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Robust-Predictive Control of (^{15}N) Isotope Separation Column

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Abstract: The chemical processes developed in the ^{15}N-isotope separation plant are very complex and many details are not yet known in totality. The authors are concerned with the problem of developing effective and readily implemental techniques for modelling and control of the isotope separation plant. In the present paper are presented a variant of Model Predictive Control (MPC) of the isotope separation plant.

Keywords: Isotope Separation Column, Model Predictive Control, Robustness

1 Introduction

In scientific research or industrial applications, the stable isotopes of O, N, C, etc are widely used [1]. In natural conditions, the ratio of concentration of the nitrogen 14N/15N is: 99.635 / 0.365 (%). In specific applications is asked a greater 'abundance' of the isotope (15N), if possible up to 99.9 (%).

There are known more methods to separate the (15N) isotope, both in laboratory scale or in large scale production. The actual paper studies the method invented by Spindel and Taylor about 1955, which is based on the 'chemical exchange' between liquid phase of nitric acid and gaseous phase of monoxide and dioxide of nitrogen:

$$\begin{cases} N^{15}O + HN^{14}O_3 \rightleftarrows N^{14}O + HN^{15}O_3 \\ N^{15}O_2 + HN^{14}O_3 \rightleftarrows N^{14}O + HN^{15}O_3 \end{cases} \tag{1}$$

Since the ratio of the reduced partition functions [3] is greater in the nitric acid molecule than in the oxide molecule, the (15N) isotope accumulates in acid.

2 The (^{15}N) Isotope Separation Column

In Figure 1 is depicted a simplified scheme of (15N) isotope separation column [1], with:

N, n – the mole fraction of 15N in liquid and pas phase [-];

L, G – specific flow of 15N in the recycled streams in [moles·s-1·m-2];

Hl, Hg – the holdup in [moles·m-3];

T – rate of transfer of 15N-isotope in [moles·s-1· m-3];

K – the transfer rate coefficient [moles·s-1·m-3];

α - the separation factor [-],

and $T = -k\left[N(1-n) - \alpha n(1-N)\right]$ [moles·s-1·m-3].

The enrichment process is governed by the equations [3]:

$$\begin{aligned} H_l \frac{\partial N}{\partial t} + L \frac{\partial N}{\partial z} &= +T \\ H_g \frac{\partial n}{\partial t} + G \frac{\partial n}{\partial z} &= -T \end{aligned} \tag{2}$$

The isotope exchange is achieved in column, endowed with a special packing [1]. The withdrawal (P) enriched in (15N) isotope is possible at the bottom side of the column. The column is fed with constant nitric acid flow (F), with a natural concentration (0.3654%) of (15N). In the bottom refluxer (R1) [2], using sulphur dioxide, the (NO) and (NO2) are 'generated' in accord to the chemical reaction:

$$\begin{cases} 2HNO_3 + 3SO_2 + 2H_2O = 3H_2SO_4 + 2NO \\ 2HNO_3 + SO_2 \qquad\qquad = H_2SO_4 + 2NO_2 \end{cases} \tag{3}$$

The top refluxer (R2), Figure 1, provides the reverse phase transformation:

$$\begin{cases} 3NO_2 + H_2O = 2HNO_3 + NO \\ NO + \frac{1}{2} O_2 = NO_2 \end{cases} \tag{4}$$

while the nitric acid flow is enriched in (15N) isotope.

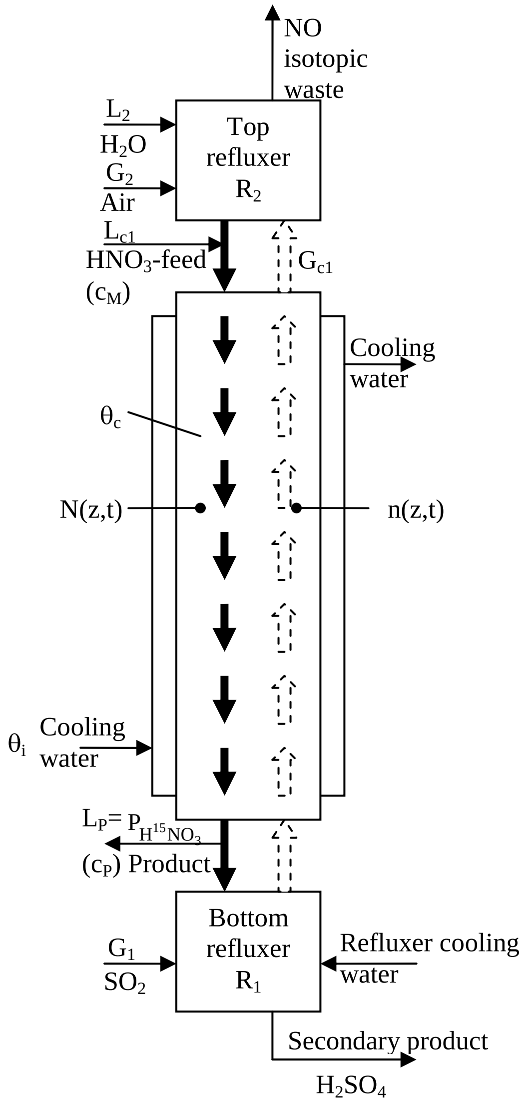

Figure 1
Simplified arrangement of the ^{15}N-isotope separation plant

3 The Simplified Mathematical Model for Control

It can be seen that the dynamics of the separation column and the adjoining equipment is rather complex. By using the traditional mass and heat balances, a high-order dynamic model can be derived. The resulting model would be highly nonlinear.

Another approach is to use a simple model of the process which describes its most important properties in combination with a robust controller.

Based on the data related in literature [2], the authors are searching for the main connections between input and output variables and between input variables and the parameters in equation (2).

The main input variables of the separation plant are [4]:

- rate-flow (Lc1) of the nitric acid with the mole ratio (cM);
- rate-flow (Lc2) of the water, or the ratio $r_{w/a} = L_{c2}/L_{c1}$
- temperature of the cooling water (θi);
- rate-flow of the sulphur dioxide (G1);
- rate-flow of the end-product (P) with the mole-fraction (cP).

The 'direct' output variables are:

- the middle temperatures in the column (θc);
- the 'position' of the reaction zone in the bottom-refluxer (hh);
- the evolution of the mole-fractions (N, n) of 15N in recycled streams (liquid and gas).

The 'indirect' outputs are the values of the parameters:

- (α) = separation factor
- and (k) = transfer rate coefficient.

Following the equations described in [5], the transfer matrix of the simplified separation plant is:

$$\begin{vmatrix}\theta_c\\ h_h\\ \Delta\alpha\\ \Delta k\end{vmatrix}=\begin{vmatrix}\dfrac{K_{\theta 1}\cdot e^{-s\tau_{m1}}}{(T_{\theta c}s+1)(T_{\theta j}s+1)} & \dfrac{K_{\theta g}\cdot e^{-s\tau_{mg}}}{T_{\theta c}s+1} & 0 & 0\\ 0 & \dfrac{K_{\theta 2}\cdot e^{-s\tau_{m2}}}{T_{\theta R}s+1} & 0 & 0\\ \dfrac{k_\theta\cdot K_{\theta 1}\cdot e^{-s\tau_{m1}}}{(T_{\theta c}s+1)(T_{\theta j}s+1)} & 0 & \dfrac{k_c}{T_\alpha s+1} & 0\\ 0 & 0 & 0 & \dfrac{\beta_k}{T_\beta s+1}\end{vmatrix}\cdot \qquad (5)$$

$$\cdot\begin{vmatrix}\theta_i\\ G_1\\ \Delta c_M\\ \Delta r_{w/a}\end{vmatrix}+\begin{vmatrix}\dfrac{-K_{\theta d}e^{-s\tau_{md}}}{T_{\theta d}s+1}\\ 0\\ 0\\ 0\end{vmatrix}\cdot\begin{vmatrix}d\\ 0\\ 0\\ 0\end{vmatrix}$$

4 The Control Problem

The control problem for isotope-separation plants differs from that of other manufacturing operations in the degree of complexity. From a theoretical point of view it is clear that the 'optimal' controller should use *all* available information (measurements of outputs and disturbances, plant model, expected model uncertainty, expected disturbances, known future reference changes, given constraints, etc.) to manipulate all 4 inputs (but avoiding large changes) to keep all 4 outputs close to their desired setpoints. Something close to this 'optimal 4×4 controller' can be realized using model predictive control (MPC). In addition to achieving better control performance, one then avoids the issue of selecting a control configuration, and the need to design special systems to handle input saturation (constraints) etc. The simulation results using MPC are presented in Figures 2 and 3.

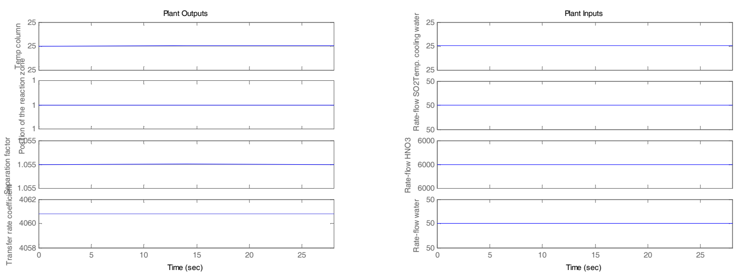

Figure 2

Simulation results of MPC control in standard operation conditions of the column

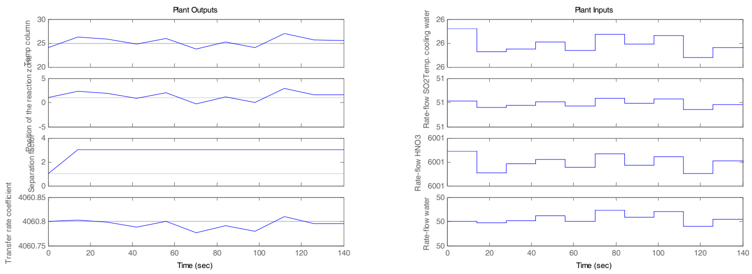

Figure 3

Simulation results of MPC control in special operation conditions of the column

It can be seen, that in some operation conditions the MPC control do not offer satifactory responses. For this reasons we derive an MPC controller that explicitly considers the external disturbances. The standard way to do this is to look at worst-case scenarios, which translates into solving a minimax problem [8]. One of the main ideas with the algorithms that are developed for the column is that they should be as close as possible to the original nominal MPC formulation. Changing from a nominal to a worst-case performance measure should not force you to leave the classical framework with finite horizon quadratic performance measures. Hence, the following minimax problem is used:

$$\begin{aligned} &\min_{u}\max_{w}\sum_{j=0}^{N-1} y_{k+j|k}^{T} Q y_{k+j|k} + u_{k+j|k}^{T} R u_{k+j|k} \\ &\text{subject to } \; u_{k+j|k} \in \boldsymbol{U}, \forall w \in \boldsymbol{W} \\ &\qquad\qquad\; x_{k+j|k} \in \boldsymbol{X}, \forall w \in \boldsymbol{W} \\ &\qquad\qquad\; w_{k+j|k} \in \boldsymbol{W} \end{aligned} \tag{6}$$

For robust performance analysis are provided LMI based functionalities.

The closed-loop responses for the proposed minimax controller and a nominal MPC controller shows that the minimax controller is successful in keeping the constrained output within its limits, in contrast to the nominal controller. The price paid is a slower step-response in the controlled output.

Conclusions

The isotope separation plant is complex equipment, nonlinear, with variable parameters and with very large equivalent time constant (of the order of days). The linear simplified mathematical model is, in opinion of authors, the single way to implement an effective control system.

The ‘optimal’ controller should use *all* available information (measurements of outputs and disturbances, plant model, expected model uncertainty, expected disturbances, known future reference changes, given constraints, etc.) to manipulate all 4 inputs (but avoiding large changes) to keep all 4 outputs close to their desired setpoints. Something close to this ‘optimal 4×4 controller’ can be realized using robust predictive control.

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