

Reaction flux versus reaction force: easy to stabilize?

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Abstract: Chemical reaction rate, also known as the reaction flux involved in chemical reactors, plays a central role as the source generating the abnormal dynamics characteristics. This paper proposes a structural approach for the stabilization of such systems through the control of the reaction flux by considering the Lyapunov stability theory within a standard thermodynamic framework. More precisely, the reaction flux is structurally considered as a nonlinear function of conjugated reaction force. The thermodynamic constraint of such a relationship is that the inherent non-negative definiteness property of the irreversible entropy production due to chemical reaction has to be fulfilled. Consequently, it allows to reexpress a large class of reaction rates described by the mass-action-law and more interestingly, the operation of the reaction system at a desired set-point consists in controlling the reaction force on the basis of an affinity-related storage function. Numerical simulations for a non isothermal continuous stirred tank reactor (CSTR) involving one reversible reaction operated with multiple steady states illustrate the application of the theoretical developments.

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1. INTRODUCTION

The state feedback control design for unstable homogeneous chemical reactors, and in particular unstable non isothermal continuous stirred tank reactors (CSTRs), is typically facing the inherent nonlinearity resulting from constitutive relations (such as chemical reaction kinetics and transport phenomena etc.). It is shown in (Georgakis (1986); Favache & Dochain (2009)) that at a given operating condition, the reaction kinetics may generate the abnormal complex dynamical behavior of the system (for example steady state multiplicity, limit cycles and chaos or non-minimum phase behaviors (Viel *et al.* (1997); Favache & Dochain (2010); Hoang *et al.* (2013b))) which gives rise to practical difficulties and theoretical challenging issues for the control design. In many industrial applications, the key motivation for the control of such systems lies in the fact that unstable chemical reactors may have to be operated at certain unstable steady state that allows to maintain some process performances to be optimal (like, for instance, an optimum tradeoff between conversion ratio and selectivity or reactor temperature etc.) (Bruns & Bailey (1975)).

Over the years, a number of control strategies have been developed to deal with the nonlinear feedback control of unstable chemical reactors. Several applications of nonlinear control methods to CSTRs can be found in a large number of references, e.g. feedback linearization for control under constraints (Viel *et al.* (1997)), Lyapunov-based

control (Antonelli & Astolfi (2003)), nonlinear PI control (Alvarez-Ramírez & Morales (2000)), port (pseudo) Hamiltonian framework (Hangos *et al.* (2001); Ramírez *et al.* (2016)), energy/power-shaping control (Favache & Dochain (2010)) and inventory control (Farschman *et al.* (1998)). In addition, the seminal results presented in (Dammers & Tels (1974); Tarbell (1977); Ydstie & Alonso (1997); Rodrigues *et al.* (2015)) are of great interest, and were dedicated to an active research area where the use of thermodynamics for both the stability analysis and control design of chemical reaction networks is extensively considered (Alonso & Ydstie (2001); Favache & Dochain (2009); Hoang *et al.* (2012)).

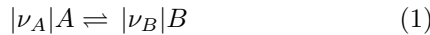
From a thermodynamic point of view, the CSTRs can be viewed as a thermodynamic system since the evolution of the system state variables is intrinsically governed by the principles of thermodynamics (Callen (1985)). In such systems, the presence of the energy and entropy transformations associated with the material transformation is an inherent property (De Groot & Mazur (1962)). While the chemical reaction takes place in the reactor, those transformations interact and they are then linked together in a systematic way (Ederer *et al.* (2011); Hoang & Dochain (2013a)). Based on the results developed in (Couenne *et al.* (2006); Favache & Dochain (2009); García-Sandoval *et al.* (2016); Hoang *et al.* (2017)), the contribution of this work is to explore further the link between irreversible thermodynamics and control systems theory usable for

the control design of non isothermal chemical reactors. More precisely, this paper presents an interesting extension of the previous works (Hoang *et al.* (2014)) aimed at providing the global exponential stabilization of a CSTR under steady state multiplicity on the basis of controlling the reaction rate only.

2. IRREVERSIBLE THERMODYNAMICS AND ITS USE FOR THE MODELING OF THE CSTR

2.1 The CSTR model

Let us consider a liquid phase CSTR under isobaric conditions involving one reversible chemical reaction of 2 chemical species A and B (with the molar masses M_A and M_B , respectively) :



The reaction stoichiometry that represents the (molar) mass invariant of the reaction (1) is given as follows :

$$\nu_A M_A + \nu_B M_B = 0 \quad (2)$$

where ν_A and ν_B are the suitable signed stoichiometric coefficients: $\nu_A < 0$ and $\nu_B > 0$ (Hoang & Dochain (2013a); Ramírez *et al.* (2016)). In that respect, it is worth noting that the net reaction rate r of reaction (1) can be expressed as follows :

$$r = r_f - r_r \quad (3)$$

where $r_f \geq 0$ and $r_r \geq 0$ are the forward and reverse reaction rates, respectively.

Throughout the paper, the following assumptions are considered :

(H1) The fluid mixture is ideal and incompressible.

(H2) The reactor is fed by the only species A with the inlet molar flow rate F_{AI} at a fixed inlet temperature T_I .

(H3) The heat flow rate \dot{Q}_J coming from the jacket is modeled by the following relation :

$$\dot{Q}_J = \lambda(T_J - T) \quad (4)$$

with $\lambda > 0$ the heat exchange coefficient. The heat flowrate \dot{Q}_J (or equivalently the jacket temperature T_J) and inlet molar flow rate F_{AI} are considered as process inputs.

(H4) The reaction rate r (3) depends only on the temperature of the reaction mixture, and on the concentrations of the involved species. The net reaction $r > 0$ is such that the species A is consumed, whereas $r < 0$ if the species A is produced. The case $r = 0$ corresponds to the chemical equilibrium. Furthermore, the reaction rate r fulfills thermodynamic constraints as follows (Sandler (1999); Favache & Dochain (2010)) :

$$\lim_{T \rightarrow 0} r = 0 \quad \text{and} \quad \lim_{T \rightarrow +\infty} r = r_{\max} \quad (5)$$

We can easily check that the above conditions hold for the mass-action-law with temperature-dependent kinetics described by the Arrhenius law :

$$k(T) = k_0 \exp\left(-\frac{E_a}{RT}\right) > 0 \quad (6)$$

where k_0 , E_a and R are the kinetic constant, activation energy and gas constant, respectively.

Notation: Let $\mathcal{I} = \{A, B\}$ be the set of chemical species involved in the reaction mixture (1).

2.2 Thermodynamically consistent CSTR modeling

In equilibrium thermodynamics, the system variables are split into extensive variables (such as the internal energy U , the entropy S , the volume V and the molar number N_i , $i \in \mathcal{I}$) and intensive ones (such as the temperature T , the pressure p and the chemical potential μ_i , $i \in \mathcal{I}$). When isobaric conditions are considered, the variation of the internal energy U is equal to that of the enthalpy H , given by considering the Gibbs' equation (Callen (1985)) :

$$dH = \sum_{i \in \mathcal{I}} \mu_i dN_i + T dS \quad (7)$$

From (7), we equivalently have :

$$dS = \sum_{i \in \mathcal{I}} \frac{-\mu_i}{T} dN_i + \frac{1}{T} dH \quad (8)$$

since the absolute temperature $T > 0$. As the entropy S is also an extensive variable, it is thus a homogeneous function of degree 1 with respect to $(N_{k \in \mathcal{I}}, H)$ ¹. By using the Euler's theorem (Callen (1985)), we get :

$$S(N_{k \in \mathcal{I}}, H) = \sum_{i \in \mathcal{I}} \frac{-\mu_i}{T} N_i + \frac{1}{T} H \quad (9)$$

(8) can then be rewritten in a compact form as follows :

$$dS = w^T dZ \quad \Rightarrow \quad w(Z)^T = \frac{\partial S(Z)}{\partial Z} \quad (10)$$

where :

$$w(Z) = \left(\frac{-\mu_{k \in \mathcal{I}}}{T}, \frac{1}{T} \right)^T, \quad Z = (N_{k \in \mathcal{I}}, H)^T \quad (11)$$

As a consequence of (9), $w(Z)$ (10) is a homogeneous function of degree 0 with respect to Z .

The system dynamics is given by considering the material and energy balance equations on the basis of the extensive variables vector Z (11) (Luyben (1990); Favache & Dochain (2009, 2010); Ramírez *et al.* (2016)) :

$$\frac{dN_A}{dt} = F_{AI} - F_A + \nu_A r V \quad (12)$$

$$\frac{dN_B}{dt} = -F_B + \nu_B r V \quad (13)$$

$$\frac{dH}{dt} = \dot{Q}_J + \sum_{i \in \mathcal{I}} (F_{iI} h_{iI} - F_i h_i) \quad (14)$$

where $(F_A, F_B)^T$, $(h_{AI}, h_{BI})^T$ and $(h_A, h_B)^T$ are the outlet flow rate vector, the inlet and outlet molar enthalpy vectors, respectively.

Let us complete the system dynamics representation (12)-(14) by the entropy balance. Indeed the hypothesis of local equilibrium applied to (10) gives (De Groot & Mazur (1962)) :

$$\frac{dS}{dt} = w^T \frac{dZ}{dt} \quad (15)$$

Nevertheless, the entropy balance can also be deduced directly from the second law of thermodynamics (Couenne

¹ Let $f : \mathcal{R}^n \rightarrow \mathcal{R}$, the function f is said to be homogeneous of degree k if $\forall x \in \mathcal{R}^n$ and $\gamma \in \mathcal{R}^{*+}$, $f(\gamma x) = \gamma^k f(x)$. In this case, γ is called the homogeneity ratio.

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