

Noise-to-state Exponentially Stabilizing (state, input)-disturbed CSTRs with Non-vanishing noise ^{*}

Yafei Lu ^a Zhou Fang ^a Chuanhou Gao ^a Denis Dochain ^b

^a*School of Mathematical Sciences, Zhejiang University, Hangzhou 310027, China*

^b*ICTEAM, UCLouvain, Bâtiment Euler, avenue Georges Lemaître 4-6, 1348 Louvain-la-Neuve, Belgium*

Abstract

In this paper, we have proposed a (state, input)-disturbed CSTR (sidCSTR) model that considers unknown but bounded fluctuations in kinetics, flow rates, and heat exchange. A feedback control law is developed that stabilizes the sidCSTR system to reach noise-to-state exponential stability (NSES). The lower bound of the distribution for the state of the NSES sidCSTR system is analyzed both from the probability domain and the time domain. The performance and the validity of the proposed method are demonstrated using a case study dealing with a second-order reversible reaction.

Key words: (state, input)-disturbed CSTR; noise-to-state exponential stability; probability domain; time domain

1 Introduction

Continuous stirred tank reactors (CSTRs) are a class of widely-used reactors in chemical industrial processes. To reach the optimal performance, the CSTR process often requires to run at unstable operation conditions. The investigations on stabilization of CSTRs around an unstable steady state thus become rather active during the past decades, including input/output feedback linearization (Adebekun & Schork, 1991), nonlinear PI control algorithm (Alvarez-Ramirez & Femat, 1999), passivity-based control (Ramirez, Gorrec, Maschke, & Couenne, 2016), Lyapunov-based control (Hoang, Couenne, Jalut, & Gorrec, 2012), etc.

The above studies on CSTRs were made basically for deterministic systems. However, in practice this process is inevitably contaminated, and moreover, affected by disturbances. Polymerization is a typical example that the disturbances can directly affect the probability distribution of the molecular weight of the polymer particle; for instance, this happens in an emulsion or mass polymer reactor. In this case, the stochastic ap-

proaches make sense, which motivates us to consider a stochastic model and stabilization strategy for CSTRs. It was Aris & Amundson (1958) that first studied a linearized CSTR model where the stochastic process with a Gaussian distribution was used to model the reactor temperature and flow rates. Later, Pell & Aris (1969) studied the local stochastic behaviors of a randomly disturbed chemical reactor through simulations on a hybrid computer. Ligon & Amundson (1981) made a further step on finding the relationship between the random input and reactor performances from the analysis of the frequency domain. Recently, Fokker-Planck (FP) theory was applied to assessing the effects of the fluctuations on the dynamics of chemical reactors (Ratto, 1998, Baratti, Tronci, Schaum, & Alvarez, 2016), for which the complicated FP equations are solved to obtain the stationary distribution.

Despite the encouraging progress made on modeling the real CSTR within the stochastic framework, little attention has been paid to the issues on how to depict the stability of general CSTRs in the stochastic sense, and further on how to control them for stabilization purpose. For these reasons, the current work builds a CSTR model with state and input subject to the fluctuations, referred to as the (state, input)-disturbed CSTR (sidCSTR) in the context, and further applies the concept of noise-to-state exponential stability (NSES) (Deng & Williams, 2000, Fang & Gao, 2020) to developing a controller. The main contributions include: (1) presenting the modeling

^{*} Corresponding author: C. Gao, Tel: +86-571-87952431, fax: +86-571-87953794.

Email addresses: 11535029@zju.edu.cn (Yafei Lu), zhou.fang@zju.edu.cn (Zhou Fang), gaochou@zju.edu.cn (Chuanhou Gao), denis.dochain@uclouvain.be (Denis Dochain).

framework for general CSTRs with kinetics (reaction temperature), flow rates and heat exchange **subject to unknown noises**; (2) based on the phenomenon of non-vanishing noise in the model of sidCSTRs, applying the notion of NSES to depicting their stable behavior and further to developing a controller for them; (3) analyzing the impact of fluctuations on the lower bound of states of the sidCSTR both from the probability domain and from the time domain. **Although no new theoretical contribution is added in comparison to the earlier work (Fang & Gao, 2020), the current study is of great significance in practice as an application paper. More specifically, the proposed method can be used to the reacting systems with the distribution function of the product, such as polymerization.**

The rest of the paper is organized as follows. Section 2 reviews some basic concepts about NSES. In Section 3, the model for sidCSTRs is set up. Then a controller is developed to stabilize sidCSTRs in the sense of NSES in Section 4. This is followed by an experimental simulation, including a case study on a sidCSTR system with a second-order reaction and comparisons with other controllers in Section 5. Finally, Section 6 concludes the paper. **The proofs of some results and more simulation cases are presented in the Supplement materials.**

Notation: \mathcal{K} : a continuous function $\gamma: \mathbb{R}_{\geq 0} \rightarrow \mathbb{R}_{\geq 0}$ is class \mathcal{K} if it is strictly increasing and $\gamma(0) = 0$. \mathcal{K}_{∞} : if $\gamma \in \mathcal{K}$ and $\lim_{x \rightarrow \infty} \gamma(x) = \infty$. \mathcal{KL} : a continuous function $\beta: \mathbb{R}_{\geq 0} \times \mathbb{R}_{\geq 0} \rightarrow \mathbb{R}_{\geq 0}$ is class \mathcal{KL} if for each fixed t , $\beta(\cdot, t) \in \mathcal{K}$ and for each fixed x , $\beta(x, \cdot)$ is decreasing and $\lim_{t \rightarrow \infty} \beta(x, t) = 0$. $\|\cdot\|_{\mathcal{F}}$: the Frobenius norm of a matrix $A \in \mathbb{R}^{m \times n}$ is defined as $\|A\|_{\mathcal{F}} = \sqrt{\sum_{i=1}^m \sum_{j=1}^n |a_{ij}|^2}$. $\mathbb{1}_{\{\cdot\}}$: $\mathbb{1}_{\{\cdot\}}$ is called indicator function, if event $\{\cdot\}$ happens, its value is 1, otherwise 0. \mathcal{W}_{-1} : \mathcal{W}_{-1} is called Lambert function in the lower branch, and for any real number $x \geq -e^{-1}$, there is $x = \mathcal{W}_{-1}(x)e^{\mathcal{W}_{-1}(x)}$ and $\mathcal{W}_{-1}(x) \geq -1$. $\mathbb{1}_p$: a p -dimensional vector with all elements being 1.

2 Preliminaries

In this section, some basic concepts and results about the noise-to-state stability are recalled. Consider a non-linear stochastic differential system written in the sense of Itô **(Mao, 2011)**:

$$d\mathbf{x} = \mathbf{f}(\mathbf{x})dt + \mathbf{h}(\mathbf{x})\Sigma(t)d\boldsymbol{\omega}, \quad (1)$$

where $\mathbf{x} \in \mathbb{R}^n$, $t \in \mathbb{R}_{\geq 0}$ are the state and time, respectively; $\mathbf{f}: \mathbb{R}^n \mapsto \mathbb{R}^n$, $\mathbf{h}: \mathbb{R}^n \mapsto \mathbb{R}^{n \times r}$ are both local Lipschitz continuous functions so that there is a local solution for the equation; $\boldsymbol{\omega}$ is a \mathbb{R}^r -valued standard Wiener process defined on a complete probability space, with **a covariance matrix** $\Sigma(t) \in \mathbb{R}^{r \times r}$ that is a non-negative definite bounded matrix but unknown.

Let $\mathbf{0}_n \in \mathbb{R}^n$ be the equilibrium of the underlying deterministic system, i.e., $\mathbf{f}(\mathbf{0}_n) = \mathbf{0}_n$. If $\mathbf{h}(\mathbf{0}_n) = \mathbf{0}_{n \times r}$, then the stochastic system has vanishing noise, and $\mathbf{0}_n$ is the equilibrium of the stochastic system. It is possible to achieve stability in probability at $\mathbf{0}_n$. However, in many cases $\mathbf{h}(\mathbf{0}_n) \neq \mathbf{0}_{n \times r}$, which means the stochastic system has non-vanishing noise, and $\mathbf{0}_n$ is not the equilibrium. It is impossible for this class of systems to be stable in probability **(Khasminskii, 2011)**. To overcome this puzzle, the notion of noise-to-state (exponential) stability, which illustrates the boundedness of the state of the system (1) in probability, is developed and presented in the following definition (Deng & Williams, 2000).

Definition 1 *The stochastic system (1) is noise-to-state stable (NSS) if $\forall \varepsilon > 0$, there exist $\beta \in \mathcal{KL}$ and $\gamma \in \mathcal{K}_{\infty}$, such that*

$$P\{\|\mathbf{x}\|_2 < \beta(\|\mathbf{x}_0\|_2, t) + \gamma_{max}\} \geq 1 - \varepsilon \quad (2)$$

for all $t \geq 0$ and any $\mathbf{x}_0 \in \mathbb{R}^n \setminus \{\mathbf{0}_n\}$, where

$$\gamma_{max} = \gamma \left(\sup_{0 \leq s \leq t} \|\Sigma(s)\Sigma(s)^{\top}\|_{\mathcal{F}} \right).$$

NSS essentially demonstrates the system state to be bounded by the amplitude of the initial state and the size of the function related to the noise covariance matrix. The following proposition provides a sufficient condition to suggest NSS.

Proposition 1 *(Adapted from (Deng & Williams, 2000)) For the stochastic system (1), suppose there exists a \mathcal{C}^2 function $V: \mathbb{R}^n \mapsto \mathbb{R}_{\geq 0}$ and \mathcal{K}_{∞} functions $\alpha_1, \alpha_2, \alpha_3$ and γ such that $\alpha_1(\|\mathbf{x}\|_2) \leq V(\mathbf{x}) \leq \alpha_2(\|\mathbf{x}\|_2)$ and for all non-negative definite matrix $\Sigma(t)$,*

$$\begin{aligned} \mathcal{L}[V(\mathbf{x})] &\triangleq \frac{\partial V}{\partial \mathbf{x}}^{\top} \mathbf{f}(\mathbf{x}) + \frac{1}{2} \text{tr} \left\{ (\mathbf{h}(\mathbf{x})\Sigma(t))^{\top} \frac{\partial^2 V}{\partial \mathbf{x}^2} \mathbf{h}(\mathbf{x})\Sigma(t) \right\} \\ &\leq -\alpha_3(\|\mathbf{x}\|_2) + \gamma(\|\Sigma(t)\Sigma(t)^{\top}\|_{\mathcal{F}}), \end{aligned} \quad (3)$$

then the system is said to be NSS and $V(\mathbf{x})$ is called a noise-to-state Lyapunov function. Particularly, if $\alpha_3(\|\mathbf{x}\|_2) \geq aV(\mathbf{x})$ for some **positive** constant a , then the system is said to be NSES.

3 Dynamics of sidCSTRs

In this section, we will set up a sidCSTR model. Consider a CSTR with constant reaction volume V , constant pressure P , p chemical components X_1, \dots, X_p and l reactions $\mathcal{R}_1, \dots, \mathcal{R}_l$ with the i th reaction written as

$$\mathcal{R}_i: \sum_{j=1}^p z_{ji} X_j \rightleftharpoons \sum_{j=1}^p z'_{ji} X_j,$$

where $z_{ji}, z'_{ji} \in \mathbb{Z}_{\geq 0}$ are the stoichiometric coefficients. To control the temperature T , the CSTR is equipped with a surrounding jacket, in which the cooling/heating fluid flows for heat exchange. The dynamics can be deduced from the mass and energy balances by considering the following assumptions:

- A.1 The reaction mixture is ideal and incompressible.
- A.2 At the inlet of the reactor, every component X_j is fed at temperature T^{in} and molar concentration c_j^{in} . In addition, there are at least $p - 1$ species **fed** to the reactor.
- A.3 The reaction kinetics obeys the Arrhenius law, i.e., the forward and backward reaction rates of \mathcal{R}_i **follow**

$$\begin{cases} r_{f_i} = k_{0f_i} \exp\left(-\frac{E_{f_i}}{RT}\right) \Pi_{j=1}^p c_j^{z_{ji}}, \\ r_{b_i} = k_{0b_i} \exp\left(-\frac{E_{b_i}}{RT}\right) \Pi_{j=1}^p c_j^{z'_{ji}}, \end{cases} \quad (4)$$

where E_{f_i}, E_{b_i} are the reaction activation energies, k_{0f_i}, k_{0b_i} are the kinetic constants, and c_j is the molar concentration of component X_j , evaluated by the molar mass N_j according to $c_j = \frac{N_j}{V}$.

- A.4 The heat flow exchanged with the jacket is represented by $\dot{Q} = \lambda S(T_w - T)$ with T_w the jacket temperature, λ the heat transfer coefficient and S the heat transfer area.
- A.5 There are slight fluctuations on every inlet and outlet volume flow rates, on every chemical backward and forward reaction rates as well as on the heat exchange. All fluctuations are supposed to be standard Wiener processes, labeled by $\boldsymbol{\omega}_1 \in \mathbb{R}^p, \boldsymbol{\omega}_2, \boldsymbol{\omega}_3 \in \mathbb{R}^l, \boldsymbol{\omega}_4 \in \mathbb{R}$, respectively. They act on the corresponding objects in proportion to their respective noise intensities $\boldsymbol{\rho}_1 : \mathbb{R}_{\geq 0} \rightarrow \mathbb{R}_{\geq 0}^{p \times p}, \boldsymbol{\rho}_2, \boldsymbol{\rho}_3 : \mathbb{R}_{\geq 0} \rightarrow \mathbb{R}_{\geq 0}^{l \times l}, \boldsymbol{\rho}_4 : \mathbb{R}_{\geq 0} \rightarrow \mathbb{R}_{\geq 0}$, which are unknown, i.e., $\Sigma(t)$ is an **arbitrary unknown but bounded matrix-valued function**.
- A.6 The total mass in the reactor is considered as a constant, i.e., there exists a positive vector $\mathbf{W} = (w_1, \dots, w_p)^\top$ and a positive constant M , such that $\mathbf{W}^\top \mathbf{N} = M$.

Basically, the assumptions A.1, A.2, A.3, A.4 and A.6 are all borrowed from the paper (Hoang, Couenne, Jallut, & Gorrec, 2011), also often emerging in many CSTR related papers. Except A.2, the others are completely the same with the reported ones. In A.2, we added a constraint that “there are at least $p - 1$ species fed into the reactor”, which looks restrictive. The main purpose is to make it convenient to design the controller. Actually, this point is also required in (Hoang, Couenne, Jallut, & Gorrec, 2011), given in Proposition 3 there, but not specially given as an assumption.

A.6 suggests $N_p = \frac{M - \sum_{j=1}^{p-1} w_j N_j}{w_p}$. For the first $p - 1$ species, we use letters with bar to identify the corresponding vectors, such as $\bar{\mathbf{N}} = (N_1, \dots, N_{p-1})^\top$, $\bar{\mathbf{c}}^{\text{in}} = (c_1^{\text{in}}, \dots, c_{p-1}^{\text{in}})^\top$. By denoting $\bar{\mathbf{z}} = [z_{ji}]_{(p-1) \times l}$, $\bar{\mathbf{z}}' = [z'_{ji}]_{(p-1) \times l}$, $\bar{\mathbf{c}}_f = \text{diag}(\Pi_{j=1}^p c_j^{z_{j1}}, \dots, \Pi_{j=1}^p c_j^{z_{jl}})$, $\bar{\mathbf{c}}_b = \text{diag}(\Pi_{j=1}^p c_j^{z'_{j1}}, \dots, \Pi_{j=1}^p c_j^{z'_{jl}})$ and $\mathbf{k}_f(T) = (k_{f_1}(T), \dots, k_{f_l}(T))^\top$, $\mathbf{k}_b(T) = (k_{b_1}(T), \dots, k_{b_l}(T))^\top$, we can write the dynamic equation (including mass balance and energy balance evaluated by the internal energy U) of the sidCSTR in the Itô form to be

$$\begin{aligned} d\mathbf{y} = & \underbrace{\begin{pmatrix} \mathbf{f}_1(\mathbf{y}) \\ 0 \end{pmatrix}}_{\mathbf{f}(\mathbf{y})} dt + \underbrace{\begin{pmatrix} \bar{\mathbf{C}}_{(p-1) \times p} - \frac{\bar{\mathbf{N}}}{V} \mathbb{1}_p^\top & \mathbb{0}_{p-1} \\ \frac{(\mathbf{U}^{\text{in}} - \mathbb{1}_p U)^\top}{V} & 1 \end{pmatrix}}_{\mathbf{g}(\mathbf{y})} \times \\ & \underbrace{\begin{pmatrix} \mathbf{q}^{\text{in}} \\ \dot{Q} \end{pmatrix}}_{\mathbf{u}(\mathbf{y})} dt + \underbrace{\begin{pmatrix} \bar{\mathbf{C}}_{(p-1) \times p} - \frac{\bar{\mathbf{N}}}{V} \mathbb{1}_p^\top & V \mathbf{A} & V \mathbf{B} & \mathbb{0}_{p-1} \\ \frac{(\mathbf{U}^{\text{in}} - \mathbb{1}_p U)^\top}{V} & \mathbb{0}_l^\top & \mathbb{0}_l^\top & 1 \end{pmatrix}}_{\mathbf{h}(\mathbf{y})} \\ & \times \underbrace{\begin{pmatrix} \boldsymbol{\rho}_1(t) & \mathbb{0}_{p \times l} & \mathbb{0}_{p \times l} & \mathbb{0}_p \\ \mathbb{0}_{l \times p} & \boldsymbol{\rho}_2(t) & \mathbb{0}_{l \times l} & \mathbb{0}_l \\ \mathbb{0}_{l \times p} & \mathbb{0}_{l \times l} & \boldsymbol{\rho}_3(t) & \mathbb{0}_l \\ 0 & 0 & 0 & \boldsymbol{\rho}_4(t) \end{pmatrix}}_{\Sigma(t)} \begin{pmatrix} d\boldsymbol{\omega}_1 \\ d\boldsymbol{\omega}_2 \\ d\boldsymbol{\omega}_3 \\ d\boldsymbol{\omega}_4 \end{pmatrix}, \quad (5) \end{aligned}$$

where $\mathbf{y} = (\bar{\mathbf{N}}^\top, U)^\top$ is state, \mathbf{A}, \mathbf{B} with each element $A_{ki} = (z_{ki} - z'_{ki}) \Pi_{j=1}^p c_j^{z_{ji}}$ and $B_{ki} = (z'_{ki} - z_{ki}) \Pi_{j=1}^p c_j^{z'_{ji}}$ ($k = 1, \dots, p - 1, i = 1, \dots, l$), $\bar{\mathbf{C}}_{(p-1) \times p}$ with the j th row vector $\bar{\mathbf{C}}_j = c_j^{\text{in}} \mathbf{e}_j^\top$, \mathbf{e}_j is a p -dimensional unit vector with the j th element to be 1 while others to be 0, \mathbf{U}^{in} is the inlet internal energy vector and $\mathbf{f}_1(\mathbf{y}) = (\bar{\mathbf{z}} - \bar{\mathbf{z}}')(\bar{\mathbf{c}}_b \mathbf{k}_b(T) - \bar{\mathbf{c}}_f \mathbf{k}_f(T))V$.

The equilibrium of the deterministic part of Eq. (5), denoted by \mathbf{y}^* , is termed as a steady state and satisfies

$$\mathbf{f}(\mathbf{y}^*) + \mathbf{g}(\mathbf{y}^*)\mathbf{u}(\mathbf{y}^*) = \mathbb{0}_p, \quad \text{and} \quad \mathbf{u}(\mathbf{y}^*) \neq \mathbb{0}_p.$$

This suggests that $\mathbf{h}(\mathbf{y})$ does not vanish at \mathbf{y}^* . Thus, this kind of sidCSTR systems is essentially a stochastic system with non-vanishing noise.

4 Noise-to-state exponential stabilization for the sidCSTR system

In what follows, we manage to realize noise-to-state exponential stabilization of sidCSTRs. The proofs of **Lemma 1, Proposition 2 and Lemma 2** are provided in the Supplement materials.

Lemma 1 The matrix $\mathbf{g}(\mathbf{y})\mathbf{g}(\mathbf{y})^\top$ is positive definite, where $\mathbf{g}(\mathbf{y})$ is defined in Eq.(5).

Proposition 2 The feedback control law

$$\mathbf{u}(\mathbf{y}) = \mathbf{g}(\mathbf{y})^\top (\mathbf{g}(\mathbf{y})\mathbf{g}(\mathbf{y})^\top)^{-1} (-\mathbf{f}(\mathbf{y}) - K\tilde{\mathbf{y}}) \quad (6)$$

stabilizes the sidCSTR system governed by Eq. (5) in the sense of NSES, where $\tilde{\mathbf{y}} = \mathbf{y} - \mathbf{y}^*$ and K is positive definite.

Remark 1 A look at the feedback law Eq. (6) implies that an analytical control law can be obtained regardless of the statistical properties of noise. Moreover, the computational burden is relatively low, mainly for a matrix inversion, which is very different from offset-free MPC that often needs to solve complicated nonlinear optimization problems with a high computational burden. In addition, the proposed method can quantitatively analyze the impact of noise on the state from the probability domain and time domain.

Remark 2 Notice that there are possibly other uncertain system parameters that are not considered during modeling, such as reaction rate constant, reaction activation energy, etc, which will result in plant-model mismatch. However, the modeling and stabilizing methods can still work if considering those parameters. By analogous setting (the standard Wiener processes assumption and bounded noise intensities assumption) to the used parameters in A.5, they are easy to be involved in the model, but the model will be more complicated. Besides, although the proposed controller lacks certain robustness to plant-model mismatch, this impact can be weakened by increasing ξ . It can be inferred from Eq. (S3) that $\gamma(\cdot)$ will decrease as ξ increases where 2ξ is the minimum eigenvalue of matrix K . That is to say, we may suppress the influence of noises by increasing ξ , i.e., using a relatively large gain matrix K . Then the state of the system may be maintained within the desired bounded area as much as possible. Naturally, it needs a high cost of control.

For the sidCSTR system that is controlled to reach NSES, we first show the probability distribution of the state is bounded by a particular function, which is related to the initial state and noise function. The following lemma is helpful to serve this purpose.

Lemma 2 For a noise-to-state exponentially stabilizing sidCSTR, for any $t \geq 0$, $\tilde{\mathbf{y}}_0 = \mathbf{y}_0 - \mathbf{y}^*$, there is

$$E[V(\tilde{\mathbf{y}}(t))] \leq \frac{1}{2e^{2\xi t}} \tilde{\mathbf{y}}_0^\top \tilde{\mathbf{y}}_0 + (1 - e^{-2\xi t}) \frac{\gamma_{\max}}{2\xi} \quad (7)$$

with $\gamma_{\max} = \frac{1}{2} \sup_{t \geq 0} \|\Sigma \Sigma^\top\|_{\mathcal{F}} \sqrt{I(\mathbf{y}^*) + (\frac{L^2}{2\xi} \sup_{t \geq 0} \|\Sigma \Sigma^\top\|_{\mathcal{F}})^{\frac{2}{3}}}$.

Then applying the Markov inequality¹ to Eq. (7), for any $r \geq \sqrt{\tilde{\mathbf{y}}_0^\top \tilde{\mathbf{y}}_0}$, we get

$$\begin{aligned} P\{\|\tilde{\mathbf{y}}\|_2 \leq r\} &= P\{V(\tilde{\mathbf{y}}(t)) \leq \frac{1}{2}r^2\} \\ &\geq 1 - \frac{E[V(\tilde{\mathbf{y}}(t))]}{\frac{1}{2}r^2} \\ &\geq 1 - \underbrace{\frac{e^{-2\xi t}[\tilde{\mathbf{y}}_0^\top \tilde{\mathbf{y}}_0 - \xi^{-1}\gamma_{\max}] + \xi^{-1}\gamma_{\max}}{r^2}}_{\phi_1(r)}. \end{aligned} \quad (8)$$

Clearly, $\phi_1(r) \geq 0$ holds when $r^2 \geq \tilde{\mathbf{y}}_0^\top \tilde{\mathbf{y}}_0$, which means the state is bounded in probability. If we set $\phi_1(r_\iota) = \iota \in (0, 1)$ and

$$r_\iota = \left(\frac{e^{-2\xi t}[\tilde{\mathbf{y}}_0^\top \tilde{\mathbf{y}}_0 - \xi^{-1}\gamma_{\max}] + \xi^{-1}\gamma_{\max}}{1 - \iota} \right)^{\frac{1}{2}}, \quad (9)$$

it depicts that at time t the state will stay in a region of radius r_ι in probability of at least ι .

From Eq. (9), we can observe that when other parameters are fixed, \mathbf{y} will be closer to \mathbf{y}^* as $\sup\|\Sigma \Sigma^\top\|_{\mathcal{F}}$ decreases at some fixed time. In addition, from Eq. (8), we can infer that as $\sup\|\Sigma \Sigma^\top\|_{\mathcal{F}}$ increases, the lower bound of the probability in which the state belongs to $\|\tilde{\mathbf{y}}\|_2 \leq r$ will become smaller. Similarly, when \mathbf{y}_0 becomes larger while other parameters are fixed, $\phi_1(r)$ will be smaller, and vice versa.

We then analyze the boundedness of the state of the controlled sidCSTR system in time domain. Based on our recent work on NSES (Fang & Gao, 2020), we introduce a distribution-like function

$$D(r) \triangleq \liminf_{T \rightarrow \infty} \int_0^T \mathbb{1}_{\|\tilde{\mathbf{x}}\|_2 < r} ds, \quad (10)$$

which estimates the mean residence time proportion of a trajectory staying in the closed ball along the whole time horizon.

Proposition 3 (Fang & Gao, 2020) For a NSES system (1), $\forall r > \alpha(a^{-1}\gamma_{\max})$ (\mathcal{K}_∞ function) we have

$$\begin{aligned} D(r) &\geq \liminf_{T \rightarrow +\infty} \int_0^T \mathbb{1}_{V(\tilde{\mathbf{y}}) < \alpha(r)} ds \\ &\geq \frac{\ln \frac{\alpha}{a^{-1}\gamma_{\max}}}{-\mathcal{W}_{-1}(-e^{-2}) + \ln \frac{\alpha}{a^{-1}\gamma_{\max}}}, \quad a.s. \end{aligned} \quad (11)$$

¹ The Markov inequality states that for any random variable X and any constant $C > 0$, $P\{\|X\| \geq C\} \leq E(\|X\|)/C$.

This theorem gives a lower bound function of $D(r)$, which is deterministic. By setting $\alpha(r') = \frac{1}{2}r'^2$ and $V(\tilde{\mathbf{y}}) = \frac{1}{2}\tilde{\mathbf{y}}^\top \tilde{\mathbf{y}}$, for any $r' > \sqrt{\xi^{-1}\gamma_{\max}}$ we have

$$D(r') \geq \underbrace{\frac{\ln \frac{r'^2}{\xi^{-1}\gamma_{\max}}}{-\mathcal{W}_{-1}(-e^{-2}) + \ln \frac{r'^2}{\xi^{-1}\gamma_{\max}}}}_{\phi_2(r')}, \quad a.s., \quad (12)$$

where $\phi_2(r')$ is a lower bound of $D(r')$. Further, a formula for the time proportion $\iota \in (0, 1)$ and the corresponding radius r'_ι is also given by

$$r'_\iota = \sqrt{\xi^{-1}\gamma_{\max} \cdot \exp\left(-\frac{\iota}{1-\iota}\mathcal{W}_{-1}(-e^{-2})\right)}, \quad (13)$$

saying that the trajectory will fluctuate in the region $\|\tilde{\mathbf{y}}\|_2 \leq r'$ with the time ratio to be no less than ι . Comparing $\phi_2(r')$ with $\phi_1(r)$, we can find the former is related to $\sup\|\Sigma\Sigma^\top\|_{\mathcal{F}}$ but independent of the initial state \mathbf{y}_0 . If other parameters are fixed, the trajectory $\mathbf{y}(t)$ will fluctuate in the region $\|\tilde{\mathbf{y}}\| \leq r'$ with a larger time proportion $\phi_2(r')$ as $\sup\|\Sigma\Sigma^\top\|_{\mathcal{F}}$ becomes smaller. This means that once $\mathbf{y}(t)$ goes out the region, it will come back to the region soon.

In summary, for the noise-to-state exponentially stabilizing sidCSTR system, its state is shown to be bounded both in **probability domain and in the time domain**. Through the specific expressions of $\phi_1(r)$ and $\phi_2(r')$, it is clear to see how the quantities (i.e., $\mathbf{y}_0, \sup\|\Sigma\Sigma^\top\|_{\mathcal{F}}$) affect the state of the sidCSTR. Therefore, the above analysis provides a theoretical basis to understand the stochastic **phenomena** of CSTRs.

5 Experimental simulation

5.1 Case study

In this section, we apply the proposed controller to a sidCSTR system where a second-order reversible reaction $A \rightleftharpoons 2B$ takes place and only the pure A component **is fed** into the reactor.

From the mass conservation $2N_A + N_B = 2c_A^{\text{in}}V$, we get $2dN_A = -dN_B$. Then the reduced dynamic equation with respect to state $\mathbf{y} = (N_A, U)^\top$ has the Itô form

$$d\mathbf{y} = \mathbf{f}(\mathbf{y})dt + \begin{pmatrix} c_A^{\text{in}} - c_A & 0 \\ \frac{U^{\text{in}} - U}{V} & 1 \end{pmatrix} \begin{pmatrix} q^{\text{in}} \\ \dot{Q} \end{pmatrix} dt + \begin{pmatrix} c_A^{\text{in}} - c_A & -c_A V & c_B^2 V & 0 \\ \frac{U^{\text{in}} - U}{V} & 0 & 0 & 1 \end{pmatrix} \Sigma d\boldsymbol{\omega}, \quad (14)$$

where $\frac{\mathbf{f}(\mathbf{y})}{V} = (k_{0b}e^{-\frac{E_b}{RT}}c_B^2 - k_{0f}e^{-\frac{E_f}{RT}}c_A, 0)^\top$, $\Sigma =$

$\text{diag}(\rho_1, \rho_2, \rho_3, \rho_4)$ and $d\boldsymbol{\omega} = (d\omega_1, d\omega_2, d\omega_3, d\omega_4)^\top$. The related parameters values are: Heat capacity $C_{P_A}, C_{P_B} = 10, 5J/K/mol$; Reference internal energy $U_{A,\text{ref}}, U_{B,\text{ref}} = 5000, 1000J/mol$; Molar gas constant $R = 8.314J/K/mol$; Reference temperature $T_{\text{ref}} = 300K$, $\lambda S = 600J/K/s$; $E_f, E_b = 10^5, 88900J$,

$$k_{0f}, k_{0b} = 1.8 \times 10^{13}s^{-1}, 3.4 \times 10^9l/(mol.s). \quad \text{The}$$

current task focuses on stabilizing the unstable steady point in the sense of NSES, which is denoted by $\mathbf{x}^* = (U^*, N_A^*, N_B^*)^\top$. The reactor operates under the initial conditions $T^{\text{in}}, T_0, T_{w_0} = 310, 330, 345.6K$; $q_0^{\text{in}} = 0.6522l/s$; $N_{A0}, N_{B0} = 1000, 1600mol$; $c_A^{\text{in}} = 18mol/l$; $V = 100l$. By setting the manipulated variables $q^{\text{in}} = 2l/s$ and $T_w = 300K$ at the steady state, it is easy to calculate the concerning steady state point to be $\mathbf{x}^* = (7.896 \times 10^6, 1164.4, 1271.2)^\top$, and $T^* = 344.6K$. We consider two cases with different noise intensities, including $\Sigma_1 = \text{diag}(0.09, 4 \times 10^{-5}, 10^{-6}, 0.2)$ in **Case 1** and $\Sigma_2 = \text{diag}(0.3, 4 \times 10^{-4}, 10^{-5}, 0.5)$ in **Case 2**.

Based on the proof of **Proposition 1**, we set $V(\tilde{\mathbf{y}}) = \frac{1}{2}[(N_A - N_A^*)^2 + (U - U^*)^2]$ and the gain matrix to be $K = \text{diag}(K_1, K_2) = \text{diag}(0.02, 0.03)$, and then design the following controller in terms of Eq. (6)

$$\begin{cases} q^{\text{in}} = V(c_A^{\text{in}} - c_A)^{-1} (-k_{0b}e^{-\frac{E_b}{RT}}c_B^2 + k_{0f}e^{-\frac{E_f}{RT}}c_A - K_1(c_A - c_A^*)) \\ \dot{Q} = -K_2(U - U^*) - q^{\text{in}}\left(\frac{U^{\text{in}} - U}{V}\right). \end{cases}$$

The manipulated variable T_w is thus calculated as

$$T_w = \frac{-K_2(U - U^*) - q^{\text{in}}\frac{U^{\text{in}} - U}{V}}{\lambda} + T.$$

Further, from Eq. (S3) we obtain

$$\mathcal{L}V \leq -2K_1V(\tilde{\mathbf{y}}) + \frac{1}{2}\|\Sigma\Sigma^\top\|_{\mathcal{F}}\sqrt{I(\mathbf{y}^*) + \left(\frac{L^2\|\Sigma\Sigma^\top\|_{\mathcal{F}}}{K_1}\right)^{\frac{2}{3}}} \quad (15)$$

where $I(\mathbf{y}^*) = \left[\left(\frac{U^{\text{in}} - U^*}{V}\right)^2 + (c_A^{\text{in}} - \frac{N_A^*}{V})^2\right]^2 + N_A^{*4} + N_B^{*8}/V^4 + 1$, $L = 4.29 \times 10^{15}$.

By combining Eqs. (15), (8) and (12), we have two lower bound functions in the sense of probability domain

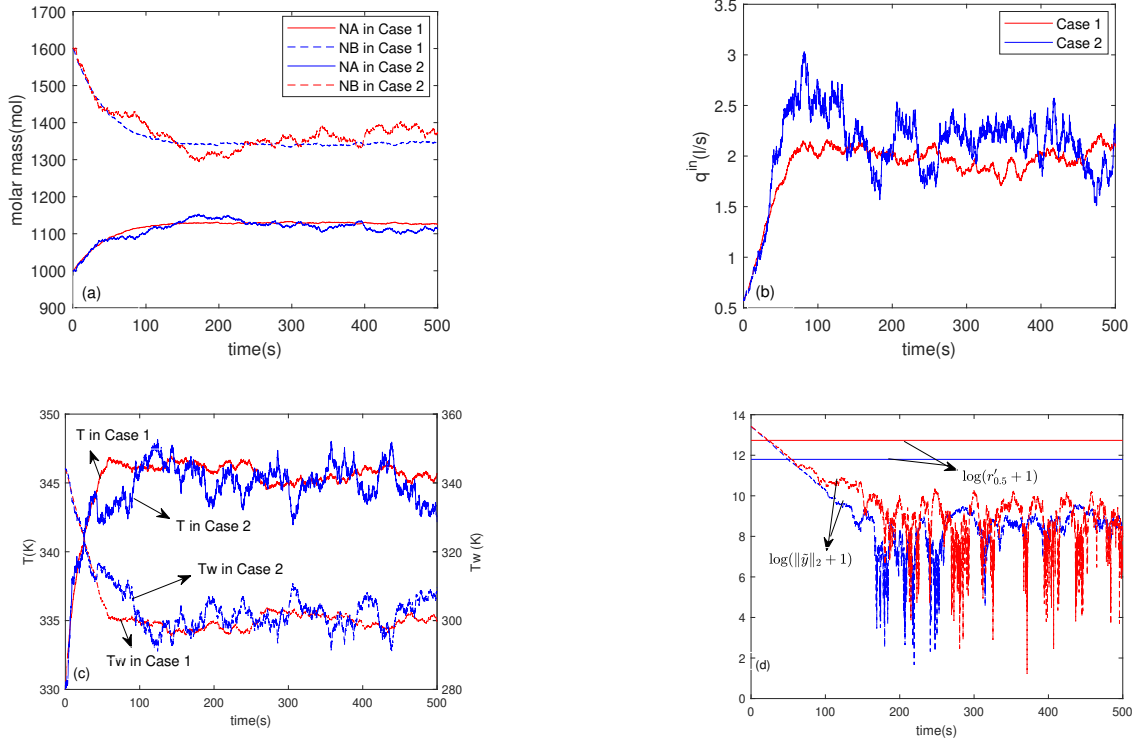


Fig. 1. Time evolution of (a) N_A and N_B , (b) input volume flow, and (c) temperature and jacket temperature in **Cases 1 and 2**. (d) Blue dashed line, red dashed line represent $\log(\|\tilde{\mathbf{y}}\|_2 + 1)$ in **Cases 1 and 2**, and blue solid line, red solid line represent the bound $\log(r'_{0.5} + 1)$ in **Cases 1 and 2**.

and of time horizon to be

$$\phi_1(r) = -\frac{e^{-K_1 t}[(N_{A0} - N_A^*)^2 + (U_0 - U^*)^2 - \sigma]}{r^2} - \frac{\sigma}{r^2} + 1 \quad (16)$$

and

$$\phi_2(r') = \frac{2 \ln r' - \ln \sigma}{-\mathcal{W}_{-1}(-e^{-2}) + 2 \ln r' - \ln \sigma}, \quad r' > \sqrt{\sigma}, \quad (17)$$

where $\sigma = \frac{1}{K_1} \sqrt{\sum_{n=1}^4 \rho_n^4} \sqrt{I(\mathbf{y}^*) + \left(\frac{L^2}{K_1} \sqrt{\sum_{n=1}^4 \rho_n^4}\right)^{\frac{2}{3}}}$.

To observe converging behaviors of this system, Fig. 1. (a) and (c) exhibit the **responses** of the molar mass and temperature to time affected by different noise intensities, respectively. As one can expect, there are small fluctuations around the steady point $(T^*, N_A^*, N_B^*)^T$ in the trajectories (T, N_A, N_B) as the controller is put into force, which shows the controlled sidCSTR is noise-to-state exponentially stable rather than converging to the steady state in probability. Basically, from $t = 200$ s the state vector (T, N_A, N_B) stays in a bounded region around the steady state. These phenomena suggest that the proposed controller is qualified for noise-

to-state exponentially stabilizing the sidCSTR system. We also present the evolution of the manipulated variables q^{in} and T_w in Fig. 1. (b) and (c). In like manner, they will fluctuate around the setting values $q^{in} \approx 2$ and $T_w \approx 300$ as the state enters into the bounded region around the steady state. Moreover, by comparing **Case 1** and **2**, and further by Eq. (16), we can find that at $t = 200$ s, $r_{50\%,1}$ approaches to 6.1296×10^4 in **Case 1** and $r_{50\%,2}$ approaches to 1.1×10^5 in **Case 2**, which illustrates the state vector (T, N_A, N_B) stays in the region $\|\tilde{\mathbf{y}}\|_2 < 6.1296 \times 10^4$ more than 50% probability at $t = 300$ s in **Case 1** while the state stays in the region $\|\tilde{\mathbf{y}}\|_2 < 1.1 \times 10^5$ more than 50% probability at the same time in **Case 2**. This result is consistent with our conclusion that the state will be more far away from the steady state as the noise intensity becomes larger at the given time. A look at the trajectory of this controlled system in the time horizon by Eq.(17) suggests $r'_{50\%,1} \approx 1.3285 \times 10^5$ and $r'_{50\%,2} \approx 3.3898 \times 10^5$ in **Case 1** and **2**, respectively, which agrees with the fact that the greater the noise intensity is, the greater fluctuations the trajectory exhibits. Besides, since the order of magnitude of $\|\tilde{\mathbf{y}}\|_2$ at $t=0$ is about 10^5 which is very large in comparison to the disturbances, the fluctuations on the evolution of $\|\tilde{\mathbf{y}}\|_2$ are not obvious. So we choose the logarithmic form of $\|\tilde{\mathbf{y}}\|_2$ instead of $\|\tilde{\mathbf{y}}\|_2$, and further, $\log \|\mathbf{y}\|_2$ is replaced by $\log(\|\tilde{\mathbf{y}}\|_2 + 1)$ as $\|\tilde{\mathbf{y}}\|_2$ may equal

to 0 at some time. Fig. 1. (d) demonstrates the evolution of $\log(\|\tilde{\mathbf{y}}\|_2 + 1)$ influenced by different noise intensities, implying the boundedness of the state in time domain, from which we can see that the states in **Case 1** and **2** are bounded by $r'_{50\%,1}$ and $r'_{50\%,2}$ with more than 50% of the time, respectively. More case studies may be found in the Supplement materials.

5.2 Comparisons with other method

The performance of the controller Eq. (6) is further exhibited by comparing it with another one. Here, we select a passivity-based controller (PBC) as a comparison. By constructing the synthetic input-output pair

$$\begin{cases} \mathcal{U} = \left(q^{\text{in}}(c_A^{\text{in}} - c_A)V + (k_b c_B^2 - k_f c_A^*)V^2, \frac{U^{\text{in}} - U}{V} q^{\text{in}} + \dot{Q} \right)^\top, \\ \mathcal{Y} = (c_A - c_A^*, U - U^*)^\top \end{cases}$$

and using the previous same Lyapunov function, we design the passivity-based controller as $\mathcal{U} = -K\mathcal{Y}$, which induces the Lyapunov function subject to

$$\begin{aligned} \mathcal{L}V &\leq \mathcal{U}^\top \mathcal{Y} - k_f V^2 (c_A - c_A^*)^2 + \frac{1}{2} K_2 \tilde{\mathbf{y}}^\top \tilde{\mathbf{y}} + \gamma_{\max} \\ &= -\mathcal{Y}^\top K \mathcal{Y} - k_f V^2 (c_A - c_A^*)^2 + \gamma_{\max} \\ &\leq -2 * \min \left(\frac{K_1}{V^2} + k_f - \frac{1}{2} K_2, \frac{1}{2} K_2 \right) V(\tilde{\mathbf{y}}) + \gamma_{\max} \end{aligned} \quad (18)$$

$$\text{with } \gamma_{\max} = \frac{1}{2} \|\Sigma \Sigma^\top\|_{\mathcal{F}} \sqrt{I(\mathbf{y}^*) + \left(\frac{L^2 \|\Sigma \Sigma^\top\|_{\mathcal{F}}}{K_2} \right)^{\frac{2}{3}}}.$$

Under the same parameter conditions given in subsection 5.1, we can compute the gain matrix to be $K = \text{diag}(K_1, K_2) = \text{diag}(173.311, 0.03)$, and further present the stabilizing results in Fig. 2. Here we only make experiments under the noise intensity of **Case 2**. From this figure, it seems difficult to say which controller is better. We further compare them from the gain cost, the convergent radius and the dependence on the assumption A.2. A large advantage of the PBC is that it can work in no need of A.2. However, it needs larger control gain K_1 : 0.02 vs. 173.31. Moreover, the convergent radius can be computed to be $6.4474 \times 10^5 / 1.2015 \times 10^6$ for Eq.(6)/the PBC using Eq. (15)/Eq. (18), beyond which $\mathcal{L}V \leq 0$. This implies that the convergence region by Eq. (6) is smaller than that by the PBC. Since the assumption A.2 is easy to operate, Eq. (6) looks more competitive than the PBC for sidCSTRs.

6 Conclusion

We present a modeling framework and a stabilizing strategy in the sense of NSES for the stochastic CSTR process. The main conclusion includes: (1) a sidCSTR

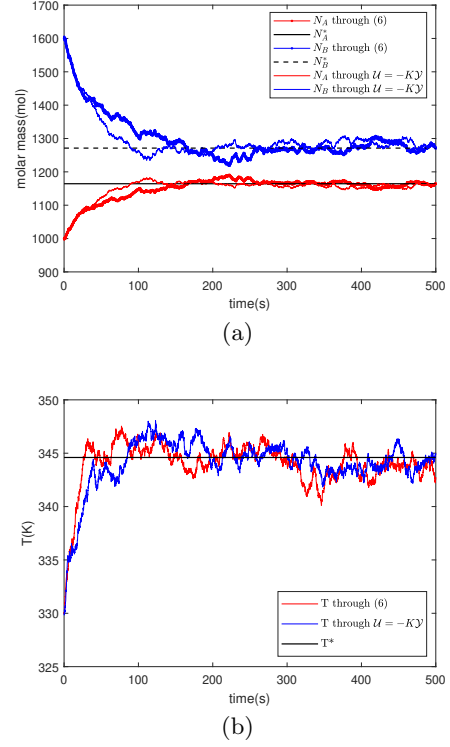


Fig. 2. Comparisons of state responses between different controllers in **Case 2**: (a) molar mass; (b) temperature.

model is proposed that considers unknown but bounded fluctuations in kinetics, flow rates and heat exchange; (2) a feedback control law is developed that stabilizes the sidCSTR system to reach NSES; and (3) the lower bound of the distribution for the state of the NSES sidCSTR system is analyzed both from the probability domain and the time domain.

Acknowledgements

This work was supported by the National Natural Science Foundation of China [Grant No. 12071428 and 62111530247] and the Zhejiang Provincial Natural Science Foundation of China [Grant No. LZ20A010002].

References

- Adebekun, A. K., & Schork, F. J. (1991). On the global stabilization of nth-order reactions. *Chemical Engineering Communications*, 100(1), 47-59.
- Alvarez-Ramirez, J., & Femat, R. (1999). Robust PI stabilization of a class of chemical reactors. *Systems & Control Letters*, 38(s 4-5), 219-225.
- Aris, R., & Amundson, N. R. (1958). An analysis of chemical reactor stability and control. *Chemical Engineering Science*, 20(3), 195-236.
- Baratti, R., Tronci, S., Schaum, A., & Alvarez, J. (2016). Dynamics of nonlinear chemical process with multiplicative stochastic noise. *IFAC-PapersOnLine*, 49 (7), 869-874.

Deng, H., Krstic, M., & Williams, R. J. (2001). Stabilization of stochastic nonlinear systems driven by noise of unknown covariance. *IEEE Transactions on Automatic Control*, 46(8), 1237–1253.

Dynkin, E. B. (1965). Markov processes. *Die Grundlehren Der Mathematischen Wissenschaften*, 121/122(9), 77–104.

Fang, Z., & Gao, C. (2020). Time-domain boundedness of noise-to-state exponentially stable systems. *ESAIM: Control, Optimisation and Calculus of Variations*, 26(4), 105, 23pp.

Hoang, H., Couenne, F., Jallut, C., & Gorrec, Y. L. (2011). The port-Hamiltonian approach to modeling and control of Continuous Stirred Tank Reactors. *Journal of Process Control*, 21(10), 1449–1458.

Hoang, H., Couenne, F., Jallut, C., & Gorrec, Y. L. (2012). Lyapunov-based control of non isothermal Continuous Stirred Tank Reactors using irreversible thermodynamics. *Journal of Process Control*, 22(2), 412–422.

Khasminskii, R. (2011). *Stochastic stability of differential equations*. Springer Science & Business Media.

Ligon, J., & Amundson, N. (1981). An analysis of chemical reactor stability and control-XVI : The stochastic stirred pot. *Chemical Engineering Science*, 36(4), 687–694.

Mao, X. (2011). *Stochastic Differential Equations and Applications (Second Edition)*. Woodhead.

Pell, T. M., & Aris, R. (1969). Some problems in chemical reactor analysis with stochastic features. *Industrial & Engineering Chemistry Fundamentals*, 8(2), 192–192.

Ramirez, H., Gorrec, Y. L., Maschke, B., & Couenne, F. (2016). On the passivity based control of irreversible processes: a port-Hamiltonian approach. *Automatica*, 64(C), 105–111.

Ratto, M. (1998). A theoretical approach to the analysis of PI-controlled CSTRs with noise. *Computers & Chemical Engineering*, 22(11), 1581–1593.

Yafei Lu received the B.Sc degree in School of mathematics and statistics from Zhengzhou university, China, in 2015. Since 2015 she has been studying for a Ph. D. degree in mathematics from Zhejiang university, China.



Her research interests are in the areas of control theory, systems and synthetic biology, and non-equilibrium thermodynamics.

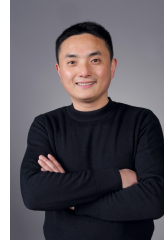
Zhou Fang received the B.Sc. and Ph. D. degrees in mathematics from Zhejiang university, China, in 2014 and 2019 respectively.



His research interests are in the areas of control theory, systems and synthetic biology, and non-equilibrium thermodynamics.

Chuanhou Gao received the B.Sc. degrees in Chemical Engineering from Zhejiang University of Technology, China, in 1998, and the Ph.D. degrees in Opera-

tional Research and Cybernetics from Zhejiang University, China, in 2004. From June 2004 until May 2006, he was a Postdoctor in the Department of Control Science and Engineering at Zhejiang University. Since June 2006, he has joined the Department of Mathematics at Zhejiang University, where he is currently a Professor. He was a visiting scholar at Carnegie Mellon University from Oct. 2011 to Oct. 2012.



His research interests are in the areas of data-driven modeling, control and optimization, chemical reaction network theory and thermodynamic process control. He is an associate editor of IEEE Transactions on Automatic Control, editor of Metallurgical Industry Automation, and guest editor of IEEE Transactions on Industrial Informatics, ISIJ International and Journal of Applied Mathematics.



Denis Dochain received his degree in Electrical engineering in 1982 from the Université Catholique de Louvain, Belgium. He completed his Ph.D. thesis and a “thèse d’agrégation de l’enseignement supérieur” in 1986 and 1994, respectively, also at the Université Catholique de Louvain, Belgium. He has been CNRS associate researcher at the LAAS (Toulouse, France) in 1989, and Professor at the Ecole Polytechnique de Montréal, Canada in 1987-88 and 1990-92. He has been with the FNRS (Fonds National de la Recherche Scientifique, National Fund for Scientific Research), Belgium since 1990. Since September 1999, he is Professor at the ICTEAM (Institute), Université Catholique de Louvain, Belgium, and Honorary Research Director of the FNRS. He has been invited professor at Queen’s University, Kingston, Canada between 2002 and 2004. He is full professor at the UCL since 2005. He is the Editor-in-Chief of the Journal of Process Control, senior editor of the IEEE Transactions of Automatic Control and associate editor of Automatica. He is active in IFAC since 1999 (Council member, Technical Board member, Publication Committee member and chair, TC and CC chair). He received the IFAC outstanding service award in 2008 and is an IFAC fellow since 2010. He received the title of Doctor Honoris Causa from the INP Grenoble on December 13, 2020. His main research interests are in the field of nonlinear systems, thermodynamics based control, parameter and state estimation, adaptive extremum seeking control and distributed parameter systems, with application to microbial ecology, environmental, biological and chemical systems, and electrical and mechanical systems. He is the (co-)author of 5 books, more than 160 papers in refereed journals and 260 international conference papers.