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An equation-oriented approach to modeling heterogeneous catalytic reactors

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HIGHLIGHTS

- A novel strategy is proposed for multiscale modeling of catalytic reactors.
- This strategy was applied for modeling tubular, monolith, fluidized bed reactors.
- Robustness of the algorithm is shown for multiscale modeling of these reactors.

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ABSTRACT

At multiscale modeling of heterogeneous catalytic reactors there appear systems comprising a large number of equations. The solution of such systems is an arduous task and available algorithms require voluminous computations.

A modeling strategy has been suggested for such systems. An effective solution algorithm has been developed for a large class of models based on the application of an identical set of numerical tools such as integro-interpolation method, method of straight lines, a special case of a second-order Rosenbrock method, tridiagonal matrix algorithm or Thomas algorithm on each scale of a multiscale reactor model. Step size control is implemented with account for the rate of change of the variables on each scale. The efficiency and robustness of this algorithm were demonstrated at multiscale modeling of heterogeneous catalytic reactors such as tubular reactors, monolith catalytic reactor, and fluidized bed reactor. When switching between reactor types at multiscale modeling it is not necessary to modify the algorithm. The algorithm can also be used for multiscale modeling of heterogeneous catalytic reactors of other types.

1. Introduction

Mathematical modeling of diverse chemical processes running in various reactors can help to reduce experimental efforts during chemical process development. In chemical reactor modeling chemical reactors are represented as space-time hierarchical structures. The space scale ranges from 10^{-11} to 10^{3} m, the associated time scale ranges from 10^{-15} to 10^{8} s [1,2]. The hierarchical multiscale approach is a backbone of the mathematical modeling of chemical reactors [1,3]. This approach consists in decomposing a complex chemical-technological process into chemical and physical components, studying these components independently and subsequently carrying out the synthesis of a general mathematical model from the models of separate parts of a complex process. Atomic-molecular processes and heat- and mass transfer in porous catalyst pellets or in the catalytic layer represent the lower two scales of the mathematical models of heterogeneous catalytic reactors. The processes of heat- and mass-transfer in the reactor, i.e. in the catalyst bed or monolith channel, represent the third scale, and modeling of a switch device with regard to the processes of mixing, heat exchange, etc – the forth scale. A multiscale mathematical model can describe the preceding levels and given level simultaneously. The history of development and application of the hierarchical approach to the modeling of catalytic processes is described in detail in [1]. Recently there have been many studies on the multiscale modeling of catalytic processes in reactors of various types.

Generally, to treat a developed mathematical model there are two groups of mathematical modeling tools based on two different approaches: block-oriented (BO) and equation-oriented (EO) [4]. The BO approaches address modeling on the flowsheet or reactor

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Nomenclature

a_i, p_i, γ_{ij}, β_{ij} parameters (Eq. (6))
$\overline{A}$, $\overline{B}$, $\overline{C}$, $\overline{f}(u)$ coefficients in equations (Fig. 1)
A_i tridiagonal matrix depend on the solution in a non-linear manner (Eq. (4))
A tridiagonal matrix of dimensionality NxN (Eq. (5))
C_i vector depend on the solution in a non-linear manner (Eq. (4)) or concentration of i-th compound in gas phase, (mol.%, vol.%, wt%)
C vector of dimensionality N (Eq. (5))
D_n matrix (Eqs. (8)–(10))
D diameter, (m)
D_{eq} equivalent catalyst pellet diameter as a plate, (m)
d_{eq} equivalent channel diameter, (mm)
f_u Jacobi matrix
f function in Table 4
I unity matrix
h stepsize
l the axial coordinate of the reactor, (m)
L length of the reactor (m)
M number of variables in Eq. (1)
N number of grid points along the reactor radius
NL number of grid points along the reactor length
NP number of grid points along the pellet radius
N_{text} number (see Subscripts)
P pressure, (atm)
ΔP pressure drop along the reactor, (atm)
R radius of reactor, (m)
S selectivity, (%)
T gas temperature, (°C, K)
t time, (s)
u linear velocity under normal temperature, (m/s)
u_i variable in Eq. (1)
X conversion, (%)
x the radial coordinate of the reactor, (m).
Y yield of product, (%)
y the axial coordinate of the reactor, (m).

Greek letters
α the heat-transfer coefficient from the tube wall to the bed, (J m^{-2} s^{-1} K^{-1})
ε porosity or accuracy of the algorithm in Eq. (11)
η effectiveness factor of catalyst pellet
λ_r radial heat conductivity in the catalyst bed, (J m^{-1} s^{-1} K^{-1})
θ intermediate species, (undimensional)

Subscripts
0 initial value
av average
cat catalyst
cool cooling agent
eqs equations
hole catalyst hole
i variable
in inlet
iter iteration
j point along reactor radius
tube tubular reactor
w tube wall

levels [5,6]. In the first case, every process is abstracted by a flow-sheet consisting of the coupled units, e.g. heat-exchanger, compressor, reactor, absorber, etc. However for the reactor most simulators provide yield based reactor models or a set of idealized models like continuous stirred tank reactor (CSTR), plug flow, etc. with limited capabilities for defining reactor kinetics [6]. In the second case, every reactor model is decomposed into simpler objects such as reaction kinetics, thermodynamics, reactor geometry, reactor configuration, etc. Data are exchanged between the individual objects. BO approaches have many advantages. However reactor configurations are limited to a fixed set of reactors with given models: adiabatic bed reactor, tubular reactor, fluidized bed reactor, and every unit or object is solved one by one in a predefined sequence. This approach is basically used for modeling processes on the same scale. Besides, BO approaches do not support the implementation of more detailed models of standard reactors or models of non-standard reactors: monolith reactor, radial flow reactor, moving bed, multiphase reactors, membrane processes etc.

EO approaches have some advantages over BO approaches, for they support the implementation of more complex reactor models and the simulators of EO process solve the system of equations simultaneously. The last-mentioned advantage indicates that EO approaches are more flexible as compared to BO approaches and,

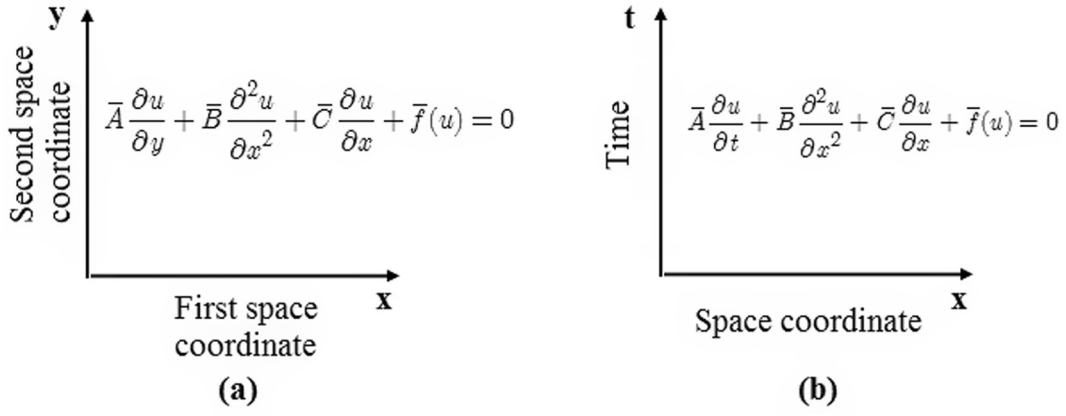

Fig. 1. Schematic representation of the structure of partial differential equations: (a) – with respect to space coordinate "y" contains only first-order derivatives, (b) – the first-order derivative with respect to time.