ANALYSIS AND PREDICTION OF THE FLOODING PHENOMENON IN THE (^{13}C) CRYOGENIC SEPARATION COLUMN

Cristina Ioana Pop, Eva-Henrietta Dulf, Cl. Festila

Technical University of Cluj-Napoca
Department of Automatic Control
26-28 Gh. Baritiu Str., 400027 Cluj-Napoca, Romania
http://aut.utcluj.ro
e-mail: clement.festila@aut.utcluj.ro

Abstract: *The industrial distillation plants and the cryogenic distillation column for (^{13}C) isotope enrichment have in common the probability to appear an undesirable phenomenon: the flooding, when the plant efficiency decreases drastically. After a short description of the separation equipment (separation column), the authors deal with the arising of the flooding process. Based on simplified equations, the evolution of an intended flooding is studied, divided in more stages: normal mode of operation, pre-flooding, "discharge" period, flooding period and flooding damping. The next step of study will analyze the possibilities to avoid the flooding process, based on online prediction methods.*

Keywords: *cryogenic isotope separation, flooding analysis and simulation, pattern recognition, prediction.*

1. INTRODUCTION

The majority of chemical elements represent mixtures between various isotopes. For example, (^{12}C) is the basic carbon element, with a concentration of 98.9%, while (^{13}C) is the "heavier" isotope, with the natural abundance of 1.1% (Vasaru, 1968; Axente, et al., 1994). If some chemical compounds with higher abundance of (^{13}C) are available, detection of compounds with higher concentration of (^{13}C) allows valuable qualitative and quantitative measurements, very important in scientific research and industrial applications. The rising of heavier isotope concentration is known as isotope separation process (Axente, et al., 1994), based on various technologies.

The column for cryogenic (^{13}C) isotope separation is the case studied in this paper, using the equipment belonging to the National Institute of Research and Development for Isotopes and Molecular Technologies (INCDTIM) Cluj Napoca (Axente, et al., 1994). The schematic representation of the isotope separation column is given in figure 1, being based on the "three elements model" (Skogestad, 1997): boiler, tower and condenser.

At very low temperatures ($\approx -190^{\circ} C$), the vapor pressure of (^{12}CO) is higher than the pressure of (^{13}CO). The basic factor is the "separation coefficient α" (Vasaru, 1968), given by the ratio of the vapor pressures:

$$\alpha = \frac{p^{o}_{^{12}CO}}{p^{o}_{^{13}CO}} > 1$$

with $\alpha = 1.011$, $p^{o}_{^{13}CO}$ the vapor pressure of ^{13}CO and $p^{o}_{^{12}CO}$ the vapor pressure of ^{12}CO. This particular isotope separation technology is called "cryogenic distillation".

By liquid and gaseous phase equilibrium, the heavier isotope (^{13}C) accumulates in the liquid phase, while the (^{12}C) isotope is evacuated as waste (with the possibility of subsequent processing).

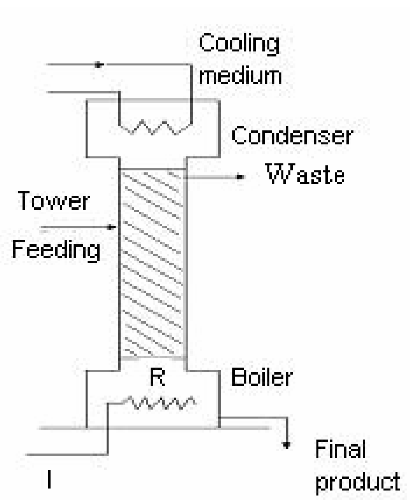

Fig. 1. Isotope separation column as a 3-element model

This principle of operation justifies the connection with the distillation process.

There exist several modes of operation of the cryogenic separation column. In the starting mode- without withdrawal- the column reaches the hydrodynamic steady state values. In the production mode, some (very small) quantity of compound (*P*), enriched in (^{13}C) isotope, is extracted. To satisfy the mass balance, the waste (*W*) and the product (*P*) flow rates equal the feeding (*F=P+W*) flow rate.

In a simplified control strategy, the manipulated variables are the boiling electrical power, the cooling efficiency and the waste flow rate (by total reflux, i.e. *P=0*). According to the structure and the dimensions of the column, the user desires a high rate of isotope separation (enrichment) and high product rate. The set-points (references) for different closed control loops are calculated in this way.

Practice has proven some limitations of the operating variables. In normal mode of operation, the gaseous phase is assumed to flow upward inside the numerous small "channels", forming the column packing material and having the same characteristic dimensions.

The liquid phase flows down the fictious walls of the "channels" and reduces the available cross-sectional area for the gas flow, thus causing a pressure drop (a differential pressure Δp). The pressure variations may be caused by other "disturbances": feed material quality/ composition, temperature distribution, flow-rates changes, etc.

An operation of the isotope separation column with as high as possible "charge" (flow rates, heating and cooling energy) is desired, but under these circumstances, the possibility to onset the flooding phenomenon increases, having disastrous consequences.

In a simplified manner, the flooding is defined as an excessive accumulation of liquid phase inside the column, caused by one of the following mechanisms (da Silva, et al., 2005; Parthasarathy, et al., 1999):

a. Excessive feed-flow, close or over the value of maximum column capacity;
b. Liquid downcomer malfunction (column dimensions, transition by different column diameters, etc.);
c. Excessive boiling power by lower condenser efficiency.

By flooding, the upward vapor velocity and pressure are high enough to "suspend" a liquid droplet. The flooding onset in column can be detected by:

a. A sharp increase in the pressure drop;
b. A sharp decline in the efficiency of the column and
c. A sharp decrease of the liquid phase in boiler.

A flooding predictor, in the **ideal** case, will keep the column point of operation close to the flooding (higher efficiency), but in a safe area. If some input/ internal variables change and the flooding can appear, the predictor alarms the operator - in open loop - or even gives a control signal- in closed loop- to avoid future flooding. So, the flooding predictor can be viewed as an **advanced process control strategy** that utilizes a pattern recognition system to identify pre-flooding conditions being possible to increase the stability and energy efficiency of the column operation.

2. HYDRODYNAMIC MODEL FOR THE FLOODING PROCESS EVOLUTION

For the hydrodynamic process it is important to analyze the evolution of the pressure, flow, temperature, etc. inside the column and the effects of the external variables (Axente, et al., 1994).

The vapor differential pressure (Δp) is a function of the hydraulic resistance given by the effective gas passing area (A_{ef}) of the evaporation rate given by the electrical heating power (P_{el}) and of the imbalance feed-waste rate (*F*, *W*) (Mphahlele and. Eldridge, 2006; da Silva, et al., 2005):

$$\Delta p = f_l(A_{ef}, P_{el}, F, W) \approx \approx \frac{k_a}{A_{ef}} + k_{el}P_{el} + k_f F - k_w W \qquad (1)$$

where ($k_a; k_{el}; k_f; k_w$) are proper coefficients.

For the sake of simplicity, the authors assume that:

$$k_f F - k_w W = 0 \qquad (2)$$

Because the effective gas passing area (A_{ef}) decreases by greater vapor pressure, the authors propose the linear dependence:

$$A_{ef} = A_0 - k_L \Delta p \qquad (3)$$

From equations (1) and (3) results:

$$\Delta p = \frac{k_a}{A_0 - k_L \Delta p} + k_{el}P_{el} + k_f F - k_w W \qquad (4)$$

Using the “long division” methods and under the assumption in (2), one approximates the previous equation, (4), to:

$$\Delta p \cong k_a \left[\frac{1}{A_0} + \frac{k_L}{A_0^2}(\Delta p) + \frac{k_L^2}{A_0^3}(\Delta p)^2 \right] + k_{el}\Delta P_{el} \qquad (5)$$

The liquid level in boiler depends on the imbalance between liquid downstream and vaporization:

$$h_B = f(L, V) \approx h_{B_0} + \varepsilon_c h_c - \varepsilon_{el} P_{el} \qquad (6)$$

where ($\varepsilon_c, \varepsilon_{el}$) are the condenser and boiler efficiency, (h_c) is the cooling level in condenser.

The liquid holdup (much greater compared to the vapor holdup) contains the boiler holdup (L_B), condenser holdup ($L_C \ll L_B$) and tower holdup (L_T). For the flooding analysis, the value (L_T) is important, for which the following approximation for the evolution is proposed:

$$L_T = L_{T_0} + \varepsilon_c h_c \qquad (7)$$

The equations (1)-(7) are valid in the time interval of liquid accumulation on the packing material. If the weight of the variable (L_T) is greater than the vapor pressure, a discharge of the packing holdup (L_T) will occur, the liquid flowing to the boiler.

During this short time period:

- the vapor pressure and the liquid holdup in tower decrease and
- the liquid level in the boiler and the effective gas passing are increase (Axente, et al., 1994; Mphahlele and. Eldridge, 2006).

Based on some experimental data (Vasaru, 1968; Axente, et al., 1994), the authors propose an exponential evolution of these variables for the discharge time period.

In order to stop the flooding process and to keep the initial presumption ($k_f F - k_w W = 0$), the electrical power (P_{el}) was drastically reduced so that the flooding process disappears. For instance, the vapor differential pressure (Δp) decreases below the operational value (Δp_0) and the liquid level in boiler reaches the maximum value.

From this moment, a very difficult and careful – usually manual- control is asked in order to “recover” the normal mode of operation of the isotope separation column.

3. SIMULATION OF THE FLOODING PROCESS EVOLUTION

The simulation using Matlab takes into account 5 phases of analysis:

1. the normal mode of operation (steady state, constant values of variables);
2. “pre - flooding”
3. “discharge”
4. typical flooding
5. flooding damping and
6. recovery process (without presentation)

The normal mode of operation, the steady state constant values are:

$(\Delta p_0, A_{ef,0} = A_0, L_{T_0}, h_{B_0}, \Delta P_{el,0})$.

For the “pre-flooding” and “typical flooding” phases, the specific equations are given in (3), (5), (6) and (7). For the “discharge” and “flooding damping” the following equations are used:

$$\begin{cases} \Delta p_k = \Delta p_{max}(\beta_{pd})^k \\ A_{ef,k} = A_{ef,min} + \beta_{Ad}(A_{ef,k-1}) \\ h_{B,k} = h_{B,min} + \beta_{Bd}(h_{B,k-1}) \\ L_{T,k} = L_{T,max} - \beta_{Td}(L_{T,k-1}) \end{cases} \quad (8)$$

The electrical power in p.u. is $P_{el} = 50$ p.u. in normal mode of operation, $P_{el} = 75$ p.u. in flooding process, $P_{el} = 0$ p.u. in the “flooding damping” phase.

Numerical values of the coefficients are:
$k_a = 9.8$; $A_{ef,0} = A_0 = 0.1$ (p.u.); $h_{B,0} = 80$ (p.u.);
$L_{T,0} = 10$ (p.u.); $k_L = 1.1 \cdot 10^{-2}$;
$\beta_{pd} = 0.96$; $\beta_{Ad} = 1.65 \cdot 10^{-2}$; $\beta_{Td} = 9 \cdot 10^{-2}$

Using the previous equations and numerical values of the coefficients, the simulated evolution of the main variables: $\Delta p(t); A_{ef}(t); L_T(t); h_B(t)$ and $P_{el}(t)$ are given in figure 3, for different time intervals:

1. Normal mode of operation: $\Delta t_1 = 15$ time units
2. Pre-flooding: $\Delta t_2 = 10$ time units
3. Discharge: $\Delta t_3 = 4$ time units
4. Typical flooding: $\Delta t_4 = 13$ time units
5. Flooding damping: $\Delta t_5 = 7$ time units

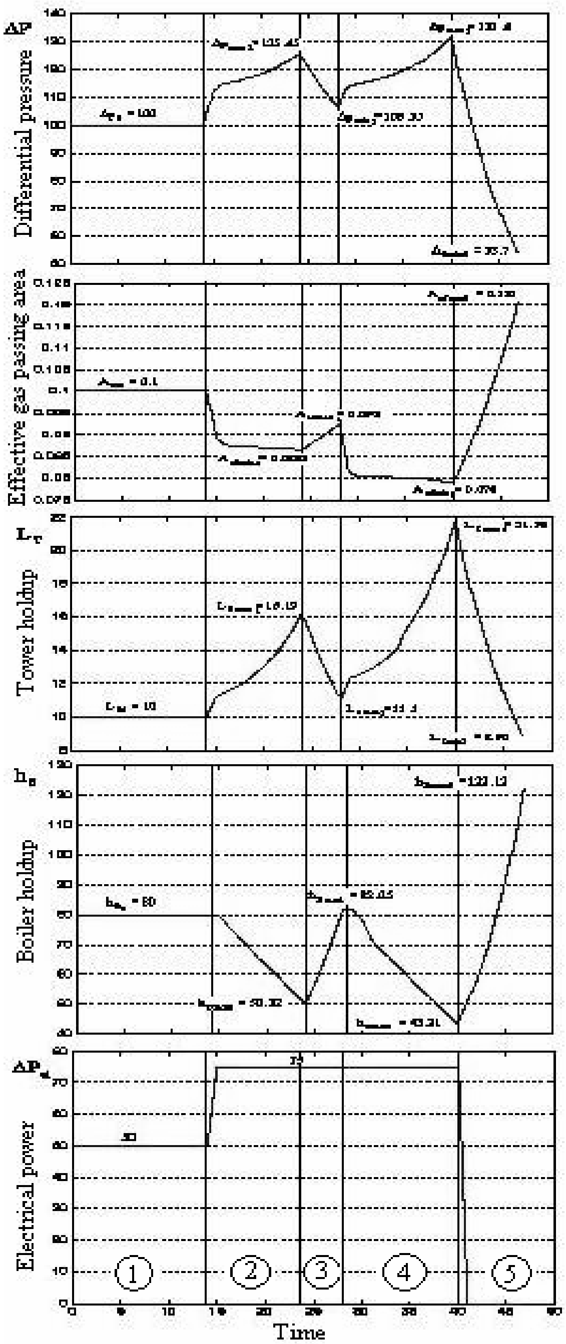

Fig. 2. Evolution of the differential pressure ($\Delta p(t)$), effective area ($A_{ef}(t)$), liquid holdup in the packing material ($L_T(t)$), liquid level in boiler ($h_B(t)$) under the control of the electrical power ($P_{el}(t)$)controlling the boiler

The column over-charging is the main cause which can start the flooding process in both cases. In the isotope separation, the enrichment process occurs during several days or weeks, in order to reach the desired concentration (6-8% for ^{13}C). The flooding leads to undesired mixture of waste material with the enriched material, thus a whole batch may be compromised.

In order to avoid this situation, in industrial distillation columns, a huge effort was made to predict and to avoid the flooding by measuring the evolution of the main variables: differential pressure, inflow and outflow, temperature distillation.

In the case of isotope separation process, the evolution of the liquid level (h_B) in boiler is another source of information which can be used in flooding prediction.

4. FLOODING PROCESS PREDICTION

If $\Phi(t)$ is the flow rate of the gaseous phase for carbon monoxide and (R_h) is the hydrodynamic resistance, in a simplified version one supposes that:

$$\Phi \approx k_{el} \cdot P_{el} \tag{9}$$

and

$$1/R_h = a - b \cdot P_{el} \tag{10}$$

where (P_{el}) is the electrical power dissipated in boiler and (k_{el}, a, b) are proper coefficients.

Using the long division method, the pressure drop is given by: the equation:

$$\Delta p = \frac{k_{el} \cdot P_{el}}{a - b \cdot P_{el}} \approx k_{el}\left(\frac{1}{a} + \frac{b}{a^2} P_{el} + \frac{b^2}{a^3} P_{el}^2 + \frac{b^3}{a^4} P_{el}^3\right) \cdot P_{el} =$$
$$= \frac{k_{el}}{a} P_{el} + k_{el} \frac{b}{a^2} P_{el}^2 + k_{el} \frac{b^2}{a^3} P_{el}^3 + k_{el} \frac{b^3}{a^4} P_{el}^4 =$$
$$= a_1 P_{el} + a_2 P_{el}^2 + a_3 P_{el}^3 + a_4 P_{el}^4 \tag{11}$$

Based on some experimental data (Vasaru, 1968; Axente, et al., 1994) by nominal feed flow rate, the differential pressure (Δp) for different values of the electrical power (P_{el}) is given in table 1.

Table 1. Differential pressure function of electrical power in the boiler

P_{el} (p.u.)	44	74	87	109
Δp (p.u.)	140	210	230	340

By polynomial interpolation, with fourth order approximation, the following values of the coefficients are obtained:

$a_1 = -1.48$;

$a_2 = 0.23$;

$a_3 = -3.63 \cdot 10^{-3}$;

$a_4 = 1.73 \cdot 10^{-5}$.

If the derivative ($\Delta p / \Delta P_{el}$) would be available, results:

$$D_p(P_{el}) = -1.48 + 0.46 P_{el} - 1.09 \cdot 10^{-2} \cdot P_{el}^2 + $$
$$+ 6.92 \cdot 10^{-5} \cdot P_{el}^3 \tag{12}$$

The technical solution of the column permits to "detect" the flooding and this was **observed** by $P_{el} \geq 85(p.u.)$.

The measured pressure evolution and the values obtained by interpolation are given in figure 3. The evolution of the derivative ($\Delta p / \Delta P_{el}$) is given in figure 4. It is not difficult to observe three different slopes:

$(\Delta p / \Delta P_{el})_1 = -0.0834$

$(\Delta p / \Delta P_{el})_2 = 0.0582$ (normal mode)

$(\Delta p / \Delta P_{el})_3 = 0.331$ (flooding)

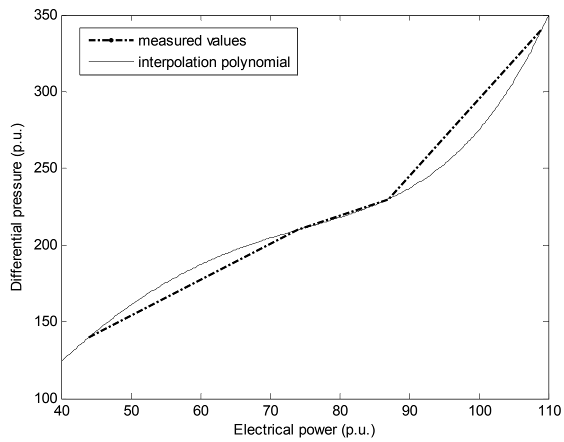

Fig. 3. Measured differential pressure evolution and the interpolation polynomial

Comparing the normal mode of operation with the flooding, the slope of the derivative has a much greater value:

0.331 >> 0.0582

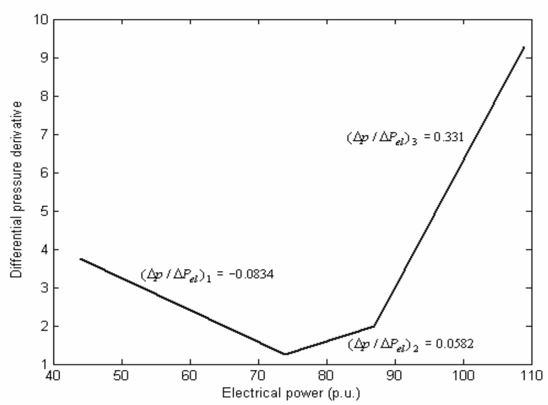

Fig. 4. Evolution of the differential pressure derivative

5. SIMPLIFIED VERSION OF THE FLOODING PREDICTOR BASED ON ESTIMATOR

In the particular case of the cryogenic isotope separation column, the flooding phenomenon may be predicted by the supervision of the slope (dp / dP_{el}) using the differential $(\Delta p)_k$ and $(\Delta P_{el})_k$.

The simplified algorithm is given by the following set of equations:

1. $\Delta p_k - \Delta p_{k-1}$ (13)
2. $P_{el,k} - P_{el,k-1}$ (14)
3. $\hat{D} = \dfrac{\Delta p_k - \Delta p_{k-1}}{P_{el,k} - P_{el,k-1}}$ (15)
4. $\hat{D} \gtrless (D)_{lim}$ (16)

Equations (13)-(16) are implemented in the structure given in figure 5.

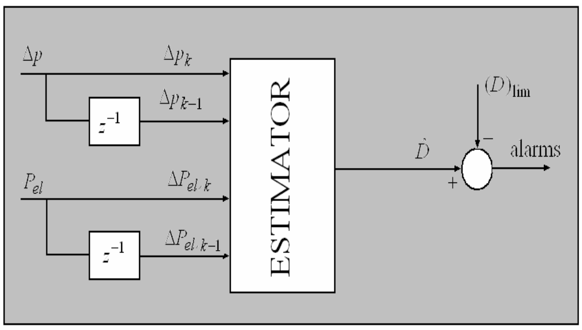

Fig. 5. Flooding predictor based on estimation of the differential

6. CONCLUSIONS

The disastrous consequences of the flooding phenomenon in distillation column justify the theoretical and practical effort of the specialists in order to predict/avoid this process. The general solution, at least according to actual references, is the predictive control theory and methods. In the particular case of the cryogenic distillation for (^{13}C) separation, where the information for the temperature distribution in the column is not available, accurate and prompt information about the differential column pressure using commercially available transducers and about the liquid level in the boiler, using a patented modern transducer (Gligan, et al., 2006), the evolution of the first and second derivative of these variables can predict and avoid the column flooding. The solution is modern, effective and low-cost.

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