



Generalization of the concept of extents to distributed reaction systems



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HIGHLIGHTS

- The concept of extents is generalized to distributed reaction systems.
- Each extent describes a rate effect subject to advection and outlets.
- Extents are defined for various tubular reactors and reactive separation columns.
- Concentrations and extents are expressed as linear transformations of each other.
- The transformations use stoichiometry and knowledge of transferring/diffusing species.

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ABSTRACT

In the chemical industry, a large class of processes involving reactions can be described by partial differential equations that depend on time and on one or more spatial coordinates. Examples of such distributed reaction systems are tubular reactors and reactive separation columns. As in lumped reaction systems, the interaction between the different rate processes (reactions, mass and heat transfers, and inlet and outlet flows) complicates the analysis and operation of distributed reaction systems. In this article, the concept of extents, which has been applied to decouple the rate processes in lumped reaction systems with one or multiple phases, is generalized to distributed reaction systems. Both the concept of extents and a linear transformation to extents are detailed for various configurations of tubular reactors and reactive separation columns, as well as for a more generic framework that is independent of the configuration and operating conditions. The application of extents to distributed reaction systems is illustrated through several case studies that show how the effect of each rate process can be expressed in terms of a corresponding extent.

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1. Introduction

The chemical industry uses chemical reactions and physical transformations to convert raw materials and energy into products. To operate these industrial chemical processes in a reliable and efficient manner, process models are typically used for design, monitoring, estimation, control and optimization. If each phase can be assumed to be well mixed, a dynamic model describes the time evolution of the system via ordinary differential equations (ODEs). The model consists of material and energy balances and includes information about the reactions (stoichiometry, kinetics, heats of reaction), the transfer of mass and energy within and between phases, and the operating conditions (inlet and outlet flows, initial conditions and exchanges with the environment). Details about

chemical reactor modeling and analysis can be found in many textbooks, for example [Levenspiel \(1999\)](#), [Froment et al. \(2010\)](#), [Rawlings and Ekerdt \(2015\)](#).

The modeling of the rate processes at work is often challenging. The difficulty arises from the coupling between the different chemical and physical effects, as in the case of two-phase reaction systems, where the different reactions are inherently coupled with each other and with the mass-transfer phenomena. For lumped reaction systems, the concept of *vessel extents* is very useful, as the transformation of the original states (concentrations and temperatures) to these extents allows isolating the contributions of the reactions, mass transfers and inlet flows ([Amrhein et al., 2010](#); [Bhatt et al., 2010](#); [Rodrigues et al., 2015](#)). The concept of extents for lumped systems has already been applied to model identification ([Bhatt et al., 2011](#); [Bhatt et al., 2012](#); [Srinivasan et al., 2012](#); [Billeter et al., 2013](#)), data reconciliation ([Srinivasan et al., 2017](#)) and state estimation ([Srinivasan et al., 2016](#)) and is currently being

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investigated for control (Rodrigues et al., 2015) and optimization (Rodrigues et al., 2016; Rodrigues et al., 2017).

In this article, the assumption of perfect mixing within each phase is relaxed. Inhomogeneity can result from either a technical flaw, as in reactors with poor mixing, or a technical choice, as in tubular reactors (Friedly, 1972), micro-reactors (Kockmann et al., 2011), reactive distillation columns (Taylor and Krishna, 2000) or reactive absorption columns (Kenig and Seferlis, 2009). These distributed reaction systems (resolved in time and space) are typically described by partial differential equations (PDEs) that couple the effects of reaction, mass transfer, diffusion, and initial and boundary conditions. The coupling of distributed reaction systems in time and space complicates their analysis. These systems are also complex to analyze due to the existence of boundary conditions, which are not present in lumped reaction systems.

The concept of extents, and of variant and invariant states in general, which aims at decoupling the various rate processes at play and thereby simplifying their analysis, has hardly been applied to distributed reaction systems. Yet, an early work describes the use of variant and invariant states for the simulation of a plug-flow reactor, resulting in reduced computational time (Waller and Mäkilä, 1981). Furthermore, a transformation to reaction-invariant compositions in reactive distillation columns was proposed to reduce the number of degrees of freedom in process design (Ung and Doherty, 1995). This transformation to reaction invariants was later applied to reactive chromatography and membrane reactors (Grüner et al., 2006).

This article considers the modeling of distributed reaction systems from a methodological standpoint. The concept of extents and the transformation of the original states (concentrations and temperature) to these extents are discussed. Emphasis is given to the conceptual understanding of extents. The introduction of the concept of extents for distributed reaction systems is expected to help design and operate this class of processes.

The article is organized as follows. Section 2 presents the extension of the concept of extents from a lumped reaction system described by ODEs to a simple plug-flow reactor described by PDEs. Section 3 describes tubular reactors in terms of extents for different situations (single-phase, multiphase, one-dimensional, two-dimensional). Section 4 introduces the concept of extents for reactive separation columns, such as packed and tray columns. Section 5 presents the concept of extents for a generic multiphase distributed reaction system and summarizes in table form all the formulations introduced in this paper. Section 6 discusses four case studies illustrating the use of extents, while Section 7 concludes the paper.

2. From lumped to distributed reaction systems

2.1. Single-phase lumped reactors

In single-phase lumped reactors, the concentrations and temperature are functions of the time t only, the spatial coordinates being eliminated under the assumption that the phase is isotropic (well mixed).

2.1.1. Material balance equations, $\mathbf{n}(t)$

Let us consider a lumped reactor. The S -dimensional vector of numbers of moles $\mathbf{n}(t)$ is described by the ODE

$$\frac{d}{dt} \mathbf{n}(t) + \omega(t) \mathbf{n}(t) = \mathbf{N}^T \mathbf{r}_v(t) + \mathbf{W}_{in}(t) \mathbf{u}_{in}(t), \quad \mathbf{n}(0) = \mathbf{n}_0, \quad (1)$$

where $\mathbf{r}_v(t) := V(t) \mathbf{r}(t)$, with $\mathbf{r}(t)$ the R -dimensional vector of reaction rates and $V(t)$ the volume, \mathbf{N} is the $R \times S$ stoichiometric matrix, $\mathbf{u}_{in}(t)$ is the p -dimensional vector of inlet mass flowrates, $\mathbf{W}_{in}(t)$ is the $S \times p$ inlet composition matrix, whose j th column is

$\mathbf{W}_{in,j}(t) := \mathbf{M}_w^{-1} \tilde{\mathbf{W}}_{in,j}(t)$, with \mathbf{M}_w the S -dimensional diagonal matrix of molecular weights and $\tilde{\mathbf{W}}_{in,j}(t)$ the S -dimensional vector of weight fractions of the j th inlet, and $\omega(t) := \frac{u_{out}(t)}{m(t)}$ is the inverse of the residence time, with $u_{out}(t)$ the outlet mass flowrate and $m(t)$ the mass. The concentrations $\mathbf{c}(t)$ can be computed as $\frac{\mathbf{n}(t)}{V(t)}$. The initial conditions \mathbf{n}_0 are denoted IC and the inlet flow conditions related to \mathbf{W}_{in} and \mathbf{u}_{in} are denoted IFC.

Remark 1. Throughout the paper, the rates associated with each rate process are modeled as signals that vary in time (and space), which hides the fact that the rates depend on concentrations and temperature that vary in time (and space). For example, in this lumped reactor, the reaction rates are written as $\mathbf{r}(t)$, but in fact they depend on $\mathbf{c}(t)$ and $T(t)$. For the sake of conciseness, the time dependence of the various variables is omitted as much as possible in the remainder.

2.1.2. Effect of the outlet on the IC and IFC

The effect of the outlet flow on the initial and inlet flow conditions can be computed as $\mathbf{n}_{ic}(t)$ by solving the ODE

$$\frac{d}{dt} \mathbf{n}_{ic} + \omega \mathbf{n}_{ic} = \mathbf{W}_{in} \mathbf{u}_{in}, \quad \mathbf{n}_{ic}(0) = \mathbf{n}_0. \quad (2)$$

2.1.3. Vessel extents of reaction $\mathbf{x}_r(t)$

The vessel extent of reaction $x_{r,i}(t)$ represents the amount of material that has been processed by the i th reaction and is in the vessel at time t , that is, discounting for the amount that has left the vessel via the outlet. These vessel extents are described by the ODE

$$\frac{d}{dt} \mathbf{x}_r + \omega \mathbf{x}_r = \mathbf{r}_v, \quad \mathbf{x}_r(0) = \mathbf{0}_R. \quad (3)$$

Eq. (1) can be reconstructed from Eqs. (2) and (3) using

$$\mathbf{n} = \mathcal{L} \mathbf{x}_r + \mathbf{n}_{ic}, \quad (4)$$

with $\mathcal{L} = \mathbf{N}^T$.

2.1.4. Transformation to vessel extents

The vessel extents \mathbf{x}_r can be computed from the numbers of moles \mathbf{n} by inversion of Eq. (4). If $\text{rank}(\mathcal{L}) = R$, then $\mathcal{L}^T \mathbf{W} \mathcal{L}$ is invertible for any positive definite weighting matrix \mathbf{W} of dimension S , and the vessel extents of reaction are given by the linear transformation

$$\mathbf{x}_r = \mathcal{T} \delta \mathbf{n}, \quad (5)$$

with $\mathcal{T} := (\mathcal{L}^T \mathbf{W} \mathcal{L})^{-1} \mathcal{L}^T \mathbf{W}$, and $\delta \mathbf{n} := \mathbf{n} - \mathbf{n}_{ic}$. Note that, for $\mathbf{W} = \mathbf{I}_S$, \mathcal{T} is the Moore-Penrose pseudo-inverse of \mathcal{L} .

Remark 2. The vessel extents $\mathbf{x}_r(t)$ described by ODE (3) and computed from $\mathbf{n}(t)$ via Eq. (5) represent the R reaction variants in the system. As shown in Rodrigues et al. (2015), one can describe and compute from $\mathbf{n}(t)$ additional vessel extents (of inlet flows and initial conditions) as well as invariant quantities.

2.1.5. Combined material and heat balance equations

The combined material and heat balance equations are described in Appendix A.1.

2.2. Single-phase plug-flow reactors

In single-phase plug-flow reactors, the concentrations and temperature are functions of the spatial coordinate z and the time t . It is assumed that the inlet of the reaction system is located at $z = 0$ and z is positive along the reactor length.

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