DYNAMICS OF PROCESS NETWORKS WITH RECYCLE AND PURGE: TIME SCALE SEPARATION AND MODEL DECOMPOSITION

Michael Baldea * **Prodromos Daoutidis** * **Aditya Kumar** **

* *Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, MN 55455*
** *GE Corporate Research and Development, P.O. Box 8, Schenectady, NY 12301*

Abstract: Process networks with recycle are well-known to exhibit complex dynamics and to present significant control challenges, due to the feedback interactions induced by the recycle streams. In this paper, we address the dynamic analysis and control of process networks with recycle and small purge streams used for removal of light inert components (feed impurities and/or reaction byproducts) from the recycle loop. We establish, through a singular perturbation analysis, that such networks exhibit a time scale separation in their dynamics, with the slow dynamics induced by the small amount of inert purged from the recycle loop. We also present a model reduction method for deriving a nonlinear low-order model of this slow dynamics which can be used to rationally address the control of the level of inerts in the network.

Keywords: singular perturbations, DAE systems, model reduction, nonlinear control

1. INTRODUCTION

Process networks consisting of reaction and separation units interconnected through material and energy recycle are the rule rather than the exception in the process industries. The dynamics and control of such networks present distinct challenges, since in addition to the nonlinear behavior of the individual units, the feedback interactions among these units, induced by recycle, typically give rise to more complex overall network dynamics (e.g. (Morud and Skogestad, 1994; Mizsey and Kalmar, 1996; Morud and Skogestad, 1998; Jacobsen and Berezowski, 1998; Bildea and Dimian, 1998; Bildea *et al.*, 2000; Pushpavanam and Kienle, 2001; Kiss *et al.*, 2002)). Design modifications (e.g. adding surge tanks between different units to attenuate disturbances propagating through the recycle) can in principle be employed to minimize these interactions, but these are not favored by the recent demands for lower capital and operating costs, and tighter process integration. At the same time, the efficient *transient operation* of such networks is becoming increasingly important, as the current environment of frequent changes in market conditions and economical objectives dictates frequent changes in operating conditions and targets (e.g. product grade transitions, feed switching, etc.) and tighter coordination of the plant-wide optimization and advanced control levels (Marquardt, 2000; Kulhavy *et al.*, 2000). A major bottleneck towards analyzing, optimizing and better controlling the dynamics of such networks is the often overwhelming size and complexity of their dynamic models, which make dynamic simulation computationally intensive, and the design of fully centralized nonlinear controllers on the basis of entire network models impractical (such controllers are almost invariably difficult to tune, expensive to implement and maintain, and sensitive to modeling er-

[1] Partial support for this work by ACS-PRF 38114-AC9 and NSF-CTS 0234440 is gratefully acknowledged

rors and measurement noise). Indeed, the majority of studies on control of networks with recycle (see e.g. (Luyben, 1993; Luyben and Floudas, 1994; Lyman and Luyben, 1996; Yi and Luyben, 1997)) are within a multi-loop linear control framework. In a different vein, a formal framework for stability analysis and stabilization of process networks, based on passivity and concepts from thermodynamics, was recently postulated in (Farschman *et al.*, 1998; Hangos *et al.*, 1999). The development of a systematic framework for analyzing the nonlinear dynamic interactions induced by recycle structures, and rationally accounting for them in the controller design clearly remains an important open problem.

In our previous work (Kumar and Daoutidis, 2002), we considered process networks with large material recycle compared to throughput. Within the framework of singular perturbations we established that the large recycle induces a time scale separation, with the dynamics of individual processes evolving in a fast time scale with weak interactions, and the dynamics of the overall system evolving in a slow time scale where these interactions become significant; this slow dynamics is usually nonlinear and of low order. Motivated by this, we proposed: i) a model reduction methodology for deriving nonlinear low-order models of the slow dynamics induced by large recycle streams, and ii) a controller design framework comprising of properly coordinated controllers in the fast and the slow time scales.

In this paper we focus on process networks with a recycle stream *and* a purge stream. The latter is typically used for the removal of inert components (feed impurities and/or reaction byproducts); the presence of the recycle can lead to accumulation of such inert components in the recycle loop, which can in turn be detrimental to the process operation (e.g. catalyst poisoning in the reactor) and the process economics (Belanger and Luyben, 1998; Luyben, 2000). Understanding the dynamics of the inert components is therefore critical and controlling the level of such components in the recycle structure can be a key operational objective.

In almost all such networks with purge streams, the magnitude of these streams is significantly smaller than the one of the throughput and/or the recycle streams, so that raw materials losses and/or pollution can be minimized. This suggests the possibility of a "core" dynamics over a much slower time scale compared to the dynamics of the individual process units and possibly the overall network dynamics. Developing an explicit nonlinear model of this slow dynamics can be beneficial both for analysis and evaluation purposes, and for model-based control.

Motivated by the above, we consider a prototype network comprising of a reactor (with gas effluent) and a separation system, with a gas recycle stream and a purge stream to remove the light inert components. Within the framework of singular perturbations we establish that such a network does exhibit a time scale separation, with the slow dynamics associated with the small purge flowrate. Furthermore, we describe a model reduction procedure which leads to an explicit nonlinear model of this slow dynamics, suitable for analysis and control, and highlight the analogies between the case of small purge and the case of large recycle treated in our previous work.

2. MODELING OF PROCESS NETWORKS WITH RECYCLE AND SMALL PURGE

Consider the network of a gas phase reactor and a condenser shown in Fig. 1.

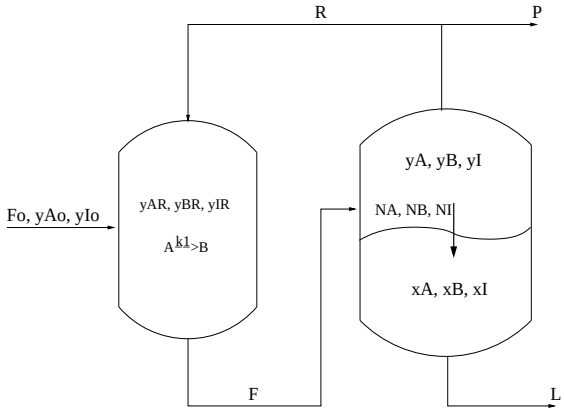

Fig. 1. Process network with recycle and purge.

Reactant A is fed at a molar flowrate F_o to the reactor, where a first-order irreversible reaction $A \rightarrow B$ takes place with a reaction rate constant k_1. The outlet stream from the reactor is fed to a partial condenser that separates the light unconverted reactant A from the heavy product B. The gas phase, rich in A is recycled back to the reactor. It is also assumed that a very volatile impurity I is present in the feed stream in small quantities. A (small) purge stream P is therefore used to remove this impurity from the recycle loop. The interphase mole transfer rates for the components A, B, I in the condenser are governed by rate expressions of the form: $N_j = k_j\alpha \left(y_j - \frac{P_j^S}{P} x_j \right)$, where $k_j\alpha$ denotes a mass transfer coefficient, y_j the mole fraction in the gas phase, x_j the mole fraction in the liquid phase, P_j the saturation vapor pressure of the component j (determined with an Antoine type relation) and P the pressure in the condenser. Assuming isothermal operation, the dynamic model of the network can be easily derived and has the form:

$$
\begin{aligned}
\dot{M}_R &= F_o + R - F \\
\dot{y}_{A,R} &= \frac{1}{M_R}[F_o(y_{0A} - y_{A,R}) + R(y_A - y_{A,R}) \\
&\quad -k_1 N_R y_{A,R}] \\
\dot{y}_{I,R} &= \frac{1}{M_R}[F_o(y_{0I} - y_{I,R}) + R(y_I - y_{I,R})] \\
\dot{M}_V &= F - R - N - P \\
\dot{y}_A &= \frac{1}{M_V}[F(y_{AR} - y_A) - N_A + y_A N] \\
\dot{y}_I &= \frac{1}{M_V}[F(y_{IR} - y_I) - N_I + y_I N] \\
\dot{M}_L &= N - L \\
\dot{x}_A &= \frac{1}{M_L}[N_A - x_A N] \\
\dot{x}_I &= \frac{1}{M_L}[N_I - x_I N]
\end{aligned} \tag{1}
$$

where $N = N_A + N_B + N_I$ and M_R, M_V, M_L denote the molar holdups in the reactor, vapor phase in the condenser and liquid phase in the condenser, respectively.

In order to facilitate a perturbation analysis of this model, the following assumptions are also introduced:

- The flowrates in the recycle loop are assumed to be $O(1)$.
- The ratio of the purge to the feed flowrate under steady state conditions is very small, or $\frac{P_s}{F_{os}} = \epsilon << 1$.
- The mole fraction of the inert in the feed is very small, or $y_{I0} = \alpha_I \epsilon$ where α_I is $O(1)$.
- The mass transfer rate for the inert component is very small, or $k_I a = \alpha_1 \epsilon^2$ where α_1 is $O(1)$.
- The inert is very volatile, or $\frac{P_I^S}{P} = \alpha_2 \frac{1}{\epsilon}$ where α_2 is $O(1)$.

Note that, based on steady state considerations, in order to remove an appreciable amount of the inert component from the recycle loop, the mole fraction of the inert in the vapor phase in the condenser, y_I, has to be $O(1)$. This implies that $O(\epsilon)$ moles of inert enter and leave the system through the feed and purge streams. Note also that the last two assumptions imply that negligible amount of inert leaves the recycle loop and exits through the liquid stream from the bottom of the condenser.

Based on the assumptions above, the dynamic model of the network takes the form:

$$
\begin{aligned}
\dot{M}_R &= F_o + R - F \\
\dot{y}_{AR} &= \frac{1}{M_R}[F_o(1 - \alpha_I \epsilon - y_{A,R}) \\
&\quad +R(y_A - y_{A,R}) - k_1 N_R y_{A,R}] \\
\dot{y}_{IR} &= \frac{1}{M_R}[F_o(\alpha_I \epsilon - y_{I,R}) + R(y_I - y_{I,R})] \\
\dot{M}_V &= F - R - (N_A + N_B) \\
&\quad -\alpha_1 \epsilon^2 y_I + \alpha_1 \alpha_2 \epsilon x_I - \epsilon F_{os} \frac{P}{P_s} \\
\dot{y}_A &= \frac{1}{M_V}[F(y_{AR} - y_A) - N_A \\
&\quad +y_A(N_A + N_B) \\
&\quad +y_A(\alpha_1 \epsilon^2 y_I - \alpha_1 \alpha_2 \epsilon x_I)] \\
\dot{y}_I &= \frac{1}{M_V}[F(y_{IR} - y_I) - (\alpha_1 \epsilon^2 y_I - \alpha_1 \alpha_2 \epsilon x_I) \\
&\quad +y_I(N_A + N_B) + y_I(\alpha_1 \epsilon^2 y_I - \alpha_1 \alpha_2 \epsilon x_I)] \\
\dot{M}_L &= (N_A + N_B) + \alpha_1 \epsilon^2 y_I - \alpha_1 \alpha_2 \epsilon x_I - L \\
\dot{x}_A &= \frac{1}{M_L}[N_A - x_A(N_A + N_B) \\
&\quad -x_A(\alpha_1 \epsilon^2 y_I - \alpha_1 \alpha_2 \epsilon x_I)] \\
\dot{x}_I &= \frac{1}{M_L}[\alpha_1 \epsilon^2 y_I - \alpha_1 \alpha_2 \epsilon x_I - x_I(N_A + N_B) \\
&\quad -x_I(\alpha_1 \epsilon^2 y_I - \alpha_1 \alpha_2 \epsilon x_I)]
\end{aligned} \tag{2}
$$

In generic form, the above model becomes:

$$
\dot{x} = f(x, u^l) + \epsilon[g(x) + g^o u^o + g^p u^p] \tag{3}
$$

where u^l denotes the scaled inputs corresponding to the *large* flow rates, u^o is a scaled input corresponding specifically to the large feed flow rate, u^p is a scaled input corresponding to the *small* purge flow rate, $g(x)$ is an $O(\epsilon)$ term corresponding to the rate of inert removal from the recycle loop by mass transfer, and f, g^o, g^p are appropriately defined vector functions.

It is evident that the above model has terms of $O(1)$ and $O(\epsilon)$ which suggests potentially a two time scale behavior. In what follows, we document the two time scale feature within the framework of singular perturbations, and address the derivation of reduced-order non-stiff approximate models of the fast and slow dynamics.

3. MODEL REDUCTION

We begin with a description of the fast dynamics. This is readily obtained by considering the dynamic model of Eq.1 in the limit as $\epsilon \to 0$:

$$
\begin{aligned}
\dot{M}_R &= F_o + R - F \\
\dot{y}_{A,R} &= \frac{1}{M_R}[F_o(1 - y_{A,R}) + R(y_A - y_{A,R}) \\
&\quad -k_1 M_R y_{A,R}] \\
\dot{y}_{I,R} &= \frac{1}{M_R}[-F_o y_{I,R} + R(y_I - y_{I,R})] \\
\dot{M}_V &= F - R - (N_A + N_B) \\
\dot{y}_A &= \frac{1}{M_V}[F(y_{AR} - y_A) - N_A \\
&\quad +y_A(N_A + N_B)] \\
\dot{y}_I &= \frac{1}{M_V}[F(y_{IR} - y_I) + y_I(N_A + N_B)] \\
\dot{N}_L &= (N_A + N_B) - L \\
\dot{x}_A &= \frac{1}{M_L}[N_A - x_A(N_A + N_B)] \\
\dot{x}_I &= -\frac{1}{M_L}[x_I(N_A + N_B)]
\end{aligned} \tag{4}
$$

or in its generic form:

$$\dot{x} = f(x, u^l) \tag{5}$$

This is a non-stiff model that approximates the dynamics in the original (fast) time scale t. The steady-state conditions for this system have the form $0 = f(x, u^l)$ or more specifically:

$$\begin{aligned}
0 &= F_o + R - F \\
0 &= F_o(1 - y_{A,R}) + R(y_A - y_{A,R}) \\
&\quad -k_1 N_R y_{A,R} \\
0 &= -F_o y_{I,R} + R(y_I - y_{I,R}) \\
0 &= F - R - (N_A + N_B) \\
0 &= F(y_{AR} - y_A) - N_A + y_A(N_A + N_B) \\
0 &= F(y_{IR} - y_I) + y_I(N_A + N_B) \\
0 &= (N_A + N_B) - L \\
0 &= N_A - x_A(N_A + N_B) \\
0 &= x_I
\end{aligned} \tag{6}$$

Note that not all of these constraints (or equivalently the differential equations in Eq.4) are linearly independent. Specifically, it can be shown that there exist only 8 linearly independent constraints. This is consistent with the fact that these constraints correspond to steady state constraints in the limit as the purge flowrate *and* the feed impurity become zero. In this limit, the inert moles leaving the reactor and the condenser are identical, hence the redundant constraint.

The above observation implies that the steady state condition in this fast time scale does not specify isolated equilibrium points, but rather a one-dimensional equilibrium manifold, which confirms the presence of the two time scale behavior.

Note also that in this time scale, only the large flowrates F, R, L affect the dynamics and can be used for addressing control objectives such as stabilization of holdups, production rate and product quality. The purge flowrate has, of course, no effect on the dynamics in this fast time scale.

Turning now to the slow dynamics, let us define a slow time scale $\tau = t\epsilon$. Considering the limit $\epsilon \to 0$, we obtain a description of the slow dynamics of the form:

$$\begin{aligned}
\frac{dM_R}{d\tau} &= lim_{\epsilon\to 0}\frac{1}{\epsilon}(F_o + R - F) \\
\frac{dy_{AR}}{d\tau} &= lim_{\epsilon\to 0}\frac{1}{\epsilon M_R}[F_o(1 - y_{A,R}) \\
&\quad +R(y_A - y_{A,R}) - k_1 N_R y_{A,R}] \\
&\quad -\frac{1}{M_R}F_o\alpha_I \\
\frac{dy_{IR}}{d\tau} &= lim_{\epsilon\to 0}\frac{1}{\epsilon M_R}[-F_o y_{I,R} \\
&\quad +R(y_I - y_{I,R})] + \frac{1}{M_R}F_o\alpha_I \\
\frac{dM_V}{d\tau} &= lim_{\epsilon\to 0}\frac{1}{\epsilon}[F - R - (N_A + N_B)] \\
&\quad -F_{os}\frac{P}{P_s} \\
\frac{dy_A}{d\tau} &= lim_{\epsilon\to 0}\frac{1}{\epsilon M_V}[F(y_{AR} - y_A) - N_A \\
&\quad +y_A(N_A + N_B)] \\
\frac{dy_I}{d\tau} &= lim_{\epsilon\to 0}\frac{1}{\epsilon M_V}[F(y_{IR} - y_I) \\
&\quad +y_I(N_A + N_B)] \\
\frac{dM_L}{d\tau} &= lim_{\epsilon\to 0}\frac{1}{\epsilon}[N_A + N_B - L] \\
\frac{dx_A}{d\tau} &= lim_{\epsilon\to 0}\frac{1}{\epsilon M_L}[N_A - x_A(N_A + N_B)] \\
\frac{dx_I}{d\tau} &= lim_{\epsilon\to 0}\frac{-1}{\epsilon M_L}[x_I(N_A + N_B)]
\end{aligned} \tag{7}$$

subject to the quasi steady state constraints of Eq.6.

Note that the $O(\epsilon)$ terms in the original system description have become $O(1)$, and thus significant, in this slow time scale. Note also that the combination of $O(1)$ terms in the original model has given rise to finite, yet indeterminate limits in this slow time scale; we will denote the vector of these unknown terms by z.

In generic form, the model of the slow dynamics has the form:

$$\begin{aligned}
\frac{dx}{d\tau} &= z + g^o u^o + g^p u^p \\
0 &= f(x, u^l)
\end{aligned} \tag{8}$$

where

$$z = lim_{\epsilon\to 0}\frac{1}{\epsilon} f(x, u^l) \tag{9}$$

The model of the slow dynamics of the system comprises thus of a set of coupled differential and algebraic equations of non-trivial index, as the variables z are implicitly fixed by the quasi steady state constraints, rather than explicitly specified in the dynamic model. Indeed, this model of the slow dynamics has a well-defined index only if the flowrates u^l which appear in the algebraic constraints that determine the constraint state-space are specified as functions of the state variables x, via a control law $u^l(x)$. It can then be shown that the index of the above DAE system is exactly 2, which implies that the dimension of the underlying ODE system is 1. This system captures the slow dynamics induced by the small purge and small feed impurity, and can be used to address the control of the impurity level using the small purge stream in this slow time scale.

An explicit ODE representation of this DAE system can be obtained be employing a coordinate change of the form:

$$\begin{bmatrix} \zeta \\ \eta \end{bmatrix} = T(x) = \begin{bmatrix} \phi(x) \\ f(x, u^l(x)) \end{bmatrix} \tag{10}$$

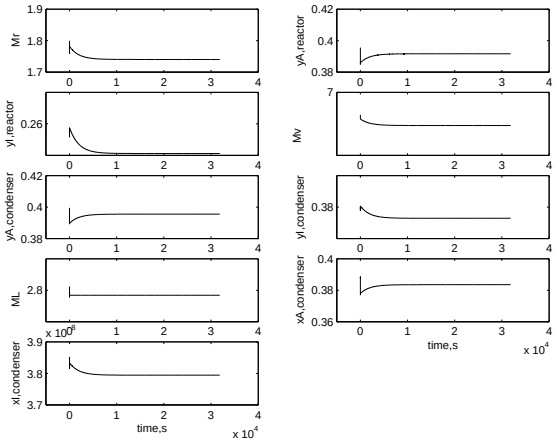

Fig. 2. Time responses of all state variables

In these new coordinates, the model of the slow dynamics has the form

$$\frac{d\zeta}{d\tau} = \frac{\partial\phi}{\partial x}z(\zeta) + \frac{\partial\phi}{\partial x}(\zeta)g^o u^o + + \frac{\partial\phi}{\partial x}(\zeta)g^p u^p \qquad (11)$$

$$\eta \equiv 0$$

Note that it is possible to chose the function $\phi(x)$ so that $\frac{\partial\phi}{\partial x}z = 0$. In this case, the variable ζ evolves independently of the 'algebraic' variables z, and is essentially a true "slow" variable in the system (whereas the original state variables exhibit both fast and slow dynamics). A meanigful choice of the function $\phi(x)$ which achieves this is $\phi(x) = M_R y_{IR} + M_V y_I$, i.e. the total impurity holdup in the recycle loop.

Notice that in the above analysis, the quantity of inert that is recycled is much larger than the inert throughput. The presence of a single slow mode associated with the inert is in complete agreement with the analysis of (Kumar and Daoutidis, 2002), which predicts a slow model of dimension equal to the number of components in the recycle loop.

4. SIMULATION STUDY

In what follows we consider a specific network of the form shown in Fig.1. The parameter values and the nominal steady states are given in Table 1. The objective is to verify the results of the analysis presented above.

As an initial simulation run, we considered an 'open-loop' experiment, whereby the three holdups are controlled by proportional controllers and we perturbed slightly the state variables from their steady state values. Fig. 2 shows the responses of all state variables. Observe that all state variables exhibit a fast transient, followed by a slow approach to steady state, which is indicative of the two time scale behavior of the system.

Table 1: Nominal values for process parameters

F_0	1.000	y_{0I}	0.0030
R	2.082	y_{AR}	0.3916
F	3.082	y_{IR}	0.2552
L	0.993	y_A	0.3955
P	$7.8{\cdot}10^{-3}$	y_I	0.3764
M_R	1.740	x_A	0.3835
M_V	6.789	x_I	$4.309 \cdot 10^{-7}$
M_L	2.784		

P-Controller gains	$K_{p,V}$	0.9	
	$K_{p,L}$	0.9	
	$K_{p,R}$	0.9	

Antoine parameters	A	B	C
for A:	15.04	273	0
for B:	15.04	273	0
for I:	17.65	273	0

ρ_L	54889	$k_A\alpha$	1
V_r	$9.25 \cdot 10^{-3}$	$k_B\alpha$	3
V_c	$5.34 \cdot 10^{-3}$	$k_I\alpha$	$1 \cdot 10^{-6}$
T_r	473	k_1	0.9
T_c	273		

Fig. 3 shows the evolution of the total inert holdup for the same simulation run; note that this variable exhibits dynamics only in the slow time scale, a fact which is consistent with it being a true slow variable.

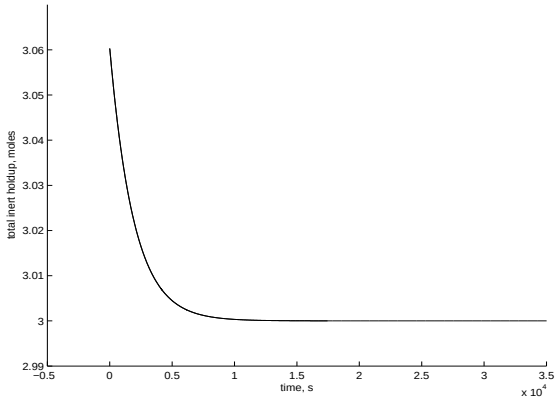

Fig. 3. Evolution of the total inert holdup

Fig. 4 illustrates the eigenspectrum of the dynamic model of the network, linearized at the nominal steady state. Observe the presence of a single eigenvalue very close to the imaginary axis, with the remaining eigenvalues further left in the complex plane.

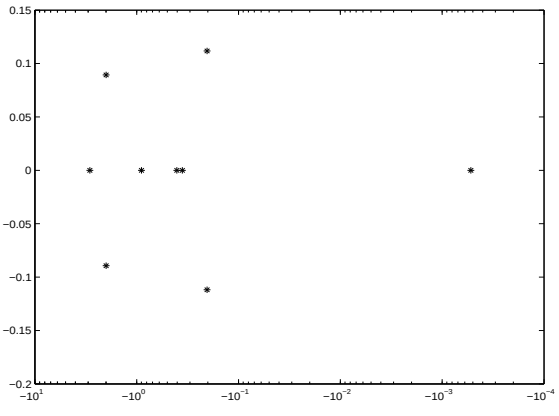

Fig. 4. Eigenspectrum of the linearized system

The model of the slow dynamics derived in the paper was used as the basis for synthesizing a nonlinear inpout/output linearizing controller which manipulates the purge flowrate to induce the following first order response for the total inert holdup ϕ :

$$\phi + \beta \frac{d\phi}{d\tau} = v \tag{12}$$

with $\beta = 4000$ and integral action imposed on the $v - \phi$ dynamics. Fig. 5 illustrates a closed-loop simulation run which illustrates the effectiveness of this controller in tracking a change in the setpoint of the inert holdup. Finally, Fig. 6 illustrates the

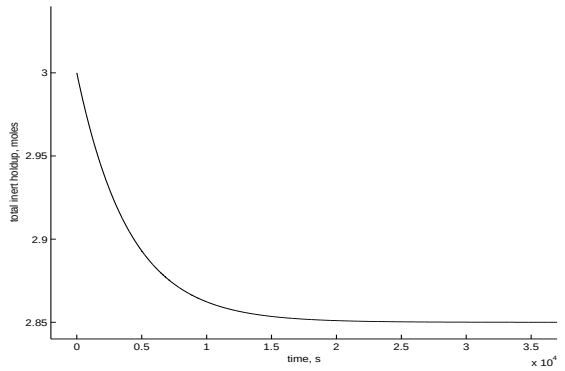

Fig. 5. Closed loop response of the controller to a 5% decrease in the inert holdup setpoint

closed-loop response of the purge flowrate for the same change in the setpoint of the inert holdup.

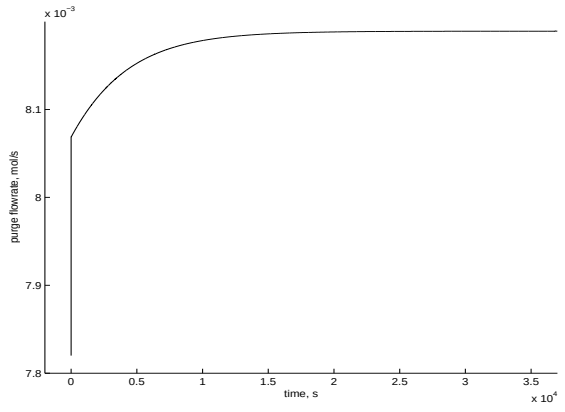

Fig. 6. Purge flowrate change for a 5% decrease in the inert holdup setpoint

5. CONCLUSIONS

In this work, we have shown that the presence of a small purge stream for the removal of an inert component from a process network with recycle, introduces a two-time scale behavior. The slow dynamics of the network was shown to be one-dimensional and directly associated with the total inert holdup. A state space realization of this slow dynamics was derived and employed in the synthesis of an input-output linearizing controller with integral action for the total inert holdup. The performance of the controller was tested by numerical simulation, indicating good setpoint tracking capabilities.

6. REFERENCES

Belanger, P. W. and W. L. Luyben (1998). Plantwide design and control of processes with inerts. 1. light inerts. *Ind. Eng. Chem. Res.* **37**(2), 516–527.

Bildea, C.S., A.C. Dimian and P.D. Iedema (2000). Nonlinear behavior of reactor-separator-recycle systems. *Comput. chem. Engng.* **24**, 209–214.

Bildea, C.S. and A.C. Dimian (1998). Stability and multiplicity approach to the design of heat-integrated pfr. *AIChE J.* **44**, 2703–2712.

Farschman, C. A., K. P. Viswanath and B. E. Ydstie (1998). Process systems and inventory control. *AIChE J.* **44**(8), 1841–1857.

Hangos, K. M., A. A. Alonso, J. D. Perkins and B. E. Ydstie (1999). Thermodynamic approach to the structural stability of process plants. *AIChE J.* **45**(4), 802–816.

Jacobsen, E. and M. Berezowski (1998). Chaotic dynamics in homogeneous tubular reactors with recycle. *Chem. Eng. Sci.* **23**, 4023–4029.

Kiss, A.A., C.S. Bildea, A.C. Dimian and P. D. Iedema (2002). State multiplicity in cstr-separator-recycle polymerization systems. *Chem. Eng. Sci.* **57**, 535–546.

Kulhavy, R., J. Lu and T. Samad (2000). Emerging technologies for enterprise optimization in the process industries. In: *Preprints of Chemical Process Control - 6.* Tucson, Arizona. pp. 411–422.

Kumar, A. and P. Daoutidis (2002). Dynamics and control of process networks with recycle. *J. Proc. Contr.* **12**, 475–484.

Luyben, M. L. and C. A. Floudas (1994). Analyzing the interaction of design and control - 2. reactor-separator-recycle system. *Comput. chem. Engng.* **18**(10), 971–994.

Luyben, W. L. (1993). Dynamics and control of recycle systems. parts 1-4.. *Ind. Eng. Chem. Res.* **32**, 466–486,1142–1162.

Luyben, W. L. (2000). Design and control of gas-phase reactor/recycle processes with riversible exothermic reactions. *Ind. Eng. Chem. Res.* **39**, 1529–1538.

Lyman, P. R. and W. L. Luyben (1996). Production rate changes in a ternary two-recycle process. *Ind. Eng. Chem. Res.* **35**(7), 2198–2203.

Marquardt, W. (2000). Fundamental modeling and model reduction for optimization based control of transient processes. In: *Preprints of Chemical Process Control - 6.* Tucson, Arizona. pp. 30–60.

Mizsey, P and I. Kalmar (1996). Effects of recycle on control of chemical processes. *Comput. chem. Engng.* **20**, S883–S888.

Morud, J. and S. Skogestad (1994). Effects of recycle on dynamics and control of chemical processing plants. *Comput. chem. Engng.* **18**, S529–S534.

Morud, J. and S. Skogestad (1998). Analysis of instability in an industrial ammonia reactor. *AIChE J.* **44**, 888–895.

Pushpavanam, S. and A. Kienle (2001). Nonlinear behavior of an ideal reactor separator network with mass recycle. *Chem. Eng. Sci.* **57**, 2837–2849.

Yi, C. K. and W. L. Luyben (1997). Design and control of coupled reactor/column systems - parts 1-3.. *Comput. chem. Engng.* **21**(1), 25–68.