

Monitoring and Control System of a Separation Column for ^{13}C Enrichment by Cryogenic Distillation of Carbon Monoxide

Eva-Henrietta Dulf, Clement Festila, and Francisc Dulf

Abstract— In incipient stage for laboratory experiments, a monitoring and control based on human operators is usual. Using the acquired experience, a computer monitoring and even control is necessary and possible. In the actual developing plan to apply the cryogenic technology for the production of the ^{13}C isotope by industrial scale, an efficient and safe operation is a strong reason to conceive and to apply a modern computer based monitoring and control strategy. The actual hardware possibilities of the computer systems and valuable interfaces enable cheap, easy-to-apply and efficient monitoring and control systems. For a complex equipment like a cryogenic isotope separation column, it was selected a common PC interfaced with a series of robust input-output modules, ICP-CON, I-7000 Series, from ICP-DAS as data acquisition and control functions. Based on general separation process and column operation descriptions, this particular application is developed using the intuitive Labview visual programming language. The front panel of the monitoring system has numerical and graphical indicators, buttons for available options, alarms to warn the user that a problem appeared in the process. The control system is conceived for the liquid nitrogen level control and the liquid carbon monoxide level control. In the actual stage, the control functions are analyzed only by simulation.

Keywords— Control system, Data acquisition, Isotope separation column, Monitoring.

I. INTRODUCTION

THE application field of the ^{13}C – carbon stable isotope – in medicine, agriculture, etc. led to the development of the specific technologies and equipment, able to produce compounds with much higher concentration than the natural abundance in the isotope of interest. For instance, based on the natural abundance $^{13}\text{C}/^{12}\text{C}$ in the ratio (1.11 at% / 98.99 at%), using the cryogenic separation method (at about -190°C) it is possible to raise appreciably the ^{13}C concentration

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processing pure carbon-monoxide. An efficient and safe operation is a strong reason to conceive and to apply a modern computer based monitoring and control strategy.

The production of the (^{13}C) isotope is based on a special case of distillation: cryogenic distillation.

Distillation is a very common method used to separate two (ore more) components in a trayed or packed distillation column. Cryogenic distillation columns are operated at extremely low temperatures (80-100°K) to separate the desired component and to “waste” useless component. For a continuous production, it is necessary to divide the column in a *stripping* section – here the upper part – and in an *enriching* (*rectifying*) section. The material of isotope natural abundance is fed at an intermediate part of the column. The depleted material (waste) is withdrawn at approximately the same rate like the feed flow rate in the upper end of the column. The enriched material is withdrawn, usually at a much smaller rate, from the bottom end of the column. The separation achieved is less under condition of steady production than it is at total reflux (no withdrawal, infinite reflux).

For a binary mixture, if (N) and (n) are the mole-fraction in the liquid phase and, respectively, in the gas, for a separating element, the “simple process separation factor” [1] is:

$$\alpha = \frac{N/(1-N)}{n/(1-n)} = \frac{N(1-n)}{n(1-N)} \quad (1)$$

In order to rise the concentration of the (^{13}C) isotope from the natural abundance (N_0) to the final product fraction (N_P), the elementary separation process is “multiplied” in the column: the overall separation factor (S) is:

$$S = \alpha^k \quad (2)$$

If the desired isotope accumulates in the bottom (B) of the column (liquid phase, L) and the waste in the top (T) section (gas, G):

$$S = \frac{\left(\frac{N_L}{N_G}\right)_{\text{bottom}}}{\left(\frac{N_L}{N_G}\right)_{\text{top}}} = \frac{\frac{C_B}{1-C_B}}{\frac{C_T}{1-C_T}} \quad (3)$$

for infinite (total) reflux, (C_B) , (C_T) are the concentrations in the bottom, respectively in the top of the column.

The (theoretical) minimum number of stages is:

$$k_{\min} = \frac{\ln(S)}{\ln(\alpha)}. \quad (4)$$

By finite reflux, in [3] is presented a generalization:

$$S = \alpha^k \frac{\left(\frac{L}{G}\right)_T^{k_T}}{\left(\frac{L}{G}\right)_B^{k_B}}, \quad (5)$$

where

(L, G) are the “internal” liquid and vapor flow rate

(k_T) is the number of stages in the top section (stripping section)

(k_B) is the number of stages in the bottom section (enriching section)

and (T, B) stay for “top” and “bottom”.

To increase the separation factor (S) , it is possible either to increase of the stage number (higher column), or to increase the energy usage with “stronger” reflux and smaller withdrawal, being accepted a trade-off: by infinity (total) reflux results (k_{\min}) ; by infinite number of separation stages, results the minimum energy usage (Q_{\min}) . Typically, one selects $k \cong 2 \cdot k_{\min}$ and the corresponding boilup rate

$V \cong 2 \cdot V_{\min}$, where $V_{\min} = \frac{Q_{\min}}{\Delta H^{\text{vap}}}$ and (ΔH^{vap}) is the heat of vaporization (assumed constant).

Now, if the height (h) equivalent to a theoretical plate (HETP) may be estimated [2] the column dimension may be approximated by:

$$Z_C \cong (k_T + k_B) \cdot h = k \cdot h. \quad (6)$$

II. EQUIPMENT DESCRIPTION. MAIN VARIABLE DEFINITION FOR MEASURE AND CONTROL DEFINITION

The column (C) is a steel packed pipe, Figure 1, feed with pure gaseous carbon-monoxide by the flow-rate (Q_m) and ^{13}C concentration (N_f) , $N_f = N_o = 1.11\%$. The gaseous carbon monoxide with lower ^{13}C concentration is evacuated as “waste” at the top side, by flow-rate (Q_{out}) and the concentration (N_w) . The efficient thermal isolation of the column is based on the external vacuum jacket.

In the column, an upward vapor (gas) stream, generated by

the evaporation in the boiler is brought into contact with a downward flowing stream of liquid produced by condensation in the condenser zone. The boiler is heated with an electrical resistor and the condenser is cooled with liquid nitrogen. During this permanent contact, the concentration of (^{13}C) increases in the liquid phase in the column basis. The packing material inside the column increases the contact surface between the liquid and the gaseous phases. In order to flow up from the boiler to the condenser, the vapor stream needs some effective area through the elements of the packing material. This area is greater by dry packing material and smaller by wet material. The gaseous upstream needs a **differential pressure** along the column. Every liquid droplet is submissive to the force of gravity and, in the opposite direction, to the pressure force of the gas. In normal mode of operation, the gravity is stronger and the liquid phase flows as droplets to the boiler.

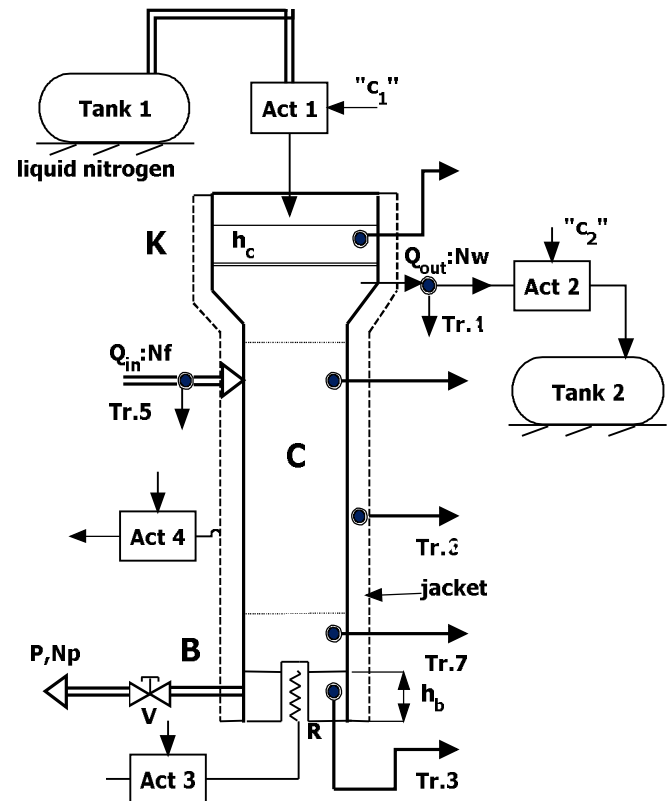


Fig. 1. Separation process scheme

By too high differential pressure, the quantity of liquid on the packing material increases and the column is no longer effective in “washing away” the undesired component from the gas [1]. The gas pressure and liquid quantity impedes proper gas transit and the liquid droplets are “suspended” on the packing material. This phenomenon - the “flooding” - reduces drastically the column efficiency and can compromise the whole process of isotope separation.

The general particularity of the isotope separation process is the huge ratio between the feed flow-rate and the final

product flow-rate ($F/P \gg$). An imposed column efficiency: shorter separation time/higher isotope enrichment, leads to a higher charge of the column: greater flow rates (F, W), more thermal energy in boiler, etc. But the column capacity is limited and by surpassing the certain values of operational variables, many undesired states may occur, the most frequent

being the flooding. Thus, the column operates “under stress” [13].

The isotope separation process may be controlled by measuring the main variables given in Table 1.

Table 1. Main variable definitions

No.	Variable name	Measurement device, sensor	Device index	Obs.
1	Liquid nitrogen level (h_c)	Resistive level transducer	Tr.1	-
2	Vacuum level	Dedicated electric device	Tr.2	-
3	Liquid carbon-monoxide level (h_b)	Capacitive level transducer	Tr.3	-
4	Output (waste) carbon-monoxide flow-rate(Q_{out})	Rotameter	Tr.4	Only as indicator
5	Input (feeding) carbon-monoxide flow-rate(Q_{in})	Rotameter	Tr.5	Only as indicator
6	Top-side column internal pressure	Pressure gage	Tr.6	Only as indicator
7	Bottom-side column internal pressure	Pressure gage	Tr.7	Only as indicator

In order to use a minimal number of data acquisition converters, the output transducer signals are scaled as DC voltages in the range of (0÷10 V).

Some of the indicated variables may be controlled using proper actuators, given in Table 2.

III. EQUIPMENT FOR MANAGEMENT AND ALARMING

The Labview project is an important step forward for the correlation of the theoretical scientific research results with the concrete information acquired from the experimental plant, and also in the analysis and better understanding of the process operation.

The front panel of the program has numerical and graphical indicators, buttons for available options, alarms to warn the user that a problem appeared in the process.

The data acquisition and transmission is based on the modules series I-7000 built by ICP CON-producer [6]. The application is based on the modules:

1) I-7017R: an 8-channel voltage and current input module with high voltage overload protection, 240Vrms, added. It has 3000V DC intra-module isolation; 24-bit sigma-delta ADC to provide excellent accuracy: $\pm 0.1\%$; input impedance 2M Ohms; input power +10 to +30 V DC; power consumption 1.3 W.

2) I-7024: a 4-channel analog output module that supports

bipolar voltage output. It accepts input power supply in the range +10 to +30 V DC; power consumption 2.4 W; accuracy: $\pm 0.1\%$; 14-bit resolution; 3000V DC intra-module isolation.

Table 2. Actuator definition

No.	Directly controlled variable	Name and type of actuator	Input variable	Obs.
1	Liquid nitrogen level (h_c)	Special electro valve (Act 1)	Voltage: TTL	Logic signal
2	Waste out-flow rate(Q_{out})	Volumetric pump (Act 2)	Continuous DC voltage -5÷+5V	-
3	Vacuum level	Dedicated two-stage vacuum pumps	Voltage: TTL	Logic signal

4	Electric dissipated power W_{el}	DC voltage controller	Continuous DC voltage 0÷10V	-
5	Feeding in-flow rate (Q_{in})	Fine-valve	-	Manual control

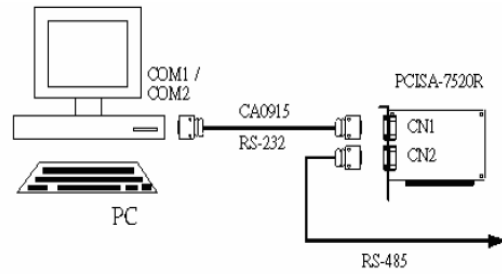


Fig. 2. Communication between the modules and the host

3) I-7044: a 4-channel isolated digital input and 8-channel isolated digital output module. The input type is Sink or Source, Isolated channel with common power or ground and the output type is Open Collector, Isolated channel with common power with 30 V maximum output voltage. The input power voltage range is +10 to +30 V DC and the power consumption is 1W.

On the front panel are shown the voltage values read from the network data acquisition module I-7017R, part of the ICP Con family of network data acquisition and control modules, providing analog-to-digital, digital-to-analog, digital input/output, timer/counter and other functions. Communication between the modules and the host is in ASCII format via an RS-485 bi-directional serial bus standard, as shown in the Figure 2.

The user can supervise the normal progress of the plant and detect an atypical value read by the sensors.

Another way to signal the user that an error occurred is the alarm system. The alarm system is activated when the limit level, previously established for the correct development of the process, is surpassed.

The instantaneous values for the variables of the process, obtained through the sensor's calibration for this parameters, are shown using the numerical indicators in engineering units and graphical from, on the real plant diagram. The charts show the time variance of the parameter's values of the process. The sampling period (freely selected) was 60 seconds and the chart history length is 1000000 samples.

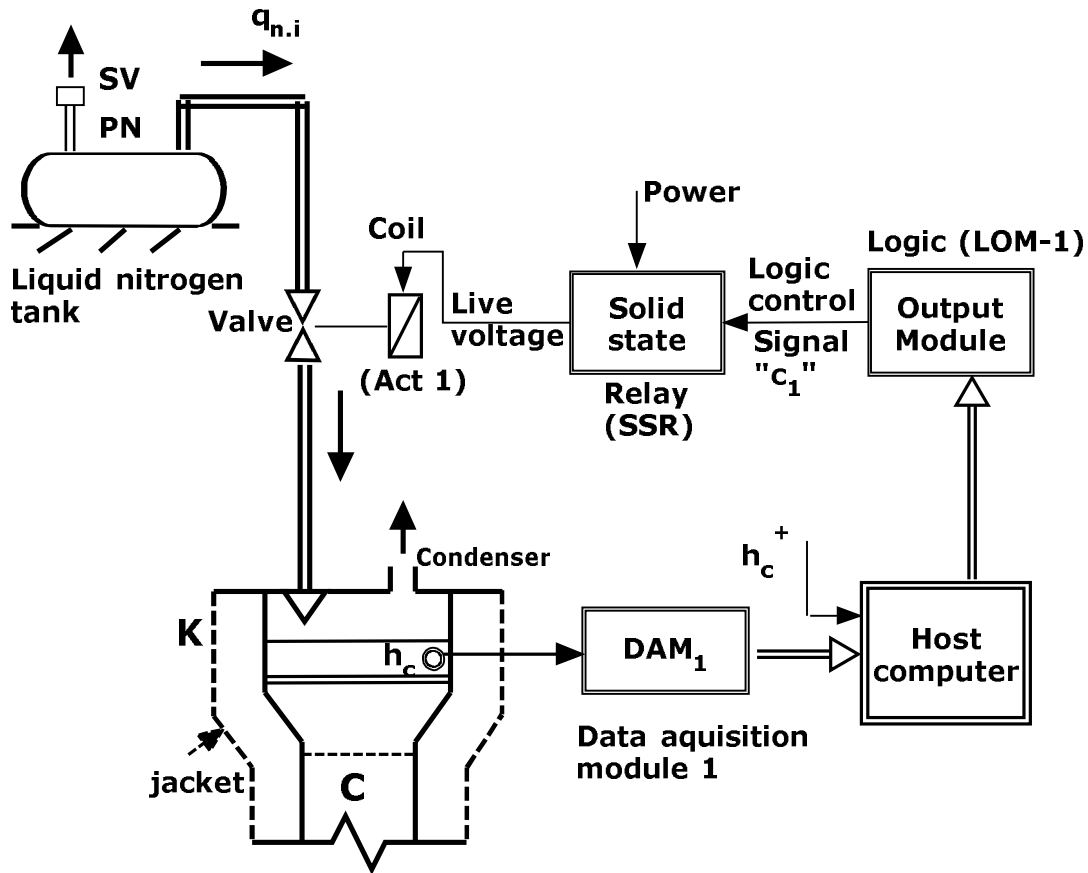


Fig. 3. Liquid nitrogen level (h_c) control diagram

The data from the process can be saved in Excel by pushing the “Save” button and typing the name and path of a file. The file has columns for the sample number, date, time and the input values read for each parameter.

IV. CONTROL EQUIPMENT AND STRATEGY

In accord to the requested control performance specifications, a broad range of control methods are recommended. This paper presents two simple versions, implemented in the computer monitoring and control system.

In the Figure 3 is presented the simplified diagram of the liquid nitrogen level control in condenser (K). The data acquisition module (DAM-1) converts the transducer output voltage into a digital size for the host computer. The control algorithm, implemented in a discrete form in computer, generates the control signal to the logic output module (LOM-1). It controls a solid-state relay (SSR) which supplies the electrovalve coil.

The control algorithm is a bang-bang type with hysteresis, Figure 4, where (h_c^*) is the set-point and ($2 \cdot \Delta h$) is the hysteresis band [7].

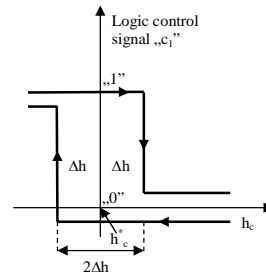


Fig. 4. Liquid nitrogen level relay control: static diagram

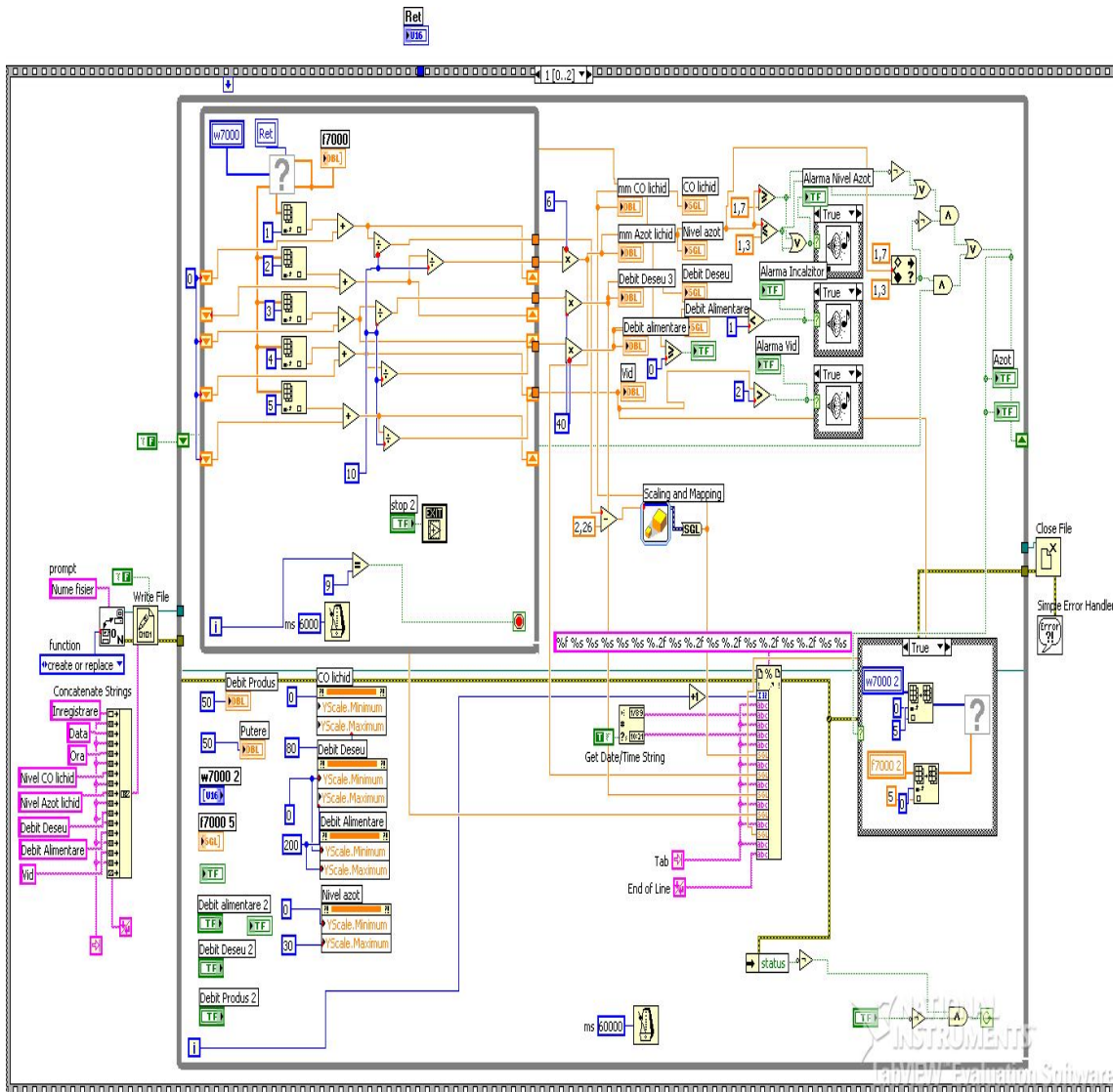


Fig. 5. The LabView Project

The logic control signal (“ c_1 ”) is given by the equation:

$$"c_1" = \begin{cases} "1" & \text{for } h_c < (h_c^* + \Delta h) \text{ and } \dot{h}_c > 0 \\ "0" & \text{for } h_c > (h_c^* + \Delta h) \text{ and } \dot{h}_c > 0 \\ "0" & \text{for } h_c > (h_c^* - \Delta h) \text{ and } \dot{h}_c < 0 \\ "1" & \text{for } h_c < (h_c^* - \Delta h) \text{ and } \dot{h}_c < 0 \end{cases} \quad (1)$$

The liquid nitrogen level (h_c) is the result of the balance between the inflow rate ($q_{n,i}$), Figure 4, and the evaporation outflow rate. If the valve-coil is energized (by the solid state relay), the inflow ($q_{n,i}$) surrounds the evaporation flow rate and the level (h_c) rises. For “ c_1 ”=0, the level (h_c) decreases. The limits of variation for $h_c(t)$ are given by the process particularities and the value of (Δh).

The Labview [5] project consists in a Stacked Sequence Structure of 3 sub-diagrams, or frames, that execute sequentially, as it is presented in figure 5.

The first sub-diagram controls the opening of COM-port 1 and sends the proper address, command, baud rate and other standard settings to start the communication of the host PC with the data acquisition and control modules.

The actual program is implemented in the second sub-diagram. In a while loop, which sets the sampling period at 60 seconds, the “analog in” sub-VI reads all the input channels at the same time and saves them in an array, which is split into components which represent the voltage values read by the input channels of the I-7017R module.

Each component of the array is plotted on a waveform chart that has a history length of 1000000 samples and a sampling period of 60 seconds.

The input data is saved in a file with a name and path chosen by the user, using the function “format into file”. This function formats string, numeric, path, or Boolean data as text and writes the text to a file. The file is opened before the while loop begins and can be changed when the program is running by pressing the “Save” button which temporary suspends the while loop. Closing the program will stop the data saving in the file.

The alarm system is implemented using comparators for the settled limits in the process and Boolean logic to enable the LEDs on the front panel and the “beep” sub-VI which causes the system to issue an audible sound.

The bang-bang controller is achieved by the Boolean logic from the alarm system which triggers a response from a Case Structure.

The Case Structure has 2 sub-diagrams, or cases, and one of these executes when the structure executes. The value wired to the selector terminal determines which case to execute and is a Boolean type.

The Case Structure sends a string to the output module with the “analog out” VI, which represents the command to switch between the two values of the output voltage needed to command the actuator to control the liquid nitrogen level.

The last sub-diagram controls the closing of COM-port 1 and sends the proper address, command, baud rate and other standard settings to stop the communication with the data acquisition and control modules. If an error occurs, this sub-diagram sends a message to the user specifying the type of the error.

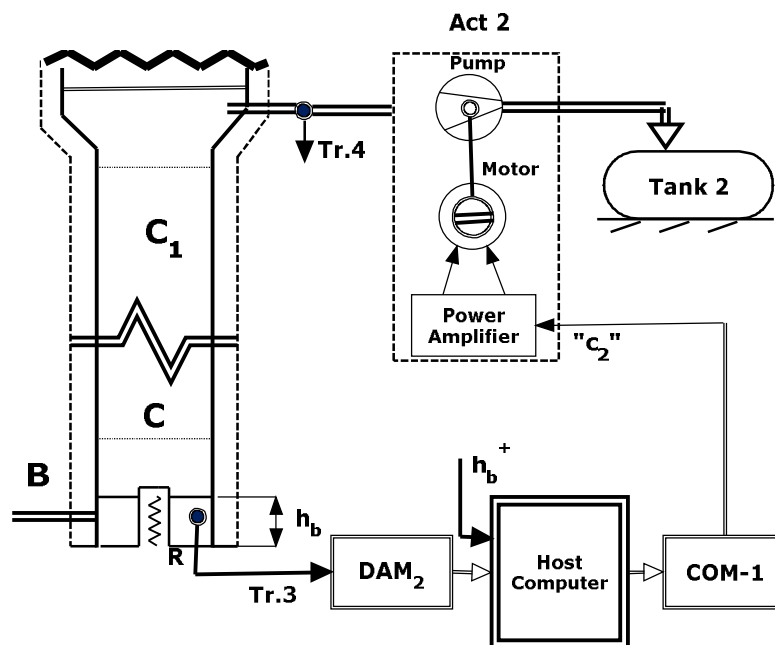


Fig. 6. Liquid carbon monoxide level control diagram

The second example illustrates the structure of the liquid carbon monoxide level (h_b) in boiler for $h_c \approx ct.$ and $W_{el} = ct.$ (Table 2). The disturbance is the inflow deviation $\Delta Q_{in}(t)$ and the manipulated variable is the “waste” outflow $Q_{out}(t)$. Under these circumstances, the level (h_b) remains constant.

The simplified level control system is illustrated in the Figure 6. The output voltage of the transducers (Tr3) controls the data acquisition module (DAM-2).

The control algorithm is in this case a continuous one in the PD form [3,11]:

$$C(s) = V_R \left(1 + \frac{\tau_d s}{T_{NS} + 1} \right); \tau_d \gg T_N. \tag{2}$$

The output continuous module (COM-1) converts the digital controller output in an analogic form “ c_2 ”, which activate the power amplifier (PA) of the actuator (Act 2), in order to change in a proper manner the outflow $Q_{out}(t)$.

Due to the special conditions of isotope separation column operation, to the complexity and dimensions of this plant, and to enable remote data transfers and data exchanges among users and to reduce the complexity in wiring connections, the authors propose in the future a networked control system. The goal is to implement the two general NCS configurations, listed as follows, and to compare the results.

A. Direct structure

The NCS in the direct structure is composed of a controller and a remote system containing the physical plant (isotope separation column), sensors and actuators (Table 2). The controller and the plant are physically placed at different locations and are directly linked by a data network in order to perform remote closed-loop control, as illustrated in Fig. 7 [8].

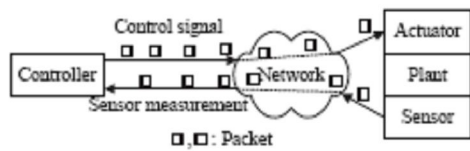


Fig. 7. NCS in direct structure

The control signal is encapsulated in a frame or a packet and sent to the plant via the network. The plant then returns the system output to the controller by putting the sensor measurement into a frame or a packet as well. In a practical implementation, multiple controllers can be implemented in a single hardware unit to manage multiple NCS loops in the direct structure.

B. Hierarchical structure

The basic hierarchical structure consists of a main controller and a remote closed loop system as depicted in Fig. 8. [8].

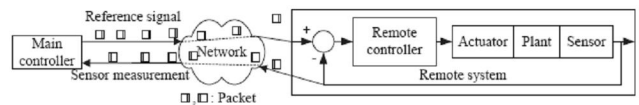


Fig. 8. NCS in hierarchical structure

Periodically, the main controller computes and sends the reference signal in a frame or a packet via a network to the remote system. The remote system then processes the reference signal to perform local closed-loop control and returns to the sensor measurement to the main controller for networked closed-loop control. The networked control loop usually has a longer sampling period than the local control loop since the remote controller supposes to satisfy the reference signal before processes the newly arrival reference signal. Similar to the direct structure, the main controller can be implemented to handle multiple networked control loops for several remote systems [9, 10].

Since an NCS operates over a network, data transfers between the controller and the remote system will induce network delays in addition to the controller processing delay. Fig. 9 shows network delays in the control loop, where r is the reference signal, u is the control signal, y is the output signal, k is the time index, and T is the sampling period. Most of networked control methodologies use the discrete-time formulation shown in Fig. 9.

Fig. 10 shows the corresponding timing diagram of network delay propagations. [8]

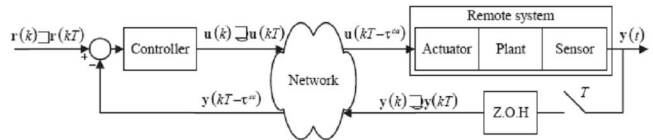


Fig.9. General NCS configuration and network delays for NCS formulations

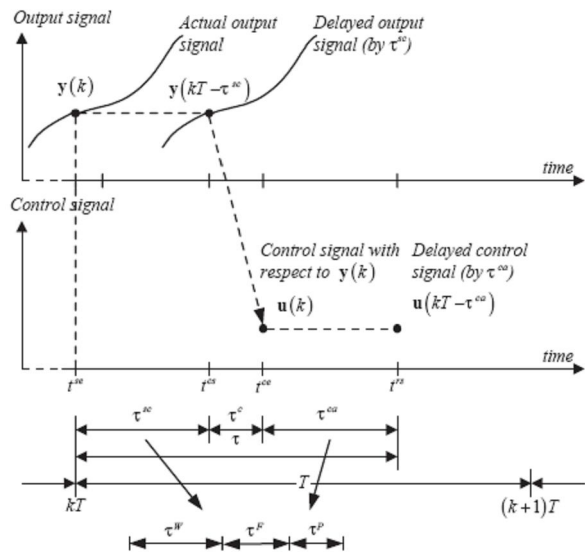


Fig.10. Timing diagram of network delay propagations

Delays in a control loop are widely known to degrade system performances of a control system, so are the network delays in an NCS. In the case of isotope separation column these delays do not affect all control loops, because ones of them are with great time delays, the control method being conceived especially for time delay systems [12].

V. APPLICATION SAMPLE, CONCLUSIONS

A short description of the column monitoring system for main variables that ensure an efficient and safe column operation is presented in Figure 9: liquid carbon monoxide level in boiler (h_b), liquid nitrogen level in condenser (h_c), vacuum jacket pressure, “waste” outflow carbon monoxide rate, electrical power dissipated in boiler.

In the present stage, the control functions are analyzed only

by simulation.

As possible results of the monitoring functions can be mentioned the record of the column operation history, a deeper understanding of the separation process and the possibility for an optimal column control as a future task [14].

One of the major challenges in this so-called networked control system (NCS) is the network-induced delay effect in the control loop. Network delays degrade the NCS control performance and destabilize the system. A significant emphasis has been on developing control methodologies to handle the network delay effect in NCS.

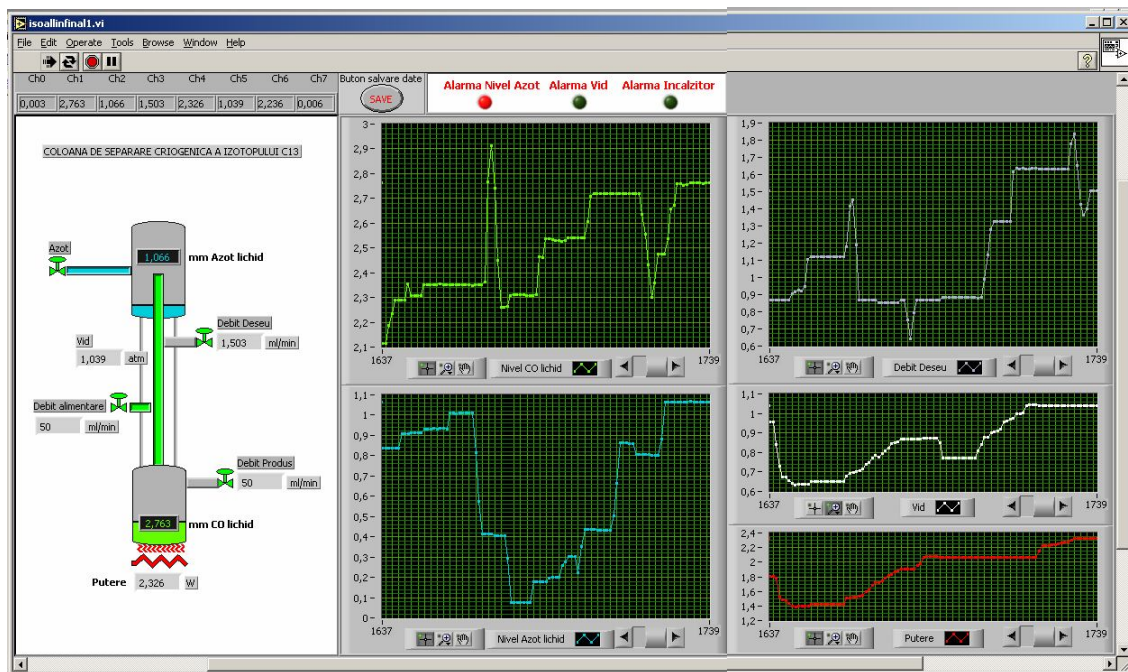


Fig. 9. Front panel of the monitoring application (sample)

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