



# Distributed control of interconnected Chemical Reaction Networks with delay

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## ABSTRACT

This paper introduces a control approach for a class of Chemical Reaction Networks (CRNs) that are interconnected through a delayed convection network. First, a control-oriented model is proposed for interconnected CRNs. Second, based on this model, a distributed control method is introduced which assures that each CRN can be driven into a desired fixed point (setpoint) independently of the delay in the convection network. The proposed algorithm is also augmented with a disturbance attenuation term to compensate the effect of unknown input disturbances on setpoint tracking performance. The control design applies the theory of passive systems and methods developed for multi-agent systems. Simulation results are provided to show the applicability of the proposed control method.

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## 1. Introduction

The control of plant-wide industrial processes is in the focus of the researchers for decades [1,2]. The decentralized or distributed control approaches are advantageous in process network applications to reduce the communication costs and possible communication hazards that could arise in the case of centralized control [3]. Because of their practical importance and challenging nature, the different nonlinear control approaches applied in process control have developed their own version of decentralized, distributed or hierarchical control architectures.

In the paper [4] the authors proposed a distributed control approach for such interconnected processes that can be modeled as linear time-invariant systems based on passivity theory. This control method also takes into consideration the transport delay in the interconnections among the processes.

A powerful control approach of interconnected process systems is based on the thermodynamic characterization of such systems. A modeling framework has been developed in [5] for networks of chemical processes considering also the thermodynamic effects.

Using the theory of cascade-connected nonlinear systems and the properties of Metzler and Hurwitz matrices, a stabilizing decentralized control approach was designed in [6] utilizing the hierarchical structure of conservation based process models.

The popular and powerful model predictive control approach is also used in distributed and in hierarchical frameworks (see the paper [7] for a review). This approach can handle nonlinear interconnected process systems, as well. A more recent review highlighting future research directions in this approach is available in [8]. Constrained control methods can be applied when input or state bounds should also be taken into consideration, see e.g. [9] or [10].

Chemical Reaction Network theory provides efficient models and techniques to describe and analyze not only the dynamics of chemical reactions [11], but a more wide class of nonlinear process systems. The study [12] deals with the modeling of interconnected reactors and shows that the transport mechanism can be described by a linear CRN model. The CRN models describe positive systems and can efficiently capture complex nonlinear dynamical phenomena. The paper [13] offers a modeling approach for CRNs with inflows and outflows and discusses the relation of these systems with the consensus dynamics. The stability of these systems is most often analyzed using entropy-based Lyapunov functions [14].

Unfortunately, however, only a few of the nonlinear process control approaches that were developed for controlling intercon-

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nected subsystems are able to explicitly handle delays in dynamic analysis and controller design. Some exceptions include the paper [15], where the synchronization problem of a class of interconnected nonlinear biochemical processes was considered by taking an input-output modeling approach. Important modeling and stability and control related results on delayed kinetic systems can be found in [16], [17], [18], [19] and [20].

In this study a control-oriented modeling approach is proposed for interconnected CRNs, relying both on compartmental systems [21], and CRN theory. The model takes into consideration both the nonlinear nature of the chemical processes and the unknown transport delays in the convection network. A distributed setpoint control algorithm is introduced which assures that the concentration levels of the chemicals in each reactor reach prescribed fixed points. The proposed control method can also attenuate the effect of unknown input disturbances on the control performance. The resulting control algorithm has an easily implementable form, it is independent of the CRNs kinetics and the delay in the convection network. The stability and tracking performance of the interconnected CRN with the proposed control is analyzed using techniques borrowed from the theory of multi-agent systems.

## 2. Interconnected passive systems

### 2.1. Passive subsystems

Consider an interconnected system consisting of  $\mathcal{C}$  subsystems in which the input of each subsystem may depend on the outputs of the other subsystems.

Each subsystem is modeled using ODEs (Ordinary Differential Equations) in the form

$$\dot{\mathbf{c}}^{(j)} = \mathbf{f}^{(j)}(\mathbf{c}^{(j)}) + \mathbf{G}^{(j)}(\mathbf{c}^{(j)})\mathbf{u}^{(j)}, \quad \mathbf{c}^{(j)}(0) = \mathbf{c}_0^{(j)}, \quad (1)$$

$$\mathbf{y}^{(j)} = \mathbf{h}^{(j)}(\mathbf{c}^{(j)})$$

where  $\mathbf{c}^{(j)} \in \mathbb{R}^n$ ,  $\mathbf{y}^{(j)} \in \mathbb{R}^m$  are the state-, output- and input vectors,  $\mathbf{f}^{(j)}(\cdot)$ ,  $\mathbf{h}^{(j)}(\cdot)$ ,  $\mathbf{G}^{(j)}(\cdot)$  are smooth mappings with appropriate dimensions such that  $\mathbf{f}^{(j)}(\mathbf{0}) = \mathbf{0}$ ,  $\mathbf{h}^{(j)}(\mathbf{0}) = \mathbf{0}$ ,  $j = 1 \dots \mathcal{C}$ .

**Definition 1.** System (1) is called *passive*, if there exists a continuously differentiable function  $S^{(j)} : \mathbb{R}^n \rightarrow \mathbb{R}$  such that

$$S^{(j)}(\mathbf{c}^{(j)}) \geq 0, \quad \forall \mathbf{c}^{(j)}, \quad (2)$$

$$S^{(j)}(\mathbf{0}) = 0, \quad (3)$$

$$\dot{S}^{(j)} \leq \mathbf{y}^{(j)T} \mathbf{u}^{(j)}, \quad \forall \mathbf{u}^{(j)}, \mathbf{c}^{(j)}. \quad (4)$$

$S$  is called the *storage function* of (1) (see, e.g. [22]).

The input-affine system (1) is passive iff the following conditions hold

$$\frac{\partial S^{(j)}}{\partial \mathbf{c}^{(j)}} \mathbf{f}^{(j)}(\mathbf{c}^{(j)}) \leq 0, \quad (5)$$

$$\frac{\partial S^{(j)}}{\partial \mathbf{c}^{(j)}} \mathbf{G}^{(j)}(\mathbf{c}^{(j)}) = \left( \mathbf{h}^{(j)}(\mathbf{c}^{(j)}) \right)^T, \quad (6)$$

see e.g. [23].

Passivity theory plays a key role in analyzing the stability of nonlinear systems as it is shown that passivity of (1) involves the stability of the autonomous system  $\dot{\mathbf{c}}^{(j)} = \mathbf{f}^{(j)}(\mathbf{c}^{(j)})$  under mild conditions [22].

### 2.2. Interconnections

The underlying graph of the interconnected system is a directed graph with  $\mathcal{C}$  vertices in which each vertex corresponds to a subsystem. There is a directed edge from the vertex  $k$  to the vertex  $j$  if

the input of the  $j$ th subsystem depends explicitly on the output of the  $k$ th subsystem.

*Neighbor set* of the  $j$ th vertex ( $\mathcal{N}_j$ ): the  $k$ th vertex belongs to  $\mathcal{N}_j$  if there is a directed edge from the vertex  $k$  to the vertex  $j$ .

Alike the modeling concepts developed for large-scale systems [3], consider the input of each subsystem ( $\mathbf{u}^{(j)}$ ) as the sum of a local control input ( $\mathbf{u}_L^{(j)}$ ) and an the interconnection input term which has the form:

$$\mathbf{i}^{(j)}(t) = \mathbf{i}^{(j)} \left( \mathbf{y}^{(k_1)}(t - T_{k_1j}), \dots, \mathbf{y}^{(k_j)}(t - T_{k_jj}) \right), \quad (7)$$

where  $\mathbf{y}^{(k_l)}$  are the outputs of the subsystems of the neighbor set  $\mathcal{N}_j$  ( $\dim(\mathcal{N}_j) = J$ ), and  $0 \leq T_{k_lj} < \infty$  is a constant transport delay from the agent  $k_l$  to the agent  $j$ .

The outputs of the subsystems in the interconnected system are *synchronized* if  $\lim_{t \rightarrow \infty} |\mathbf{y}^{(j)}(t) - \mathbf{y}^{(k)}(t - T_{kj})| \rightarrow \mathbf{0}$ ,  $\forall j, k$ .

Assume the interconnection inputs in the form (*synchronization protocol*)

$$\mathbf{i}^{(j)}(t) = \sum_{k \in \mathcal{N}_j} w_{kj} (\mathbf{y}^{(k)}(t - T_{kj}) - \mathbf{y}^{(j)}(t)), \quad w_{kj} > 0, \quad (8)$$

and  $\mathbf{u}_L^{(j)} = \mathbf{0}$  element-wise. For the analysis of the interconnected systems with such subsystem inputs the following functional can be applied:

$$S_\Sigma = \sum_{i=1}^N S^{(i)} + \sum_{j=1}^N \sum_{k \in \mathcal{N}_j} \int_{t-T_{kj}}^t \mathbf{y}^{(j)T}(\tau) \mathbf{y}^{(j)}(\tau) d\tau. \quad (9)$$

It is shown in [24,25] that, under certain assumptions on the underlying graph and the subsystems, the interconnection input (8) assures the synchronization of the subsystems.

## 3. Basic modeling notions

### 3.1. Chemical Reaction Networks and their stability

Chemical Reaction Networks (abbreviated as CRNs) are composed of *elementary irreversible reactions*  $\mathcal{R}_k : C_i \rightarrow C_j$ ,  $k = 1 \dots R$ , where  $C_j$ ,  $j = 1 \dots m$  are the so called complexes. A *complex*  $C_j$  is formally a linear combination of species  $X_i$ ,  $i = 1 \dots K$ , such that  $C_j = \sum_{i=1}^K \beta_{ij} X_i$ , for  $j = 1 \dots m$ , where  $\beta_{ij}$  is the nonnegative stoichiometric coefficient corresponding to species  $X_i$  in complex  $C_j$ .

The concentrations of the species are collected into a vector  $\mathbf{c} \in \mathbb{R}^K$  so that  $\mathbf{c}_i = [X_i]$  for  $i = 1 \dots K$ . The dynamics of a CRN describing the time evolution of the concentrations of the species induced by the reactions can be written in the following form assuming constant volume and temperature (see, e.g. [26]):

$$\dot{\mathbf{c}} = M\varphi(\mathbf{c}) = Y A_\kappa \varphi(\mathbf{c}), \quad \mathbf{c}(0) = \mathbf{c}_0. \quad (10)$$

where  $\mathbf{c}_0$  is strictly positive element-wise, and  $Y \in \mathbb{R}^{K \times m}$  is the complex composition matrix the  $j$ th column of which contains the stoichiometric coefficients of complex  $C_j$ , i.e.  $Y_{ij} = \beta_{ij}$ ,  $\forall i, j$ . Moreover,  $\varphi_i(\mathbf{c}) = \prod_{i=1}^K c_i^{Y_{ik}}$  is the mass action vector and  $A_\kappa \in \mathbb{R}^{m \times m}$  is the *Kirchhoff matrix*:

$$A_\kappa(i, j) = \begin{cases} \kappa_{ji}, & \text{for } j \neq i \\ -\sum_{\ell \neq j} \kappa_{j\ell}, & \text{if } j = i. \end{cases} \quad (11)$$

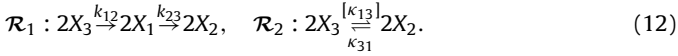
where  $\kappa_{ji}$  is the rate constant of the reaction  $\mathcal{R}_k : C_j \rightarrow C_i$ .

The reaction vector of  $\mathcal{R}_k$  is formed by the corresponding stoichiometric vectors, such that  $\mathbf{e}_k = \mathbf{Y}_{\cdot i} - \mathbf{Y}_{\cdot j}$ . The span of the reaction vectors defines the stoichiometric subspace of the CRN:

$\mathcal{S}_c = \text{span}\{\mathbf{e}_k\}$ . The positive stoichiometric compatibility classes of a CRN are represented by  $\mathcal{S}_{c_0} = (\mathbf{c}_0 + \mathcal{S}_c) \cap \mathcal{R}^{ns+}$ .

The general CRN model (10) may have multiple (even infinite number of) steady states in the whole state space. Therefore, the structure and number of equilibria are most often studied by restricting the dynamics to the stoichiometric compatibility classes corresponding to different initial conditions.

**Example 1** (A simple CRN). Let us consider a Chemical Reaction Network consisting of the following reactions:



The model contains three species:  $X_1, X_2, X_3$ , and three complexes:  $C_1 = 2X_3, C_2 = 2X_1, C_3 = 2X_2$ . From these, the complex composition matrix can be written as

$$Y = \begin{bmatrix} 0 & 2 & 0 \\ 0 & 0 & 2 \\ 2 & 0 & 0 \end{bmatrix} \quad (13)$$

We can see from (12) that the network contains four elementary reactions. The rate coefficients of these reactions are the non-zero off-diagonal elements of the Kirchhoff matrix which is given by

$$A_K = \begin{bmatrix} -(\kappa_{12} + \kappa_{13}) & 0 & \kappa_{31} \\ \kappa_{12} & -\kappa_{23} & 0 \\ \kappa_{13} & \kappa_{23} & -\kappa_{31} \end{bmatrix} \quad (14)$$

The pair  $(Y, A_K)$  is called a *realization* of a kinetic system with a given coefficient matrix  $M$  and reaction-monomial vector  $\varphi$ , where the complex composition matrix  $Y$  is determined by  $\varphi$ . It is important to note that a realization of a kinetic system may not be unique, i.e. there may exist more than one Kirchhoff matrix  $A_K$  for a kinetic dynamics given by  $M$  and  $\varphi$  [27].

There are important structural properties of a CRN realization that can be used for determining the stability properties of the dynamics, that are the deficiency and the reversibility properties. The *deficiency* of a CRN realization is defined as  $\delta = \dim(\text{Ker} Y \cap \text{Im} A_K)$ . A CRN is *weakly reversible* if the existence of a directed path (i.e. reaction sequence) from the complex  $C_i$  to the complex  $C_j$  implies the existence of a directed path from  $C_j$  to  $C_i$ .

Although weak reversibility and zero deficiency is a realization property of a CRN, they have important implications on the stability of the CRN system. If a CRN is weakly reversible and has zero deficiency then the system (10) has exactly one equilibrium point ( $\mathbf{c}^*$ ) in each positive stoichiometric compatibility class [11] that is at least locally stable with the following Lyapunov function:

$$\tilde{S}(\mathbf{c}) = \sum_{i=1}^K \left[ c_i \left( \ln \frac{c_i}{c_i^*} - 1 \right) + c_i^* \right] \quad (15)$$

An equilibrium point  $\mathbf{c}^*$  of the CRN (10) is called *complex balanced* if

$$A_K \varphi(\mathbf{c}^*) = \mathbf{0} \quad (16)$$

It is also known that if there exists a complex balanced equilibrium in a CRN, then all other equilibria are complex balanced, too [26]. Therefore, complex balance is a system property once the CRN structure and parameters are fixed, and thus a CRN itself can be called complex balanced if (16) is fulfilled. However, a kinetic differential equation may have several complex balanced and non-complex-balanced realizations [27]. It is important that complex balance implies weak reversibility. Complex balance is strongly related to the stability of kinetic systems. According to the well-known Global Attractor Conjecture (GAC), complex balanced CRNs are globally stable with the Lyapunov function (15). The GAC was

proved for several special cases, most remarkably for CRNs with one reaction graph component [28], and a proof for the general problem has been reported in [29]. A significant result in the theory of CRNs is that any deficiency zero weakly reversible network is complex balanced independently of the values of the rate constants [30]. This ensures a robust stability property which can be important in the general theory of nonnegative systems [31].

In order to have passive agents for the analysis, we consider the following sub-class of CRNs.

**Assumption 1.** The CRN (10) is complex balanced.

**Assumption 2.** The CRN (10) is persistent.

Persistence means that the trajectories of the system (10) do not approach the boundary  $\partial \mathbb{R}_+^K$  arbitrarily close, i.e.  $\forall i = 1 \dots K$  it stands:  $c_i(t) > 0$  if  $c_{i0} > 0$  and  $t \geq 0$ . It is important to note that Assumptions 1 and 2 are strongly related. In the case of CRNs with one graph component, complex balance implies persistence [28]. We recall the important special case that deficiency zero weakly reversible networks are complex balanced for any positive rate coefficients [30]. Moreover, according to [29] (which is not published officially at the time of writing) the dynamics of any complex balanced CRN is persistent.

### 3.2. Network of CRNs connected by convection with delay

It is considered that the mass action CRNs are located in continuously stirred tank reactors (CSTRs) that are connected through static connections. In order to have a usual CRN model considered in Section 3.1, we assume constant volume, constant temperature and constant physico-chemical properties in each CSTR.

As we shall see later in Section 4.1, the above assumptions together with Assumptions 1 and 2 ensure that the local CSTRs are passive with a certain input-output pair. In practice the constant temperature assumption – that implies constant physico-chemical properties with constant pressure – is approximately valid for most of the biochemical applications, where also the complex balanced and persistent nature of the reaction network is also valid. The constant volume assumption, however, puts severe restrictions on the convection network as it will be described later in Section 3.2.2.

Each CSTR has an inlet and an outlet port with volumetric flow rates  $v_{li}$  and  $v_i$  such that  $v_{li} = v_i$ ,  $i = 1 \dots \mathcal{C}$ , where the number of CSTRs is denoted by  $\mathcal{C}$ .

We also introduce a *pseudo-CSTR* (CSTR0) for describing the environment. Because of the constant volume assumption of each internal CSTR, this assumption also holds for the environment, such that  $v_{l0} = v_0$ .

**Assumption 3.** In CSTR0 the concentration vector ( $\mathbf{c}^{(0)}$ ) is constant and strictly positive element-wise.

In the usual practically important cases the variation of the inlet feed – usually its composition but some times even its flow rate is changing – is the major disturbance to a plant, therefore, the constant concentrations assumption in CSTR0 that represents the environment does not always hold in practice. However, one can relax this assumption if a disturbance rejective extension of the control scheme is developed: this technique is used for our proposed control method in Section 4.3.

#### 3.2.1. Open CRN model

A number of  $\mathcal{C}$  mass-action Chemical Reaction Networks are considered in the system under investigation. Let  $Y^{(j)}$  and  $A_K^{(j)}$  be the stoichiometric and Kirchhoff matrices of the ODE model of the CRN that takes place in the  $j$ th CSTR:

$$\dot{\mathbf{c}}^{(j)} = Y^{(j)} A_K^{(j)} \varphi^{(j)}(\mathbf{c}^{(j)}). \quad (17)$$

Then the component mass balance containing the in- and out-flow convective terms of the  $j$ th CSTR reads as

$$\frac{d\mathbf{c}^{(j)}}{dt} = \frac{1}{V_j} \left( \sum_{\ell=0}^c \alpha_{\ell j} v_{\ell} \mathbf{c}^{(\ell)} - v_j \mathbf{c}^{(j)} \right) + Y^{(j)} A_k^{(j)} \varphi(\mathbf{c}^{(j)}) \quad (18)$$

where  $j = 1 \dots \mathcal{C}$ .

### 3.2.2. The convective connections

Connections are set up between the reactors such that the outlet of the  $i$ th reactor is divided into fractions with the fraction coefficients  $\alpha_{ij}$  that are fed into the  $j$ th reactor. This means that

$$\sum_{\ell=0}^c \alpha_{i\ell} = 1, \quad i = 0, \dots, \mathcal{C}, \quad (19)$$

$$v_{ij} = v_j = \sum_{\ell=0}^c \alpha_{\ell j} v_{\ell}, \quad j = 0, \dots, \mathcal{C}. \quad (20)$$

Because constant volume is assumed in every region, the sum of convective inflows

$$v_0 = \sum_{\ell=0}^c \alpha_{0\ell} v_{\ell} \quad (21)$$

is equal to the sum of the convective outflows of the process system. It is important to note that the composite system consisting of the original process system and its environment is closed with  $\mathcal{C} + 1$  regions each of constant volume.

With the notations above we can formulate the convection matrix as follows:

$$C_c = \begin{bmatrix} -(1 - \alpha_{00})v_0 & \alpha_{10}v_1 & \alpha_{20}v_2 & \dots & \alpha_{c0}v_c \\ \alpha_{01}v_0 & -(1 - \alpha_{11})v_1 & \alpha_{21}v_2 & \dots & \alpha_{c1}v_c \\ \dots & \dots & \dots & \dots & \dots \\ \alpha_{0c}v_0 & \alpha_{1c}v_1 & \alpha_{2c}v_2 & \dots & -(1 - \alpha_{cc})v_c \end{bmatrix} \quad (22)$$

This matrix will be termed *Kirchhoff convection matrix*.

The constant volume assumption implies that  $C_c \mathbf{1} = \mathbf{0}$  which is also a consequence of Eq. (19). Here  $\mathbf{1} = (1 \dots 1)^T$  and  $\mathbf{0} = (0 \dots 0)^T$ . Moreover, Eq. (20) implies that  $\mathbf{1}^T C_c = \mathbf{0}^T$ .

The above two equations and the sign pattern of  $C_c$  shows that Kirchhoff convection matrices are both row and column conservation matrices. When there is no flow from the  $i$ th CSTR to the  $j$ th CSTR, the parameter  $\alpha_{ij} = 0$  in  $C_c$ . The Kirchhoff matrix describes the structure of the underlying directed graph  $\mathcal{G}_c$  of the convection network: the weighted Laplacian of  $\mathcal{G}_c$  is  $-C_c$ . On  $\mathcal{G}_c$  the following assumption is made:

**Assumption 4.**  $\mathcal{G}_c$  contains a directed spanning tree with root CSTR0.

This assumption means that the supply from the environment reaches each individual reactor, there are no reactors which are unreachable from the CSTR0 in the process network.

This assumption is easy to verify in practice based on the flow sheet of the plant, but it may not hold in all practical cases for the overall plant. At the same time, if the supply is really necessary for the production, then the plant can be naturally decomposed into sub-plants that obey Assumption 4 individually, and the controller design can be done for them separately.

### 3.2.3. Connected model with convection delays

Consider that transport delays are present in the interconnections among the CSTRs. Denote the delay value between the  $\ell$ th and

$j$ th CSTR as  $T_{\ell j}$ . Then the state equation of (18) obtains the modified form:

$$V_j \frac{d\mathbf{c}_i^{(j)}}{dt} = \sum_{\ell=0}^c \alpha_{\ell j} v_{\ell} \mathbf{c}_i^{(\ell)}(t - T_{\ell j}) - v_j \mathbf{c}_i^{(j)} + V_j Y^{(j)} A_k^{(j)} \varphi(\mathbf{c}^{(j)}), \quad (23)$$

$$\mathbf{c}_i^{(\ell)}(\tau) = \xi_i^{(\ell)}(\tau), \quad -T_{\ell j} \leq \tau \leq 0.$$

Here  $j = 1 \dots \mathcal{C}$  and  $\xi_i^{(\ell)}(\tau) \in \mathbb{C}^+$  is an initial condition function.

## 4. Distributed controller design

The design of the distributed control scheme is based on the passivity analysis of the interconnected CSTRs that is given in the next section. Thereafter the distributed setpoint control design will be introduced and finally, its disturbance rejective version is described.

### 4.1. Storage function and passivity analysis of open CRNs

Let us consider that the input of the  $j$ th open CRN (23) is the vector  $\mathbf{u}^{(j)} \in \mathbb{R}^K$  so that:

$$\frac{d\mathbf{c}^{(j)}}{dt} = Y^{(j)} A_k^{(j)} \varphi(\mathbf{c}^{(j)}) + \frac{1}{V_j} \mathbf{u}^{(j)}. \quad (24)$$

Note that the above general form is derived from Eq. (18) by considering the difference between the convective component mass in- and outflow term as an input. However, because of the constant volume assumption in each CSTR one can arbitrarily manipulate only the local inlet concentrations from the environment, but not the flow rates.

Choose the *storage function* for the  $j$ th CRN as the weighted form of the Lyapunov function (15):

$$S^{(j)} = \zeta V_j \left( (\text{Lnc}^{(j)} - \text{Lnc}^{(j)*})^T \mathbf{c}^{(j)} - \mathbf{1}^T (\mathbf{c}^{(j)} - \mathbf{c}^{(j)*}) \right). \quad (25)$$

Here  $\zeta \in \mathbb{R}_+$  is a positive finite constant,  $\text{Ln}$  is the natural logarithm applied element-wise to a vector, and  $\mathbf{c}^{(j)*}$  is a (generally initial condition dependent) equilibrium point of the  $j$ th CRN.

For the passivity the output mapping of the subsystem has to satisfy the relation (6). From the model (24) it results that the output of the  $j$ th subsystem has to have the form:

$$\mathbf{y}^{(j)} = \frac{1}{V_j} \left( \frac{\partial S^{(j)}}{\partial \mathbf{c}^{(j)}} \right)^T. \quad (26)$$

By Eqs. (25) and (26) yields the *passive output vector* of the  $j$ th CRN:

$$\mathbf{y}^{(j)} = \zeta (\text{Lnc}^{(j)} - \text{Lnc}^{(j)*}). \quad (27)$$

**Lemma 1.** The open CRN system (24) with Assumption 1 is passive from the input  $\mathbf{u}^{(j)}$  to the output  $\mathbf{y}^{(j)}$  in Eq. (27).

**Proof.** Consider the storage function (25). The time derivative of  $S^{(j)}$  reads as:

$$\dot{S}^{(j)}(\mathbf{c}^{(j)}) = \zeta V_j \left( (\text{Lnc}^{(j)} - \text{Lnc}^{(j)*} + \mathbf{1})^T \dot{\mathbf{c}}^{(j)} - \mathbf{1}^T \dot{\mathbf{c}}^{(j)} \right), \quad (28)$$

$$\dot{S}^{(j)}(\mathbf{c}^{(j)}) = \zeta V_j (\text{Lnc}^{(j)} - \text{Lnc}^{(j)*})^T \left( \frac{1}{V_j} \mathbf{u}^{(j)} + Y^{(j)} A_k^{(j)} \varphi(\mathbf{c}^{(j)}) \right). \quad (29)$$

By Assumption 1, if  $\mathbf{u}^{(j)} = \mathbf{0}$ ,  $\dot{S}^{(j)}$  is non-increasing, see e.g. the study [11]. Hence,  $(\text{Lnc}^{(j)} - \text{Lnc}^{(j)*})^T Y^{(j)} A_k^{(j)} \varphi(\mathbf{c}^{(j)}) \leq 0$ . It yields:

$$\dot{S}^{(j)}(\mathbf{c}^{(j)}) \leq \mathbf{y}^{(j)T} \mathbf{u}^{(j)}, \quad (30)$$

i.e. the system (24) is passive.  $\square$



**Remark 1.** For passivity analysis of open CRNs a similar approach was taken in [32]: the input was chosen proportional to the positive input flow rate and the passive output was taken as  $(\text{Ln } \mathbf{c} - \text{Ln } \mathbf{c}^*)^T (\mathbf{c} - \mathbf{c}_{IN})$ , where  $\mathbf{c}_{IN}$  is the inlet concentration. However, for control purposes it is more beneficial to choose the input  $\mathbf{u}^{(j)}$  as in (24) since the control can be implemented by modifying the inlet concentration.

## 4.2. Distributed setpoint control

### 4.2.1. Control problem statement

Consider a process network consisting of  $\mathcal{C}$  subsystems (CRNs) that are interconnected through a delayed convection network. The reactions that take place in the reactors are described by the open CRN model (24).

**Control objective:** Let the setpoint of the  $j$ th CSTR be  $\mathbf{c}_{SP}^{(j)}$  that belongs to the equilibrium point set of (17). Design a distributed controller for each CRN such to assure that  $\mathbf{c}^{(j)} \rightarrow \mathbf{c}_{SP}^{(j)}$  as  $t \rightarrow \infty$ ,  $\forall j = 1 \dots \mathcal{C}$ .

### 4.2.2. Local control input

To implement the control, augment the input of each CRN with a local control inflow ( $v_{Lj} \mathbf{c}_L^{(j)}$ ) in which the species' concentrations ( $\mathbf{c}_L^{(j)}$ ) are specifiable and they are used as local control inputs, and  $v_{Lj} \in \mathbb{R}_+$  is the constant local input flow rate,  $j = 1 \dots \mathcal{C}$ ;  $v_{L0} = 0$ .

As it has already been noted before, the constant volume assumption in each CSTR implies that the flow rates cannot be freely manipulated, but they are related. Therefore, the local inlet species concentrations, that are different from the overall feed concentration assumed to be constant in Assumption 3, are the manipulable local inlet concentrations. In practical schemes, the volume (or overall mass) of each CSTR is held constant on a lower level of a hierarchical control scheme by manipulating the output flow rates [6,33] while the concentrations are controlled on a higher level: that is considered in this paper.

In the controlled process network the input vector of the  $j$ th open CRN has the form:

$$\mathbf{u}^{(j)} = \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_{\ell} \mathbf{c}^{(\ell)} (t - T_{\ell j}) + a_{Lj} \tilde{v}_j \mathbf{c}_L^{(j)} - \tilde{v}_j \mathbf{c}^{(j)} \quad (31)$$

where  $\tilde{v}_j = v_j + v_{Lj}$ ,  $a_{Lj} = v_{Lj} / \tilde{v}_j$ .

In the controlled process network the balance equations in the convection network (similar to Eq. (19)) read as:

$$\sum_{\ell=0}^{\mathcal{C}} a_{j\ell} = 1, \quad j = 0, \dots, \mathcal{C}, \quad (32)$$

$$\tilde{v}_j = \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_{\ell} + a_{Lj} \tilde{v}_j, \quad j = 0, \dots, \mathcal{C}. \quad (33)$$

Let us distinguish in the inflow vector  $\mathbf{u}^{(j)}$  the interconnection term ( $\mathbf{i}^{(j)}$ ) and the local control term ( $\mathbf{u}_L^{(j)}$ ) as follows:

$$\mathbf{u}^{(j)} = \mathbf{i}^{(j)} + \mathbf{u}_L^{(j)}, \quad (34)$$

$$\mathbf{i}^{(j)} = \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_{\ell} \mathbf{c}^{(\ell)} (t - T_{\ell j}) - (1 - a_{Lj}) \tilde{v}_j \mathbf{c}^{(j)}, \quad (35)$$

$$\mathbf{u}_L^{(j)} = a_{Lj} \tilde{v}_j (\mathbf{c}_L^{(j)} - \mathbf{c}^{(j)}). \quad (36)$$

The interconnection term contains the flows from - and to the other open CRNs. The value of local control flow can be specified through  $\mathbf{c}_L^{(j)}$ .

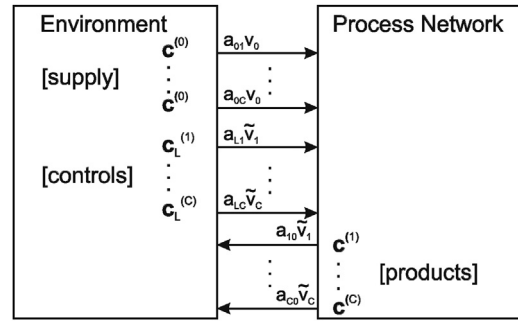


Fig. 1. Interconnections between the environment and process network.

The flows between the controlled process network and the environment are shown in Fig. 1.

### 4.2.3. The concept of synchronization-based control

The properties of the balance equations (32) and (33) can be exploited to achieve the distributed control objective by tracing back the formulated control problem to the synchronization problem, discussed in Section 2.

If the synchronization can be reached in the process network, the steady-state outputs of the CRNs take the same value, i.e.  $\mathbf{y}^{(j)} = \mathbf{y}^{(k)}$  as  $t \rightarrow \infty$ ,  $\forall j, k = 1 \dots \mathcal{C}$ .

As the supply has constant concentration vector, i.e.  $\mathbf{c}^{(0)} = \mathbf{c}^{(0)*}$  it can be considered that  $\mathbf{y}^{(0)}(t) = \mathbf{0}$ , see (27).

If the outputs of all CRNs are synchronized ( $\mathbf{y}^{(j)} = \mathbf{y}^{(i)} \forall i, j$ ), it yields that  $\mathbf{y}^{(j)} = \mathbf{y}^{(0)} = \mathbf{0} \forall j \in \mathcal{N}_0$ , i.e.  $\mathbf{c}^{(j)} = \mathbf{c}^{(j)*} \forall j \in \mathcal{N}_0$ . Moreover, if Assumption 4 holds,  $\mathbf{y}^{(j)} = \mathbf{0}$ , i.e.  $\mathbf{c}^{(j)} = \mathbf{c}^{(j)*} \forall j = 1 \dots \mathcal{C}$ .

According to the synchronization protocol (8) the input  $\mathbf{u}^{(j)}$  of the subsystem has to be designed as a function of the passive output ( $\mathbf{y}^{(j)}$ ) of the subsystem and the neighboring subsystems.

However, in the process network the interconnection input ( $\mathbf{i}^{(j)}$ ) depends on the state vectors ( $\mathbf{c}^{(j)}$ ), see Eq. (35).

Define the *desired input* vector  $\mathbf{u}_y^{(j)}$  of the CRNs as a function of the passive outputs:

$$\mathbf{u}_y^{(j)} = \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_{\ell} \mathbf{y}^{(\ell)} (t - T_{\ell j}) - (1 - a_{Lj}) \tilde{v}_j \mathbf{y}^{(j)}. \quad (37)$$

Here  $\mathbf{y}^{(j)} = \zeta (\text{Ln } \mathbf{c}^{(j)} - \text{Ln } \mathbf{c}_{SP}^{(j)})$  where  $\mathbf{c}_{SP}^{(j)}$  is the prescribed equilibrium point (setpoint)  $\forall j = 1 \dots \mathcal{C}$ .

The local control inputs  $\mathbf{u}_L^{(j)}$  of the CRNs in (34) have to be formulated such that they compensate the differences between the desired interconnection (necessary to reach the synchronization with the prescribed setpoint) and the real interconnections.

From the relations (35), (36), (37) we can compute the explicit form of the local control input which satisfies  $\mathbf{u}^{(j)} = \mathbf{i}^{(j)} + \mathbf{u}_L^{(j)} = \mathbf{u}_y^{(j)}$ :

$$\mathbf{c}_L^{(j)} = \mathbf{y}^{(j)} + \frac{1}{a_{Lj} \tilde{v}_j} \left( \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_{\ell} (\mathbf{y}^{(\ell)} (t - T_{\ell j}) - \mathbf{c}^{(\ell)} (t - T_{\ell j})) - \tilde{v}_j (\mathbf{y}^{(j)} - \mathbf{c}^{(j)}) \right). \quad (38)$$

It is visible from (38) that the computation of the control law of each subsystem requires the knowledge of the outputs of the neighboring (connected) subsystems. This necessitates the local communication of the controllers in a straightforward implementation. Therefore, the proposed approach can be classified as distributed control design as it is defined e.g. in [7,34].

#### 4.2.4. Stability and steady-states of the closed loop system

**Theorem 1.** Consider a system of interconnected CRNs in which each subsystem model is given by (24), (34), (35), (36) and the interconnection parameters are defined as in (32) and (33). If Assumptions 1–4 hold and  $\mathbf{c}_L^{(j)}$  in (36) can be chosen such that  $\mathbf{u}^{(j)} = \mathbf{u}_y^{(j)}$  element-wise  $\forall j = 1 \dots \mathcal{C}$ , then  $\mathbf{c}^{(j)}(t)$  is bounded for  $t \geq 0$  and  $\mathbf{c}^{(j)} \rightarrow \mathbf{c}_{SP}^{(j)}$  as  $t \rightarrow \infty \forall j > 0$ .

**Proof.** Based on the storage function  $S^{(j)}$ , given in (25), define the following Lyapunov-Krasovskii functional:

$$S_\Sigma = 2 \sum_{j=0}^{\mathcal{C}} S^{(j)} + \sum_{j=0}^{\mathcal{C}} \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_\ell \int_{t-T_{\ell j}}^t \mathbf{y}^{(\ell)T} \mathbf{y}^{(\ell)} d\xi. \quad (39)$$

The time derivative of it reads as

$$\dot{S}_\Sigma = 2 \sum_{j=0}^{\mathcal{C}} \dot{S}^{(j)} + \sum_{j=0}^{\mathcal{C}} \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_\ell \left( \mathbf{y}^{(\ell)T} \mathbf{y}^{(\ell)} - \mathbf{y}^{(\ell)T}(t - T_{\ell j}) \mathbf{y}^{(\ell)}(t - T_{\ell j}) \right). \quad (40)$$

By Lemma 1 it yields that  $\dot{S}^{(j)} \leq \mathbf{y}^{(j)T} \mathbf{u}^{(j)} \forall j$ . Accordingly:

$$\begin{aligned} \dot{S}_\Sigma &\leq 2 \sum_{j=0}^{\mathcal{C}} \mathbf{y}^{(j)T} \mathbf{u}^{(j)} + \sum_{j=0}^{\mathcal{C}} \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_\ell \left( \mathbf{y}^{(\ell)T} \mathbf{y}^{(\ell)} \right. \\ &\quad \left. - \mathbf{y}^{(\ell)T}(t - T_{\ell j}) \mathbf{y}^{(\ell)}(t - T_{\ell j}) \right). \end{aligned} \quad (41)$$

By choosing  $\mathbf{c}_L^{(j)}$  such that  $\mathbf{u}^{(j)} = \mathbf{i}^{(j)} + a_{Lj} \tilde{v}_j \left( \mathbf{c}_L^{(j)} - \mathbf{c}^{(j)} \right) = \mathbf{u}_y^{(j)} \forall j = 1 \dots \mathcal{C}$ , and since  $\mathbf{y}^{(0)} = \mathbf{0}$ , it yields:

$$\dot{S}_\Sigma \leq 2 \sum_{j=0}^{\mathcal{C}} \mathbf{y}^{(j)T} \left( \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_\ell \mathbf{y}^{(\ell)}(t - T_{\ell j}) - (1 - a_{Lj}) \tilde{v}_j \mathbf{y}^{(j)} \right) \quad (42)$$

$$+ \sum_{j=0}^{\mathcal{C}} \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_\ell \left( \mathbf{y}^{(\ell)T} \mathbf{y}^{(\ell)} - \mathbf{y}^{(\ell)T}(t - T_{\ell j}) \mathbf{y}^{(\ell)}(t - T_{\ell j}) \right).$$

As  $\tilde{v}_j = a_{Lj} \tilde{v}_j + \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_\ell = \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_\ell$  (the sum of inflows is equal to the sum of the outflows), it results that

$$\begin{aligned} \dot{S}_\Sigma &\leq \sum_{j=0}^{\mathcal{C}} \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_\ell \left( -\mathbf{y}^{(j)T} \mathbf{y}^{(j)} + 2\mathbf{y}^{(j)T} \mathbf{y}^{(\ell)}(t - T_{\ell j}) \right. \\ &\quad \left. - \mathbf{y}^{(\ell)T}(t - T_{\ell j}) \mathbf{y}^{(\ell)}(t - T_{\ell j}) \right), \\ \dot{S}_\Sigma &\leq - \sum_{j=0}^{\mathcal{C}} \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_\ell \left( \mathbf{y}^{(j)} - \mathbf{y}^{(\ell)}(t - T_{\ell j}) \right)^T \left( \mathbf{y}^{(j)} - \mathbf{y}^{(\ell)}(t - T_{\ell j}) \right) \leq 0. \end{aligned} \quad (43)$$

Let us introduce the notation

$$e_i^{(j,\ell)}(t) = y_i^{(j)}(t) - y_i^{(\ell)}(t - T_{\ell j}). \quad (44)$$

As  $\dot{S}_\Sigma \leq 0$  it yields that  $S_\Sigma(\infty) = \lim_{t \rightarrow \infty} S_\Sigma(t) < \infty$  for finite  $S_\Sigma(0)$ . Accordingly, by (43) it yields that:

$$\begin{aligned} \sum_{j=0}^{\mathcal{C}} \sum_{\ell=0}^{\mathcal{C}} a_{\ell j} \tilde{v}_\ell \sum_{i=1}^K \int_{t=0}^{\infty} \left( y_i^{(j)}(\xi) - y_i^{(\ell)}(\xi - T_{\ell j}) \right)^2 d\xi &\leq S_\Sigma(0) \\ -S_\Sigma(\infty) &< \infty. \end{aligned} \quad (45)$$

Hence,  $e_i^{(j,\ell)} \in L_2 \forall i, j, \ell$ .

Since  $S_\Sigma(t) < \infty$  and  $c_{SPi}^{(j)}$  is a finite, strictly positive constant  $\forall i, j$ , it yields that  $c_i^{(j)}$  and consequently  $e_i^{(j,\ell)}, y_i^{(j)} \in L_\infty \forall i, j, \ell$ . Hence, all the entries of  $\mathbf{c}^{(j)}$  are bounded  $\forall j$ .

$c_i^{(j)} \in L_\infty$  and  $y_i^{(j)} \in L_\infty$  implies that  $u_i^{(j)} \in L_\infty$ . By Eq. (24) it can be seen that  $\dot{c}_i^{(j)} \in L_\infty \forall i, j$ . The time derivative of  $y_i^{(j)}$  reads as  $\dot{y}_i^{(j)} = \zeta \dot{c}_i^{(j)} / c_i^{(j)}$ . By Assumption 2 it yields that  $\dot{y}_i^{(j)} \in L_\infty$ . Hence  $\dot{e}_i^{(j,\ell)} \in L_\infty \forall i, j, \ell$ .

As  $e_i^{(j,\ell)} \in L_2, \dot{e}_i^{(j,\ell)} \in L_\infty$  and  $\dot{e}_i^{(j,\ell)} \in L_\infty$ , by Barbalat's lemma, it yields that  $\lim_{t \rightarrow \infty} e_i^{(j,\ell)} = 0 \forall i, j, \ell$ .

Since  $T_{\ell j}$  are finite,  $\lim_{t \rightarrow \infty} y_i^{(j)}(t) = \lim_{t \rightarrow \infty} y_i^{(\ell)}(t), \forall i, j, \ell$ . From Assumption 3 it results that  $y_i^{(0)} = 0$ . Hence, by Assumption 4, it yields that  $\lim_{t \rightarrow \infty} c_i^{(j)}(t) = c_{iSP}^{(j)} \forall i, j$ .  $\square$

It is important to note that  $S_\Sigma$  in (39) and the local control input (38) are independent of the kinetic parameters of the CRN subsystems.

#### 4.3. Distributed setpoint control with disturbance attenuation

The control law, presented in the previous subsections, requires the knowledge of the convection network parameters. The augmented control proposed in this subsection follows the idea of high-gain control methods to attenuate the effects of unmodelled disturbances and uncertainties in the interconnections on the control performances: in stable control loops, with sufficiently high feedback gain the effect of the bounded disturbances on steady-state performances can be made arbitrarily small (see e.g. [35]).

Consider that the CRNs in the interconnected system, originally described by the model (24), are subject to additive disturbances, i.e. they can be modelled as

$$\frac{d\mathbf{c}^{(j)}}{dt} = Y^{(j)} A_K^{(j)} \varphi^{(j)}(\mathbf{c}^{(j)}) + \frac{1}{V_j} \mathbf{u}^{(j)} + \mathbf{d}^{(j)}, \quad j = 1 \dots \mathcal{C} \quad (46)$$

where the disturbance input  $\mathbf{d}^{(j)}(t) \in \mathbb{R}^K$ .

It is important to notice that the above general form is also derived from Eq. (18) by considering the difference between the convective component mass in- and outflow term as an input, and by separating the inflow term originating from the environment as the disturbance, i.e.  $\mathbf{d}^{(j)} = \alpha_{0j} \nu_0 \mathbf{c}^{(0)}$ . By assuming the disturbance changing in time, we have relaxed the constant environment concentration conditions in Assumption 3 in order to design a controller that rejects its effect on the controlled output.

Note that the passivity property, discussed in Lemma 1, is preserved from  $\mathbf{d}^{(j)}$  to  $\mathbf{y}^{(j)}$  when  $\mathbf{u}^{(j)} = \mathbf{0}$ . It is because the disturbance input vector has the same dimension and the same linear input structure as the local control input vector.

**Assumption 5.** The disturbance input  $\mathbf{d}^{(j)}$  is continuous and

$$\|\mathbf{d}^{(j)}\|_2 \leq d_M^{(j)} \quad (47)$$

where  $d_M^{(j)} \in \mathbb{R}_+$  is a finite constant.

Define the input and output vectors of all the subsystems

$$\mathbf{u} = (\mathbf{u}^{(1)T} \dots \mathbf{u}^{(c)T})^T, \quad (48)$$

$$\mathbf{d} = (\mathbf{d}^{(1)T} \dots \mathbf{d}^{(c)T})^T, \quad (49)$$

$$\mathbf{y} = (\mathbf{y}^{(1)T} \dots \mathbf{y}^{(c)T})^T. \quad (50)$$

By Assumption 5 it yields

$$\|\mathbf{d}\|_2 \leq d_M, \quad \text{where } d_M = \sqrt{\sum_{j=1}^{\mathcal{C}} \left( d_M^{(j)} \right)^2}. \quad (51)$$

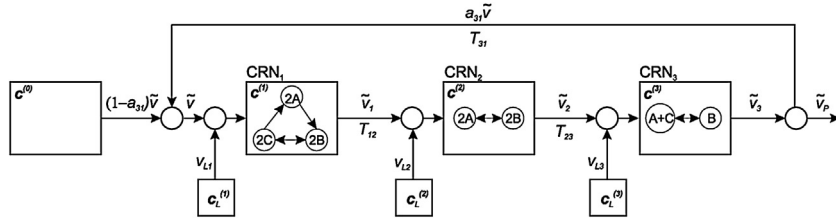


Fig. 2. Example of controlled interconnected CRNs.

To attenuate the effect of disturbances, augment the desired input, originally given in (37), as

$$\mathbf{u}_d^{(j)} = \sum_{\ell=0}^c a_{\ell j} \tilde{v}_\ell \mathbf{y}^{(\ell)}(t - T_{\ell j}) - (1 - a_{1j}) \tilde{v}_j \mathbf{y}^{(j)} - \frac{\gamma}{2} \mathbf{y}^{(j)} \quad (52)$$

where  $\gamma \in \mathbb{R}_+$  is the constant, finite control gain.

**Theorem 2.** Consider a system of interconnected CRNs in which each subsystem model is given by (46), (34), (35), (36) and the interconnection parameters are defined as in (32) and (33). If Assumptions 1–5 hold and  $\mathbf{c}_L^{(j)}$  in (36) can be chosen such that  $\mathbf{u}^{(j)} = \mathbf{u}_d^{(j)}$  element-wise  $\forall j = 1 \dots \mathcal{C}$  with

$$\gamma > 1 + \frac{d_M}{\varepsilon}, \quad 0 < \varepsilon < \infty, \quad (53)$$

then  $\mathbf{y}$  converges toward the set  $\{\mathbf{y} \mid \|\mathbf{y}\|_2 \leq \varepsilon\}$ .

**Proof.** Consider the Lyapunov function candidate  $S_\Sigma$  given in (39).

With the CRN model (46), by following similar arguments as in the proof of Theorem 1, the time derivative of  $S_\Sigma$  reads as:

$$\dot{S}_\Sigma \leq - \sum_{j=0}^c \sum_{\ell=0}^c a_{\ell j} \tilde{v}_\ell \mathbf{e}^{(j, \ell)T} \mathbf{e}^{(j, \ell)} - \gamma \|\mathbf{y}\|_2^2 + 2\mathbf{y}^T \mathbf{d} \quad (54)$$

where  $\mathbf{e}^{(j, \ell)} = \mathbf{y}^{(j)}(t) - \mathbf{y}^{(\ell)}(t - T_{\ell j})$ .

By applying that  $\mathbf{y}^T \mathbf{d} \leq \|\mathbf{y}\|_2 \|\mathbf{d}\|_2 \leq (\|\mathbf{y}\|_2^2 + \|\mathbf{d}\|_2^2)/2$ , and by Assumption 5 it yields:

$$\begin{aligned} \dot{S}_\Sigma &\leq - \sum_{j=0}^c \sum_{\ell=0}^c a_{\ell j} \tilde{v}_\ell \mathbf{e}^{(j, \ell)T} \mathbf{e}^{(j, \ell)} \\ &\quad + (d_M + (\gamma - 1)\|\mathbf{y}\|_2)(d_M - (\gamma - 1)\|\mathbf{y}\|_2). \end{aligned} \quad (55)$$

Since  $\gamma > 1 + \frac{d_M}{\varepsilon}$ , it yields that  $d_M + (\gamma - 1)\|\mathbf{y}\|_2 > 0$  and, if  $\|\mathbf{y}\|_2 > \varepsilon$ ,  $d_M - (\gamma - 1)\|\mathbf{y}\|_2 < 0$ . Accordingly,  $\dot{S}_\Sigma < 0$  i.e.  $S_\Sigma$  is a decreasing storage function if  $\|\mathbf{y}\|_2 > \varepsilon$ .

By (39) the decrease of  $S_\Sigma$  involves the decrease of  $S^{(j)}$  or the decrease of  $\int_{t-T_{\ell j}}^t \mathbf{y}^{(\ell)T} \mathbf{y}^{(\ell)} d\xi$  terms.

As  $S^{(j)} = 0$  iff  $\mathbf{y}_i^{(j)} = 0$ ,  $i = 1 \dots K$ ,  $j = 1 \dots \mathcal{C}$  the decrease of  $S^{(j)}$  involves that some  $\mathbf{y}_i^{(j)}$  tend to 0.

As  $T_{\ell j}$  are finite and positive  $\forall \ell, j$ , the decrease of  $\int_{t-T_{\ell j}}^t \mathbf{y}^{(\ell)T} \mathbf{y}^{(\ell)} d\xi$  also involves that some  $\mathbf{y}_i^{(j)}$  tend to 0.

The convergence of  $\mathbf{y}_i^{(j)}$  towards zero persists until  $\|\mathbf{y}\|_2 \geq \varepsilon$ , i.e. until the relation (53) is satisfied.  $\square$

#### 4.4. Restriction on convection flow rates: positivity

During the control design the physical meaning of the control inputs, that are concentrations, should be taken into account. This implies, that the control should always be positive, i.e.  $\mathbf{c}_L^{(j)} \geq \mathbf{0}$  element-wise  $\forall j = 1 \dots \mathcal{C}$ . These positivity conditions imply restrictions on the achievable setpoints that will be derived below.

Consider the equations used for control design  $\mathbf{u}^{(j)} = \mathbf{i}^{(j)} + \mathbf{u}_L^{(j)} = \mathbf{u}_Y^{(j)}$ , and  $\mathbf{i}^{(j)} + \mathbf{u}_L^{(j)} = \mathbf{u}_d^{(j)}$  respectively. In steady-state  $\mathbf{u}_Y^{(j)} = \mathbf{u}_d^{(j)} = \mathbf{0}$  since  $\mathbf{y}^{(j)} = \mathbf{0} \forall j = 1 \dots \mathcal{C}$ . Hence, the steady-state value of the control is

$$\mathbf{c}_L^{(j)*} = \frac{1}{a_{1j} \tilde{v}_j} (-\mathbf{i}_{SP}^{(j)} + a_{1j} \tilde{v}_j \mathbf{c}_{SP}^{(j)}) = \frac{1}{a_{1j} \tilde{v}_j} \left( -\sum_{\ell=0}^c a_{\ell j} \tilde{v}_\ell \mathbf{c}_{SP}^{(\ell)} + \tilde{v}_j \mathbf{c}_{SP}^{(j)} \right). \quad (56)$$

The positiveness of the control input in steady state is assured if the following inequalities hold element-wise:

$$\sum_{\ell=0}^c a_{\ell j} \tilde{v}_\ell \mathbf{c}_{SP}^{(\ell)} \leq \tilde{v}_j \mathbf{c}_{SP}^{(j)}, \quad \forall j, \ell = 1 \dots \mathcal{C}. \quad (57)$$

The above inequality expresses the component mass conservation conditions, similarly to those for the overall mass in Eq. (33). Eq. (57) puts a condition on the achievable set point in the  $j$ th CSTR depending on the set point in the CSTRs connected to its incoming flows.

The relation (56) can also be applied to derive relations among the setpoints of the controlled subsystem and the steady state upper bound of the control inputs.

## 5. A case study

### 5.1. Interconnected CRN model for simulations

An interconnected CRN network was considered consisting of three different CRNs and the environment. The CRNs in the three subsystems were chosen as

$$\text{CRN}_1 : \mathcal{R}_{11} : 2C \xrightarrow{K_{31}} 2A \xrightarrow{K_{12}} 2B, \quad \mathcal{R}_{12} : 2C \xrightleftharpoons[K_{23}]{K_{32}} 2B. \quad (58)$$

$$\text{CRN}_2 : \mathcal{R}_2 : 2A \xrightleftharpoons[K_{21}]{K_{12}} 2B. \quad (59)$$

$$\text{CRN}_3 : \mathcal{R}_3 : A + C \xrightleftharpoons[K_{54}]{K_{45}} B. \quad (60)$$

It is easy to see that all three CRNs are reversible or weakly reversible deficiency zero networks with a single graph component (linkage class). Therefore, based on [28], Assumptions 1 and 2 (complex balance and thus persistence of the dynamics) are valid for them independently of their reaction rate coefficients. It is important to remark that complex balance is often fulfilled in practice for thermodynamical reasons, especially in the case of pure chemical reactions [36].

For the simulations, the rate parameters were chosen to be  $\kappa_{ij} = 1 \text{ mol/m}^3/\text{s}$ ,  $\forall i, j$ . The reactions take place in reactors having constant volumes ( $V_j = 1 \text{ m}^3$ ). For each CRN the state vector, incorporating the concentration of species A, B and C, is defined as  $\mathbf{c}^{(j)} = (c_A \ c_B \ c_C)^T$ ,  $\forall j$ .

The interconnections among the CRNs were implemented as it is presented in Fig. 2 with the following parameters  $\tilde{v} = 0.1 \text{ m}^3/\text{s}$ ,  $a_{31} = 0.1$ . The delays in the interconnections were chosen  $T_{ij} = 10 \text{ s}$ ,  $i, j = 1, 2, 3$ .

The flow rates in the interconnections between the environment and the process network are: from the supply to CRN<sub>1</sub> the flow rate is  $(1 - a_{31})\tilde{v}$ ; the control flow rates of the CRNs are  $v_{L1}$ ,  $v_{L2}$ ,  $v_{L3}$ ; the product flow rate from the CRN<sub>3</sub> to the environment is  $\tilde{v}_p = (1 - a_{31})\tilde{v} + v_{L1} + v_{L2} + v_{L3}$ .

The outflows of the open CRNs are:  $\tilde{v}_1 = \tilde{v} + v_{L1}$ ,  $\tilde{v}_2 = \tilde{v}_1 + v_{L2}$ ,  $\tilde{v}_3 = \tilde{v}_2 + v_{L3}$ .

The constant supply concentration was chosen

$$\mathbf{c}^{(0)} = (0.02 \ 0.02 \ 0.02)^T \text{ mol/m}^3.$$

The initial states of the CRNs were chosen as

$$\mathbf{c}^{(j)}(0) = (0.05 \ 0.1 \ 0.15)^T \text{ mol/m}^3, j = 1, 2, 3.$$

The setpoints were chosen as:

$$\mathbf{c}_{SP}^{(1)} = (\sqrt{0.016} \ \sqrt{0.032} \ \sqrt{0.016})^T \text{ mol/m}^3,$$

$$\mathbf{c}_{SP}^{(2)} = (0.18 \ 0.18 \ \sqrt{0.016})^T \text{ mol/m}^3,$$

$$\mathbf{c}_{SP}^{(3)} = (0.875 \ 0.875^2 \ 0.875)^T \text{ mol/m}^3.$$

## 5.2. Simulation experiments

### 5.2.1. No control case

The simulations were performed in Matlab/Simulink environment. First, the behavior of the CRN network was examined without control ( $\mathbf{u}_L^{(j)} = \mathbf{0}$ ,  $j = 1, 2, 3$ ). Fig. 3 shows the trajectories for this case. The behaviors of the interconnected CRNs depend both on the dynamics of the individual chemical reactions and on the flows through the interconnections among the reactors. The trajectories of the uncontrolled subsystems interconnected with the convection network converge to initial value dependent equilibrium states.

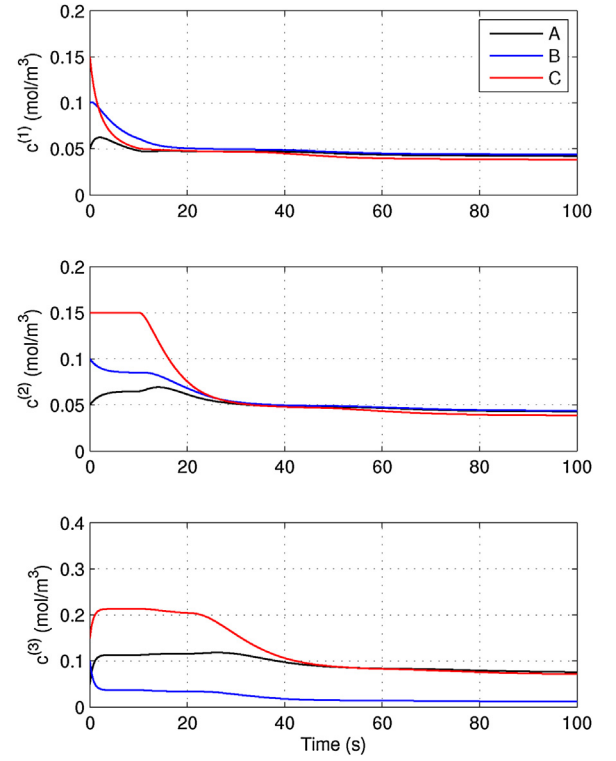


Fig. 3. Simulation results – interconnected CRNs without control.

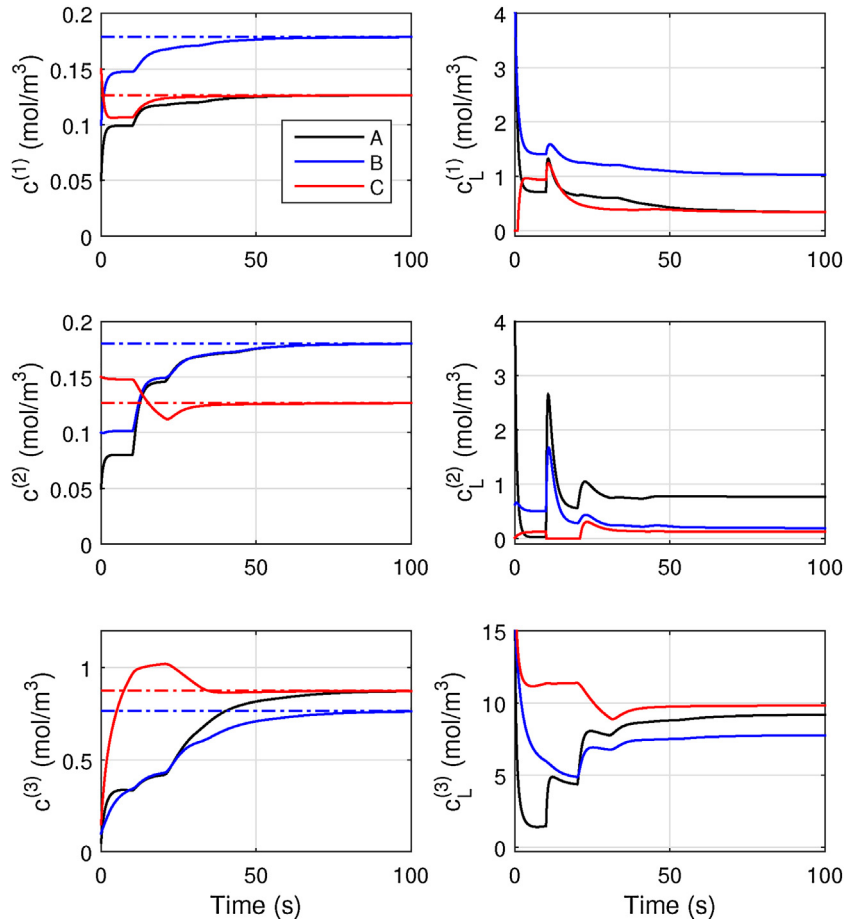


Fig. 4. Simulation results – interconnected CRNs with distributed setpoint control.



### 5.2.2. Distributed setpoint control

Second, the distributed control was computed and implemented based on the relation  $\mathbf{u}_L^{(j)} = \mathbf{u}_y^{(j)} - \mathbf{i}^{(j)}$  with  $v_{Lj} = 0.01 \text{ m}^3/\text{s}$   $j = 1, 2, 3$ , see the control signal defined in (38). During this simulation experiment, no disturbance was considered. The controlled trajectories of the CRNs are presented in Fig. 4. Fig. 4 shows that, with the proposed control, the prescribed setpoints are reached. The output calibration constant in (27) was chosen  $\zeta = 0.8 \text{ mol/m}^3$ .

The parameter  $\zeta$ , introduced in the output Eq. (27), acts as a gain in the controller. Its effect on the control transient performances is presented in Fig. 5. This example presents the trajectory of the controlled concentration  $c_A^{(1)}$  for different values of  $\zeta$ .

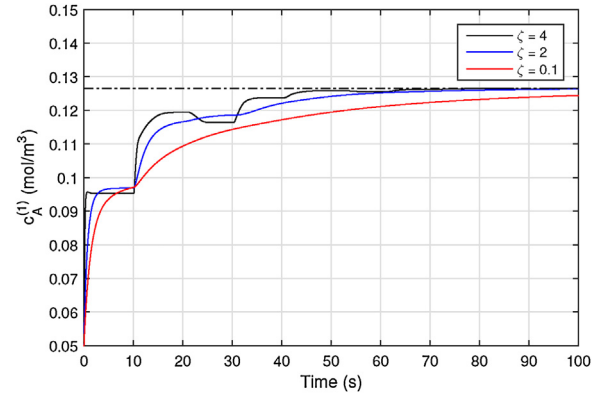


Fig. 5. Simulation results – effect of  $\zeta$  on distributed setpoint control.

### 5.2.3. Comparison with a model predictive controller

For the same interconnected process network a Model Predictive Controller (MPC) was designed using the MPC Controller Simulink block of Matlab MPC Toolbox [37]. This performs the linearization around the setpoint, and the discretization of the linearized model. The sampling period for discretization was chosen 0.1 s. The MPC block solves a constrained optimization problem in each sampling period to obtain the control input, see [38]. All the output weights and input control rate weights of the cost function were chosen 0.1. The lower bound constraints for all the outputs and control inputs were chosen 0.

The simulation results with the MPC is presented in Fig. 6. By comparing the simulations results of the proposed control with the MPC, it can be affirmed that similar settling times and tracking control performances can be obtained. However, for the implementation of the proposed distributed control algorithm only the convection network parameters should be known. For the implementation of the MPC, beside it, the parameters of the CRN subsystems should also be known.

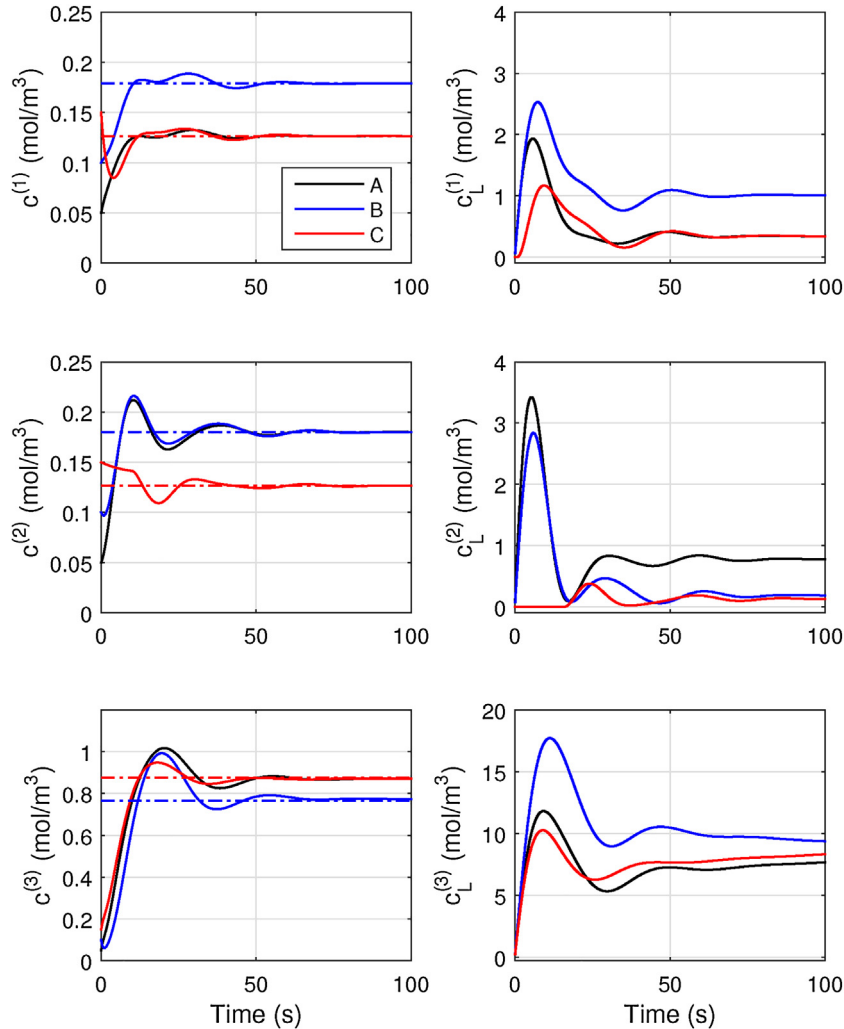


Fig. 6. Simulation results – interconnected CRNs with MPC.

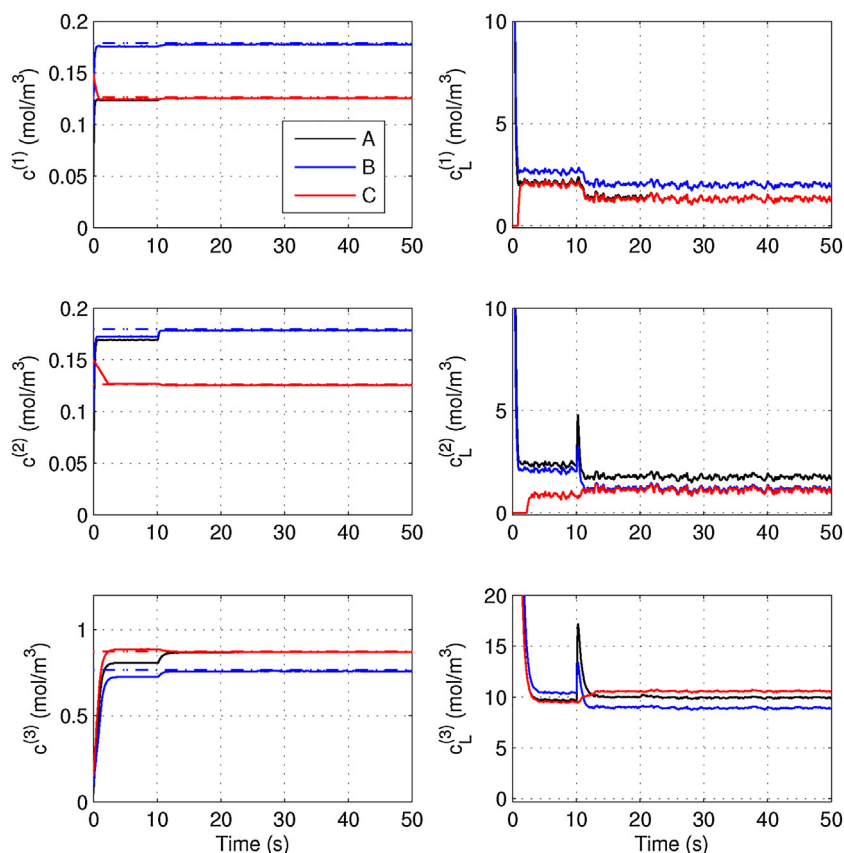


Fig. 7. Simulation results – interconnected CRNs with disturbance attenuated setpoint control.

#### 5.2.4. Distributed setpoint control with disturbance attenuation

Third, it was considered that each CRN has a disturbance input flow in the form  $\mathbf{d}^{(j)} = (d_1^{(j)} d_2^{(j)} d_3^{(j)})^T$ ,  $d_i^{(j)} = 0.01 \cdot (1 + w_i^{(j)}(t))$  mol/m<sup>3</sup>/s where  $w_i^{(j)}(t)$  is a uniformly distributed random signal such that  $0 \leq w_i^{(j)}(t) \leq 1$ ,  $\forall t$ .

The augmented distributed control, introduced in Section 4.3, was computed based on the relation  $\mathbf{u}_L^{(j)} = \mathbf{u}_d^{(j)} - \mathbf{i}^{(j)}$  with controller gain parameter  $\gamma = 4$ . As it can be seen in Fig. 7, the distributed control algorithms augmented with disturbance attenuation terms assure the setpoint tracking. The consequence of the high gain feedback is not just the compensation of the effect of disturbances on the steady state but it also ensures better settling time for the controlled trajectories.

## 6. Conclusions

The similarities between the communication models in multi-agent systems theory and the convective interconnections in the process networks can be explored to develop novel modeling and control approaches for complex process systems based on the theory of multi-agent systems. In this study, the setpoint control problem of interconnected chemical reactions was lead back to the synchronization problem of multi-agent systems. To deal with the nonlinear nature of the chemical reactions, the theory of the passive systems was applied for control design: the passive output of each reactor was defined such that it depends on the difference between the state vector and a given fixed point of the Chemical Reaction Network. The local controller of each CRN was formulated in function of its passive output and it can solve the setpoint regulation based only on information that is available at the corresponding reactor. A Lyapunov-Krasovskii functional based analysis

shows that the setpoint regulation can be achieved in the presence of constant transport delay without any knowledge of the delay values. The resulting control does not depend on the nonlinear terms and on the reaction rate constants in the model of the addressed CRN subsystems; only the parameters of the convection network are necessary for the implementation. An augmented version of the proposed control was also introduced for such cases when the CRNs are subject to input disturbance flows. The performed simulation investigations confirm the efficiency of the proposed control approach.

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