined between the function $F_t(t)$ and u(t) - the final input signal.

The evolution of concentration depending on the input flow is implemented in F_H1(H) subsystem, and it can be approximated by:

$$F_{H1}(H) = (y_0 \cdot \alpha - 1) + e^{\frac{H}{C + K \cdot (F_{in}(t)) - F_{in0})}}, \quad (8)$$

Where C = 430 cm is the equivalent of a SC constant an K = 0.7527 (cm·h)/ml, implying the definition of the constant H (the length):

$$H = C + K \cdot (F_{in}(t) - F_{in0}). \tag{9}$$

The concentration of the process can be determined at any height in the separation column, depending on the ethanolamine input flow $F_{in}(t)$ by:

$$y_{an}(t, H) = y_0 + F_t(t) \cdot F_H(H, F_{in}(t)).$$
(10)

III. CONTROL STRUCTURES

A. Relay Controller

The control structure based on the bipositional relay controller strategy [7] is presented in Fig. 4 - the purpose is to control the concentration of the ¹³C isotope.

It is obviously that the input reference of the control model is the desired concentration of ¹³C isotope to be acquired. The direct channel of the system consists of the relay and the process; the error and the controller's signal are also monitored.



Fig. 3. Simulink plant implementation.

In real life, the process cannot be simulated from 0 initial conditions, this being the reason why another constant is added in the system and subtracting the output of the relay.

The presented control structure is expected to result in an oscillating output.



Fig. 4. Implementation of the relay controller.



Fig. 5. Implementation of the PID controller.

B. PID Controller

The relay controller is expected to be less accurate and a PID controller is also implemented [8], for the same control strategy: to acquire the desired concentration of the ¹³C isotope, with a rational input flow. The Simulink representation is presented in Fig 5.

The PID controllers' parameters were tuned using the Ziegler-Nichols method which requires a closed loop system. In order to implement the specific formulas, the process was brought to its stability limit using a bipositional relay, the same principle that is presented in Fig. 4, by varying the hysteresis parameter. The identification of the result was performed and it is presented in Fig 6, where $eps = \pm 0.1$ -

the hysteresis and d = 100 - the output. Two parameters are significant to be read from Fig. 6, the oscillation period $T_c = 37.318(h)$ and the amplitude of the signal $A_c = 2.39(\%)$. Table I presents the calculus for the PID controller parameters.

TABLE I. PID PARAMETERS

Parameter	Formula	Value
K _c	$4 \cdot d/(\mathbf{\pi} \cdot \mathbf{A}_c)$	53.2736
K_p	$0.6 \cdot K_c$	31.9642
T_i	T _c /2	18.6590
T_{d}	Tc/8	4.6648
\mathbf{K}_{i}	K_p/T_i	1.7131
K_{d}	$Kp \cdot T_d$	149.1049

The PID controller is computed as it follows:

$$PID(s) = K_p + K_i \cdot \frac{1}{s} + K_d \cdot s.$$
(11)

IV. SIMULATION RESULTS

A. Plant Simulation

The plant simulations [6] are performed in Matlab/Simulink. Table II presents the steady-state values of the concentration of the ¹³C isotope $(y(t_f,H_f))$ when the ethanolamine input flow increases. It is considered that H= H_f. The variations for the input flow $F_{in} \in \{100, 180, 367, 414, 500\}$, representing a step type variation.



TABLE II: VARIATION OF THE 13C ISOTOPE CONCENTRATION

F _{in} [ml/h]	$y(t_{f},H_{f})$ [%]
100	3.9248
180	2.9403
367	2.1282
427	2.0011
550	1.8153

TABLE III: VARIATION OF THE 13C ISOTOPE	
CONCENTRATION-1	

H [cm]	$y(t_f,H_f)$ [%]
$\mathrm{H_{f}}$	2.7988
$H_{f}/2$	1.7560
$H_{\rm f}/3$	1.5080
$H_{\rm f}/4$	1.3982





Fig. 8. Step response of a PID controller - reference concentration: 2.3%.



These results were compared to the experimental data in [6] and it can be concluded that the implementation of the equations is correct. It can be remarked that the input flow of ethanolamine affects the resulted concentration - an increased flow results in a smaller concentration.

The open loop simulation of the plant where the input flow was considered constant F_{in} =300 ml/h, for different inputs for

 $H \in \{H_f, H_f/2, H_f/3, H_f/4\}$ is centralised in Table III.

It can be remarked that the decrease of H results in a decrease of concentration.

B. Bipositional Relay Controller

The parameters of the bipositional relay controller were set experimentally. The desired concentration (2.3%) was reached in the shortest amount of time when the hysteresis was set to $eps = \pm 0.01$ and d = 100. The simulation is presented in Fig. 7.

It can be remarked that the step response of the system has a small oscillation around the imposed concentration value. For this reason, a PID controller was designed.

C. PID Controller

The PID controller was set to impose reference concentration of 2.3% to a constant input ethanolamine flow of 250 (ml/h). From Fig. 8 it can be remarked that the concentration is reached after 350 h. The response stabilisation requires additional time, but the effect of oscillations was removed.

The efficiency of the PID controller was tested to a different concentration. The reference was chosen to be 2.8 - the PID controller successfully reached the imposed concentration; Fig 9 presents the second simulation of the step response over the new reference concentration.

V. CONCLUSION

This paper implements the mathematical model solution for the technological process of the ¹³C isotope separation. The control of its concentration is acquired by two methods, with a bipositional relay with hysteresis and with a PID controller.

The mentioned control structures are implemented in order to achieve the desired concentration of ¹³C isotope using a normal input flow of ethanolamine. Further development of the process may imply advanced control strategies, such as the Internal Model Control (IMC).

The validity of the implemented model and the efficiencies of the control structures are proven in chapter IV. As it can be observed in Fig 7-9, the imposed concentration was reached by the two controllers; whilst the oscillation of the relay controller may damage the pump (by closing and opening it), the PID controller is preferred.

CONFLICT OF INTEREST

The author declares no conflict of interest.

AUTHOR CONTRIBUTIONS

R.M implemented the equations and simulations in Simulink, tuned the two controllers and wrote the paper. V.M analysed the results obtained from the simulations and M.A. obtained the experimental data and the interpretation of the process in the phenomenological perspective.

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