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Liquid residence time distribution of multiphase horizontal flow in packed bed milli-channel: Spherical beads versus open cell solid foams

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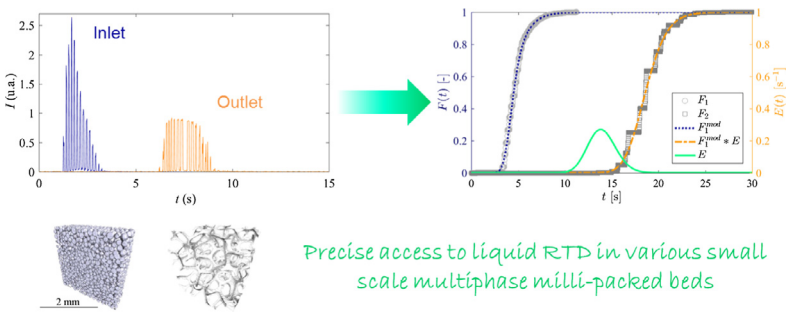
HIGHLIGHTS

- Robust access to liquid RTD in small-scale porous packings under multiphase flow conditions is presented.
- Dense micro-packed beds are compared to open cell solid foams.
- A modified liquid hold up correlation is developed and is valid for both packings.
- RTD broadening is explained by a combination of convective dispersion and mass transfer to a fraction of immobile liquid.

GRAPHICAL ABSTRACT



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ABSTRACT

A robust approach to access liquid residence time distribution (RTD) adapted to multiphase flow in porous media is presented. It is tailored to meet specific requirements of small scale systems (centimeter, millimeter or less) with Taylor segmented flow feed. The method involves direct visualization using fluorescence microscopy close to both extremities of a porous packing. Critical image treatment steps and optimization of a versatile discrete model with 4 parameters are detailed and discussed. They allow the precise and rapid determination of RTD curves and their 1st- and 2nd-order moments. The application of the method is successfully illustrated with dense micro-packed beds of sub-millimeter particles and highly porous media like open cell solid foams undergoing a preformed G-L segmented (Taylor) flow. Original results regarding the effect of fluid flowrates and different confined porous media are discussed and lead to a single two-parameter liquid hold up correlation, which is valid for both packings. As usual, RTD broadening is treated as a combination of convective dispersion and mass transfer to a fraction of immobile liquid. The predominant role of mass transfer is underlined with an analysis of characteristic times.

1. Introduction

Micro- or milli-packed beds (MPBs) of small catalyst particles (10–200 μm) became established and efficient lab-tools in the case of heterogeneously catalyzed gas–liquid (G-L) reactions (Losey et al., 2001; van Herk et al., 2005; Al-Rifai et al., 2016; Moulijin et al., 2016, Faridkhou et al., 2016). Their attractive mass and heat transfer performances and their low material inventory are the driving reasons for this development. Overall, they help speeding up the access to intrinsic chemical activity (van Herk et al., 2009) and optimizing operating conditions in potentially unconventional and risky domains where macroscale reactors are difficult to operate (Inoue et al., 2007). Additionally, these tools can be of great

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Nomenclature

$CV_{\tilde{t}_{exp}}$	variation coefficient of the 1st-order moment of the RTD [%]
$CV_{\sigma^2_{exp}}$	variation coefficient of the 2nd-order moment of the RTD [%]
d	spherical particle diameter [m]
d_{pore}	mean cell (OCSFs) or pore (MPB) diameter [m]
$E(t)$	residence time distribution function [s^{-1}]
$\bar{E}(s)$	Laplace transform of the RTD [-]
$E_R(\theta)$	RTD in reduced time coordinate [–]
$\overline{E_R}(s)$	Laplace transform of the RTD in reduced time coordinate [–]
$F_{I,\,i}\,(t)$	cumulative raw tracer intensity [a.u.]
$F_i\,(t)$	cumulative tracer intensity after baseline correction [a.u.]
$F_i^{mod}(t)$	modelled cumulative tracer intensity [a.u.]
$\overline{F_i^{mod}}(s)$	modelled cumulative tracer intensity in Laplace domain [–]
h	channel height [m]
$I_i(t)$	raw tracer intensity [a.u.]
J	number of mixing cells in series [–]
K_{im}	ratio of immobile to mobile liquid fractions [–]
of	objective function [–]
Q_i	volumetric flowrate of fluid i [m^3/s]
r	radius of the gas injection channel [m]
s	Laplace variable [s^{-1}]
S_L	liquid saturation in the porous media [–]
$\tilde{t}$	first-order moment of the RTD [s]
$\tilde{t}_i$	first-order moment of curve i [s]
t_m	residence time of the mobile liquid phase [s]
t_M	mass transfer time between mobile and stagnant zones [s]
t_D	characteristic time of hydrodynamic dispersion in the mobile zone [s]
t_T	characteristic time of mass transfer between static and dynamic zones [s]
u_i	superficial velocity of the fluid i [m/s]
u_{DF}	drift flux velocity [m/s]
V	volume of liquid [m^3]
$\vec{v}_i$	Eigenvectors of the variance-covariance matrix (i = 1–4) [–]
V_R	volume of the reactor [m^3]

Greek symbols:

α, β	parameters of the drift flux model by Molga and Westerterp (1997) [–]
α', β'	parameters of the adapted drift flux model [–]
γ	surface tension [N/m]
δ	distance from channel wall [m]
δ_F	residuals between the model and experimental outlet F_2 curves [–]
ε	volume averaged foam or bed porosity [–]
$\varepsilon\delta$	mean porosity at a distance δ from the channel walls [–]
ε_{bulk}	mean porosity far from the walls [–]
ε^*	reduced porosity, $\varepsilon\delta/\varepsilon_{bulk}$ [–]
ε_L	liquid hold up of the reactor [–]
ε_L^{mod}	liquid hold up estimated from the drift flux model [–]
θ	reduced time coordinate, $t/\tilde{t}$ [–]
θ_m	fraction of mobile liquid phase [–]
θ_{im}	fraction of immobile or stagnant liquid phase [–]
μ	viscosity [–]
ρ	density [kg/m^3]
σ^2	variance of the RTD [s^2]
σ_i^2	variance of the curve i [s^2]
σ'^2	reduced variance of the RTD [s^2]

Subscripts:

0	initial values
1	inlet
2	outlet
DF	drift flux
exp	experimental
G	gas
L	liquid
min	minimum
max	maximum
mod	model
R	reactor, reduced.
S	solid

Abbreviations:

$CSTR_4$	series of continuously stirred tank reactor model with 4 parameters
FFT	fast Fourier transform
LHS	Latin hypercube sampling
MPB	micro packed bed
OCSF	open cell solid foam
PPI	pores per inch
RTD	residence time distribution

interest to mimic and understand the complex two-phase flow patterns occurring at intergranular pore scale in larger scale reactors.

For production purpose, this concept faces scalability and operability issues inherent to multiple channel geometries. Important pressure drop, clogging, uniform two-phase distribution, reproducible loading, catalytic bed replacement and heat exchange integration are among them. Nonetheless, several examples are reported in the literature with reliable small production throughput (Plucinski et al. 2005; Lerou et al., 2010; Deshmukh et al. 2010; Inoue et al. 2015).

Recently, milli-channels containing alternative porous structures like pillar arrays (de Loos et al., 2010; Yang et al., 2015), open-cell solid foams (Saber et al. 2012; Liu et al. 2013; Tourvieille et al. 2015, 2015a) or ordered porous media (Häfeli et al., 2013; Elias and von Rohr, 2016; Potdar et al. 2017) have been reported for multiphase applications. All these internals still exhibit very good mass transfer and reaction performances but with drastically reduced pressure drop which can be attractive for future lab-tools or millireactor concepts.

At millimeter scale, in a wide range of feeding conditions and in empty channels, gas-liquid flows are often self-organized as segmented flows (also well-known as Taylor flows) (Kreutzer et al., 2005a, 2005b). A global understanding of the progressive disorganization of this flow entering porous beds (spheres, foams, etc.) is of fundamental importance because it governs the phase ratio, local mixing, the gas-liquid contact surface area and eventually the mass transfer performance. Residence Time Distribution (RTD) measurements provide precious and meaningful information (mean residence time and phase hold up of each fluid phase, dispersive behavior, presence of dead-zones or by-pass, etc.) about these issues. It helps handling and modeling the spreading of the reactants and products along the reactor which can lead to selectivity and conversion issues.