STABILITY ANALYSIS FOR NON-ISOTHERMAL CONTINUOUS STIRRED TANK REACTOR-NCSTR

Dumitru POPESCU[1], Ştefania COLBU[2], Daniel BĂNCILĂ[3], Severus OLTEANU[4]

Rezumat. *Procesele chimice și petrochimice non-izoterme reprezintă o categorie particulară conform exploatării tehnologice datorită unor posibile fenomene de instabilitate termică. Scopul lucrării este de a analiza stabilitatea regimului staționar de funcționare în cazul unui reactor neizoterm cu parametri concentrați - NCSTR, pentru a garanta siguranța și performanțele dorite prin soluții adecvate de control. În acest scop, se estimează modelul matematic neliniar bazat pe evaluarea bilanțului de masă și de energie termică. Din bilanțul de energie termică evaluat în punctul de funcționare staționar, se determină caracteristicile termice pentru studiul stabilității și condițiile geometrice necesare stabilității interne a NCSTR. Modelul dinamic neliniar de stare este liniarizat în jurul punctului staționar și se deduc specificațiile de funcționare stabilă. Se demonstrează condițiile necesare și suficiente de stabilitate internă a regimului exoterm al NCSTR, folosind teorema Lyapunov de stabilitate pentru sisteme liniare.*

Abstract. *The non-isothermal chemical and petrochemical processes represent a particular technological position in exploitation, by a possible thermal instability evolution. The aim of our paper is to analyse the stationary regime's stability, for the Non-isothermal Continuous Stirred Tank Reactor- NCSTR, to guaranty the imposed security and performances by an adequate control solution. The mathematical nonlinear model based on the mass and thermal balance is estimated. From the thermal balance equation corresponding to the stationary operating point, the heat characteristics for the stability analysis and the necessary geometrical conditions of internal stability for NCSTR are deduced. The nonlinear dynamic state model is linearized around the stationary point and the stable operating specifications are inferred. To demonstrate the necessary and sufficient conditions for the internal stability of the exothermal regime of NCSTR, the Lyapunov stability theorem for the linear systems is established.*

Keywords: non-isothermal processes, mathematical nonlinear model of NCSTR, geometrical stability, internal Lyapunov stability analysis.
DOI https://doi.org/10.56082/annalsarsciinfo.2022.1-2.5

1. Introduction

A Continuous Stirred Tank Reactor (CSTR) is a relevant installation in many industrial chemical processes that demand uninterrupted addition and withdrawal

[1]Prof., PhD, University POLITEHNICA of Bucharest, Romania (e-mail: dumitru.popescu@upb.ro).
[2]Eng., PhD Student, University POLITEHNICA of Bucharest, Romania, (colbucristiana@gmail.com).
[3]Eng., PhD Student, University POLITEHNICA of Bucharest, Romania, (danbancila95@gmail.com).
[4]Prof., PhD, University POLITEHNICA of Bucharest, Romania, (severus.olteanu@upb.ro).

of reactants and products. To improve efficiency and to obtain optimal production in these plants, the reactors are regulated at very high conversion rates. The operating points set should illustrate a stable steady state behavior under the impact of the disturbances as well. Based on the contributions of the mass and heat balance charts and on reactor's stability analysis, studies and research were elaborated over the upcoming decade. In the references [1-3], important results related to the complexity and nonlinearity of mathematical models associated with the dynamics of NCSTR are developed. An analysis of the previous simulations performed on NCSTR was described by V. Chandra and al. in [4]. Decisive outcomes related to the internal stability of nonlinear chemical processes by linearization, established around of the Lyapunov stability criteria and advanced control design, are presented by Lu and al. in [5-6].

The main contributions by M. Tertisco and al. for reactor dynamics in [7] and the Van Heerden stability on heat balance diagrams were developed to determine the performance of a non-isothermal reactor. A problem which incorporates notions of chemical reaction theory and geometric stability characterization for temperature control, and analysis of the possible equilibrium of unstable and stable operation points of NCSTR with autonomous evolution was presented by H.Hoang and al. in [8]. Results obtained on the internal stability of nonlinear chemical processes by linearization technique based on Lyapunov stability criterion were presented in [9], to demonstrate the performances of the non-isothermal reactors. In the papers [10-13], different control strategies from PID control, feed-forward control and IMC to Adaptive, Optimal and Intelligent control and advanced solutions for design and implementation possibilities for non-isothermal reactors were proposed.

The current paper is structured on five chapters. Subsequent to the introduction, in the second chapter the model of the non-isothermal reactor is estimated as a nonlinear one, based on the ma balance equations and thermal balance equations. In the third chapter, the model is used to explore the static and dynamic evolution of the non-isothermal process. From the thermal balance equation corresponding to the stationary operating region, the heat characteristics for the stability analysis of the NCSTR are defined. Afterwards the geometric stability conditions are deduced. In the fourth chapter, the internal stability of the stationary operating region of the NCSTR is deduced. The obtained state model is linearized about a fixed operating point, and comparable stability conditions to those provided by the geometric approach are inferred. Stable operating specifications for the reactor are estimated by these conditions and acquired on the linear model by minor variations at about a stable operating point. Additionally, research on the stability of a stationary state of the process represented by the linearized mathematical model near to the examined stationary point is proposed. To demonstrate the necessary and sufficient conditions for the stability of the NCSTR, the Lyapunov stability technique is provided. Finally, the fifth chapter includes the conclusions and perspectives.

2. Mathematical models of the Non-isothermal CSTR

We take into account the situation of a reactor in which a singular decomposition reaction a reactant A at constant volume and pressure, as in Figure 1.

$$A \xrightarrow{k} B : with\ v_{R_A} = k_0 e^{-E/RT} C_A \tag{1}$$

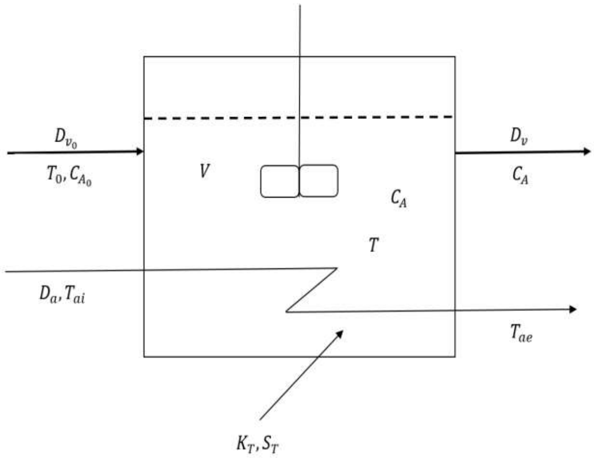

Fig. 1. Non-isothermal Continuous Stirred Tank Reactor- NCSTR

The chemical reaction is characterized by the following variables:

a) independent variables: feed flow $\left(D_{v0}\right)$, reactant concentration in the feed $\left(C_{A0}\right)$, feed temperature $\left(T_0\right)$, thermal agent flow $\left(D_a\right)$;

b) the state variables (output variables): the reactant A concentration in the reaction mixture $\left(C_A\right)$ and the reaction temperature $\left(T\right)$.

The reactor's mathematical model consists of material balance equations for reactant A and thermal energy balance.

The material balance equation of reactant A is given by:

$$\frac{d(C_A V)}{dt} = D_{v0} C_{A0} - D_v C_A - v_{R_A} V \tag{2}$$

where v_{R_A} is the speed of transformation of reactant A and V the constant volume of the reaction mixture. Considering from (2) equality between input and output flows $D_v = D_{v0}$, we obtain the relation (3):

$$V\frac{dC_A}{dt} = D_{v0}\left(C_{A0} - C_A\right) - v_{R_A}V \tag{3}$$

In the case of the thermal energy conservation equation, the thermal transfer from the thermal agent's flow, occurs without accumulation in the dividing wall. In this case, the thermal balance equation can be written as:

$$mc_p\frac{dT}{dt} = D_a c_p\left(T_0 - T\right) + v_{R_A}V\left(-\Delta H_{R_A}\right) + K_T S_T\left(T_a - T\right) \tag{4}$$

where c_p is the mixture's specific heat, m is the mixture's mass, D_a is the feed flow, T_a is the average temperature of the thermal agent, and:

$$Q = K_T S_T\left(T_a - T\right) \tag{5}$$

is the heat transferred between the reaction environment and the thermal agent.

From the relation (4) the amount of heat generated by the chemical reaction q_G is defined and q_T the sum of the quantities of consumed energy to heat the reactants from the supply temperature to the reaction temperature and respectively transferred to the thermal agent:

$$q_G\left(T_S\right) = v_{R_A}\left(C_{AS}, T_S\right)V\left(-\Delta H_{R_A}\right) \tag{6}$$

$$q_T\left(T_S\right) = D_m c_p\left(T_S - T_0\right) + K_T S_T\left(T_S - T_a\right) \tag{7}$$

The relations (6) and (7) show that the function q_G is strongly nonlinear by the exponential term $\left(-\Delta H_{R_A}\right)$ and function q_T is linear in relation with the temperature T_S.

3. Geometrical stability analysis of the stationary operating points, for NCSTR

In our case of the chosen single reaction, the state of the chemical reaction mixture is completely characterized by the conversion values of the reference reactant and process temperature. A stationary state defined by the pair of values of concentration and temperature is represented in plane by $\left(C_A, T_S\right)$ as a stationary operating point. Operation in industrial working conditions is characterized by different random disturbances $\left(D_{v0}, C_{A0}, T_0, T\right)$ that have the effect of moving the reactor from the point of stationary operation. A stationary operating point for exothermal reactor is stable, if the reaction system returns autonomously without any adjustment action to that stationary point; otherwise, the stationary operating point is unstable. Figure 2 shows the diagram $(q-T)$ for the case of exothermic reaction, with three possible stationary state points to temperatures T_M, T_N and T_P. The stationary regime points corresponding to the temperatures T_M and T_P are stable, as to positive changes in temperature the reaction system reaches the area where $q_T > q_G$, from where, after the disappearance of the disturbance, it returns, by cooling, to the starting point. Similarly, at negative changes in temperature (cooling), the system reaches the area where $q_G > q_T$, from which, after the disappearance of the disturbance, returns by heating to the stationary point. The intermediate state of temperature T_N doesn't respect these conditions and is an unstable stationary state. From the diagram, it is found that at the points corresponding to the stable stationary states, P and M, the slope of the heat generated is less than the slope of the heat transferred while at the intermediate point N, which represents an unstable state, the situation is inverse.

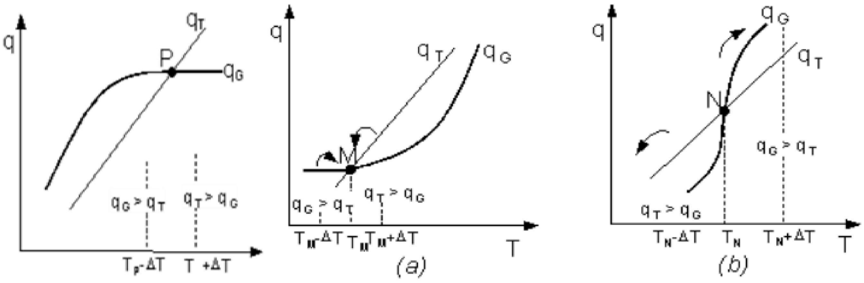

Fig. 2. Geometrical stability analysis in q-T diagram

This analysis can serve as geometrical criteria for the stability of a stationary reactor and leads to the following results:

a) for deviations $\Delta T < 0$ (cooling in relation to stationary state) the reaction system returns by heating at the stationary point $(q_G > q_T)$ if,

$$\left(\frac{dq_G}{dT}\right)_s \prec \left(\frac{dq_T}{dT}\right)_s \tag{8}$$

b) for $\Delta T > 0$ (heating in relation to the stationary state) the reaction system also returns by cooling $(q_G < q_T)$, at the stationary point, if the relationship (8) is fulfilled.

Inequality (8) is a necessary condition in the geometrical meaning, for the stability of the stationary state exothermal regime.

In the case of singular reactions, inequality (8) can be brought to an explicit form in relation to the sizes that characterize the process, by the following expressions:

$$\frac{dq_G}{dT} = \left(-H_{R_A}\right) V \frac{dv_{R_A}}{dT} \tag{9}$$

where,

$$\frac{dv_{R_A}}{dT} = \frac{\partial v_{R_A}}{\partial X_A}\frac{dX_A}{dT} + \frac{\partial v_{R_A}}{\partial T} \tag{10}$$

From the material balance equation of the reactant in stationary regime:

$$t_0 = \frac{V}{D_{v0}} = \frac{C_{A0} X_A}{v_{R_A}} \tag{11}$$

$$X_A = \frac{t_0}{C_{A0}} v_{R_A}, \quad \frac{dX_A}{dT} = \frac{t_0}{C_{A0}} \frac{dv_{R_A}}{dT} \tag{12}$$

From (10) and (11) it results:

$$\frac{dv_{R_A}}{dT} = \frac{\partial v_{R_A}}{\partial T}\left(1 - \frac{t_0}{C_{A0}}\frac{\partial v_{R_A}}{\partial X_A}\right)^{-1} \tag{13}$$

Replacing in (9):

$$\frac{dq_G}{dT} = \left(-\Delta H_{R_A}\right) V \frac{\partial v_{R_A}}{\partial T}\left(1 - \frac{t_0}{C_{A0}}\frac{\partial v_{R_A}}{\partial X_A}\right)^{-1} \tag{14}$$

On the other hand, from (7) it results:

$$\frac{dq_T}{dT} = D_m c_p + \frac{dQ_T}{dT} = D_m c_p\left(1 + \frac{dQ_c}{dT}\right) \tag{15}$$

$$Q_c = \frac{Q_T}{D_m c_p} = \frac{K_T S_T \left(T - T_a\right)}{D_m c_p} \tag{16}$$

Replacing (14) and (15) in (8) results in equivalent inequality:

$$\left(1 - \frac{t_0}{C_{A0}}\frac{\partial v_{R_A}}{\partial X_A}\right)_s \left(1 + \frac{\partial Q_c}{\partial T}\right)_s > \frac{\Delta T_{ad}}{C_{A0}} t_0 \left(\frac{\partial v_{R_A}}{\partial T}\right)_s \tag{17}$$

4. Study of the internal stability on the exothermal CSTR

The application of the graphic method described above requires the design of the $(q-T)$ diagram, which is a disadvantage of the technique. More general and direct methods of studying the stability of the stationary state of the reactor with perfect continuous mixing can be developed based on the general theory of nonlinear systems.

The mathematical model of the reactor in the dynamic regime, consisting of balance sheet equations (3) and (4) can be written in the vector form and becomes for the material balance equation (3):

$$\frac{dX_A}{d\theta} = -X_A + \frac{t_0}{C_{A0}} v_{R_A}\left(X_A, T\right) \tag{18}$$

and the thermal balance equation (4):

$$\frac{dT}{d\theta} = T_0 - T + \frac{t_0 \Delta T_{ad}}{C_{A0}} v_{R_A}\left(X_A, T\right) - Q_c\left(T\right) \tag{19}$$

If X_{A_s} and T_s are two values that define a stationary operating point, the equations (18) and (19) can be approximated by linearized forms:

$$\frac{dx_1}{d\theta} = -Hx_1 + \frac{F}{\Delta T_{ad}} x_2;\ x_1 = X_A - X_{A_s};\ x_2 = T - T_s \tag{20}$$

$$\frac{dx_2}{d\theta} = \Delta T_{ad}\left(1-H\right)x_1 + \left(F-G\right)x_2 \tag{21}$$

$$H = 1 - \frac{t_0}{C_{A0}}\left(\frac{\partial v_{R_A}}{\partial X_A}\right)_\theta;\ G = \left(\frac{\partial Q_c}{\partial T}\right)_\theta + 1;\ F = \frac{t_0 \Delta T_{ad}}{C_{A0}}\left(\frac{\partial v_{R_A}}{\partial T}\right)_\theta \tag{22}$$

In the above relationships H, G, F the derivatives are estimated in the stationary regime.

Entering the notations:

$$X = \begin{pmatrix} X_1 \\ X_2 \end{pmatrix};\ \mathbf{A} = \begin{pmatrix} -H & F/\Delta T_{ad} \\ \Delta T_{ad}\left(1-H\right) & F-G \end{pmatrix} \tag{23}$$

equations (20) and (21) are rewritten cumulatively into the matrix equation:

$$\frac{dX}{d\theta} = \mathbf{A}X \tag{24}$$

The solution of the equation (24) is in the form:

$$x_i\left(t\right) = C_{1_i} e^{\lambda_1 t} + C_{2_i} e^{\lambda_2 t}, i = 1:2 \tag{25}$$

The eigenvalues of matrix $\mathbf{A}$ are calculated by solving the equation:

$$\det(\mathbf{A}-\lambda\mathbf{I})=0 \tag{26}$$

where $\mathbf{I}$ - unit matrix, having the same dimensions as state matrix $\mathbf{A}$.

The equation (26) is written:

$$\det\begin{pmatrix} -H-\lambda & F/\Delta T_{ad} \\ \Delta T_{ad}(1-H) & F-G-\lambda \end{pmatrix}=0 \tag{27}$$

or

$$\lambda^2+(H+G-F)\lambda+GH-F=0 \tag{28}$$

The stability condition of the stationary state defined by the values X_{A_S}, T_s, is defined as the need that after a temporary disturbance $X_A(t)$ and $T(t)$ to return to the values X_{A_S}, T_s, i.e.:

$$\lim_{t\to\infty} X_A(t)=X_{A_S};\ \lim_{t\to\infty} T(t)=T_s \tag{29}$$

or in deviation variables x_1, x_2:

$$\lim_{\theta\to\infty} x_1(\theta)=0;\ \lim_{\theta\to\infty} x_2(\theta)=0 \tag{30}$$

The condition of stability (30) is satisfied if the eigenvalues are real and negative, or complex conjugated with the real negative axis. In terms of equation coefficients (28), these requirements are translated into inequalities derived from the relationships between roots and coefficients:

$$H+G-F>0 \tag{31}$$

Inequalities in (31) represent conditions of stability, valid for small amplitudes disturbances, which maintain at an acceptable level the errors introduced by linearizing the balance sheet equations. By replacing the definition expressions for $H, G\ and\ F$ in these inequalities, the conditions of stability become (32):

$$2-\frac{t_0}{C_{A_0}}\left[\left(\frac{\partial v_{R_A}}{\partial X_A}\right)_s+\Delta T_{ad}\left(\frac{\partial v_{R_A}}{\partial T}\right)_s\right]+\left(\frac{\partial Q_c}{\partial T}\right)_s>0 \tag{32}$$

The last inequality (32) offers the necessary and sufficiently internal stability conditions for CSTR, around a stationary point.

Conclusions

Conclusion (1). The paper addresses the stability analysis of the operating regime for the non-isothermal processes, based on the dynamic model of the NCSTR.

Conclusion (2). The nonlinear model of the NCSTR is computed and the necessary geometric stability conditions are evaluated in a stationary plain $(q-T)$.

Conclusion (3). The nonlinear steady state dynamic model is linearized around a stationary operating point for studying the internal stability of non-isothermal reactor.

Conclusion (4). For the linearized mathematical model, the Lyapunov stability analysis offer stable operating conditions for the nonlinear reactor by minor variations around a stable stationary point.

Conclusion (5). The internal stability conditions obtained on the linearized dynamic state model prove the necessary and sufficient stability conditions for the exothermal reactor.

Conclusion (6). The future research will by oriented to evaluate of the maximum stability region of non-isothermal CSTR Moreover, the region of stability will be estimated based on the Lyapunov functions and a performant LQR optimal control solution can by proposed for this kind of chemical processes.

R E F E R E N C E S

[1] D. Popescu, R. Dobrescu, F.Ionescu, D. Stefanoiu, *Modeling in Industrial Processes* (AGIR Publishing House, Bucharest, Romania, 2011).

[2] D.Popescu, N.Paraschiv, C.Patrascioiu, *Automatica Vol. 3*(Romanian Academy, Bucharest, Romania, 2015) Chap.33, pp. 40.

[3] L. Foulloy, D. Popescu, G. Dauphin-Tanguy, *Modélisation, Identification et commande des Systèmes*(Romanian Academy Publishing House, Bucharest, Romania, 2004)

[4] V. Chandra, E.A.J.F. Peters, J.A.M. Kuipers, *Direct numerical simulation of a non-isothermal non-adiabatic packed bed reactor* (Chemical Engineering Journal, 2020) Vol. **385**.

[5] Lu, S. Das, E.A.J.F. Peters, J.A.M. Kuipers, *Direct numerical simulation of fluid flow and mass transfer in dense fluid-particle systems with surface reactions* (Chemical Engineering Science, 2018) Vol. **176**, pp. 1-18.

[6] Lu, E.A.J.F. Peters, J.A.M. Kuipers, *Direct numerical simulation of fluid flow and dependently coupled heat and mass transfer in fluid-particle systems* (Chemical Engineering Science, 2019) Vol. **204**, pp. 203-219.

[7] M.Tertisco, D.Popescu, B. Jora, I.Russ, *Control for Industrial Continuous Processes* (Didactical Publishing House, Bucharest, Romania, 1991).

[8] N.H. Hoang, F. Couenne, C. Jallut, Y. Le Gorrec, *Thermodynamics based stability analysis and its use for nonlinear stabilization of the CSTR* (Computers & Chemical Engineering, 2013) Vol. **58**, pp. 156-177.

[9] S. Colbu, F.Hamidi, D.Popescu, H.Jerbbi, D.Bancila, *Stability Analisis and Control for Nonisothermal reactors* (IEEE Xplore, 2022).

[10] T. Pan, S. Li, W.J. Cai, *Lazy learning-based online identification and adaptive PID control: a case study for CSTR process*(Industrial & Engineering Chemistry Research, 2007) Vol. **46**, pp. 472-480.

[11] Y. Zhai and S. Ma, *Controller design for continuous stirred tank reactor based on feedback linearization* (2012 Third International Conference on Intelligent Control and Information Processing, 2012) pp. 378-381.

[12] D. Bruns, J. E. Bailey, *Nonlinear feedback control for operating a nonisothermal CSTR near an unstable steady state* (Chemical Engineering Science, 1977) Vol. **32**, No. 3, pp. 257-264.

[13] H. Jerbi, F.Hamidi, S.Olteanu, D. Popescu, *Lyapunov-based Methods for Maximizing the Domain of Attraction* (International Journal of Computers, Communications & Control-IJCCC, 2020).