Nonlinear model-based control of a solution copolymerization continuous stirred tank reactor

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SIMULATION OF THE NONLINEAR CONTROL OF A SOLUTION COPOLYMERIZATION CONTINUOUS STIRRED TANK REACTOR (CSTR)

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Abstract – The control of solution copolymerization continuous stirred tank reactor is one of the most complex problems of chemical industry. As a first approach, this system had a mathematical model of eight interactive nonlinear equations, which require an appropriate decoupling and linearization strategy. To do so, we use state feedback linearization. Moreover, three equations have been added in order to enable the control of the number and weight average molecular weights.

Four cases were distinguished in the present study, depending on the considered outputs set. From an industrial application perspective, the robustness of the controllers as well as the change of the set point have received a particular attention. The solution copolymerization of methyl methacrylate and vinyl acetate is used as an illustrative example where ethyl acetate is the solvent and azobisisobutyronitrile (AIBN) is the initiator.

Key words: Continuous Solution Copolymerization, State Feedback Linearization, Input Multiplicity, Robustness.

1. INTRODUCTION

Solution copolymerization is an important industrial process in the manufacture of commodity and engineering plastics. However, the control of copolymerization and polymerization reactors is a tough and complex problem. Some of the reasons of this complexity are:

- Kinetic and modelling inherent difficulties;
- Operating phenomenons such the “gel effect”;
- The extreme sensitivity of the steady state to small changes in parameter values or operating conditions;
- The highly interactive non-linear dynamic behaviour.

Since polymerization and copolymerization are closely linked; series of papers [1-12] dealing with polymerization were used. Moreover, we recalled few papers [12-13] dealing with semicontinuous emulsion copolymerization. The rather sparse prior literature on solution copolymerization has been discussed previously [16] so that only a few points will be noted here. As a matter of fact the papers of J. P. Congaldis, J. R. Richards, W. H. Ray (1989) and S. Padilla, J. Alvarez (1997) are the only works in which, the problem of solution copolymerization control has been addressed.

From a practical point of view the key control objectives of the copolymerization reactor are:

- The linearization;
- The compensation of interaction;
- The preclusion of input multiplicity;
- The robustness of the controller (i.e., tolerance to modelling and tuning errors).

The previous works indicate that the input multiplicity issue should be the central point in the study of the solution copolymerization control problem.

It is worth noting that the copolymer composition, conversion and chain length distribution determine the properties and the quality attributes of the product. As a result, those parameters must be controlled directly or indirectly.

In this short paper the guidelines of solution copolymerization control are given including the mathematical model of the process, the control strategy and some simulation results. On the other hand, we start by extending the main work, in which a systematic linearization and the zero dynamics analysis have been carried out [16].

We emphasize that unlike the previous work of S. Padilla, J. Alvarez (1997), a model including the equation of the second moment was used. This improvement aims at enabling the control of the number average weight.

DEVELOPEMENT OF PROCESS MODEL

Let us consider a reactor where two monomers A and B are continuously introduced as well as the initiator and the solvent. This reactor is assumed to be a jacketed, well-mixed tank. A coolant flows through the jacket to remove the heat of copolymerization. Copolymer, solvent,
unreacted monomers, and eventually initiator flow out of the reactor to a separator.

\[ M_{1c} = \frac{q_1}{V} M_{1e} = \frac{M_1}{V} (q_1 e_{m1} + q_2 e_{m2} + q_S e_S) - R_1 - M_1 (e_1 R_1 + e_2 R_2) \]  
\[ M_2 = \frac{q_2}{V} M_{2e} = \frac{M_2}{V} (q_1 e_{m1} + q_2 e_{m2} + q_S e_S) - R_2 - M_2 (e_1 R_1 + e_2 R_2) \]  
\[ \dot{I} = \frac{q_1}{V} I_c - \frac{I}{V} (q_1 e_{m1} + q_2 e_{m2} + q_S e_S) - R_1 - I (e_1 R_1 + e_2 R_2) \]  
\[ \dot{S} = \frac{q_S}{V} S_c - \frac{S}{V} (q_1 e_{m1} + q_2 e_{m2} + q_S e_S) - S (e_1 R_1 + e_2 R_2) \]  
\[ \dot{P}_1 = -\frac{P_1}{V} (q_1 e_{m1} + q_2 e_{m2} + q_S e_S) + R_1 - P_1 (e_1 R_1 + e_2 R_2) \]  
\[ \dot{P}_2 = -\frac{P_2}{V} (q_1 e_{m1} + q_2 e_{m2} + q_S e_S) + R_2 - P_2 (e_1 R_1 + e_2 R_2) \]  
\[ \dot{T} = q_1 \Omega_1 + q_2 \Omega_2 + q_S \Omega_S - \varphi (T - T_C) + \beta_1 + \beta_2 \]  
\[ \dot{V} = e_{m1} q_1 + e_{m2} q_2 + e_S q_S - q + V (e_1 R_1 + e_2 R_2) \]  

The kinetic equations are based on the “terminal model” [15]. The free radical kinetic mechanism includes initiation, propagation, termination by coupling, termination by disproportionation, and chain transfer to monomer [16].

Both monoisothermal operation and variable material and energy balances, while the equations of the zeroth, first, and second moments of the molecular weight distribution are obtained from the generating function technique [10], [16].

Finally the mathematical model which describes the process [18], is given by the following equations:

\[ \dot{\eta}_0^p = \frac{\eta_0^p - \eta_0^p}{\theta} + \frac{k_{i11}}{2} (\eta_0^M_0 + \eta_0^M_0) + \frac{k_{i12}}{2} (\eta_0^M_0 + \eta_0^M_0) + L_1 \eta_0^M_0 + L_2 \eta_0^M_0 \]  
\[ \dot{\eta}_1^p = \frac{\eta_1^p - \eta_1^p}{\theta} + k_{i11} (\eta_1^M_0 + \eta_0^M_0 + \eta_1^M_0 + \eta_0^M_0) + L_1 \eta_0^M_0 + L_2 \eta_0^M_0 \]  
\[ \dot{\eta}_2^p = \frac{\eta_2^p - \eta_2^p}{\theta} + k_{i12} (\eta_2^M_0 + \eta_0^M_0 + \eta_2^M_0 + \eta_0^M_0 + \eta_1^M_0 + \eta_0^M_0) + k_{i11} (\eta_1^M_0 + \eta_0^M_0 + \eta_1^M_0 + \eta_0^M_0) + L_1 \eta_0^M_0 + L_2 \eta_0^M_0 \]  

It appears from the different equations that the mathematical model is strongly interactive and non-linear. So, it requires an appropriate decoupling and linearization strategy, before the control implementation.

**REACTOR CONTROL PROBLEM**

To address the control problem we must define the control (manipulated) inputs and the control (regulated) outputs. First, there are eleven states in the mathematical model and four control inputs \((q_1, q_2, q_3, q_4)\).

Two problems appear as a result of the difference between the number of states and the number of control inputs:

1. The choice of the control outputs;
2. The dynamics of the residual states (zero dynamics)

We remind that the issue is to control the following outputs or product attributes:

\[ P_r = \left( \frac{M_{1c} P_1 + M_{2c} P_2}{P_c + P_2} \right) q \]  
\[ C_1 = \frac{P_1}{P_1 + P_2} \]  
\[ X_{av} = \frac{P_1 + P_2}{M_1 + M_2 + P_1 + P_2} \]  
\[ M_{pw} = \frac{q_p}{q_0} \]  
\[ M_{pn} = \frac{q_p}{q_0} \]

where:

\[ C_1 : \text{instantaneous copolymer composition of monomer I} \]
\[ P_r : \text{nominal production rate, kg/s} \]
\[ M_{pw} : \text{average molecular weight, kg/mmol} \]
\[ M_{pn} : \text{number average weight, kg/mmol} \]
\[ X_{av} : \text{overall conversion} \]

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It should be noted that the last parameters depend mainly on: $M_0$, $M_2$, $P_2$, $P_2$. 

From practical reasons, we specify three control outputs ($S$, $T$, $V$) and the first two are to be determined between $M_0$, $M_2$, $P_2$, $P_2$, “Fig.2”.

To summarize, the study of the control problem of continuous free-radical copolymerization reactor in the present work is conducted under the following assumptions and requirements:

1. Temperature $T$, volume $V$, solvent concentration $S$, and two concentration (to be determined) from the set ($M_0$, $M_2$, $P_2$, $P_2$), are available for feedback control.

2. Regulation of the nominal production rate, the overall conversion, the copolymer composition of monomer 1, the number average, and average molecular weights must be accomplished indirectly.

3. The resulting control scheme must give a robust linear feedback controller.

4. The tuning of the multivariable (five inputs, five outputs) controller must be translated into the tuning of five closed-loop, non-interactive, linear single-input, single-output (SISO) systems.

On-line measurements of $T$ and $V$ can be done with standard instrumentation. The compositions $S$, $M_1$, $M_2$, $P_1$, and $P_2$ can be determined from on-line measurements of density, refractive index, boiling point, and viscosity.

According to the preceding assumptions we distinguish four cases:

First case (I) outputs : $P_1$, $P_2$, $S$, $T$, $V$

Second case (II) outputs : $M_1$, $M_2$, $S$, $T$, $V$

Third case (III) outputs : $M_2$, $P_1$, $S$, $T$, $V$

Fourth case (IV) outputs : $M_1$, $P_2$, $S$, $T$, $V$

For each case we implement a state feedback linearization according to “Fig.3”. The non-linear system (reactor model) gives a state map from which we obtain simple outputs or outputs functions. A state feedback to the linearizing and decoupling law gives an input-output non-interactive and linearized system. Finally, a simple feedback control can be applied using linear controller.

The output dynamics is linear, non-interactive, and pole assignable.

To handle modeling errors, the state feedback controller was equipped with integral action. So we used proportional integral (PI) controllers whose parameters are determined for an underdamped responses in conjunction with the dominant dynamics (zero dynamics), [17].

Since the details of the linearization process and the study of the zero dynamics are quite lengthy, only the results are presented here for the sake of brevity. However we give in the appendix the main solvability conditions applied to our copolymerization reactor. For more details we recommend to see Isidori’s (1989) and Khalil’s (1989) textbooks.

For the first case (I), the choice of regulating the composition $P_1$ and $P_2$ seems, at first sight, justified for regulating the overall conversion and the copolymer composition. However, after testing the first condition of the proposition (appendix), one concludes that the decoupling matrix $A(x)$ is singular. So case (I) has been rejected. For the other cases the zero dynamics analysis are required to study the eventual input multiplicity phenomenon.

APPLICATION

In this section, we consider our illustrative copolymerization reactor. As mentioned in the abstract the copolymerization system is methyl methacrylate MMA (monomer 1), vinyl acetate VA (monomer 2), ethyl acetate EA (solvent), azobisisobutyronitrile AIBN (initiator).

The operation conditions are such as to make a comparison between our results and the previous works:

$q_1 = 36$ kg/h  $T_1 = 350$ K
$q_2 = 90$ kg/h  $T_2 = 300$ K
$q_3 = 0.36$ kg/h $V = 2$ m$^3$
$q_4 = 36$ kg/h

The simulations have been done using Matlab/Simulink, where the kinetic parameters are obtained from the literature.
For the operating conditions the nominal steady state (which satisfies the static version of the model) is:

\[
\bar{\mathbf{x}} = \begin{bmatrix} M_1, M_2, I, S, P_1, P_2, T, C_1, \eta_0^n, \eta_1^n, \eta_2^n \end{bmatrix} = \begin{bmatrix} 0.3293 \text{ kmol/m}^3, 5.3829 \text{ kmol/m}^3, 0.0032 \text{ kmol/m}^3, 2.5334 \text{ kmol/m}^3, 1.8985 \text{ kmol/m}^3, 1.0920 \text{ kmol/m}^3, 347.69 \text{ K}, 2 \text{ m}, 0.0382, 292.53, 1.105 \times 10^7 \end{bmatrix}
\]

The nominal production attributes are:

\[
\bar{P}_w = 45.41 \text{ kg/h} \quad \bar{C}_1 = 0.6348 \quad \bar{M}_{pw} = 37813 \text{ kg/kmol} \quad \bar{M}_{pn} = 7663 \text{ kg/kmol}
\]

It should be emphasized that the first case has been rejected. Therefore, we only represent the simulation results of the second, third, and fourth cases.

In “Fig.4” we illustrate the first eight states dynamics which have a physical signification ( unlike the last three), while the product attributes dynamics are represented in “Fig.5”. Control schemes II and IV steer the reactor to its nominal design value. Otherwise, control scheme III takes the reactor to its nominal outputs ( \( M_2, P_1, S, T, V \) ) but ( \( M_3, P_2, I \) ) reach unwanted values. Likewise, the corresponding product attributes in case III reach erroneous values.
As a matter of fact the nominal steady state is not an attractor of the zero dynamics of case III, and there is input multiplicity (the state-input pair reaches unwanted value).

To study the robustness of the controllers, we have considered that the gel effect coefficient, the initiator efficiency, the heat transfer coefficient and the activation energy of the heat production propagation rates have uncertain values. Those parameters were underestimated by 10% to test the robustness of the controllers.

In the industrial practice, it is very important to change the operating conditions as safely as quickly as possible in order to minimize the production of off-specification polymer. From this perspective, a set point change was tested assuming that the reactor is operating with modelling errors.

The simulations of the last two operations (robustness study and the change of the set point) are not represented here. However, they showed the robustness of the controller which is considered as a perfect compensator of non-linear interactions and modelling errors. Moreover, the change of the set point have been successfully implemented showing one more time, the efficiency of the controllers of case IV.

CONCLUSIONS

The control problem of a continuous solution copolymerization reactor has been addressed in conjunction with considerations from the few previous works.

First, the mathematical model has been improved by adding the equations of the molecular weight distribution moments, which have been obtained from generating function technique. In spite of dealing with more complex model this approach enabled us to assess and to control number average and average molecular weights.

It was established how the kind of copolymerization kinetics, the operating conditions and above all, the choice of the control outputs, determines the solvability of the control problem. If the problem was not solvable, the methodology gave guidelines to choose a different set of outputs and/or to redesign the reactor and operating condition.

With the proposed state feedback linearization converts our interactive non-linear multivariable system into five single-input single-output linear systems. The parameters of the PI controllers depend on the settling time of the dominant dynamic (zero dynamics).

From the study of the four cases the first was rejected, and the third case (III) represented input-multiplicity phenomenon. For industrial application we recommend to use the fourth case which manifested the best performance. Moreover, it is possible in industrial practice to use an inhibitor such as m-dinitrobenzene (m-DNB) in order to limit or to stop copolymerization reactions. In the actual processes an on-line separating is used. The separator often involves a series of steps, which may include dryers and distillation columns. Unreacted monomers and solvent can be then recycled back to the reactor. This framework needs in fact another control approach.

NOTATIONS

\[ I \] : initiator concentration, kmol/m³
\[ I_o \] : initiator feed concentration, kmol/m³
\[ M_1, M_2 \] : monomer 1, 2 concentrations, kmol/m³
\[ M_{1w}, M_{2w} \] : monomer 1, 2 feed concentrations, kmol/m³
\[ M_1, M_2 \] : monomer 1, 2 molecular weights, kg/kmol
\[ P_1, P_2 \] : converted monomers concentrations, kmol/m³
\[ q_{1o}, q_{2o} \] : monomer 1, 2 feed rates, m³/s
\[ q_{1}, q_{2} \] : solvent and initiator feed rates, m³/s
\[ q \] : output flow rate, m³/s
\[ R_1, R_2 \] : monomer 1, 2 reaction rates, kmol/m³.s
\[ R_i \] : initiator reaction rate, kmol/m³.s
\[ S \] : solvent concentration, kmol/m³
\[ S_o \] : solvent feed concentration, kmol/m³
\[ T, T_r, T_c \] : reactor, feed-rate and cooling temperatures, K
\[ V \] : reactor volume, m³
\[ \eta_0, \eta_1, \eta_2 \] : zeroth, first, and second moments of the molecular weight distribution
\[ \phi \] : overall heat transfer coefficient, kJ/m².s.K
\[ \eta_m, \eta_{m2}, \eta_1 \] : monomer 1,2 and solvent contraction factors
\[ \epsilon_1, \epsilon_2 \] : converted monomers contraction factors
\[ \beta_1, \beta_2 \] : temperature variation due to monomer 1 and monomer 2 reactions
\[ \rho \] : average density, kg/m³
\[ \rho_1, \rho_2, \rho_3 \] : density of monomer 1,2 and solvent, kg/m³
\[ \Omega_1, \Omega_2, \Omega_3 \] : monomer 1, 2 and solvent heat contributions, K/m³

REFERENCES


APPENDIX

Proposition :

The reactor control problem is solvable if and only if there are two positive integers $k_1$ and $k_2$, such that the output set $(h_1(x), h_2(x), S, T, V)$ meet the following conditions :

- **a).** the decoupling matrix :

$$ A(x) = \begin{bmatrix}
L_{g1}L_{j1}^{-1}h_1 & L_{g2}L_{j2}^{-1}h_1 & L_{g3}L_{j3}^{-1}h_1 & 0 & 0 \\
L_{g1}L_{j1}^{-1}h_2 & L_{g2}L_{j2}^{-1}h_2 & L_{g3}L_{j3}^{-1}h_2 & 0 & 0 \\
-S \varepsilon_m V^{-1} & -S \varepsilon_m V^{-1} & (S c, S \varepsilon) V^{-1} & 0 & 0 \\
\Omega_1 & \Omega_2 & \Omega_3 & \phi & 0 \\
\varepsilon_m & \varepsilon_m & \varepsilon \delta & 0 & -1
\end{bmatrix} $$

is non-singular in the zero-dynamics manifold $X_z$.

- **b).** There is a complementary map:

$$ \phi_H = [\phi_{k1}(x), ..., \phi_{k}(x)]^T $$

such that the map

$$ \phi(x) = \begin{bmatrix} \phi_1(x) \\ \phi_H(x) \end{bmatrix} $$

is invertible on some strip neighbourhood $N(X_z)$ of the zero dynamics manifold $X_z$.

c). the nominal steady-state $x$ is the unique A-stable attractor of the $[8 - (k_1 + k_2 +3)]$ dimensional zero dynamics

$$ \dot{x} = f(x) - G(x)A(x)^{-1}b(x), \quad x \in X_Z $$

Where the five entries of $b(x)$ are:

- $b_1(x) = L_{j1}^{k_1} h_1$
- $b_2(x) = L_{j2}^{k_2} h_2$
- $b_3(x) = -S(\varepsilon_1 R_1 + \varepsilon_2 R_2)$
- $b_4(x) = \beta_1 + \beta_2 - \phi T$
- $b_5(x) = V(\varepsilon_1 R_1 + \varepsilon_2 R_2)$