Helmholtz-Zentrum Dresden-Rossendorf (HZDR)



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	<u>Robert Timaeus</u> ^{a, *} , Eckhard Schleicher ^a , André Bieberle ^a , Uwe Hampel ^{a,b} , Markus
4	Schubert ^{a, *}
5	
6	^a Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstraße 400,
7	01328 Dresden, Germany.
8	^b 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.

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

1 Introduction

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

for reactions with fast kinetics, which are controlled by the gas transfer (Mills and Duducović', 1980). Here, the liquid film on the solid catalysts acts as mass transfer barrier (Boelhouwer et al., 2002a). Several reactor concepts and operation strategies 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 α -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

rapidly decay along the fixed-bed (Boelhouwer et al., 2002b), which eliminatesbeneficial 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 α -methylstyrene hydrogenation and returned almost 71 doubled conversion rates compared to the conventional TBR.

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



77

Figure 1: a) IRR concept with concentric inner tube and (b) minimum and maximum liquid filling level $h_{\rm L}$, in the cross-section of the IRR indicating the beneficial process window with (c) corresponding local liquid saturation $\beta_{\rm L}$ and product concentration at the catalyst surface $C_{\rm P}$.

82

This advanced reactor design enables full utilization of the catalyst as long as the liquid filling level h_L is between lower and upper heights ($h_{L,\min} \le h_L \le h_{L,\max}$) 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 α -methylstyrene hydrogenation based on a heterogeneous continuum model revealed the potential of the advanced design (Timaeus et al., 88 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 $D_{\rm T}$ 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.

Configuration	Tube diameter	Particle diameter	Gap-to-particle-
	(mm)	(mm)	diameter ratio
А	50	4.5	5.5
В	50	2.5	10
С	30	4.5	7.8

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

116



119

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

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 $h_{\rm L}$ 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 $\beta_{\rm L}$ are considered as socalled reduced liquid saturations, i.e. dynamic liquid fractions related to the interstitial
void space of the fixed-bed reduced by the static liquid. The allocation of involved
phases in the IRR to certain fractions is exemplarily shown in Figure 3.

162



163

Figure 3: a) Phase distribution in the cross-section of the IRR with marked liquid filling level $h_{\rm L}$ and b) allocation of occurring phases in the IRR to certain fractions, which are: gas: continuous gas phase of the stratified flow pattern, static liquid: external liquid trapped between the catalyst, internal liquid: liquid inside the catalyst pores, dynamic liquid: external flowing liquid, solid: skeletal aluminum structure of the 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

$$V_{\log} = a \cdot \ln(\varepsilon_{\rm X} + \varepsilon_{\rm P}) + b, \tag{1}$$

174 where, a and b are geometry-related constants, ε_x and ε_p are the permittivities 175 accounting for fluid phases and solid packing, respectively. Recently, it was shown 176 that the permittivity $\varepsilon_{\rm P}$ of a constant solid fraction can be neglected as the deviation is 177 less than 2% for the cross-sectionally averaged $\beta_{\rm L}$ 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 $\beta_{\rm L}$ 180 and the measured permittivity ε_{Meas} is assumed (Bierberle et al., 2010; Prasser et al., 181 1998; da Silva et al., 2007) and $\beta_{\rm L}$ can be determined as

$$\beta_{\rm L} = \frac{\varepsilon_{\rm Meas} - \varepsilon_{\rm G}}{\varepsilon_{\rm L} - \varepsilon_{\rm G}},\tag{2}$$

where $\epsilon_{\rm L}$ and $\epsilon_{\rm G}$ are the permittivies of gas and liquid phases, respectively. This way, $\beta_{\rm L}$ is calculated for each crossing point exposed to the flow. Subsequently, average liquid saturation and corresponding liquid filling level $h_{\rm L}$ are determined from 7500 frames obtained for a measurement duration of 300 seconds with a frequency of 25 Hz. Further details of the capacitance WMS and the data processing are summarized in Timaeus et al. (2019b).

188

189 2.3 Porosity of the annular packed bed

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

$$\varepsilon(r) = \begin{cases} \varepsilon_{\rm b} \left(1 + C \exp\left(-N\frac{r - R_{\rm T}}{d_{\rm P}}\right) \right) & \text{for } R_{\rm T} \le r \le \frac{R_{\rm T} + R_{\rm O}}{2}, \\ \varepsilon_{\rm b} \left(1 + C \exp\left(-N\frac{R_{\rm O} - r}{d_{\rm P}}\right) \right) & \text{for } \frac{R_{\rm T} + R_{\rm O}}{2} \le r \le R_{\rm O}, \end{cases}$$
(3)

where *r* is the radial coordinate, $R_{\rm T}$ is the inner tube radius, R_0 is the reactor radius, $\epsilon_{\rm b}$ is the bulk porosity of the packed bed and *C* and *N* are fitting parameters. According to (Vortmeyer and Schuster, 1983; Cheng and Hsu, 1986; Hunt and Tien, 1990), *C* is used to fulfill

$$\varepsilon(r = R_{\rm T}) = \varepsilon(r = R_0) = 1. \tag{4}$$

Furthermore, *N* describes the slope of the porosity in the vicinity of the walls and a wide range for *N* is reported in the literature (du Toit, 2008; Vortmeyer and Schuster, 1983; Cheng and Hsu 1986). For example, Cheng and Hsu (1986) applied N = 2, 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 ¹³⁷Cs 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 $A_{\rm CT}$ of Configuration A is shown in Figure 4a.

213



214

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

219

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

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

where, ρ is the fitted density and m_{bed} is the mass of the annular packed bed. The porosity of the packed bed is defined as

$$\varepsilon(r) = \frac{\rho_{\rm P} - \rho_{\rm bed}(r)}{\rho_{\rm P} - \rho_{\rm G}},\tag{6}$$

where ρ_{P} , ρ_{bed} and ρ_{G} are the densities of particles, packed bed and gaseous phase. Eventually, the bulk porosity ε_{b} is obtained from the radial porosity profile, which enables the determination of the parameter *N* by a least square fit. In Figure 5a the porosity distribution of Configuration A obtained by the procedure described above is exemplarily shown.

233





Figure 5: a) Porosity profile of Configuration A ($D_{\Gamma} = 5 \text{ cm}$, $d_{P} = 4.5 \text{ mm}$) obtained tomographically and predicted with Eq. 3 for various *N* proposed in the literature and b) deviations between measured and predicted average porosities.

238

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

Configuration	\mathcal{E}_{ave} (mv ³ mr ⁻³)	\mathcal{E}_{b} (mv ³ mr ⁻³)	С(-)	N(-)
А	0.392	0.362	1.76	8.02
В	0.375	0.352	1.84	5.99
С	0.380	0.359	1.78	8.37

Table 2: Porosity data and determined parameters of the exponential approximationsbased on gamma-ray CT scans.

249 **3 Results**

The hydrodynamic characterization of the new IRR design comprises phase 250 251 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.



Figure 6: Cross-sectional saturation distribution for Configuration A ($D_{\rm T} = 5$ cm, $d_{\rm P} = 4.5$ mm) at $u_{\rm L} = 0.01$ m s⁻¹, $u_{\rm G} = 0.05$ m s⁻¹ and different rotational velocities *n* and inclination angles α . Gravity vector \vec{g} and gas-liquid interface of the stratified flow patterns (dashed line) are also shown.

265

The liquid filling level $h_{\rm L}$, i.e. height of the gas-liquid interface position for the stratified flow patterns, is determined according to the procedure explained in Section 2.2 (highlighted with a horizontal dashed red line). The gravity vector indicates the bottommost position of the reactor cross-section. 270 At low inclination angles ($\alpha = 15^{\circ}$ and 30°) 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° to 75° at a rotational velocity of 1 rpm for a superficial gas 281 velocity of 0.05 m s⁻¹. The beneficial hydrodynamic process window is defined by 282 inner tube and reactor diameters (D_{Γ} 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





Figure 7: a) Normalized liquid filling level $H_{\rm L}$ and (b) specific pressure drop $\Delta p L^{-1}$ depending on superficial liquid velocity $u_{\rm L}$ and inclination angle α (n = 1 rpm, 290 291 $u_{\rm G} = 0.05 \text{ m s}^{-1}$, $D_{\rm T} = 5 \text{ cm}$, $d_{\rm P} = 4.5 \text{ mm}$). The shaded area indicates the

hydrodynamic process window for the IRR (same notation is used in the figuresbelow).

294

Logically, $H_{\rm L}$ increases with increasing superficial liquid velocity regardless of the inclination angle. The same trend is obtained for the specific pressure drop. Here, higher solid-fluid phase interactions occur at higher superficial liquid velocities due to less accessible cross-section per fluid phase. Considering the stratified flow patterns, the specific pressure drop is most sensitive to the liquid load at inclination angles of 75° , where $H_{\rm L}$ reaches the highest values.

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

For the highest superficial liquid velocity of 0.02 m s^{-1} at an inclination angle of 75° *H*_L exceeds the inner tube. Thus, parts of the catalyst packing are excluded from the advantageous wetting intermittency and rotate permanently within the liquid phase 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 $H_{\rm L}$ can be adjusted to $H_{\rm L,min}$ 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 m s⁻¹ 0.05 m s⁻¹, respectively, for Configuration A.

327



328

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

332

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

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

341 At a rotational velocity of 0.4 rpm and an inclination angle of 30° $H_{\rm L}$ is below the inner 342 tube, thus, parts of the catalyst packing are excluded from the wetting intermittency and rotate permanently within the gas phase without access to the liquid bulk phase.Such operating conditions cause severe liquid educt depletion.

345

346 3.3 Effect of superficial gas velocity

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

351



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

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

367



368

Figure 10: Effect of the superficial gas velocity on the gas-liquid distribution $(D_{\rm T} = 5 \text{ cm}, d_{\rm P} = 4.5 \text{ mm}, n = 1 \text{ rpm}, u_{\rm L} = 0.01 \text{ m s}^{-1}).$

371

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



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

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.

Furthermore, the cross-sectional gas-liquid patterns reveal a smaller process window for Configuration B. A significant entrainment of liquid is already observed at an inclination angle of $\alpha = 60^{\circ}$ and a rotational velocity of n = 10 rpm for the small particle packing, while distinct stratification is obtained at the same operating 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 higher405 fluid-solid interactions caused by the smaller particles as explained above.

406



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 ($D_{\Gamma} = 5$ cm, n = 10 rpm, $u_L = 0.01$ m s⁻¹, $u_G = 0.05$ m s⁻¹).

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 $D_{\rm T}$ narrows the 414 hydrodynamic process window. Nevertheless, for reactions or process conditions 415 with better performance at higher $H_{\rm L}$ (0.35 < $H_{\rm L}$ <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.



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

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

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

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

462	Symbols us	ed
-----	------------	----

463	а	[-]	geometry-related constant 1
464	Α	[m]	normalized attenuation coefficient
465	b	[-]	geometry-related constant 2
466	С	[-]	fitting parameter 1, porosity correlation
467	$d_{\mathbb{P}}$	[m]	particle diameter
468	D	[m]	diameter
469	∆e	[-]	deviation between experiment and correlation

470	g	[m s ⁻²]	gravitational accelaration
471	h	[m]	filling level
472	Η	[-]	normalized filling level
473	L	[m]	axial length of reactor
474	n	[rpm]	rotational velocity
475	Ν	[-]	fitting parameter 2, porosity correlation
476	p	[Pa]	pressure
477	r	[m]	radial coordinate
478	R	[m]	radius
479	u	[m s ⁻¹]	superficial velocity
480	V	[V]	voltage
481	Ζ	[m]	axial coordinate
482			
483	Greek	symbols	
484	α	[°]	inclination angle towards the gravity
485	β	[m ³ m ⁻³]	reduced saturation
486	$\Delta p L^{-1}$	[kPa m ⁻¹]	specific pressure drop
487	ρ	[kg m ⁻³]	density
488	ε	[-]	relative permittivity
489	ε	$[m_V{}^3 m_R{}^{-3}]$	porosity
490	Φ	[°]	azimuthal coordinate

492 Sub- and Superscripts

493	а	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	0	outer tube
503	Р	particle
504	R	reactor
505	т	inner tube
506	V	void
507	х	fluid contributors
508		
509	Abbreviations	
510	СТ	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 Banchero, 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

Bieberle, A., Schubert, M., da Silva, M. J., Hampel, U., 2010, Measurement of liquid
distributions in particle packings using wire-Mesh sensor versus transmission
tomographic imaging, Ind. Chem. Res. 49, 9445 – 9453.
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

- Boelhouwer, J. G., Piepers, H. W., Drinkenburg, A. A. H., 2002b, Liquid-induced
 pulsing flow in trickle-bed reactors, Chem. Eng. Sci. 57, 3387 3399.
 https://doi.org/10.1016/S0009-2509(02)00210-5
- Castellari, A. T., Haure, P. M., 1995, Experimental study of the periodic operation of a
 trickle-bed reactor, AIChE 41, 1593 1597.
 https://doi.org/10.1002/aic.690410624
- Cheng, P., Hsu, C. T., 1986, Fully-developed, forced convective flow through an
 annular packed-sphere bed with wall effects, Int. J. Heat Mass Tran. 29, 1843 –
 1853. https://doi.org/10.1016/0017-9310(86)90003-7
- 537 Dietrich, W., Grünewald, 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
- Edouard, D., Lacroix, M., Pham, C., Mbodji, M., Pham-Huu, C., 2008, Experimental
 measurements and multiphase flow models in solid SiC foam beds, AIChE 54,
 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

- Haase, S., Murzin, D. Y., Salmi, T., 2016, Review on hydrodynamics and mass
 transfer in minichannel wall reactors with gas–liquid Taylor flow, Chem. Eng.
 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
- Hunt, M. L., Tien, C. L., 1990, Non-Darcian flow, heat and mass transfer in catalytic
 packed-bed reactors, Chem. Eng. Sci. 45, 55 63. https://doi.org/10.1016/00092509(90)87080-C
- Hunt, M. L., Tien, C. L., 1988, Non-darcian convection in cylindrical packed beds, J.
 Heat Transfer 110, 378 384. https://doi.org/10.1115/1.3250495
- Khadlikar, M. R., Al-Dahhan, M. H., Duduković, M. P., 1999, Parametric study of
 unsteady-state flow modulation in trickle-bed reactors, Chem. Eng. Sci. 54, 2585
 2595. https://doi.org/10.1016/S0009-2509(98)00426-6
- Lange, R., Hanika, J., Stradiotto, D., Hudgins, R. R., Silveston, P. L., 1994,
 Investigations of periodically operated trickle-bed reactors, Chem. Eng. Sci. 49,
 563 5615 5621. https://doi.org/10.1016/0009-2509(94)00363-7
- Mills, P. L., Duducović, M. P., 1980, Analysis of catalyst effectiveness in trickle-bed
 reactors processing volatile or nonvolatile reactants, Chem. Eng. Sci. 35, 2267 –
 2279. 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
- Nijhuis, T. A., Kreutzer, M. T., Romijn, A. C. J., Kapteijn, F., Moulijn, J. A., 2001,
 Monolithic catalysts as efficient three-phase reactors, Chem. Eng. Sci. 56, 823 –
 829. 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

- 575 Schubert, M., 2018, DE 10 2018 110 091.4.
- da Silva, M. J., Schleicher, E., Hampel, U., 2007, Capacitance wire-mesh sensor for
 fast measurement of phase fraction distributions, Meas. Sci. Technol. 18, 2245 –
 2251. https://doi.org/10.1088/0957-0233/18/7/059
- Sodré, J. R., Parise, J. A. R., 1998, Fluid flow pressure drop through an annular
 packed bed of spheres with wall effects, Exp. Therm. Fluid Sci. 17, 265 275.
 https://doi.org/10.1016/S0894-1777(97)10022-X
- Timaeus, R., Hampel, U., Schubert, M., 2019a, Heterogeneous modeling approach
 for gas-limited reactions in an inclined rotating fixed bed reactor, Chem. Ing.
 Tech. 91, 637 644. https://doi.org/10.1002/cite.201800183
- Timaeus, R., Berger, R., Schleicher, E., Hampel, U., Schubert, M., 2019b, Liquid flow
 visualization in packed-bed multiphase reactors: wire-mesh sensor design and
 data analysis for rotating fixed beds, Chem. Ing. Tech. 91, 1 11.
 https://doi.org/10.1002/cite.201900117
- Timaeus, R., Schleicher, E., Bieberle, A., Hampel, U., Schubert, M., 2019c,
 Hydrodynamic data of an advanced inclined rotating fixed-bed reactor, RODARE
 repository. https://doi.org/10.14278/rodare.204
- du Toit, C. G., 2008, Radial variation in porosity in annular packed beds, Nucl. Eng.
 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
- Wallenstein, M., Kind, M., Dietrich, B., 2014, Radial two-phase thermal conductivity
 and wall heat transfer coefficient, Int. J. Heat Mass Transf. 79, 486 495,
 http://dx.doi.org/10.1016/j.ijheatmasstransfer.2014.08.003

- Zalucky, J., Wagner, M., Schubert, M., Lange, R., Hampel, U., 2017a,
 Hydrodynamics of descending gas-liquid flows in solid foams: Liquid holdup,
 multiphase pressure drop and radial dispersion, Chem. Eng. Sci. 168, 480 494.
 http://dx.doi.org/10.1016/j.ces.2017.05.011
- 2017 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