

**Identification of the process windows of inclined rotating fixed-bed reactors with concentric tube – a hydrodynamic analysis**

Timaeus, R.; Schleicher, E.; Bieberle, A.; Hampel, U.; Schubert, M.;

Originally published:

July 2020

**Chemical Engineering Science 220(2020), 115637**

DOI: <https://doi.org/10.1016/j.ces.2020.115637>

Perma-Link to Publication Repository of HZDR:

<https://www.hzdr.de/publications/Publ-30121>

Release of the secondary publication  
on the basis of the German Copyright Law § 38 Section 4.

CC BY-NC-ND

1     **Identification of the process windows of inclined rotating fixed-bed reactors**  
2                     **with concentric tube – a hydrodynamic analysis**

3     Robert Timaeus<sup>a,\*</sup>, Eckhard Schleicher<sup>a</sup>, André Bieberle<sup>a</sup>, Uwe Hampel<sup>a,b</sup>, Markus  
4                     Schubert<sup>a,\*</sup>

5  
6             <sup>a</sup> Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstraße 400,  
7                     01328 Dresden, Germany.

8             <sup>b</sup> TU Dresden, Institute of Power Engineering, 01069 Dresden, Germany.

9  
10            \* Corresponding authors: r.timaeus@hzdr.de, m.schubert@hzdr.de

11  
12     **Abstract:** The inclined rotating fixed-bed reactor with inner tube is a promising  
13     process intensification concept for gas-limited reactions. In order to take full  
14     advantage of the reactor concept the installation of an inner concentric displacement  
15     tube is proposed to support the wetting intermittency of the whole fixed-bed at  
16     different liquid filling levels. The effects of operating conditions and design  
17     parameters on flow stratification, liquid filling level and specific pressure drop are  
18     analysed to identify the process window. In particular, the influence of superficial  
19     phase velocities, inclination angle, rotational velocity, particle diameter and inner tube  
20     diameter are studied. The liquid phase distribution is characterized with a  
21     capacitance-based wire-mesh sensor, which is adapted to cope with organic liquid,  
22     porous alumina catalyst packings and reactor rotation. Furthermore, the radial  
23     porosity distribution in the annular fixed-beds is determined using gamma-ray  
24     computed tomography.

25  
26     **Keywords:** Inclined rotating fixed-bed, Process intensification, Capacitance wire-  
27     mesh sensor, Multiphase reactor

28  
29     **1 Introduction**

30     The performance of multiphase reactors is often limited by mass transfer processes  
31     (Utikar and Ranade, 2017). Especially, trickle-bed reactors (TBR) are frequently used

32 for reactions with fast kinetics, which are controlled by the gas transfer (Mills and  
33 Duducović, 1980). Here, the liquid film on the solid catalysts acts as mass transfer  
34 barrier (Boelhouwer et al., 2002a). Several reactor concepts and operation strategies  
35 have been proposed to intensify the mass transfer (Nigam and Larachi, 2005).

36 Structured reactors with novel packings were suggested to intensify the gas-liquid-  
37 solid contact. For example, Taylor flow in monolith reactors was found to feature low  
38 pressure drop, high specific surface area and improved catalyst wetting (Haase et al.,  
39 2016). For example, Nijhuis et al. (2001) obtained higher productivities for  $\alpha$ -  
40 methylstyrene and benzaldehyde hydrogenations in a pilot-scale monolith reactor  
41 compared to a conventional trickle-bed reactor. Open-cell solid foams are another  
42 promising replacement for conventional fixed-bed reactors with randomly packed  
43 beds. Here, the solid material network provides high void space and specific surface  
44 area, while the pressure drop is low (Edouard et al., 2008; Zalucky et al., 2017a). The  
45 skeletal structure of the solid foams leads also to a higher thermal conductivity  
46 enhancing the radial heat transfer (Wallenstein et al., 2014), which is especially  
47 beneficial for exothermic reactions. Applying the limiting current technique, Zalucky et  
48 al. (2017b) confirmed also higher liquid-solid mass transfer coefficients in solid  
49 foams.

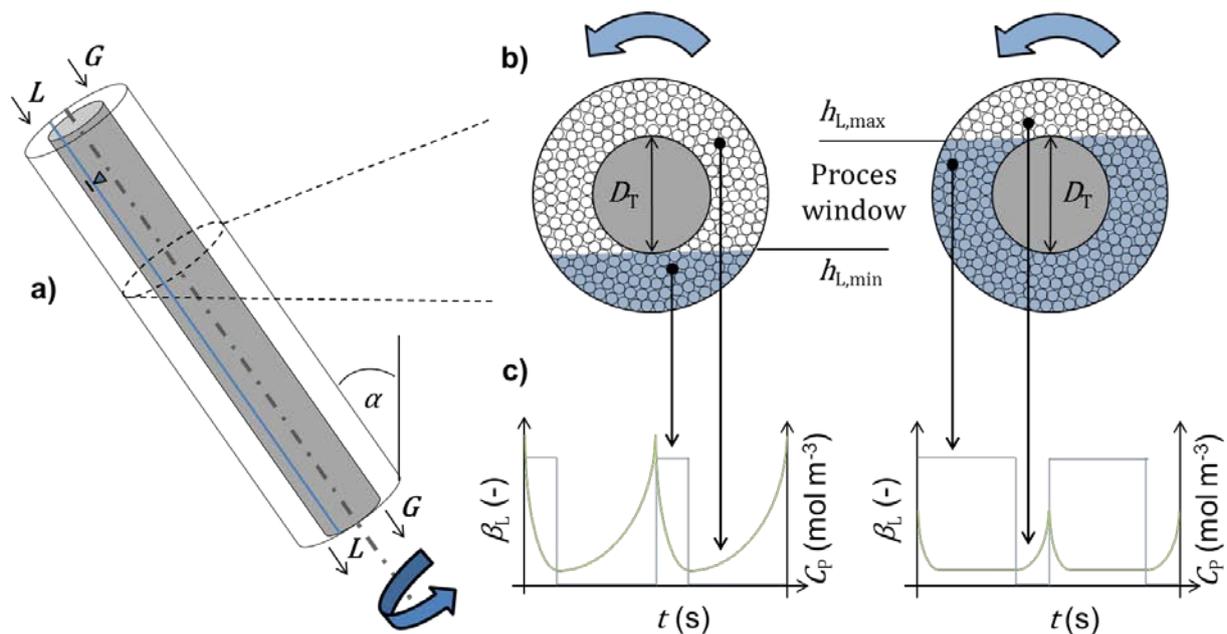
50 Recently, Gelhausen et al. (Gelhausen et al., 2017) proposed a siphon reactor to  
51 increase gas-solid and gas-liquid mass transfer. Here, the catalyst is alternatingly  
52 exposed to gas and liquid based on a customized self-sustaining liquid filling-draining  
53 cycle in packing segments. Beyond the adjustable gas-solid or liquid-solid contact  
54 time, stagnant liquid is effectively discharged at each cycle. Compared to a  
55 conventional TBR, a twofold increase of the space-time yield (STY) was obtained for  
56 the hydrogenation of 2-butyne-1,4-diol.

57 Periodic operation of TBRs was proposed as a process intensification method (Lange  
58 et al., 1994; Castellari and Haure, 1995). Here, the liquid flow rate is cycled at the  
59 reactor inlet to induce a temporal modulation of the liquid holdup along the fixed-bed  
60 in order to decrease the liquid film thickness surrounding the catalyst. This way, the  
61 access of gaseous educts to the catalysts at low local liquid holdups is facilitated.  
62 Many lab-scale studies confirmed significantly increasing STY via flow modulation  
63 (Banchero et al., 2004; Khadlikar et al., 1999). However, such induced liquid pulses

64 rapidly decay along the fixed-bed (Boelhouwer et al., 2002b), which eliminates  
 65 beneficial conditions at lower reactor positions (Dietrich et al., 2005).

66 Thus, Härting et al. (2015) introduced the inclined rotating fixed-bed reactor (IRR)  
 67 concept, where the superposition of reactor inclination and rotation at moderate  
 68 rotational velocities shape a stratified flow. Such flow pattern yields a spatial  
 69 modulation of the liquid holdup along the whole fixed bed. The process intensification  
 70 potential was studied for  $\alpha$ -methylstyrene hydrogenation and returned almost  
 71 doubled conversion rates compared to the conventional TBR.

72 However, a stratified flow, which ensures a binary wetting cycle of the whole catalyst  
 73 bed, i.e. ideal flow stratification with liquid filling level of exactly 50%, can hardly be  
 74 adjusted. To enlarge the beneficial process window towards lower liquid holdups and  
 75 to adjust the respective durations of the wetting and draining parts, Schubert (2018)  
 76 proposed an advanced IRR design with a concentric inner tube (Figure 1a).



77

78 **Figure 1:** a) IRR concept with concentric inner tube and (b) minimum and maximum  
 79 liquid filling level  $h_L$ , in the cross-section of the IRR indicating the beneficial process  
 80 window with (c) corresponding local liquid saturation  $\beta_L$  and product concentration at  
 81 the catalyst surface  $C_P$ .

82

83 This advanced reactor design enables full utilization of the catalyst as long as the  
 84 liquid filling level  $h_L$  is between lower and upper heights ( $h_{L,min} \leq h_L \leq h_{L,max}$ ) of the

85 inner tube (Figure 1b). Accordingly, inner tube diameter and reactor diameter are  
86 crucial geometric parameters of this advanced reactor concept. A recent numerical  
87 analysis for the  $\alpha$ -methylstyrene hydrogenation based on a heterogeneous  
88 continuum model revealed the potential of the advanced design (Timaeus et al.,  
89 2019a). Compared to the previous concept of Härting et al. (2015), a further increase  
90 of the STY was obtained at lower gas-liquid interface positions (Figure 1b, left). Here,  
91 the catalyst particles experience the wetting intermittency with a longer period at  
92 drained conditions, which intensifies the gas mass transfer to the catalysts even more  
93 (Figure 1c, left).

94 The objective of this work is to identify the process windows for the stratified flow in  
95 the IRR with concentric inner tube to fully utilize the potential of that reactor concept.  
96 In particular, flow pattern, liquid filling level, and specific pressure drop are analysed  
97 depending on superficial phase velocities, inclination angle, rotational velocity,  
98 particle diameter and inner tube diameter. For the flow visualization, capacitance-  
99 based wire-mesh sensors (WMSs) are applied to distinguish liquid (cumene) and gas  
100 (nitrogen) in the fixed-bed (porous alumina particles) based on the relative  
101 permittivities (Timaeus et al., 2019b).

102

## 103 **2 Material and methods**

### 104 *2.1 Experimental setup*

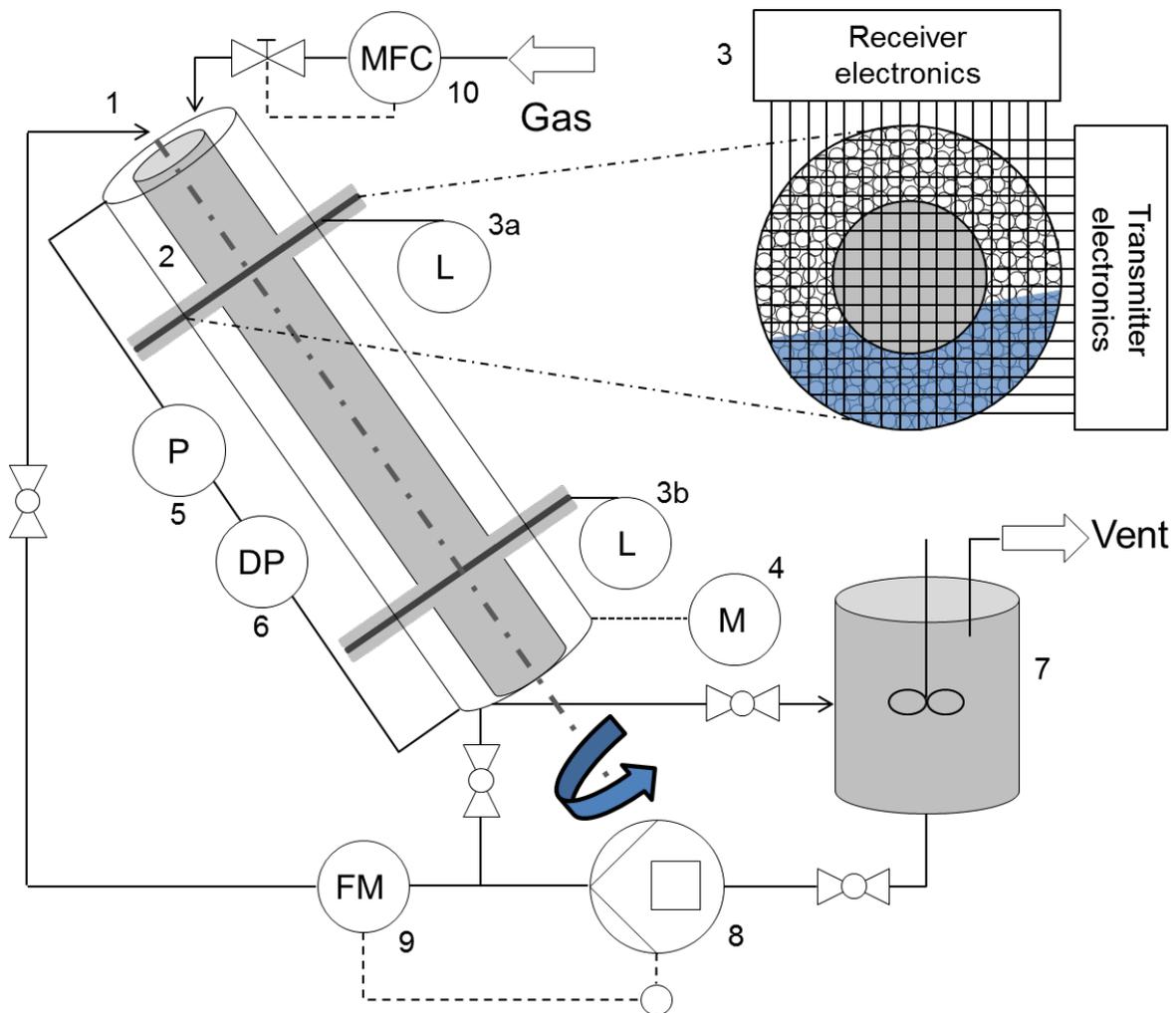
105 The experimental setup of the IRR (1) with concentric inner tube (2) is schematically  
106 shown in Figure 2. Reactor and concentric inner tube of the IRR are made of three  
107 stainless steel segments. The upper and lower segments have a length of 0.3 m,  
108 while the middle segments have a length of 1.0 m. The inner reactor diameter  $D_0$  is  
109 0.1 m and the interchangeable inner tubes have outer diameters  $D_T$  of 0.03 m or  
110 0.05 m. The resulting annular gaps of 25 mm or 35 mm, respectively, are filled with  
111 porous alumina particles of 2.5 mm or 4.5 mm mean diameter. To prevent both  
112 particle motion relative to the IRR rotation and particle abrasion, two grids installed at  
113 reactor inlet and outlet immobilize the particle packing in between. Three  
114 configurations of the IRR are installed, which are defined by concentric tube diameter  
115 and particle diameter as summarized in Table 1.

116

117 **Table 1:** Configurations of the IRR with inner tube and corresponding dimensions.

Configuration	Tube diameter (mm)	Particle diameter (mm)	Gap-to-particle- diameter ratio
A	50	4.5	5.5
B	50	2.5	10
C	30	4.5	7.8

118



119

120 **Figure 2:** Inclined rotating fixed-bed reactor setup with capacitance wire-mesh  
 121 sensors (1 – reactor, 2 – inner tube, 3a,b – capacitance wire-mesh sensors with  
 122 autarkic sensor electronics, 4 – hollow shaft rotary actuator, 5 – absolute pressure  
 123 transducer, 6 – difference pressure transducer, 7 – gas-liquid separator, 8 – lobe  
 124 rotor pump, 9 – Coriolis liquid flow meter, 10 – gas mass flow controller).

125

126 The reactor is installed in an inclinable rack and can be adjusted at arbitrary  
127 inclination angles from horizontal to vertical orientation. The capacitance WMSs (3)  
128 are flange-mounted between the reactor segments. A hollow shaft rotary actuator (4,  
129 DG130R-ASAC, Oriental Motor) with a stepper motor is used to rotate the IRR. To  
130 cope with the rotating reactor, wireless power supply and data acquisition are utilized.  
131 Two power banks are mounted on the reactor to operate the WMSs as well as  
132 absolute (5, PAA23SY, Omega) and differential (6, PD23, Omega) pressure  
133 transducers. Moreover, a WLAN module is fixed at the IRR to connect the sensors to  
134 a measurement computer for the data acquisition. The liquid mass flow is adjusted  
135 via lobe rotor pump (8, MDL0230, Waukesha Cherry-Burrell) controlled by a Coriolis  
136 liquid flow meter (9, Optimass 1300C, Krohne) and the gas flow rate is controlled via  
137 mass flow controller (10, FMA-2611A, Omega). It should be noted that the superficial  
138 velocities provided below refer to the cross-sectional area of the annuli available for  
139 the flow. Gas (nitrogen) and liquid (cumene, purity of 99.9 %, Acros Organics) are co-  
140 currently introduced at top of the IRR and a gas-liquid separator is installed  
141 downstream the IRR (7). Here, gas is released to the in-house ventilation system and  
142 liquid is recycled. The measurements were conducted at ambient pressure and room  
143 temperature.

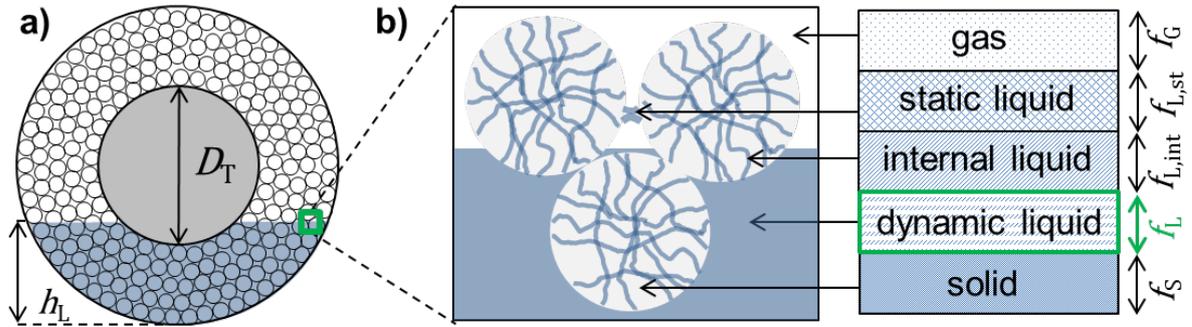
144

## 145 2.2 Capacitance wire-mesh sensors

146 Low-intrusive capacitance WMSs are used to visualize the flow and to determine the  
147 liquid filling level  $h_L$  in the IRR. Each WMS consists of 16 receiver and 16 transmitter  
148 wires orthogonally arranged with a small axial offset of 2.5 mm. 176 out of the 256  
149 wire crossing points are located within in the cross-section of the annulus. Their  
150 spatial separation is 6.25 mm. The sensor data are read out with a temporal  
151 resolution of 25 Hz, which is sufficient for the evolving quasi-stationary flows in the  
152 IRR. Naturally, same liquid distributions are obtained at both sensor positions, thus,  
153 only data of the lower WMS, i.e. 0.3 m upstream the reactor outlet, are processed  
154 and discussed below. It should be noted that the second WMS could be used in the  
155 future for transient tracer studies to reveal the liquid residence time and dispersion  
156 behavior. Liquid absorbed in the pore network of the catalyst particles and static  
157 liquid trapped between catalyst particles cannot be distinguished from the flowing  
158 liquid by the WMS. Hence, the obtained local liquid fractions  $\beta_L$  are considered as so-

159 called reduced liquid saturations, i.e. dynamic liquid fractions related to the interstitial  
 160 void space of the fixed-bed reduced by the static liquid. The allocation of involved  
 161 phases in the IRR to certain fractions is exemplarily shown in Figure 3.

162



163

164 **Figure 3:** a) Phase distribution in the cross-section of the IRR with marked liquid  
 165 filling level  $h_L$  and b) allocation of occurring phases in the IRR to certain fractions,  
 166 which are: gas: continuous gas phase of the stratified flow pattern, static liquid:  
 167 external liquid trapped between the catalyst, internal liquid: liquid inside the catalyst  
 168 pores, dynamic liquid: external flowing liquid, solid: skeletal aluminum structure of the  
 169 catalyst.

170

171 The measuring principle of the capacitance WMS is based on the logarithmic relation  
 172 between the output voltage  $V_0$  of the transimpedance amplifier of the WMS and the  
 173 permittivity of a sensing point according to

$$V_{\log} = a \cdot \ln(\varepsilon_x + \varepsilon_p) + b, \quad (1)$$

174 where,  $a$  and  $b$  are geometry-related constants,  $\varepsilon_x$  and  $\varepsilon_p$  are the permittivities  
 175 accounting for fluid phases and solid packing, respectively. Recently, it was shown  
 176 that the permittivity  $\varepsilon_p$  of a constant solid fraction can be neglected as the deviation is  
 177 less than 2% for the cross-sectionally averaged  $\beta_L$  at stratified flow (Timaeus et al.,  
 178 2019b). Thus, the unknown constants  $a$  and  $b$  can be obtained via two reference  
 179 scans (flooded and drained fixed-bed). Accordingly, a linear dependence between  $\beta_L$   
 180 and the measured permittivity  $\varepsilon_{\text{Meas}}$  is assumed (Bierberle et al., 2010; Prasser et al.,  
 181 1998; da Silva et al., 2007) and  $\beta_L$  can be determined as

$$\beta_L = \frac{\varepsilon_{\text{Meas}} - \varepsilon_G}{\varepsilon_L - \varepsilon_G}, \quad (2)$$

182 where  $\varepsilon_L$  and  $\varepsilon_G$  are the permittivities of gas and liquid phases, respectively. This way,  
 183  $\beta_L$  is calculated for each crossing point exposed to the flow. Subsequently, average  
 184 liquid saturation and corresponding liquid filling level  $h_L$  are determined from  
 185 7500 frames obtained for a measurement duration of 300 seconds with a frequency  
 186 of 25 Hz. Further details of the capacitance WMS and the data processing are  
 187 summarized in Timaeus et al. (2019b).

188

### 189 2.3 Porosity of the annular packed bed

190 The porosity of the annular packed bed near the walls can significantly deviate from  
 191 the bulk porosity (Sodré and Parise, 1998; du Toit, 2008). Accordingly, higher  
 192 interstitial phase velocities near the walls affect mass transfer and residence time  
 193 behavior of the IRR. Cheng and Hsu (Cheng and Hsu, 1986) proposed an  
 194 exponential approximation of the porosity in an annulus packed with spheres  
 195 according to

$$\varepsilon(r) = \begin{cases} \varepsilon_b \left( 1 + C \exp\left(-N \frac{r - R_T}{d_p}\right) \right) & \text{for } R_T \leq r \leq \frac{R_T + R_O}{2}, \\ \varepsilon_b \left( 1 + C \exp\left(-N \frac{R_O - r}{d_p}\right) \right) & \text{for } \frac{R_T + R_O}{2} \leq r \leq R_O, \end{cases} \quad (3)$$

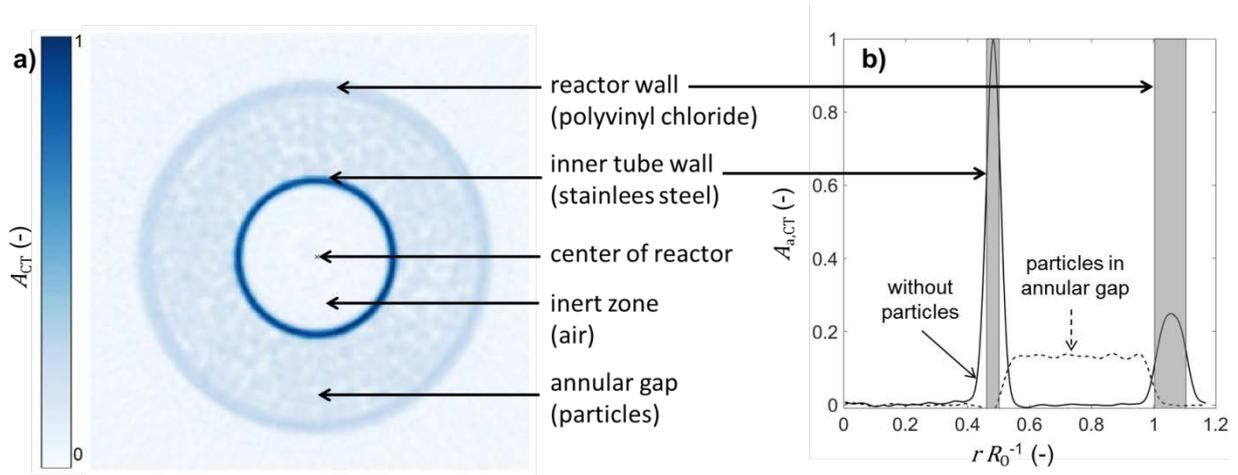
196 where  $r$  is the radial coordinate,  $R_T$  is the inner tube radius,  $R_O$  is the reactor radius,  $\varepsilon_b$   
 197 is the bulk porosity of the packed bed and  $C$  and  $N$  are fitting parameters. According  
 198 to (Vortmeyer and Schuster, 1983; Cheng and Hsu, 1986; Hunt and Tien, 1990),  $C$  is  
 199 used to fulfill

$$\varepsilon(r = R_T) = \varepsilon(r = R_O) = 1. \quad (4)$$

200 Furthermore,  $N$  describes the slope of the porosity in the vicinity of the walls and a  
 201 wide range for  $N$  is reported in the literature (du Toit, 2008; Vortmeyer and Schuster,  
 202 1983; Cheng and Hsu 1986). For example, Cheng and Hsu (1986) applied  $N = 2$ ,  
 203 while Hunt and Tien (1988) proposed  $N = 8$ . Sodre and Parise (1998) described a

204 linear relation between particle-to-annulus diameter ratio and  $N$ , however, non-linear  
 205 behavior is rather expected at wider parameter range. Thus,  $N$  for the IRR is derived  
 206 from the porosity profile obtained via gamma-ray computed tomography (CT) with a  
 207 collimated  $^{137}\text{Cs}$  source (662 keV gamma-photon energy). Details about the non-  
 208 invasive gamma-ray imaging and corresponding data processing can be found at  
 209 Bieberle et al. (2010). The tomographic experiments were conducted with annular  
 210 packed beds of 0.3 m length for the configurations listed in Table 1 (see Section 2.1).  
 211 Exemplarily, the cross-sectional distribution of the normalized attenuation coefficients  
 212  $A_{\text{CT}}$  of Configuration A is shown in Figure 4a.

213



214

215 **Figure 4:** a) Cross-sectional distribution of the normalized attenuation coefficients  $A_{\text{CT}}$   
 216 of the IRR (Configuration A) and b) corresponding radial profiles of the azimuthally  
 217 averaged  $A_{\text{a,CT}}$  with and without packed bed (the latter profile is utilized to locate the  
 218 position of the packed bed).

219

220 Additionally, measurements without particles were carried out to detect the exact  
 221 positions of the annuli. Here, the local maxima define the centers of the inner and  
 222 outer tube walls. The radial profiles of the two associated normalized attenuation-  
 223 coefficients  $A_{\text{a,CT}}$  for Configuration A are shown in Figure 4b. To assign densities to  
 224 the attenuation coefficients, the total weight of the packing was considered and a  
 225 least square optimization was performed according to

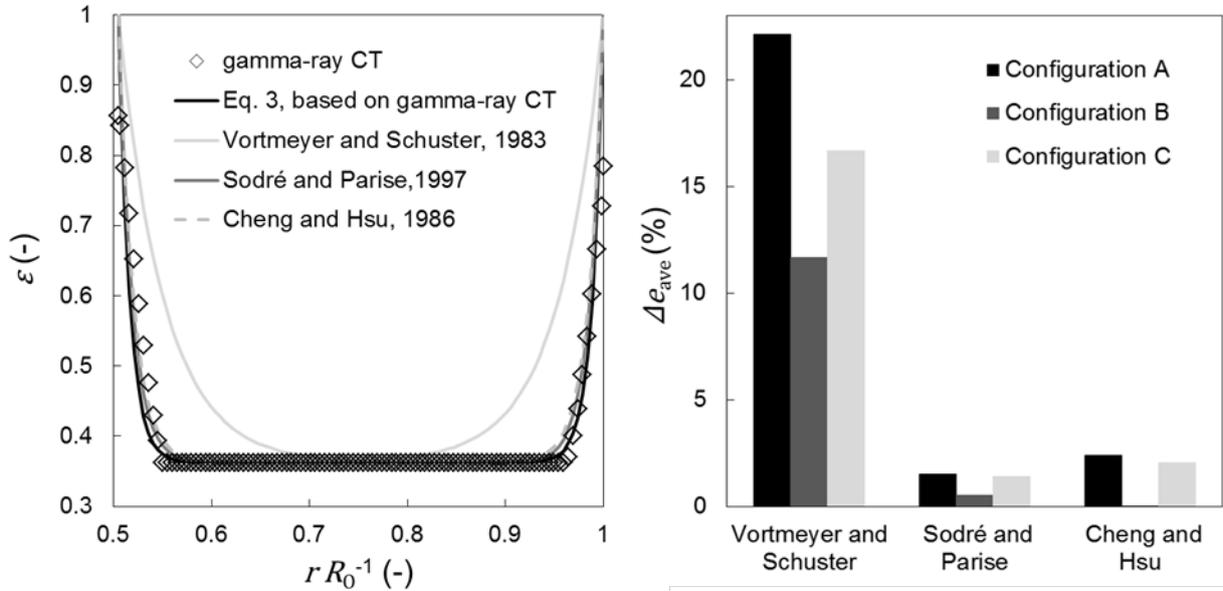
$$\min_{\rho} \left( \iiint A_{\text{a,CT}}(r) \rho r dr d\varphi dz - m_{\text{bed}} \right)^2, \quad (5)$$

226 where,  $\rho$  is the fitted density and  $m_{\text{bed}}$  is the mass of the annular packed bed. The  
 227 porosity of the packed bed is defined as

$$\varepsilon(r) = \frac{\rho_P - \rho_{\text{bed}}(r)}{\rho_P - \rho_G}, \quad (6)$$

228 where  $\rho_P$ ,  $\rho_{\text{bed}}$  and  $\rho_G$  are the densities of particles, packed bed and gaseous phase.  
 229 Eventually, the bulk porosity  $\varepsilon_b$  is obtained from the radial porosity profile, which  
 230 enables the determination of the parameter  $N$  by a least square fit. In Figure 5a the  
 231 porosity distribution of Configuration A obtained by the procedure described above is  
 232 exemplarily shown.

233



234

235 **Figure 5:** a) Porosity profile of Configuration A ( $D_T = 5$  cm,  $d_P = 4.5$  mm) obtained  
 236 tomographically and predicted with Eq. 3 for various  $N$  proposed in the literature and  
 237 b) deviations between measured and predicted average porosities.

238

239 Additionally, predictions for the porosity profiles using Eq. 2 are shown for  $N$  fitted to  
 240 the experimental data and proposed in the literature. Figure 5b shows the deviations  
 241  $\Delta e$  between the various predictions for the average porosity and the experimental  
 242 value. The determined parameters for each configuration of the IRR are summarized  
 243 in Table 2. The shown attenuation coefficients and the porosity data are available in  
 244 RODARE: 10.14278/rodare.204 [dataset] (Timaeus et al., 2019).

245

246 **Table 2:** Porosity data and determined parameters of the exponential approximations  
247 based on gamma-ray CT scans.

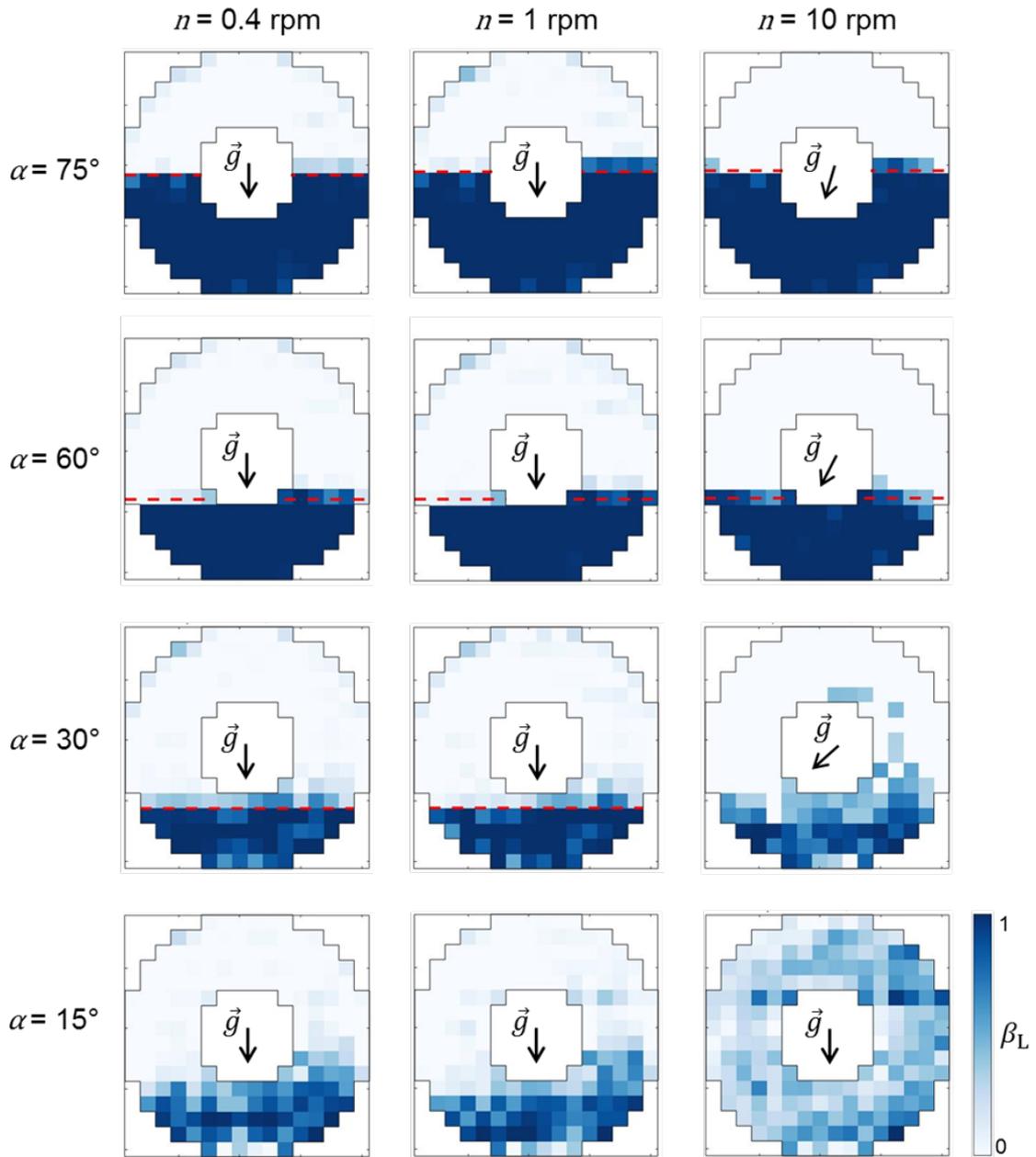
Configuration	$\varepsilon_{ave}$ ( $mV^3 mR^{-3}$ )	$\varepsilon_b$ ( $mV^3 mR^{-3}$ )	$C$ (-)	$N$ (-)
A	0.392	0.362	1.76	8.02
B	0.375	0.352	1.84	5.99
C	0.380	0.359	1.78	8.37

248

### 249 **3 Results**

250 The hydrodynamic characterization of the new IRR design comprises phase  
251 distribution, liquid filling level and specific pressure drop. The operating conditions  
252 are chosen to adjust the stratified flow, ensuring the beneficial wetting intermittency.  
253 Here, low to moderate rotational velocities are of particular interest as observed by  
254 Härting et al. in reaction studies with the first generation design of the IRR (Härting et  
255 al., 2015). Initially, the cross-sectional phase distribution in the IRR is visualized for  
256 each operating point to identify the flow regime using the WMS sensor 3b. Figure 6  
257 summarizes the patterns for different rotational velocities and inclination angles for  
258 Configuration A.

259



260

261 **Figure 6:** Cross-sectional saturation distribution for Configuration A ( $D_T = 5$  cm,  
 262  $d_p = 4.5$  mm) at  $u_L = 0.01$  m s $^{-1}$ ,  $u_G = 0.05$  m s $^{-1}$  and different rotational velocities  $n$   
 263 and inclination angles  $\alpha$ . Gravity vector  $\vec{g}$  and gas-liquid interface of the stratified flow  
 264 patterns (dashed line) are also shown.

265

266 The liquid filling level  $h_L$ , i.e. height of the gas-liquid interface position for the stratified  
 267 flow patterns, is determined according to the procedure explained in Section 2.2  
 268 (highlighted with a horizontal dashed red line). The gravity vector indicates the  
 269 bottommost position of the reactor cross-section.

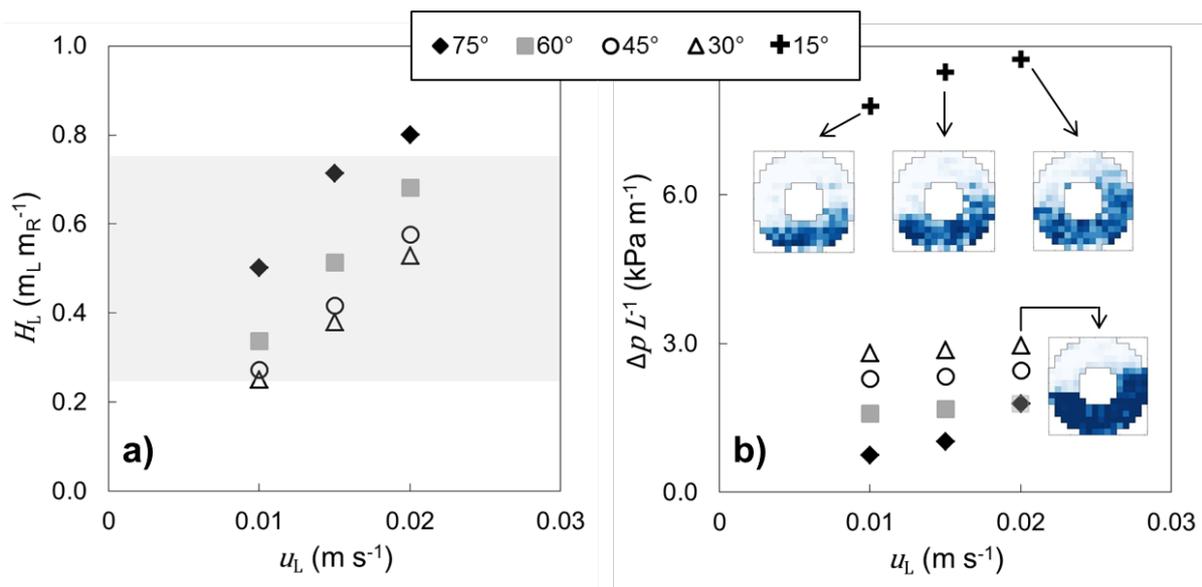
270 At low inclination angles ( $\alpha = 15^\circ$  and  $30^\circ$ ) and high rotational velocities ( $n = 10$  rpm)  
 271 the flow stratification vanishes. Here, the effects of the centrifugal force excel the  
 272 gravitational force counterparts and the operating conditions result in liquid filling  
 273 levels, which are beyond the beneficial IRR process window. Below, the effects of  
 274 superficial phase velocities, inclination angle, rotational velocity, particle size and  
 275 inner tube diameter on normalized liquid filling level ( $H_L = h_L / D_0$ ) and specific  
 276 pressure drop are discussed in detail.

277

### 278 3.1 Effects of superficial liquid velocity and inclination angle

279 Figure 7a shows  $H_L$  as function of the superficial liquid velocity  $u_L$  for inclination  
 280 angles between  $15^\circ$  to  $75^\circ$  at a rotational velocity of 1 rpm for a superficial gas  
 281 velocity of  $0.05 \text{ m s}^{-1}$ . The beneficial hydrodynamic process window is defined by  
 282 inner tube and reactor diameters ( $D_r$  and  $D_0$ ), which set minimum and maximum  
 283 normalized liquid filling levels ( $H_{L,\min}$  and  $H_{L,\max}$ ), enabling a wetting intermittency for  
 284 the whole fixed-bed. For Configurations A and B,  $H_{L,\min}$  and  $H_{L,\max}$  are 0.25 and 0.75,  
 285 respectively (indicated by the shaded area in Figure 7a). Additionally, the  
 286 corresponding specific pressure drop  $\Delta p L^{-1}$  is shown in Figure 7b.

287



288

289 **Figure 7:** a) Normalized liquid filling level  $H_L$  and (b) specific pressure drop  $\Delta p L^{-1}$   
 290 depending on superficial liquid velocity  $u_L$  and inclination angle  $\alpha$  ( $n = 1$  rpm,  
 291  $u_G = 0.05 \text{ m s}^{-1}$ ,  $D_r = 5 \text{ cm}$ ,  $d_p = 4.5 \text{ mm}$ ). The shaded area indicates the

292 hydrodynamic process window for the IRR (same notation is used in the figures  
293 below).

294

295 Logically,  $H_L$  increases with increasing superficial liquid velocity regardless of the  
296 inclination angle. The same trend is obtained for the specific pressure drop. Here,  
297 higher solid-fluid phase interactions occur at higher superficial liquid velocities due to  
298 less accessible cross-section per fluid phase. Considering the stratified flow patterns,  
299 the specific pressure drop is most sensitive to the liquid load at inclination angles of  
300  $75^\circ$ , where  $H_L$  reaches the highest values.

301  $H_L$  also increases with increasing inclination angle due to the lower downhill driving  
302 force. This increases the residence time of the liquid. The effect of the inclination  
303 angle on  $H_L$  decreases with lower inclination angles. Contrary, the specific pressure  
304 drop decreases with increasing inclination angles. Here, the gas-liquid interactions  
305 reduce with more pronounced phase stratification. In particular, a strong increase of  
306 the specific pressure drop occurs from  $30^\circ$  to  $15^\circ$ , which can be explained by the  
307 transition from stratified to sickle flow as illustrated in Figure 7b.

308 For the highest superficial liquid velocity of  $0.02 \text{ m s}^{-1}$  at an inclination angle of  $75^\circ$   $H_L$   
309 exceeds the inner tube. Thus, parts of the catalyst packing are excluded from the  
310 advantageous wetting intermittency and rotate permanently within the liquid phase  
311 without access to the gas phase.

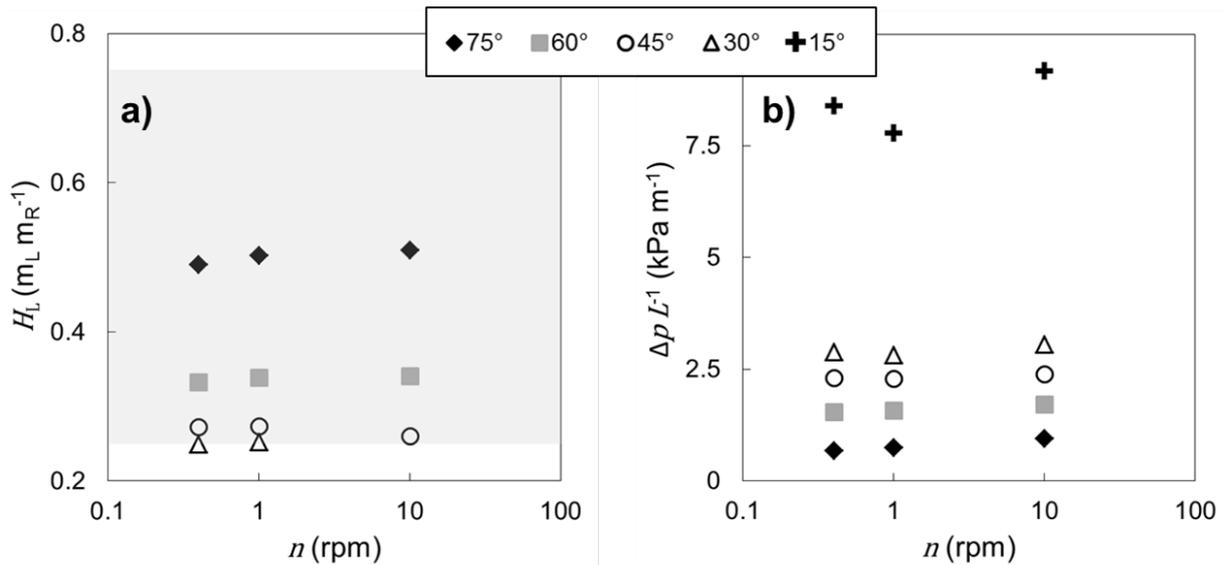
312 The results reveal that the selection of inclination angle and superficial liquid velocity  
313 enables an arbitrary adjustment of the gas-liquid interface position to tune the  
314 duration of the catalyst immersion per reactor rotation, also referred to as 'split' in  
315 periodically operated trickle-bed reactors. From a reaction engineering point of view,  
316  $H_L$  can be adjusted to  $H_{L,\min}$  to increase the gas-solid mass transfer for reaction  
317 systems with excess of liquid educt. At the same time, the residence time of the liquid  
318 phase is decreased, which leads to a trade-off between high residence time and  
319 beneficial mass transfer conditions. Contrary, if liquid educt depletion occurs  $H_L$  can  
320 be increased accordingly.

321

322 3.2 Effect of rotational velocity

323 The immersion and draining durations of the catalysts are adjusted by the rotational  
 324 velocity. Here, a range between 0.4 rpm and 10 rpm was investigated. Figure 8  
 325 shows  $H_L$  and  $\Delta p L^{-1}$  as a function of the rotational velocity at superficial liquid and  
 326 gas velocity of  $0.01 \text{ m s}^{-1}$   $0.05 \text{ m s}^{-1}$ , respectively, for Configuration A.

327



328

329 **Figure 8:** Effect of the rotational velocity on a) liquid filling level  $H_L$  (only data for  
 330 stratified flow are shown) and b) specific pressure drop  $\Delta p L^{-1}$  for different inclination  
 331 angles ( $u_G = 0.05 \text{ m s}^{-1}$ ,  $u_L = 0.01 \text{ m s}^{-1}$ ,  $D_T = 5 \text{ cm}$ ,  $d_p = 4.5 \text{ mm}$ ).

332

333  $H_L$  slightly increases with increasing rotational velocity as a consequence of higher  
 334 liquid residence time at higher rotational velocities. Here, increasing transversal  
 335 forces act against the bulk flow. However, the effect of the rotational velocity is low  
 336 compared to the effects of inclination angle and superficial liquid velocity.

337 The same trend is observed for the specific pressure drop as long as stratified flow is  
 338 present. The transition from stratified to dispersed flow at an inclination angle of  $15^\circ$   
 339 results in a large increase of the specific pressure drop due to significantly higher  
 340 interactions between the fluid phases.

341 At a rotational velocity of 0.4 rpm and an inclination angle of  $30^\circ$   $H_L$  is below the inner  
 342 tube, thus, parts of the catalyst packing are excluded from the wetting intermittency

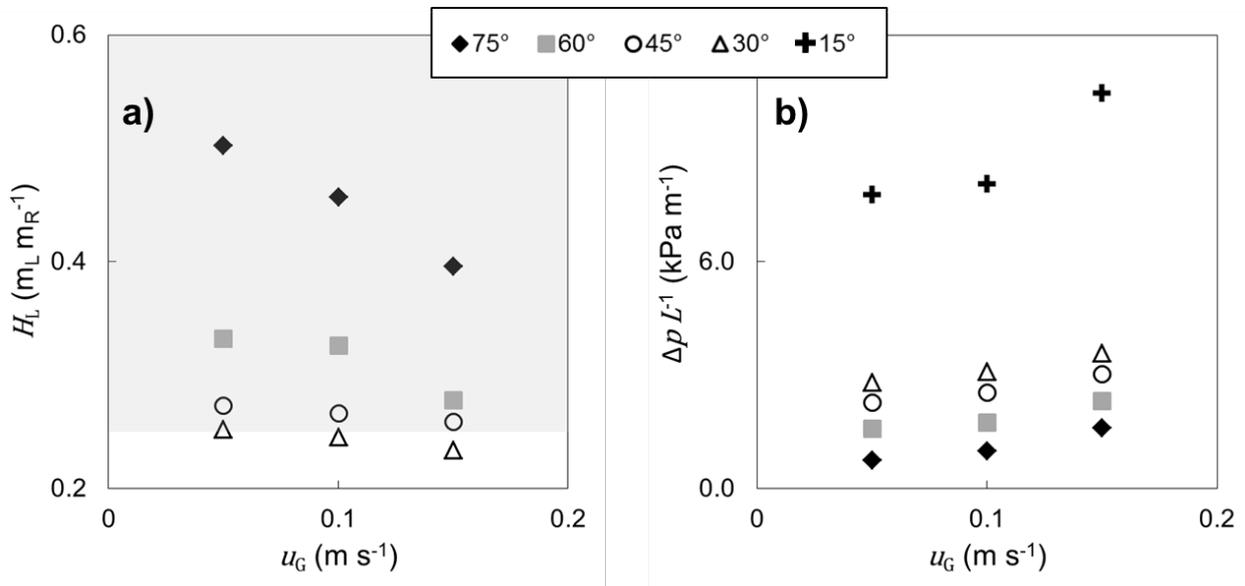
343 and rotate permanently within the gas phase without access to the liquid bulk phase.  
 344 Such operating conditions cause severe liquid educt depletion.

345

### 346 3.3 Effect of superficial gas velocity

347 The superficial gas velocity is crucial for the gas-solid mass transfer in the drained  
 348 sections of the catalyst packings. In Figure 9,  $H_L$  and  $\Delta p L^{-1}$  are shown as a function  
 349 of the superficial gas velocity ranging from  $0.05 \text{ m s}^{-1}$  to  $0.15 \text{ m s}^{-1}$ , for  
 350 Configuration A.

351



352

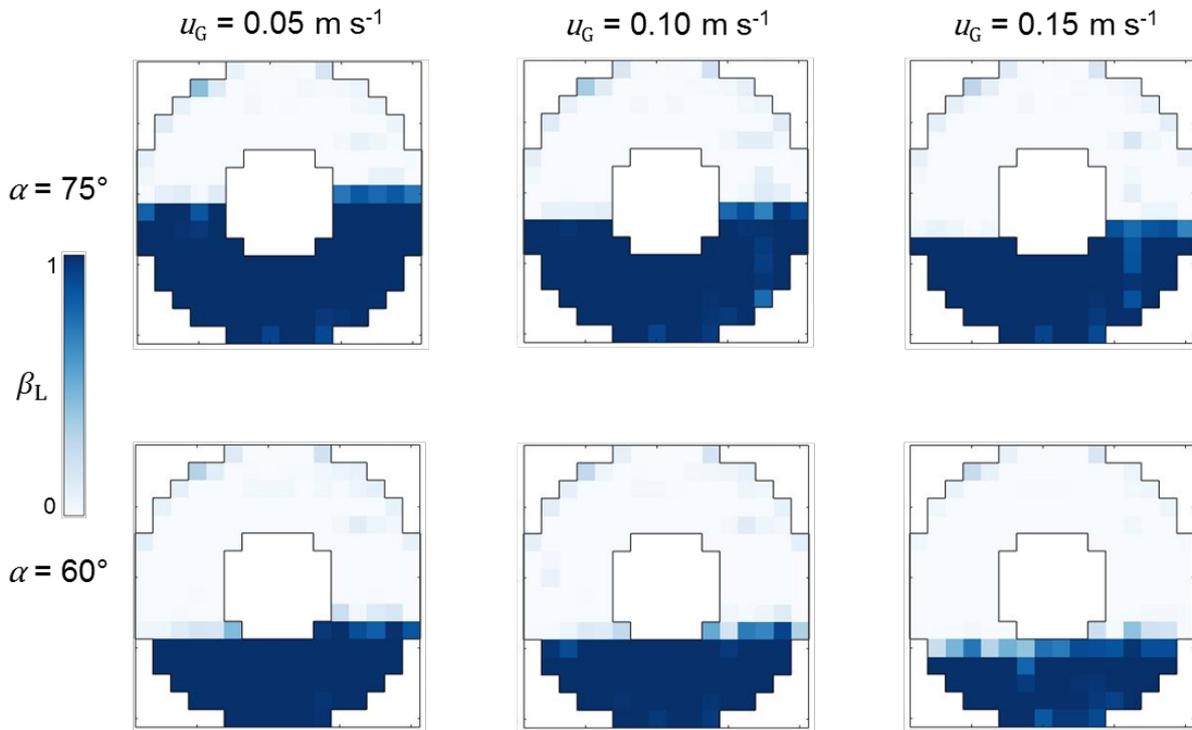
353 **Figure 9:** a) Liquid filling level  $H_L$  and b) specific pressure drop  $\Delta p L^{-1}$  as a function of  
 354 the superficial gas velocity ( $D_T = 5 \text{ cm}$ ,  $d_p = 4.5 \text{ mm}$ ,  $n = 1 \text{ rpm}$ ,  $u_L = 0.01 \text{ m s}^{-1}$ ).

355

356  $H_L$  decreases with increasing superficial gas velocity for all cases. The higher shear  
 357 rate at the gas-liquid interface accelerates the liquid. This effect is very pronounced  
 358 at the highest inclination angle as a result of the high interstitial phase velocity  
 359 difference. Here, the axial bulk velocity of the liquid phase is comparably low as the  
 360 liquid height is built up, while the remaining area available for the gas phase  
 361 decreases, causing a higher interstitial gas velocity. Logically, the specific pressure  
 362 drop increases with higher superficial gas velocity due to increasing gas-solid and  
 363 gas-liquid interactions.

364 At the same time, the phase distinction for the stratified flow patterns remains stable  
365 without significant liquid entrainment towards the drained sections as shown in  
366 Figure 10.

367



368

369 **Figure 10:** Effect of the superficial gas velocity on the gas-liquid distribution  
370 ( $D_T = 5 \text{ cm}$ ,  $d_p = 4.5 \text{ mm}$ ,  $n = 1 \text{ rpm}$ ,  $u_L = 0.01 \text{ m s}^{-1}$ ).

371

372 High superficial gas velocities in the IRR foster the gas-solid mass transfer, and in  
373 turn, the reaction rate for gas-limited reactions. Following this, the superficial gas  
374 velocity should be increased to reach saturation of the gaseous educts at the catalyst  
375 surfaces as long as stratification is maintained. On the other hand the gas-solid  
376 interactions should not exceed conditions at which particle pores start to dry out and  
377 depletion of liquid educt leads to a limitation of the reaction rate.

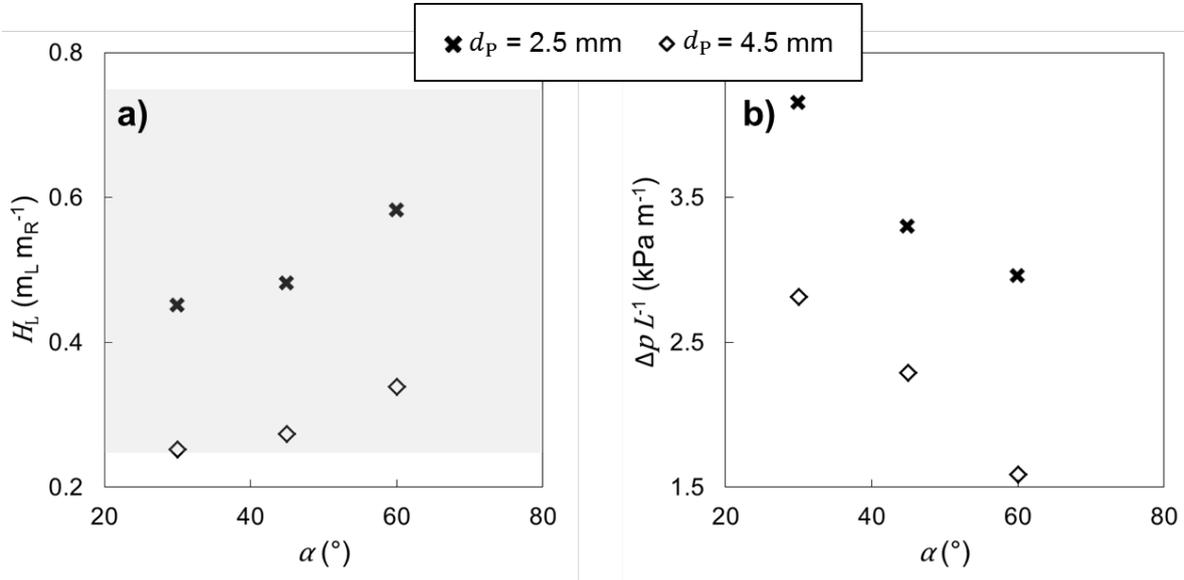
378

### 379 3.4 Effects of particle size and inner tube diameter

380 In fixed-bed reactors the particle diameter is decisive for liquid saturation, pressure  
381 drop and fluid-particle mass transfer. Thus, the effect of different spherical porous  
382 alumina particles on the hydrodynamics of the IRR is analysed for the

383 Configurations A and B. In Figure 11  $H_L$  and  $\Delta p L^{-1}$  are shown for different inclination  
 384 angles.

385



386

387 **Figure 11:** Comparison of a) liquid filling level  $H_L$  and b) specific pressure drop  $\Delta p L^{-1}$   
 388 for Configurations A ( $d_p = 4.5$  mm) and B ( $d_p = 2.5$  mm) depending on the inclination  
 389 angle ( $n = 1$  rpm,  $u_L = 0.01$  m s $^{-1}$ ,  $u_G = 0.05$  m s $^{-1}$ ,  $D_T = 5$  cm)

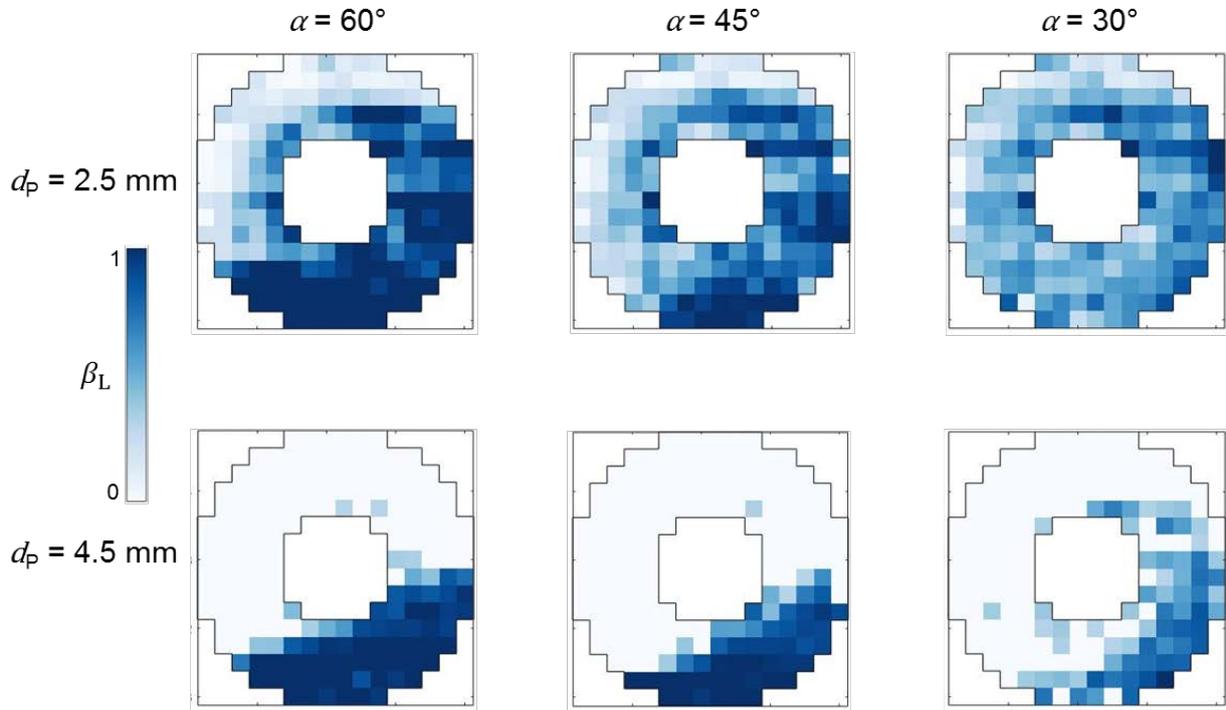
390

391 Under the same operating conditions  $H_L$  is significantly higher for Configuration B  
 392 compared to Configuration A regardless of the inclination angle. Here, the higher  
 393 specific surface area of Configuration B causes significant higher fluid-solid  
 394 interactions. Thus, the liquid velocity decreases and liquid accumulates. Moreover,  
 395 the average porosity of Configuration B is lower as a result of the lower bulk porosity  
 396 (see Table 2). Consequently, less void space is accessible for the fluids, which yields  
 397 a higher displacement of the liquid in Configuration B. Accordingly, the pressure drop  
 398 is significantly higher for Configuration B.

399 Furthermore, the cross-sectional gas-liquid patterns reveal a smaller process window  
 400 for Configuration B. A significant entrainment of liquid is already observed at an  
 401 inclination angle of  $\alpha = 60^\circ$  and a rotational velocity of  $n = 10$  rpm for the small  
 402 particle packing, while distinct stratification is obtained at the same operating  
 403 conditions for the larger particles up to an inclination angle of  $\alpha = 45^\circ$  (Figure 12). The

404 earlier transition to sickle and dispersed flow for Configuration B results from higher  
405 fluid-solid interactions caused by the smaller particles as explained above.

406

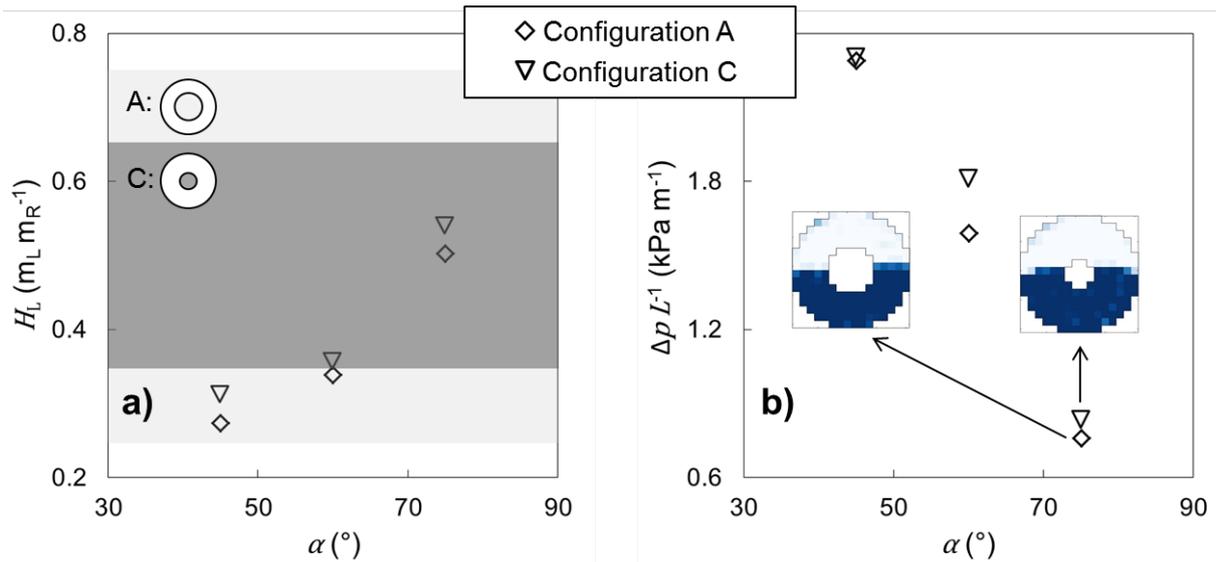


407

408 **Figure 12:** Effect of the particle diameter (lower row: Configuration A with  
409  $d_p = 4.5$  mm, upper row: Configuration B with  $d_p = 2.5$  mm) on the gas-liquid  
410 distribution ( $D_T = 5$  cm,  $n = 10$  rpm,  $u_L = 0.01$  m s<sup>-1</sup>,  $u_G = 0.05$  m s<sup>-1</sup>).

411

412 A basic design parameter of the IRR is the inner tube diameter  $D_T$ , which defines the  
413 hydrodynamic process window. Obviously, the reduction of  $D_T$  narrows the  
414 hydrodynamic process window. Nevertheless, for reactions or process conditions  
415 with better performance at higher  $H_L$  ( $0.35 < H_L < 0.7$ ), smaller inner tubes might be of  
416 advantage since the absolute mass flow is higher (at same superficial fluid  
417 velocities), i.e. lower investment costs compared to designs with larger inner tube  
418 diameter. Figure 13 shows the effect of different inner tube diameters on  $H_L$  for  
419 different inclination angles.



420

421 **Figure 13:** Effect of the inner tube diameter (Configuration A:  $D_T = 5$  cm,  
 422 Configuration C:  $D_T = 3$  cm) on a) normalized liquid filling level  $H_L$  and b) specific  
 423 pressure drop  $\Delta p L^{-1}$  for different inclination angles ( $d_p = 4.5$  mm,  $n = 1$  rpm,  
 424  $u_L = 0.01$  m s $^{-1}$ ,  $u_G = 0.05$  m s $^{-1}$ ). The shaded areas denote the hydrodynamic process  
 425 windows of Configuration A (bright grey) and C (dark grey), respectively.

426

427 Slightly smaller values for liquid filling level  $H_L$  and specific pressure drop are  
 428 obtained for Configuration C with the smaller inner tube. The obtained differences are  
 429 caused by the slightly lower porosity of Configuration C due to the smaller wall effect  
 430 (see Table 2). The lower interstitial void space between the catalysts reduces the  
 431 liquid residence time and increases the accumulation of the liquid.

432 All datasets analyzed in this Section are available in RODARE: 10.14278/rodare.204  
 433 [dataset] (Timaeus et al., 2019).

434

## 435 4 Conclusions

436 In this work, the hydrodynamic process windows, i.e. essential stratified flow pattern  
 437 and range of the liquid filling level, and the specific pressure drop of an inclined  
 438 rotating fixed-bed reactor with inner tube were studied for different design  
 439 parameters. Besides, the wall effect of the additional inner tube on the porosity profile  
 440 was characterized via gamma-ray CT. A capacitance wire-mesh sensor was used to  
 441 visualize the liquid phase distribution in packings of porous alumina particles. Here,

442 the liquid filling level for the stratified flow patterns was derived averaging the local  
443 liquid saturations at the 176 crossing points.

444 Only minor effects of the rotational velocity were obtained at stratified flow conditions,  
445 while superficial phase velocities and inclination angle were found to have crucial  
446 effects. Arbitrary positions of the gas-liquid interface are adjustable in the IRR to  
447 adapt the durations for the liquid-solid and gas-solid contact depending on the  
448 prevailing mass transfer limitation of the reaction system. While the installation of a  
449 smaller tube revealed no significant influence on the investigated hydrodynamic  
450 parameters, smaller particle diameters were found to significantly increase the liquid  
451 filling level and the specific pressure drop, respectively.

452 Reaction studies will be carried out in another study to quantify the intensification  
453 potential of the inclined rotating fixed-bed reactor with inner tube for the located  
454 process window. Additionally, the new hydrodynamic data obtained in this study will  
455 further help to develop and validate a numerical model for a CFD-based reactor  
456 design as a next step.

457

## 458 **Acknowledgement**

459 The authors are grateful to the German Research Foundation for financial support  
460 (DFG SCHU 2421/2-3)

461

## 462 **Symbols used**

463	$a$	[-]	geometry-related constant 1
464	$A$	[m]	normalized attenuation coefficient
465	$b$	[-]	geometry-related constant 2
466	$C$	[-]	fitting parameter 1, porosity correlation
467	$d_p$	[m]	particle diameter
468	$D$	[m]	diameter
469	$\Delta e$	[-]	deviation between experiment and correlation

470	$g$	[m s <sup>-2</sup> ]	gravitational acceleration
471	$h$	[m]	filling level
472	$H$	[-]	normalized filling level
473	$L$	[m]	axial length of reactor
474	$n$	[rpm]	rotational velocity
475	$N$	[-]	fitting parameter 2, porosity correlation
476	$p$	[Pa]	pressure
477	$r$	[m]	radial coordinate
478	$R$	[m]	radius
479	$u$	[m s <sup>-1</sup> ]	superficial velocity
480	$V$	[V]	voltage
481	$z$	[m]	axial coordinate

482

483 **Greek symbols**

484	$\alpha$	[°]	inclination angle towards the gravity
485	$\beta$	[m <sup>3</sup> m <sup>-3</sup> ]	reduced saturation
486	$\Delta p L^{-1}$	[kPa m <sup>-1</sup> ]	specific pressure drop
487	$\rho$	[kg m <sup>-3</sup> ]	density
488	$\varepsilon$	[-]	relative permittivity
489	$\varepsilon$	[m <sub>V</sub> <sup>3</sup> m <sub>R</sub> <sup>-3</sup> ]	porosity
490	$\phi$	[°]	azimuthal coordinate

491

492 **Sub- and Superscripts**

493	a	azimuthal
494	ave	average
495	b	bulk
496	bed	packed bed with porous alumina spheres
497	G	gas
498	L	liquid
499	max	maximum
500	Meas	measured
501	min	minimum
502	O	outer tube
503	P	particle
504	R	reactor
505	T	inner tube
506	V	void
507	X	fluid contributors

508

509 **Abbreviations**

510	CT	gamma-ray computed tomography
511	IRR	inclined rotating fixed-bed reactor
512	STY	space-time yield
513	TBR	trickle-bed reactor
514	WMS	wire-mesh sensor

515

516 **References**

- 517 Banchemo, M., Manna, L., Sicardi, S., Ferri, A., 2004, Experimental investigation of  
518 fast-mode liquid modulation in a trickle-bed reactor, *Chem. Eng. Sci.* 59, 4149 –  
519 4154. <https://doi.org/10.1016/j.ces.2004.03.048>
- 520 Bieberle, A., Schubert, M., da Silva, M. J., Hampel, U., 2010, Measurement of liquid  
521 distributions in particle packings using wire-Mesh sensor versus transmission  
522 tomographic imaging, *Ind. Chem. Res.* 49, 9445 – 9453.  
523 <https://doi.org/10.1021/ie100900g>
- 524 Boelhouwer, J. G., Piepers, H. W., Drinkenburg, B. A. H., 2002a, Advantages of  
525 forced non-steady operated trickle-Bed reactors, *Chem. Eng. Technol.* 25, 647 –  
526 650. [https://doi.org/10.1002/1521-4125\(200206\)25:6<647::AID-  
527 CEAT647>3.0.CO;2-9](https://doi.org/10.1002/1521-4125(200206)25:6<647::AID-CEAT647>3.0.CO;2-9)
- 528 Boelhouwer, J. G., Piepers, H. W., Drinkenburg, A. A. H., 2002b, Liquid-induced  
529 pulsing flow in trickle-bed reactors, *Chem. Eng. Sci.* 57, 3387 – 3399.  
530 [https://doi.org/10.1016/S0009-2509\(02\)00210-5](https://doi.org/10.1016/S0009-2509(02)00210-5)
- 531 Castellari, A. T., Haure, P. M., 1995, Experimental study of the periodic operation of a  
532 trickle-bed reactor, *AIChE* 41, 1593 – 1597.  
533 <https://doi.org/10.1002/aic.690410624>
- 534 Cheng, P., Hsu, C. T., 1986, Fully-developed, forced convective flow through an  
535 annular packed-sphere bed with wall effects, *Int. J. Heat Mass Tran.* 29, 1843 –  
536 1853. [https://doi.org/10.1016/0017-9310\(86\)90003-7](https://doi.org/10.1016/0017-9310(86)90003-7)
- 537 Dietrich, W., Grünwald, M., Agar, D. W., 2005, Dynamic modelling of periodically  
538 wetted catalyst particles, *Chem. Eng. Sci.* 60, 6254 – 6261.  
539 <https://doi.org/10.1016/j.ces.2005.03.054>
- 540 Edouard, D., Lacroix, M., Pham, C., Mbodji, M., Pham-Huu, C., 2008, Experimental  
541 measurements and multiphase flow models in solid SiC foam beds, *AIChE* 54,  
542 2823 – 2832. <https://doi.org/10.1002/aic.11594>
- 543 Gelhausen, M. G., Yang, S., Cegla, M., Agar, D. W., 2017, Cyclic mass transport  
544 phenomena in a novel reactor for gas–liquid–solid contacting, *AIChE* 63, 208 –  
545 215. <https://doi.org/10.1002/aic.15532>

546 Haase, S., Murzin, D. Y., Salmi, T., 2016, Review on hydrodynamics and mass  
547 transfer in minichannel wall reactors with gas–liquid Taylor flow, *Chem. Eng.*  
548 *Res. Des.* 113, 304 – 329. <https://doi.org/10.1016/j.cherd.2016.06.017>

549 Härting, H. U., Lange, R., Larachi, F., Schubert, M., 2015, A novel inclined rotating  
550 tubular fixed bed reactor concept for enhancement of reaction rates and  
551 adjustment of flow regimes, *Chem. Eng. J.* 281, 931 – 944.  
552 <http://dx.doi.org/10.1016/j.cej.2015.06.092>

553 Hunt, M. L., Tien, C. L., 1990, Non-Darcian flow, heat and mass transfer in catalytic  
554 packed-bed reactors, *Chem. Eng. Sci.* 45, 55 – 63. [https://doi.org/10.1016/0009-](https://doi.org/10.1016/0009-2509(90)87080-C)  
555 [2509\(90\)87080-C](https://doi.org/10.1016/0009-2509(90)87080-C)

556 Hunt, M. L., Tien, C. L., 1988, Non-darcian convection in cylindrical packed beds, *J.*  
557 *Heat Transfer* 110, 378 – 384. <https://doi.org/10.1115/1.3250495>

558 Khadlikar, M. R., Al-Dahhan, M. H., Duduković, M. P., 1999, Parametric study of  
559 unsteady-state flow modulation in trickle-bed reactors, *Chem. Eng. Sci.* 54, 2585  
560 – 2595. [https://doi.org/10.1016/S0009-2509\(98\)00426-6](https://doi.org/10.1016/S0009-2509(98)00426-6)

561 Lange, R., Hanika, J., Stradiotto, D., Hudgins, R. R., Silveston, P. L., 1994,  
562 Investigations of periodically operated trickle-bed reactors, *Chem. Eng. Sci.* 49,  
563 5615 – 5621. [https://doi.org/10.1016/0009-2509\(94\)00363-7](https://doi.org/10.1016/0009-2509(94)00363-7)

564 Mills, P. L., Duducović, M. P., 1980, Analysis of catalyst effectiveness in trickle-bed  
565 reactors processing volatile or nonvolatile reactants, *Chem. Eng. Sci.* 35, 2267 –  
566 2279. [https://doi.org/10.1016/0009-2509\(80\)87004-7](https://doi.org/10.1016/0009-2509(80)87004-7)

567 Nigam, K. D. P., Larachi, F., 2005, Process intensification in trickle-bed reactors,  
568 *Chem. Eng. Sci.* 60, 5880 – 5894. <https://doi.org/10.1016/j.ces.2005.04.061>

569 Nijhuis, T. A., Kreutzer, M. T., Romijn, A. C. J., Kapteijn, F., Moulijn, J. A., 2001,  
570 Monolithic catalysts as efficient three-phase reactors, *Chem. Eng. Sci.* 56, 823 –  
571 829. [https://doi.org/10.1016/S0009-2509\(00\)00294-3](https://doi.org/10.1016/S0009-2509(00)00294-3)

572 Prasser, H. M., Böttger, A., Zschau, J., 1998, A new electrode-mesh tomograph for  
573 gas–liquid flows, *Flow. Meas. Instrum.* 9, 111 – 119.  
574 [https://doi.org/10.1016/S0955-5986\(98\)00015-6](https://doi.org/10.1016/S0955-5986(98)00015-6)

575 Schubert, M., 2018, DE 10 2018 110 091.4.

576 da Silva, M. J., Schleicher, E., Hampel, U., 2007, Capacitance wire-mesh sensor for  
577 fast measurement of phase fraction distributions, *Meas. Sci. Technol.* 18, 2245 –  
578 2251. <https://doi.org/10.1088/0957-0233/18/7/059>

579 Sodré, J. R., Parise, J. A. R., 1998, Fluid flow pressure drop through an annular  
580 packed bed of spheres with wall effects, *Exp. Therm. Fluid Sci.* 17, 265 – 275.  
581 [https://doi.org/10.1016/S0894-1777\(97\)10022-X](https://doi.org/10.1016/S0894-1777(97)10022-X)

582 Timaeus, R., Hampel, U., Schubert, M., 2019a, Heterogeneous modeling approach  
583 for gas-limited reactions in an inclined rotating fixed bed reactor, *Chem. Ing.  
584 Tech.* 91, 637 – 644. <https://doi.org/10.1002/cite.201800183>

585 Timaeus, R., Berger, R., Schleicher, E., Hampel, U., Schubert, M., 2019b, Liquid flow  
586 visualization in packed-bed multiphase reactors: wire-mesh sensor design and  
587 data analysis for rotating fixed beds, *Chem. Ing. Tech.* 91, 1 – 11.  
588 <https://doi.org/10.1002/cite.201900117>

589 Timaeus, R., Schleicher, E., Bieberle, A., Hampel, U., Schubert, M., 2019c,  
590 Hydrodynamic data of an advanced inclined rotating fixed-bed reactor, RODARE  
591 repository. <https://doi.org/10.14278/rodare.204>

592 du Toit, C. G., 2008, Radial variation in porosity in annular packed beds, *Nucl. Eng.  
593 Des.* 238, 3073 – 3079. <https://doi.org/10.1016/j.nucengdes.2007.12.018>

594 Utikar, R. P., Ranade, V., V., 2017, Intensifying multiphase reactions and reactors:  
595 strategies and examples, *ACS Sustain. Chem. Eng.* 5, 3607 – 3622.  
596 <https://doi.org/10.1021/acssuschemeng.6b03017>

597 Vortmeyer, D., Schuster, J., 1983, Evaluation of steady flow profiles in rectangular  
598 and circular packed beds by a variational method, *Chem. Eng. Sci.* 38, 1691 –  
599 1699. [https://doi.org/10.1016/0009-2509\(83\)85026-X](https://doi.org/10.1016/0009-2509(83)85026-X)

600 Wallenstein, M., Kind, M., Dietrich, B., 2014, Radial two-phase thermal conductivity  
601 and wall heat transfer coefficient, *Int. J. Heat Mass Transf.* 79, 486 – 495,  
602 <http://dx.doi.org/10.1016/j.ijheatmasstransfer.2014.08.003>

- 603 Zalucky, J., Wagner, M., Schubert, M., Lange, R., Hampel, U., 2017a,  
604 Hydrodynamics of descending gas-liquid flows in solid foams: Liquid holdup,  
605 multiphase pressure drop and radial dispersion, Chem. Eng. Sci. 168, 480 – 494.  
606 <http://dx.doi.org/10.1016/j.ces.2017.05.011>
- 607 Zalucky, J., Schubert, M., Lange R., Hampel, U., 2017b, Dynamic liquid–solid mass  
608 transfer in solid foam packed reactors at trickle and pulse flow, Ind. Eng. Chem.  
609 Res. 56, 13190 – 13205. <https://doi.org/10.1021/acs.iecr.7b01578>