International Journal of Multiphase Flow 86 (2016) 56–66

Contents lists available at ScienceDirect

International Journal of Multiphase Flow

journal homepage: www.elsevier.com/locate/ijmultiphaseflow

A study of two-phase flow in monoliths using ultrafast single-slice X-ray computed tomography

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ARTICLE INFO

Article history:
Available online 29 July 2016

Keywords:
Ultrafast X-ray tomography
Two-phase flow
Monolith
Holdup
Maldistribution
Flow pattern
Flow visualization

ABSTRACT

Running chemical reactions in monolithic structures is being considered as highly promising for intensifying industrial reaction processes. A potential pitfall of such structures is the difficulty to achieve homogeneous and well defined gas/liquid distributions patterns with economically feasible distribution mechanisms. Experimental studies on gas/liquid distribution in monoliths are often hampered by missing measurement and visualization techniques to disclose the two-phase flow inside the narrow and opaque channels.

This paper presents results of a study carried out with ultrafast single-slice X-ray tomography, a novel imaging technique, which can overcome these limitations. We investigated two-phase flow in two different types of square-channel monolith structures, one with high cell density of 400 cpsi and one with low cell density of 39 cpsi. Our study discloses in-channel flooding and draining behavior via extraction of characteristic distribution parameters, such as averaged and channel-linked liquid holdup, two-phase flow patterns and liquid maldistribution from X-ray images using advanced image processing techniques.

1. Introduction

Multiphase reactors comprising internal monolithic structures, that are, structures with multiple parallel straight channels coated with a catalyst, are a promising concept in the field of process intensification of heterogeneous multiphase reactions (Boger et al., 2004a, Cybulski et al., 2010, Edvinsson et al., 1994, Nijhuis et al., 2001). The potential for using monolithic structures in multiphase reactors has not yet been fully explored. Monolithic structures have been used extensively for hydrogenations, oxidations and enzymatic reactions (Albers et al., 2001, Deugd et al., 2003, Irandoust et al., 1990, Klinghoffer et al., 1998, Quan et al., 2003). Recent studies have shown that they could be a suitable replacement for conventional packed beds in gas-liquid-solid configurations (e.g. for hydrogenation reactions) (Bauer et al., 2011, Boger et al., 2003, Crynes et al., 1995, Edvinsson et al.,1995, Kapteijn et al., 2001), specifically because they have minimal axial dispersion and backmixing in the slug flow regime. Monolithic structured bed reactors have also been applied for hydrodesulfurization of heavy gas oil fractions (Kallinikos et al., 2007) and for the abatement of NOx and CO emissions from automobile engines (Cybulski et al., 1999). The application of monolithic catalysts as an alternative to slurry systems in the hydrogenation of edible oils has been investigated (Boger et al., 2004b). Structured fixed-bed reactors are an interesting alternative to intensify multiphase reactions in processes without rapid catalyst deactivation (Haase et al., 2013). The applicability of structured packed columns for application in reactive distillation and hydro-treating has also been investigated (Ellenberger et al., 1999). It was found that for fast gas-liquid-solid catalyzed reactions, the monolith reactor can also be an attractive alternative to mechanically agitated slurry reactors, which are widely used in the fine chemicals production (Edvinsson et al.,1996). Another study presented an in-line monolithic reactor as a novel reactor concept of a compact gas-liquid catalytic converter (Stankiewicz, 2001).

Monolith reactors have several advantages over conventional multiphase reactors (e.g. packed beds or trickle beds), such as lower pressure drop, shorter diffusions paths and minimal axial dispersion as well as higher mass transfer rates. This enables to run reactions at higher flow rates which means higher capacities and enhanced space-time-yield. Monoliths can be characterized by geometric parameters including the hydraulic diameter of the channels d_h, cell density (number of cells per unit area), open frontal area and geometric surface area (Roy et al., 2004). The hydrodynamics is characterized by pressure drop, superficial

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http://dx.doi.org/10.1016/j.ijmultiphaseflow.2016.07.008
0301-9322/

Symbols

Latin symbols

C	Coefficient (-)
ch	Channel (-)
d	Diameter (m)
E	Extinction (-)
f	Flooding holdup (-)
G	Superficial gas flow rate (kg s^{-1} m^{-2})
h	Two-phase holdup (-)
H	Liquid holdup (-)
$\bar{H}$	Time averaged holdup (-)
i	First dimension of voxel grid (-)
I	Intensity (W m^{-2})
j	Second dimension of voxel grid (-)
k	Samples (-)
l	Length (m)
L	Superficial liquid flow rate (kg s^{-1} m^{-2})
m	Number of voxels in first dimension (-)
M	Mapping matrix (-)
n	Number of voxels in second dimension (-)
N	Number (-)
P	Probability distribution (-)
Q	Quantity (-)
u	Superficial velocity (m s^{-1})
X, x	Duration (s)

Greek symbols

μ	Linear attenuation coefficient (m^{-1})
ρ	Density (kg m^{-3})

Subscripts

0	Initial
ch	Channel
d	Distance
$frames$	Frames
G	Gas
h	Hydraulic
i	Index
L	Liquid
liq	Liquid
$long$	Longitudinal
m	Middle
mal	Maldistribution
ray	Ray
S	Structure
$total$	Cross-sectional
TPF	Two-phase flow

liquid and gas velocities, mean two-phase flow velocity, gas-to-liquid ratio (void fraction, dynamic gas holdup, liquid saturation), mean bubble length and mean bubble velocities. The flow regime is controlled by the gas and liquid superficial velocities, but is also dependent on the hydraulic diameters of the monolith channels and the liquid and gas properties. Bubbly flow can be observed in monoliths at low gas and high liquid superficial velocities whereas slug flow, churn flow and annular flow occur at high gas and low liquid superficial velocities (Roy et al., 2004). A favorable flow regime for gas-liquid reactions seems to be the slug flow regime because it is characterized by a good mixing within the slugs due to recirculation and short diffusive paths through the liquid film at the catalyst wall (Abiev et al., 2011). Furthermore, slug flow regime enables to enhance the mass transfer rates in reactive multiphase flows by controlling the interfacial area via the flow pattern (Leclerc et al., 2010). Four different characteristic mass transfer phenomena appearing in monoliths have to be considered: (1) gas-liquid mass transfer from the gas slugs through the liquid film to the monolith walls, (2) liquid-solid mass transfer (liquid reactant to catalyst), (3) gas-solid mass transfer (gaseous reactant to catalyst), and (4) diffusion of reactants inside the catalyst pores.

Monolith reactors can be easily scaled-up (in terms of a numbering up), which is an important benefit. However, for the application of this reactor concept to industrial processes, a uniform phase distribution is an essential performance criterion (Kreutzer et al., 2005). The performance of monolith reactors can be affected by several conditions. These are for example the initial distribution of the liquid and gas phase, irregular slug length, compression of gas slugs and aeration of liquid slugs (Mewes et al., 1999, Reinecke et al., 1996, Sederman et al., 2003). Detailed knowledge about these influencing conditions is required to enhance the performance of monolith reactors. In order to address this lack of understanding we present a series of experimental investigations of the two-phase flow in two typical monoliths with established cell densities, and using different gas-liquid distributors. Using the latest ultrafast single-slice X-ray tomography (XCT) we investigated averaged and channel-linked liquid holdup, two-phase flow patterns, liquid maldistribution as well as in-channel flooding and draining behavior.

Table 1
Operational range and specifications of the experimental facility.

Operational range:	
Gas superficial velocities	0.1 to 3.0 m/s
Liquid superficial velocities	0.018 to 0.07 m/s
Conditions:	
Operating pressure	1 bar (open system)
Operating temperature	20°C (room temperature)
Gas phase	air
Liquid phase	aqueous tenside solution

2. Experimental setup

The gas-liquid two-phase flow inside the monolithic structures was performed using a bespoke experimental rig designed for compatibility with ultrafast single-slice XCT (Fig. 1a). The facility is operated in co-current down-flow mode, with adjustable phase fraction ratios of liquid and gas. Liquid flow rates are controlled by a frequency-controlled gear pump VGS060 (Verder) whereas the gas flow rates are adjusted by several rotameters (Krohne). The monolith test section consists of a vertical DN 40 tube in which the monolith is placed (Fig. 1a,b).

Two different monoliths made of cordierite ceramics with different cell densities of square channels were investigated in this work (Fig. 1c): one with 39 cells per square inch (cpsi) and 32 channels ($d_h = 3.60$ mm), and one with 400 cpsi and 357 channels ($d_h = 1.09$ mm). A single-stage phase separator is placed downstream the monolith test section holding the monolith in place and controlling the discharge of the two-phase flow. The operating conditions and the operational range are listed in Table 1. Both fluids are fed into the system via an adaptable distribution module with exchangeable gas/liquid distributors. Either a spray nozzle distributor or a novel needle distributor is used to spread the fluids homogeneously over the cross section of the reactor. The spray nozzle distributor consists of a single phase full cone spray nozzle of 45° spray angle (MC GmbH) for the liquid phase and is placed at that distance l_d that the spray cone ends on the reactor as seen in Fig. 2a. The gas is injected above the nozzle from two radial inlets. The second distributor, which has been specifically designed for application with organic liquids, is a proprietary needle distributor (Fig. 2b) which has been developed and tested at TU Dresden