



Semi-analytical solutions for tubular chemical reactors



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HIGHLIGHTS

- The 1-dimensional tubular reactor model with advection and axial diffusion is studied.
- Semi-analytical solutions are found for any initial/boundary conditions and kinetics.
- Concentrations are expressed as integrals to analyze effects of earlier conditions.
- The effects of initial/boundary conditions are separated from the effect of reactions.
- Former and latter effects are solved analytically and numerically, respectively.

ARTICLE INFO

Article history:

Received 7 February 2017

Received in revised form 2 June 2017

Accepted 5 June 2017

Available online 8 June 2017

Keywords:

Partial differential equations

Tubular reactors

Analytical solutions

System decoupling

ABSTRACT

The one-dimensional tubular reactor model with advection and possibly axial diffusion is the classical model of distributed chemical reaction systems. This system is described by partial differential equations that depend on the time t and the spatial coordinate z . In this article, semi-analytical solutions to these partial differential equations are developed regardless of the complexity of their initial and boundary conditions and reaction kinetics. These semi-analytical solutions can be used to analyze the effect on the concentrations at the current coordinates z and t of (i) the initial and boundary conditions, and (ii) the reactions that took place at an earlier time. A case study illustrates the application of these results to tubular reactors for the two cases, without and with diffusion.

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1. Introduction

Industrial chemical processes typically rely on process models for design, monitoring, control and optimization. Distributed chemical reaction systems correspond to processes involving reactions with phases that are not well mixed, thus resulting in spatial dependencies. Tubular reactors (Friedly, 1972) and reactive separation columns (Taylor and Krishna, 2000) are examples of such systems. The dependent variables of these models are typically concentrations and temperatures. These variables, which depend on time and spatial coordinates, are described by partial differential equations (PDEs) consisting of material and heat balances that couple the effects of advection, reaction, diffusion, conduction, and initial and boundary conditions. The coupling in time and space makes the analysis of distributed reaction systems more complex. In addition, depending on the type of boundary conditions, these systems can be more or less difficult to analyze (Parulekar and Ramkrishna, 1984).

Each combination of dynamic effects results in a different class of physical problems. Two important classes are the *advection-reaction* and the *advection-diffusion-reaction* problems. It has been shown that, for a single-phase one-dimensional system representing an advection-diffusion-reaction problem with a first-order reaction, an analytical solution can be obtained for arbitrary boundary conditions (Logan and Zlotnik, 1995). An extensive list of analytical solutions for the single-phase one-dimensional advection-diffusion-reaction problem with various specific initial and boundary conditions and zeroth- and first-order reaction kinetics is available (van Genuchten and Alves, 1982). These solutions have also been obtained for several particular cases of spatially and temporally dependent velocities and diffusion coefficients (Kumar et al., 2010). Besides the semi-infinite spatial domain considered in this paper, it is also possible to find solutions for the case of finite spatial domain (Guerrero and Skaggs, 2010). However, the influence of and interaction between the different dynamic effects become more difficult to understand when reactions with arbitrarily complex kinetics are considered, or for higher-dimensional problems. Nevertheless, solutions for two- and three-dimensional advection-diffusion-reaction problems

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with a first-order decay reaction are available (Zoppou and Knight, 1999).

This paper focuses on understanding the influence of and interaction between dynamic effects for the case of single-phase one-dimensional tubular reactors subject to advection, axial diffusion and reaction. It uses a novel approach to gain insight into the solutions to PDEs describing concentration profiles. Instead of generating *analytical solutions* for specific initial and boundary conditions and simple reaction kinetics, one obtains *semi-analytical solutions* for arbitrary initial and boundary conditions and arbitrarily complex reaction kinetics. The terminology *semi-analytical* comes from the fact that the effect of the initial and boundary conditions can be determined analytically, whereas complex reaction kinetics generally lead to a numerical solution, because the reaction rates depend on the concentrations themselves. These semi-analytical solutions quantify the effect on the concentrations at the current coordinates of (i) the initial and boundary conditions, and (ii) the reactions that took place at an earlier time, taking into account the transport by advection and diffusion.

These solutions use a transformation of the original PDE system that separates the effects of the initial and boundary conditions from those of the reactions. This transformation has similarities with the transformation to extents for distributed reaction systems that has been suggested recently (Rodrigues et al., 2015a,b, 2017). This paper focuses on the conceptual understanding of the solution to PDE systems that describe one-dimensional tubular reactors, rather than on the discussion of potential applications of semi-analytical solutions. This understanding results from the separation and quantification of the different effects at play in the reactor.

The article is organized as follows. Section 2 reviews the dynamic model (in terms of material balance equations) of single-phase one-dimensional tubular reactors. Section 3 presents semi-analytical solutions for the two cases without and with diffusion. Section 4 discusses a case study where these semi-analytical solutions are used to analyze the dynamics of a tubular reactor, while Section 5 concludes the paper.

2. One-dimensional tubular reactor model

In single-phase one-dimensional tubular reactors, the concentrations and temperature are functions of the spatial coordinate z and the time t . It is assumed that the inlet of the system is located at $z = 0$ and z is positive along the reactor length. Since the concentrations and the temperature vary with the spatial coordinate, the concentration and temperature gradients might lead to significant diffusion when the flow of material is not dominated by advection. Hence, the general formulation of the material and energy balances for a single-phase one-dimensional tubular reactor considers the system from the standpoint of an advection-diffusion-reaction problem.

Let us consider a single-phase one-dimensional tubular reactor containing S species involved in R independent reactions. The S -dimensional vector of concentrations $\mathbf{c}(z, t)$ is described by the PDEs

$$\frac{\partial}{\partial t} \mathbf{c}(z, t) + v_z \frac{\partial}{\partial z} \mathbf{c}(z, t) = \boldsymbol{\phi}_r(z, t) + \mathbf{D} \frac{\partial^2}{\partial z^2} \mathbf{c}(z, t), \quad (1)$$

where v_z is the constant advective velocity, $\boldsymbol{\phi}_r(z, t) := \mathbf{N}^T \mathbf{r}(\mathbf{c}(z, t), T(z, t), \boldsymbol{\theta})$ is the S -dimensional vector of reaction contributions affecting the S species, with \mathbf{N} the $R \times S$ stoichiometric matrix and $\mathbf{r}(\mathbf{c}(z, t), T(z, t), \boldsymbol{\theta})$ the R -dimensional vector of reaction rates depending on the rate parameters $\boldsymbol{\theta}$, \mathbf{D} is the S -dimensional diagonal matrix of constant diffusion coefficients, and the temperature $T(z, t)$ is described by the PDE

$$\frac{\partial}{\partial t} T(z, t) + v_z \frac{\partial}{\partial z} T(z, t) = \psi(z, t) + \alpha \frac{\partial^2}{\partial z^2} T(z, t), \quad (2)$$

where $\psi(z, t) := \frac{-\Delta \mathbf{H}^T \boldsymbol{\phi}_r(z, t) + \phi_{ex}(z, t)}{\rho c_p}$ represents the contribution of reactions and heat exchange to the temperature, with $\Delta \mathbf{H}$ the S -dimensional vector of enthalpies of formation at temperature T_{ref} , $\phi_{ex}(z, t)$ the rate of heat exchange with the environment and ρc_p the constant product of density and specific heat capacity for temperatures between T_{ref} and any $T(z, t)$, and $\alpha := k/(\rho c_p)$ is the thermal diffusivity, with k the constant thermal conductivity.

For the sake of simplicity, only the concentrations will be considered and the temperature will be omitted in the remainder of this article. However, all the results are valid for both isothermal and nonisothermal reactors, since the dependence of the reaction term $\boldsymbol{\phi}_r$ on the temperature does not need to be taken into account in the following developments.

The assumption of constant advective velocity requires the same velocity for all axial and radial positions in the tubular reactor. In physical terms, this implies that (i) the advective flow must not be laminar (since it is well known that under this regime the velocity would be a parabolic function of the radial position), and (ii) the density of the mixture must be constant along the reactor (to ensure the conservation of mass). Moreover, the assumption of constant diffusion coefficients implies that the variation of composition and temperature within the reactor does not affect significantly these diffusion coefficients.

Eq. (1) is subject to the following initial conditions (IC) and boundary conditions (BC):

$$\mathbf{c}(z, 0) = \mathbf{c}_0(z), \quad \forall z > 0 \text{ (IC)}, \quad (3)$$

$$\mathbf{c}(0, t) = \mathbf{c}_{in}(t) + \frac{\mathbf{D}}{\beta v_z} \frac{\partial \mathbf{c}}{\partial z}(0, t), \quad \forall t \geq 0 \text{ (BC)}, \quad (4)$$

$$\lim_{z \rightarrow \infty} \mathbf{D} \frac{\partial \mathbf{c}}{\partial z}(z, t) = \mathbf{0}_S, \quad \forall t \geq 0 \text{ (BC)}. \quad (5)$$

Eqs. (4) and (5) assume that the boundary conditions are located at $z = 0$ and $z \rightarrow \infty$ for a semi-infinite spatial domain. Note that $\beta \rightarrow \infty$ if the advective boundary conditions are of the *first type* (Dirichlet boundary conditions), whereas $\beta = 1$ if the advective boundary conditions are of the *third type* (Robin boundary conditions) (Farlow, 1993). The boundary conditions of the third type may be more appropriate if the flow is discontinuous at the inlet and hinders diffusion at that point, which is the case when the fluid falls into the reactor, for example.

According to Rodrigues et al. (2015b, 2017), the solution to Eq. (1) has the form

$$\mathbf{c}(z, t) = \mathbf{c}_{ibc}(z, t) + \delta \mathbf{c}(z, t). \quad (6)$$

The variable \mathbf{c}_{ibc} denotes the effect of advection and diffusion on the initial and boundary conditions and is defined in its differential form as

$$\frac{\partial}{\partial t} \mathbf{c}_{ibc}(z, t) + v_z \frac{\partial}{\partial z} \mathbf{c}_{ibc}(z, t) = \mathbf{D} \frac{\partial^2}{\partial z^2} \mathbf{c}_{ibc}(z, t), \quad (7)$$

with $\mathbf{c}_{ibc}(z, 0) = \mathbf{c}_0(z), \forall z > 0$ (IC), $\mathbf{c}_{ibc}(0, t) = \mathbf{c}_{in}(t) + \frac{\mathbf{D}}{\beta v_z} \frac{\partial \mathbf{c}_{ibc}}{\partial z}(0, t), \forall t \geq 0$ (BC), and $\lim_{z \rightarrow \infty} \mathbf{D} \frac{\partial \mathbf{c}_{ibc}}{\partial z}(z, t) = \mathbf{0}_S, \forall t \geq 0$ (BC).

On the other hand, the variable $\delta \mathbf{c}$ denotes the effect of advection and diffusion on the reactions, and is defined in its differential form as

$$\frac{\partial}{\partial t} \delta \mathbf{c}(z, t) + v_z \frac{\partial}{\partial z} \delta \mathbf{c}(z, t) = \boldsymbol{\phi}_r(z, t) + \mathbf{D} \frac{\partial^2}{\partial z^2} \delta \mathbf{c}(z, t), \quad (8)$$

with $\delta \mathbf{c}(z, 0) = \mathbf{0}_S, \forall z > 0$ (IC), $\delta \mathbf{c}(0, t) = \frac{\mathbf{D}}{\beta v_z} \frac{\partial \delta \mathbf{c}}{\partial z}(0, t), \forall t \geq 0$ (BC), and $\lim_{z \rightarrow \infty} \mathbf{D} \frac{\partial \delta \mathbf{c}}{\partial z}(z, t) = \mathbf{0}_S, \forall t \geq 0$ (BC).

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