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Experimental investigation of gas-liquid distribution in periodic open cellular structures as potential catalyst supports

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ABSTRACT

Radial and axial liquid distribution was measured in a packed column of 500 mm length and 100 mm inner diameter under various gas- and liquid superficial velocities to achieve trickle flow regime. Liquid maldistribution was determined using a liquid collector at the outlet consisting of 21 collection zones of equal area. In this work, as packings inside the column, periodic open cellular structures (POCS) were manufactured and applied. Such POCS can be used as structured catalyst support and/or liquid distributor.

The hydrodynamic behavior of packings with Kelvin cell, Diamond cell and a hybrid combination of both unit cells was studied to identify unit cells with high potential for achieving homogeneous liquid distribution over the entire reactor cross-section. The liquid superficial velocity had a strong influence on the liquid distribution, while an increasing gas flow rate did not change the flow patterns. Kelvin cell packings tend to distribute the liquid towards the center whereas Diamond cell packings predominantly spread the fluid towards the rim. As a consequence of these observations, in this work a novel POCS structure is proposed: The combination of Kelvin and Diamond unit cells, called hybrid DiaKel unit cell. With POCS consisting of this unit cell type, a significantly improved liquid distribution can be achieved. Thus, this structure features high potential as liquid distributor and/or catalyst support in gas-liquid reaction systems.

1. Introduction

Multiphase reactions (e.g. gas-liquid-solid) are often carried out in trickle bed reactors. In this reactor setup, gas and liquid phases are fed co- or countercurrently into the reactor containing the fixed catalyst bed [1]. Depending on the gas- and liquid flow rate, the flow regime of the multiphase flow can be adjusted to realize either a low or a high interaction regime. The low interaction regime is also known as trickle flow, whereas the high interaction regime comprises pulse, spray, and bubble flow [2–4]. The prevailing flow regime determines decisively important characteristics of the multiphase reactor such as, e.g., the two-phase pressure drop, the liquid holdup, and heat and mass transfer [5–7].

One of the main challenges in multiphase reactor design, scale-up and operation is the realization of sufficient contacting of the phases. Heterogeneously catalyzed gas-liquid reactions such as, e.g., hydrodesulfurization (HDS) of gas oil, are commonly carried out in trickle bed reactors. The performance of this reactor system strongly depends on a preferably homogeneous gas-liquid distribution over the entire cross-section of the catalyst bed [8,9]. Liquid maldistribution due to ineffective initial distribution at the reactor inlet and/or anisotropy of the packing inside the reactor cause "dry" areas or result in the formation of rivulets, which leads to poor catalyst utilization. To ensure an even liquid distribution initial distribution as well as homogeneous flow inside the catalyst packing both play an important role [10,11].

In this regard, the use of structured reactors of well-defined geometry offers high potential. In recent years, several studies were performed concerning the use of monolithic honeycomb structures or reticulated open-cell foams which consist of an irregular arrangement of pores that represent an interconnected three-dimensional pore space. To mention exemplarily a few of these

Abbreviations: ABS, Acrylonitrile butadiene styrene; cpi, Cells per inch; FDM, Fused deposition modelling; HDS, Hydrodesulfurization; MP, Multipoint; op, Operating point; PE/PP/$BaSO_4$, Polyethylene/Polypropylene/Bariumsulfate; POCS, Periodic open cellular structures; SEBM, Selective electron beam melting; SLA, Stereolithography; SLM, Selective laser melting; SP, Singlepoint.

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Nomenclature

List of symbols

A	Length of one unit cell (mm)
d_s	Strut diameter (mm)
H	Packing length (mm)
1_s	Strut length (mm)
M	Mass (kg)
M_f	Maldistribution factor
N	Number of equal areas
Q_1	Liquid flow rate ($l\,min^{-1}$)
S_v	Specific surface area ($m^2\,m^{-3}$)
U	Superficial velocity (ms^{-1})
x	Liquid holdup

Subscripts

c	Container
G	Gas
HS	Hollow struts
i	Compartment of the liquid collector
L	Liquid
L,D	Dynamic liquid holdup
L,S	Static liquid holdup
Mean	Mean
mp	Multipoint distributor at operating point i
op	At operating point i
P	Packing
uw	Unwetted structure
w	Wetted structure

Superscripts

30	Drainage of the packings after 30 min
EtOH	Ethanol
in air	Weighing of the packings in air
in EtOH	Weighing of the packings in ethanol

Greek symbols

β_l	Total liquid saturation
ε_H	Hydrodynamic porosity
μ	Surface tension ($mN\,m^{-1}$)
ρ	Density ($kg\,m^{-3}$)

studies, Mohammed et al. [12] investigated the gas-liquid distribution in reactors with open-cell foam packings, Stemmet et al. [13] focused on the hydrodynamics of counter-current flow in solid foam packings, and Roy et al. [14] characterized the flow distribution in monolithic reactors. One important finding is that foam packings and monolithic structures can be used as catalyst support and/or liquid distributor as long as a good initial distribution is provided and suitable operation points are adjusted.

Periodic open cellular structures (POCS) can be classified into a category in between those investigated reticulated open-cell foams and monolithic structures [15]. POCS consist of a regular arrangement of periodic unit cells and thus feature a well-defined geometry compared to the random geometry of reticulated foams. Moreover, due to the interconnected unit cells POCS, in contrast to monolithic honeycomb structures, allow for heat and mass transfer not only in axial but also in radial direction [16]. By applying additive manufacturing techniques it is nowadays possible to produce periodic open cellular structures for a variety of materials from polymeric materials (by, e.g., fused deposition modelling (FDM) or stereolithography (SLA)) to metals (by, e.g., selective electron beam melting (SEBM) or selective laser melting (SLM)). Such kinds of packings represent multifunctional devices and can be used as catalyst support as well as liquid (re) distributor. Furthermore, owing to their regular cell geometry they allow for a systematic investigation which can contribute to an improved understanding of the complex hydrodynamic behavior of gas-liquid flow [16,17]. Finally, POCS offer a unique combination of adjustable properties such as low pressure drop due to high porosity, high thermal conductivity and high specific surface area [18,19].

2. Materials and methods

Experiments were performed in a laboratory scale test rig (see Fig. 1(a)). The column had a length of 750 mm and an inner diameter of 100 mm. The setup consisted of a gas-liquid distributor at the top, a cellular packing and a liquid fraction collector at the bottom of the column. 100 mm segments of POCS were stacked to form a total packing length of 500 mm. The radial and axial spreading of the liquid was determined using a liquid collector with 21 collection zones of equal collection area, and based on this a liquid maldistribution factor was calculated. The assembly of equal collections zones of the liquid collector can be seen from Fig. 2. The collector was attached at the bottom of the acrylic column and the cell packings were stacked directly above the collector to avoid radial liquid flow and/or coalescence of small liquid streams in between the packing and collector. The volumetric flow rate of each zone was simultaneously determined with 21 Digmesa FHKSC 1.00 mm flow sensors within the operating range of 0.0560–0.4044 $l\,min^{-1}$. After reaching steady state flow conditions for each operation point, the measurement time was 60–120 s. Steady state conditions were reached when the course of online flow rate measurements had a slope of zero. The liquid flow rate was controlled by a Digmesa FHK flow meter (0...3.5 $l\,min^{-1}$) and the gas flow rate (air) was adjusted by a MASS-STREAM™ mass flow controller D-6361-FGD-44-AV-99-D-S-DR (4...200 Nl min^{-1}).

The liquid stream was circulated with an EDEN 140 centrifugal pump. To study the influence of viscosity three different binary mixtures of water and glycerol were used in the experiments (10, 30, and 50 wt.-% glycerol in water). Measurements of the viscosity were carried out with a Fungilab rotational viscometer at 25 °C. The viscosity significantly increased with the amount of glycerol: 10 wt.-% (1.41 mPa s), 30 wt.-% (2.45 mPa s) and 50 wt.-% (4.95 mPa s).

For the initial liquid distribution two different perforated trays (singlepoint (SP) and multipoint (MP)) with gas risers were applied (according to Ref. [20]). Fig. 1(b) and (c) show technical drawings with the dimensions of multipoint and singlepoint liquid distributors. Before starting with the experiments all drip points of the perforated tray were checked for homogeneous initial distribution. Drip points with lower flow rates were carefully drilled out until all drip points featured the same liquid flow rate. Reactor internals and POCS for these cold-flow experiments were fabricated with a MakerBot Replicator 2X 3D-printer and consisted of acrylonitrile butadiene styrene (ABS). The principle of this additive manufacturing technique (FDM) is the deposition of molded polymer on a heated plate to build up a three dimensional structure [21]. CAD files were created with Solid Edge ST6 and replicated to a cylindrical packing with 100 mm in diameter and 100 mm height using the software Magics 17.02. Kelvin cell and diamond cell packings of different cell sizes with or without directly attached printed wall (see Fig. 3(c)) were manufactured, and their gas-liquid distribution performance was compared to a random sphere packing as state-of-the-art reference system.

For the determination of the dynamic and static liquid holdup of the periodic open cellular structures and the sphere packing, the drainage method was applied [4,22]. The liquid storage tank (see Fig. 1) was placed on a balance (Sartorius AZ 6101) and the weight