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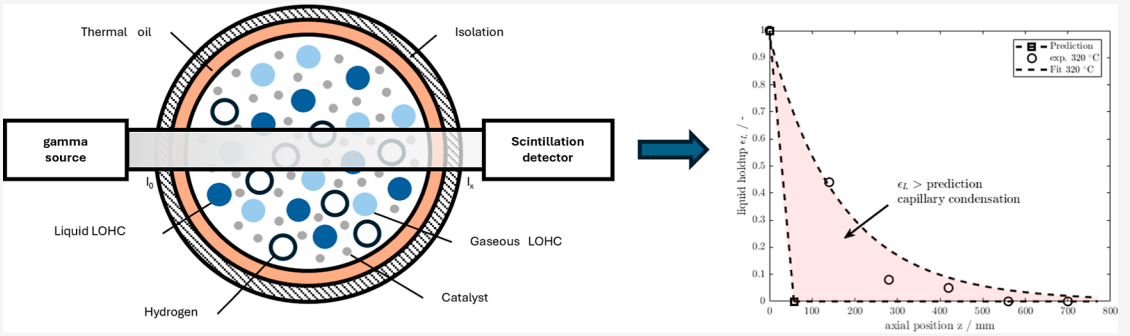
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Experimental Investigation of the Axial Liquid Holdup Profile in a Liquid Organic Hydrogen Carrier Dehydrogenation Reactor Using Densitometry

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ABSTRACT: In the dehydrogenation of perhydro benzyltoluene, one of the most promising liquid organic hydrogen carrier (LOHC) molecules, in a fixed-bed reactor, hydrogen release induces pronounced volumetric expansion and progressive evaporation of the carrier, leading to a transition from a liquid−solid to a gas−liquid−solid and finally to a gas−solid reaction system. Since the axial liquid holdup profile governs both heat transfer and catalyst stability, quantitative knowledge of the spatial distribution under reactive conditions is essential for process optimization and predictive reactor modeling. However, such data are not available for technically relevant LOHC systems. In the present study, the axially resolved liquid holdup is determined in operando in a fixed-bed LOHC dehydrogenation reactor over a wide range of reaction conditions using a non-destructive densitometric measurement technique. While a pronounced axial decrease in liquid holdup with increasing shell temperature was observed, all experimentally determined liquid holdup values significantly exceeded predictions based on bulk vapor−liquid equilibrium. This discrepancy is attributed to the increased capillary pressure in the pores resulting in condensation of LOHC within the pores. Based on the acquired experimental data, a first empirical correlation for the radially averaged local liquid holdup is proposed, thus providing a first quantitative basis for incorporating realistic liquid holdup profiles into multiphase LOHC reactor models.

1. INTRODUCTION

The liquid organic hydrogen carrier (LOHC) technology offers a promising solution for large-scale, long-term storage and transportation of hydrogen under ambient conditions.[1,2] Herein, hydrogen is chemically bound in a heterogeneously catalyzed reaction to a hydrogen-lean carrier compound in an exothermic hydrogenation reaction. On hydrogen demand, hydrogen is released later or at another location through an endothermic dehydrogenation reaction. An ideal LOHC system is nontoxic and widely available, provides high volumetric and gravimetric hydrogen storage density, and remains liquid in its hydrogen-rich and hydrogen-lean form, enabling the utilization of the existing fuel infrastructure for storage and transport.

The benzyltoluene/perhydro benzyltoluene (H0-BT/H12-BT) LOHC system is a well-known candidate that has gained particular interest due to its favorable thermophysical and storage properties.[3,4] With a boiling point of approximately 280 °C (H0-BT) and 270 °C (H12-BT) at atmospheric pressure, this LOHC system remains liquid across a wide operational window with low viscosity. A gravimetric hydrogen storage density of 6.2 wt % ensures sufficient hydrogen capacity for many interesting technical use cases.[5]

Typically, the dehydrogenation of H12-BT is carried out in fixed-bed reactors that are operated in cocurrent upflow mode.[6−8] Typical dehydrogenation conditions are at temperatures between 290 and 340 °C and pressures ranging from 3

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bar_a to 5 bar_a.[6,7,9] Often the pressure is chosen above the vapor pressure of H12-BT to ensure liquid feeding of H12-BT into the dehydrogenation reactor. A defining characteristic of H12-BT dehydrogenation is the extremely large volumetric expansion associated with hydrogen release. Up to 670 mL of hydrogen can be released per mL of H12-BT at ambient conditions. Due to this massive volume change in the reactor, the released hydrogen significantly alters the axial gas−liquid ratio, which leads to a significant change of the hydrodynamics in the dehydrogenation reactor. The release of hydrogen decreases the partial pressure of all benzyltoluene species (H*x*-BT) and results in evaporation of H*x*-BT. Complete evaporation of H*x*-BT is predicted by vapor−liquid-equilibrium (VLE) estimations under bulk conditions for all technically relevant operating conditions.[8,10]

Consequently, the hydrodynamics in the dehydrogenation reactor shift from a two-phase reaction system consisting of the liquid LOHC mixture and solid catalyst to a multiphasic system with a solid catalyst, liquid LOHC, evaporated LOHC, and gaseous hydrogen to, finally, a gas−solid system. This transition has a significant impact on catalyst stability, activity, and on the heat transfer from the reactor walls into the catalyst bed.[10,11] The presence of liquid H*x*-BT enables removal of coke precursors by washing them from the catalyst surface, preventing deactivation by catalyst coking.[12] In contrast, in the gas−solid reaction system, evaporation of coke precursors is difficult, which leads to an accelerated catalyst deactivation.[11,13] In addition to influencing the catalyst lifetime, the presence of a liquid phase enhances heat transfer from the reactor walls into the endothermic dehydrogenation zone. In packed-bed LOHC reactors, heat transfer is predominantly controlled by the prevailing phase distribution, and thus by the local liquid holdup.[6] Close to the reactor inlet, where liquid LOHC and hydrogen dominate, heat transfer coefficients of up to 1500 W m^{-2} K^{-1} are achieved. As hydrogen is released and the LOHC compounds evaporate along the reactor axis, the heat transfer coefficient decreases by almost an order of magnitude, i.e., reaching values of around 150 W m^{-2} K^{-1}.[6,8] This results in a pronounced axial heat transfer gradient, directly coupling the hydrodynamics and heat transfer. A schematic representation of the evolving hydrodynamic regimes in a H12-BT dehydrogenation reactor is shown in Figure 1.

A number of alternative reactor concepts have been proposed to enhance the liquid holdup within the dehydrogenation reactor, including reactive distillation,[14−16] coated wall reactors,[17−19] and membrane reactors.[20,21] However, scaling of these reactor concepts is challenging and relies on experimental data of prototypes. Consequently, the fixed-bed reactor remains the most viable and robust option for the short-term commercial roll-out of LOHC technology.

According to the VLE, evaporation mainly depends on pressure, temperature, and the ratio of hydrogen to H*x*-BT. Kadar et al.[8] developed an equation to determine the degree of evaporation assuming that H*x*-BT can be described as a pseudocomponent and no hydrogen is dissolved in liquid H*x*-BT (see eq 1). The Degree of Dehydrogenation (DoDH) is defined as the ratio of released hydrogen to the maximum releasable amount of H_2 and can be used to describe the ratio of hydrogen to H*x*-BT in the reactor.

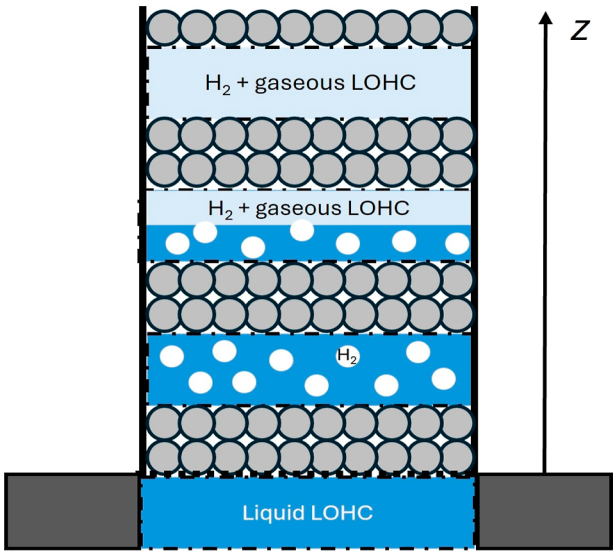


Figure 1. Visual representation of different hydrodynamic regimes along the reaction length axis. Liquid LOHC compounds (dark blue) enter the reaction zone. In contact with the catalyst (gray), hydrogen (white) is released from the LOHC system forming a two-phase flow. Further LOHC evaporation ultimately results in pure gas-phase dehydrogenation (light blue).

$$\text{Degree of evaporation} = \frac{\text{DoDH} \times 6 \times \frac{P^{LV}_{0,\text{Hx-BT}}}{P}}{1 - \frac{P^{LV}_{0,\text{Hx-BT}}}{P}} \quad (1)$$

In Figure 2, the estimated degree of evaporation according to eq 1 is shown. For the relevant catalyst temperatures,

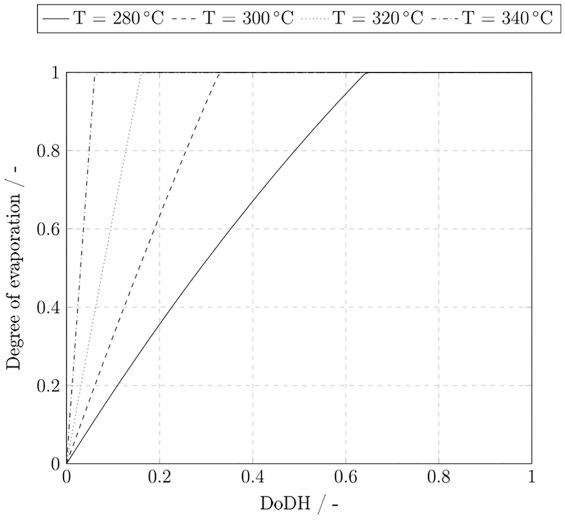


Figure 2. Estimated degree of evaporation as a function of the DoDH according to eq 1 for constant temperatures between 280 and 340 °C at a 5 bar_a pressure.

complete evaporation is expected from theory. It must be stressed, however, that this equilibrium-based estimation provides no information about the actual liquid holdup within the packed bed. In particular, the gas and liquid flows have different velocities, and thus a degree of evaporation of 0.5 does not correspond to a liquid holdup of 0.5. Consequently, quantitative insight into the axial liquid distribution cannot be derived from equilibrium considerations alone, highlighting the

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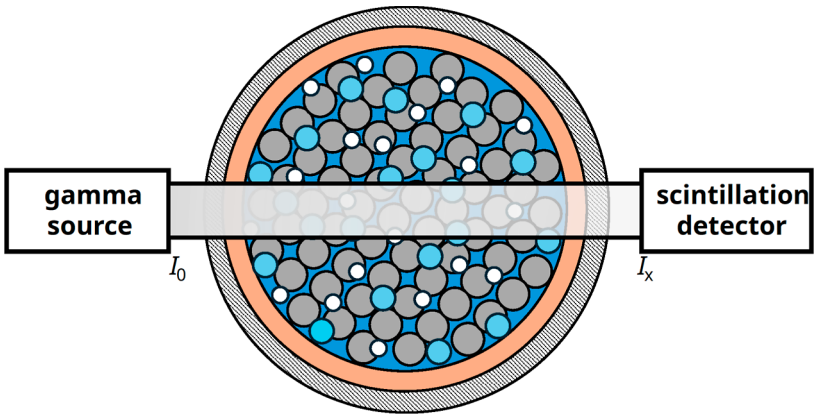


Figure 3. Top view of the densitometric measurement system and the dehydrogenation reactor sketch. The isotopic source emits gamma photons with intensity I_0 and a scintillation detector behind the reactor measures the attenuated radiation intensity I_x as the gamma photons have to pass the constant insulation (gray), thermal oil (orange), and stainless-steel reactor walls (black). In the reaction volume an immovable catalyst packing consisting of catalyst particles (gray) are in a flow of liquid LOHC (dark blue), evaporated LOHC (light blue), and released hydrogen (white).

need for a direct, spatially resolved experimental determination of the liquid holdup under realistic reaction conditions.

There is currently no quantitative correlation that describes the axial holdup profile, despite its significant influence on the design of a dehydrogenation reactor. This is primarily due to a lack of quantitative information on the liquid−gas distribution, particularly the local liquid holdup under realistic reaction conditions. Although numerous empirical correlations to predict the liquid holdup in packed beds exist,[22−28] these are largely restricted to trickle beds and simplified model systems such as air−water or ambient conditions. Consequently, their applicability to the highly dynamic, high-temperature and coupled reactive environment in a dehydrogenation reactor is limited. So, direct experimental determination of the local liquid holdup under realistic operating conditions is of critical importance to understand, model, and improve the liquid holdup in a dehydrogenation reactor.

In systems with porous particles, the total liquid holdup $\varepsilon_{liq,tot}$ is subdivided into (a) the external liquid holdup $\varepsilon_{liq,ext}$, describing the liquid occupying the void volume, and (b) the internal liquid holdup $\varepsilon_{liq,int}$ accounting for liquid within the particle pores. The external liquid holdup is further divided into a static component $\varepsilon_{liq,stat}$ and a dynamic component $\varepsilon_{liq,dyn}$.[29]

$$\varepsilon_{liq,tot} = \varepsilon_{liq,int} + \varepsilon_{liq,ext} = \varepsilon_{liq,int} + \varepsilon_{liq,stat} + \varepsilon_{liq,dyn} \quad (2)$$

There are several approaches reported to measure liquid holdup in multiphase flow systems. Global holdup values can be determined using established draining[25,26,29−32] or weighing[27,33−37] techniques. However, these methods fail to capture the significant axial changes in liquid holdup distribution due to the release of hydrogen. Local measurement techniques, including electrical capacitance tomography,[38] X-ray tomography,[39] and gamma-ray tomography,[40−42] are generally limited by the harsh operating conditions in the dehydrogenation reactor, including temperatures above 300 °C, elevated pressures, and being restricted in spatial resolution.

In the present study, the liquid holdup in a fixed-bed LOHC dehydrogenation reactor is determined at different axial positions using gamma-radiation based densitometry. This noninvasive technique enables axially resolved liquid holdup measurements with flexible positioning and high accuracy, even under extreme thermal and chemical conditions. Although gamma-ray densitometry was previously applied for multiphase flow characterization under demanding operating conditions,[43−47] its application to a reactive LOHC system at system temperatures of up to 340 °C represents an additional challenge as temperature-related density changes are no more negligible. The present work demonstrates the applicability of this noninvasive technique for axially resolved liquid holdup measurements under these demanding LOHC dehydrogenation conditions.

In this study, the feasibility and reproducibility of the in situ determination of the axial holdup profile in the dehydrogenation of H12-BT are evaluated. First, reference measurements and determination of the standard deviation are discussed. Then, first experimental results for the axially resolved liquid holdup under realistic LOHC dehydrogenation conditions are provided. These data are used to develop a first correlation describing quantitatively the axial progress of the liquid holdup in a dehydrogenation reactor. The results obtained imply an important step toward modeling multiphasic dehydrogenation reactors and for the development of process intensification strategies.

2. MATERIAL AND METHODS

2.1. Gamma-Ray Densitometry

The liquid holdup within the LOHC reactor is determined by γ-ray densitometry, as this measurement technique can be applied as in situ analytic without any disturbance of the multiphasic flow. Densitometric measurement systems are well-established for two-phase flow investigations in technical devices and provide reliable operation.[48] However, careful data analysis is required if the measurement system is operated under fluctuating environmental conditions, e.g., temperature and pressure.

In this study, a Cs-137 isotopic source with an activity of around 20 mCi and a single γ photon energy of 662 keV is used. The radiation source is housed in a cylindrical protective container made of tungsten (diameter = 70 mm) with a 5 mm diameter opening, which focuses the radiation field into a pencil beam aligned along the cross-sectional diameter of the LOHC reactor. On the opposite side, there is a scintillation detector with an active area of 10 mm^2 and a similar beam collimation. A sketch of the measurement system and the reactor cross section is given in Figure 3, where the pencil beam penetrating the reactive system can be seen.

The radiation detector counts those γ photons passing the LOHC reactor without interaction. For this purpose, the multichannel analyzer MCA-527 (GBS electronics) is used with 1024 channel

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resolution, continuous interval measurement of 5 min, and the "Live Time" mode to compensate detection losses within a fixed measuring interval. As Cs-137 offers a single γ photon energy line at 662 keV, the obtained pulse height spectra (PHS) provide clearly the typical energy characteristics, i.e., the photo peak and the Compton edge. As γ photons deposit either their full or partial energy into the detector, γ photons passing the LOHC reactor without any interaction can securely be identified as these events are within the photopeak area.

The radiation attenuation strictly follows the exponential attenuation law

$$I_x = I_0 \times \exp(\mu \times d) \tag{3}$$

with I_x and I_0 being the measured intensity, i.e., number of registered photo peak events, at the radiation detector with and without object, respectively, as well as μ and d representing the attenuation coefficient and the length of the materials penetrated. To quantify the liquid holdup ϵ from a densitometric two-phase flow scan I_{tpf}, two additional reference scans must be performed for a pure gas−solid system I_{gas} and a pure liquid−solid system I_{liq}, respectively. Assuming that the liquid fraction along the radiation pencil beam is only influencing beam intensity, the liquid holdup

$$\epsilon = \frac{\ln(I_{gas}) - \ln(I_{tpf})}{\ln(I_{gas}) - \ln(I_{liq})} = \ln\left(\frac{I_{gas}}{I_{tpf}}\right) / \ln\left(\frac{I_{gas}}{I_{liq}}\right) \tag{4}$$

can be calculated without a blank measurement I_0. As the density of the catalyst bed, the reactor wall, and its thermal heat insulation is not changing during the experiments, eq 4 is valid. Unfortunately, this is not the case for the thermal oil and the LOHC fluid as shown in Table 1.

Table 1. Densities of the Thermal Oil and the LOHC Fluids (kg m^{-3}) at Various Temperatures[a]

	210 °C	300 °C	360 °C
liquid H0-BT[4]	850.7	781.9	736.1
liquid H12-BT[4]	740.1	681.5	639.7
gaseous H0-BT[a]	18.1	15.3	13.8
gaseous H12-BT[a]	19.3	16.3	14.8
thermal oil Marlotherm SH[49]	912.0	843.0	792.0

[a]Gaseous LOHC densities are calculated via ideal gas law at 4 bar$_a$.

Thus, both reference measurements should actually be performed at corresponding two-phase flow conditions, i.e., at the same temperature and pressure. However, liquid reference scans cannot be performed at dehydrogenation conditions, as hydrogen release has to be prevented. Thus, the existing density differences

$$K = \frac{\rho_{liq}^{(\text{thermal oil})} + \rho_{liq}^{(\text{H}x\text{-BT})}}{\rho_{tpf}^{(\text{thermal oil})} + \rho_{tpf}^{(\text{H}x\text{-BT})}} \tag{5}$$

need to be considered in eq 4 that results in

$$\epsilon = \frac{\ln(I_{gas}) - \ln(I_{tpf})}{\ln(I_{gas}) - \ln(I_{liq})} \times K \tag{6}$$

Note, compared to liquid LOHC the temperature-related absolute density changes of the gaseous LOHC are maximal at 4% and, therefore, negligible. Moreover, despite liquid intermediate products (i.e., the partially dehydrogenated benzyltoluene H6-BT) formed during dehydrogenation,[50,51] a pseudo two-component mixture of H12-BT and H0-BT is assumed for liquid density calculation of Hx-BT as the density of the reaction mixture does not differ significantly from the density of the pseudo two-component mixture, which was proven for the comparable LOHC system dibenzyltoluene/perhydro dibenzyltoluene (H0-DBT/H18-DBT).[52]

$$\rho^{(\text{H}x-\text{BT})} = \text{DoDH} \times \rho^{(\text{H0-BT})} + (1 - \text{DoDH}) \times \rho^{(\text{H12-BT})} \tag{7}$$

Another key concern for densitometric measurement systems is that environmental temperature changes need to be considered. As the LOHC reactor operates up to 340 °C and is installed in a closed housing, the temperature in the housing increases over the experiment. This leads to a signal amplification of the photomultiplier (see Figure 4). Thus, the position of the photo peak area changes in the PHS. This effect must be considered by adjusting the boundaries accordingly. Therefore, a Gaussian fitting curve of the photo peak area is automatically generated and its mean value μ is determined. Finally, the upper and lower borderlines are set to $\mu \pm 0.14\ \mu$, respectively, and the radiation intensity is calculated by summing all photo peak events.

Eventually, the statistical deviation of the isotopic source must be considered to provide reliable measurement accuracy values. Because the decay process is Poisson-distributed, the measured radiation intensity I (i.e., the count rate at the detector $\dot{N}$) differs for fixed material arrangements and scanning intervals. The deviation $\pm e_\epsilon$ of the calculated liquid holdup ϵ is calculated by

$$e_\epsilon^2 = \left(\frac{\delta\epsilon}{\delta I_{liq}} e_{I_{liq}}\right)^2 + \left(\frac{\delta\epsilon}{\delta I_{gas}} e_{I_{gas}}\right)^2 + \left(\frac{\delta\epsilon}{\delta I_{tpf}} e_{I_{tpf}}\right)^2 \tag{8}$$

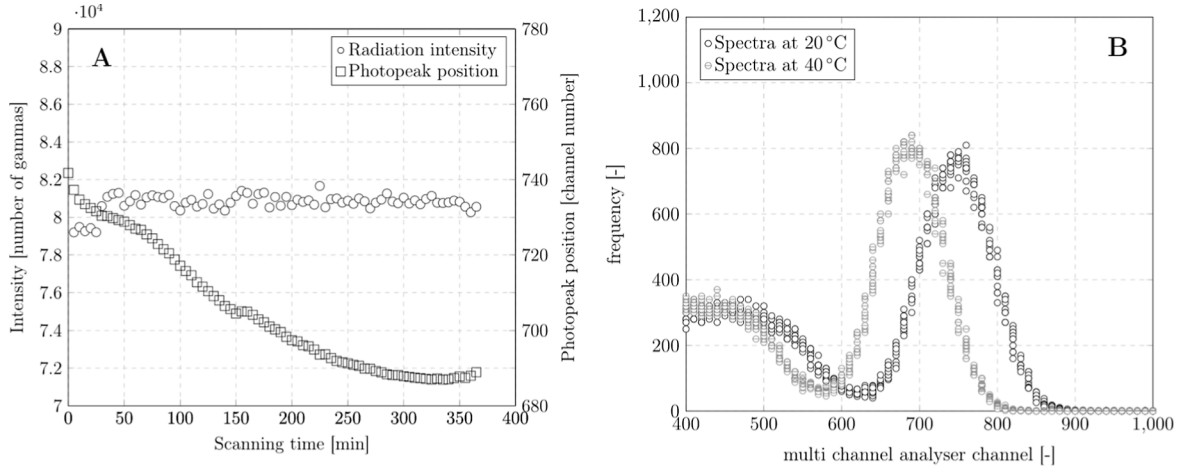


Figure 4. (A) Position of the photo peak and calculated radiation intensity over time with increasing temperature in the housing. Measured with a dry catalyst bed at 320 °C shell temperature and hydrogen flow of 2 NL min^{-1}. (B) Spectra obtained with the multichannel analyzer (MCA-527) at both extreme temperatures of the housing.

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For which eq 4 is partially derived according to all measured radiation intensities, i.e.

$$\frac{\delta\epsilon}{\delta I_{liq}} = \frac{\ln(I_{gas}) - \ln(I_{tpf})}{I_{liq} \times (\ln(I_{gas}) - \ln(I_{liq}))^2} \quad (9)$$

$$\frac{\delta\epsilon}{\delta I_{gas}} = \frac{\ln(I_{tpf}) - \ln(I_{liq})}{I_{gas} \times (\ln(I_{gas}) - \ln(I_{liq}))^2} \quad (10)$$

and

$$\frac{\delta\epsilon}{\delta I_{tpf}} = \frac{-1}{I_{tpf} \times (\ln(I_{gas}) - \ln(I_{liq}))} \quad (11)$$

and multiplied by their respective Poisson deviations $e_{I_x} = \sqrt{I_x}$. Please note, this measurement accuracy investigation is related to the isotopic sources' decay only. Practically, these values are used to ensure a better measurement accuracy than the expected deviation of the process to be investigated. To determine correct liquid holdups only measurements at steady state are selected to minimize the deviation caused by the statistic of the isotopic decay.

2.2. LOHC Dehydrogenation Reactor

The dehydrogenation setup consists of a vertically oriented tubular packed-bed reactor with an inner diameter of 27.9 mm and a catalyst bed height of 770 mm. The reactor is instrumented with five type-K thermocouples and sampling ports at defined axial positions of z = 140 mm, 280 mm, 420 mm, 560 mm, and 700 mm. In addition, three Fiber-Bragg sensors are implemented in the reaction zone, located at radial positions of 0 mm, 5 mm, and 10 mm, each with 12 Bragg gratings positioned at equidistant axial intervals of 70 mm, enabling high resolution axial and radial temperature measurements. The reactor is heated with the commercially available heat transfer oil Marlotherm SH. A comprehensive description of the reactor and instrumentation is given in Willer et al.[5] The catalyst bed consists of 347.4 g of the commercially available dehydrogenation catalyst Elemax D102 from Clariant (0.3 wt % Pt on Al_2O_3, sulfur doped). The spherical catalyst particles provide a mean diameter of 2.8 mm, resulting in a packing porosity of approximately 0.38. The specific pore volume of the catalyst used is approximately 0.47 $cm^3\ g^{-1}$, which gives a total pore volume of 163.3 cm^3. This is in the same order of magnitude as the void volume of 178.9 cm^3. H12-BT was obtained by in-house hydrogenation of Hx-BT from previous experiments.

The DoDH of all liquid samples was determined by using an Agilent 8890 gas chromatography system equipped with a capillary column (Rxi-17Sil MS column, l = 30 m, di = 250 μm) and a flame ionization detector (FID). Samples were prepared by diluting a 30 μL sample volume in 1000 μL of 2-propanol (VWR chemicals, CAS: 67-63-0, Lot: 24I094015). An injection volume of 1 μL was introduced using split injection with a split ratio of 150:1. The injector temperature was kept at 250 °C. For pure hydrocarbon-based LOHC systems, the FID response can be assumed to be directly proportional to the total amount of carbon atoms in a molecule. Consequently, a uniform response factor is applied to all Hx-BT isomers. The DoDH can then be calculated directly with the peak areas of the respective compounds A_i obtained by GC measurements according to eq 12.[51]

$$\mathrm{DoDH} = 1 - \frac{A_{H12\text{-}BT} + \frac{1}{2}A_{H6\text{-}BT}}{A_{H12\text{-}BT} + A_{H6\text{-}BT} + A_{H0\text{-}BT}} \quad (12)$$

A variation of shell temperature, reaction pressure, and liquid feed rate was investigated. The reaction pressure was set to 4 bar_a and 5 bar_a, respectively. The liquid feed rate was varied between 5 mL min^{-1} and 25 mL min^{-1}, which equals a liquid hourly space velocity (LHSV) of 0.6 h^{-1} to 3.2 h^{-1}. The shell temperature was varied between 280 and 340 °C to provide operation points with minimal, strong, and complete evaporation.

Measurements were performed sequentially on different experimental days from the lowest axial position (z = 140 mm) toward the reactor outlet. This is because the γ-ray densitometry system cannot easily be axially moved during experiments. Thus, it remains at a fixed axial position for the whole experimental day. This means that the axial holdup profile was recorded for a specific combination of reaction parameters on different experimental days. As the local temperature and the liquid density information are required for accurate data evaluation, the already existing sampling ports of the LOHC reactor were selected to determine the axial DoDH profile. To ensure reproducibility, a defined shut-down and startup protocol was used, and the released hydrogen flow and the axial temperature profile were compared for each set of reaction parameters, and no significant deviation from the previous tests could be detected. Therefore, the tests with identical reaction parameters on different experimental days were evaluated as being reproducible.

To ensure comparable hydrogen release rates and stable catalyst activity, a defined startup and shutdown protocol was applied, which is described in the following. During startup the reactor was heated to

- a shell temperature of T_{Shell} = 320 °C,
- a reactor pressure of p = 1 bar_a and
- under a constant pure hydrogen flow rate of $\dot{V}_{H2}$ = 2 $NLmin^{-1}$ and no LOHC flow.

Once steady-state conditions were achieved, a densitometric reference measurement of the dry catalyst, i.e., gas-filled, bed was performed for 15 min. Afterward, a standard operating point was set at

- a shell temperature of T_{Shell} = 320 °C,
- a reactor Pressure of p = 4 bar_a and
- with a liquid feed rate of $\dot{V}_{in}$ = 15 $mLmin^{-1}$.

This standard operating point was always operated first to ensure a consistent catalyst activity and to assess reproducibility of the measured liquid holdup. At the end of each experimental run, the reactor was cooled down using

- a shell temperature of T_{Shell} = 210 °C,
- a reactor pressure of p = 5 bar_a and
- a liquid feed rate of $\dot{V}_{in}$ = 25 $mLmin^{-1}$.

At these conditions no hydrogen is released due to thermodynamic equilibrium limitations, resulting in a catalyst bed completely filled with liquid LOHC serving as a reference experiment. In addition, this high liquid volume flow $\dot{V}_{in}$ facilitates the removal of accumulated byproducts and coke precursors from the catalyst surface, avoiding coking of the catalyst. This regeneration procedure has been successfully applied in previous studies in our group.[6] After acquiring the liquid reference data, the reactor is reheated again to

- a shell temperature of T_{Shell} = 320 °C,
- a reactor pressure of p = 5 bar_a and
- with a constant pure hydrogen flow rate of $\dot{V}_{H2}$ = 5 $NLmin^{-1}$ and no LOHC flow for at least 30 min.

This drying procedure ensures that the catalyst bed is not nucleation-inhibited on the next experimental day.[53,54] Once the catalyst bed is completely dried, a repeated reference measurement of the dry catalyst bed is carried out. The cross-check of this last measurement with the first measurement at the beginning of the day acts as proof that all liquid Hx-BT has been removed from the catalyst bed and that the catalyst bed or any other reactor components have not changed over the day. Following this startup and shutdown protocol, no significant catalyst deactivation could be detected. The average released hydrogen flow rate at standard operating conditions was 5.7 NL min^{-1} with a standard deviation of less than 4%.

3. RESULTS

3.1. Influence of External Temperature on the Densitometric Measurement System

Because the entire dehydrogenation setup is enclosed within a housing to ensure safe operation, waste heat from the dehydrogenation setup leads to a temperature increase of up

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to 45 °C within the housing. To assess whether this gradual temperature increase affects the accuracy and stability of the densitometric measurements, the detector signal was monitored over several hours, i.e., from the starting sequence at ambient temperature of 20 °C followed by the startup of the reactor resulting in an increasing temperature of the housing. Therefore, gamma photon spectra were seamlessly acquired in intervals of 5 min and the aforementioned data processing was applied to dynamically select the photo peak area. Note that the selected measuring interval length means both (a) sufficient statistics to securely detect the photo peak area and (b) a justifiable time for scanning the reactor under steady-state conditions.

Figure 4A shows the extracted photo peak positions as well as the corresponding radiation intensities over time. For constant process conditions, a systematic shift of the photo peak position is observed corresponding with the external housing temperature increase. For confirmation, spectra at 20 and 40 °C are shown in Figure 4B. Importantly, despite the shift of 25 channel positions per 10 K, the integrated radiation intensity remains constant over time applying our data processing (see Figure 4A for details). In detail, the determined standard deviation of the measured radiation intensity was below 0.3% and, thus, within the statistical deviation of the isotopic source.

The intensity changes during the first 25 min of the experiment, i.e., during the heating phase of the initially cold catalyst bed to the desired shell temperature of 320 °C, are not related to detector or housing temperature effects. It is much more attributed to the density decrease of the thermal oil surrounding the reactor during heat-up. This observation directly confirms the necessity of density corrections according to eqs 5 and 6. Overall, these results demonstrate that the gamma-ray densitometric measurement system exhibits a high degree of robustness with respect to environmental temperature variations. As the determination of liquid holdup relies exclusively on the integrated photo peak area rather than its channel position, the observed peak shift has no impact on the estimation of the liquid holdup.

3.2. Reproducibility of the Densitometric Measurement System

Since the isotope source had to be stored securely every night, new reference measurements had to be taken every day. Figure 5 shows the calculated liquid holdup for a representative experimental day. First, the attenuated intensity of the dry catalyst bed I_{gas} was measured as a reference resulting in a liquid holdup of 0 according to eq 6 (red circle symbols). The dry catalyst reference was repeated at the end of the day as a cross-check to the first dry reference measurement.

Furthermore, the attenuated intensity of a flooded catalyst bed I_{liq} (green diamond symbols) was measured at the end of the experimental sequence (violet square symbols) and resulted in a liquid holdup of 1 according to eq 6. By using the measured intensity of both reference measurements, i.e., I_{gas} at the beginning and I_{liq} at the end of the sequence, the maximal radiation attenuation E_{liq}

$$E_{liq} = \mu_{liq} \times d_{liq} = \ln(I_{gas}/I_{liq}) \tag{13}$$

is determined. The maximum attenuation of radiation was the same at every measurement position, which demonstrates satisfactory reproducibility of the references. Moreover, the comparison of the estimated liquid holdup of the dry catalyst

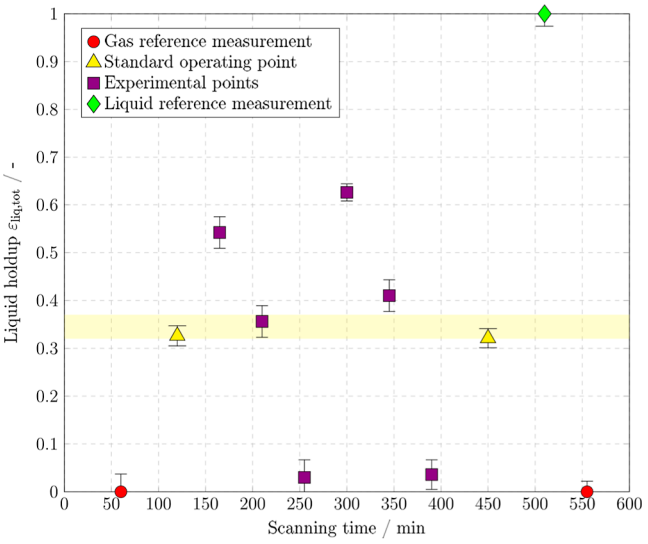


Figure 5. Applied measuring sequence including two standard operating points (yellow triangles), dry reference measurements (red circles), liquid reference measurement (green diamond), and multiphasic experimental points (violet squares). (Experimental conditions for violet squares: T_{Shell} = 300 °C, p_{rea} = 4 bar_a to 5 bar_a, $\dot{V}_{in}$ = 5 $mLmin^{-1}$ to 25 mL min^{-1}, 347.4 g of catalyst (0.3 wt % Pt/Al_2O_3)). The yellow band indicates the absolute deviation of the liquid holdup at standard operating conditions. (Experimental conditions for yellow triangles: T_{Shell} = 320 °C, p_{rea} = 4 bar_a to 5 bar_a, $\dot{V}_{in}$ = 5 $mLmin^{-1}$ to 25 mL min^{-1}, 347.4 g of catalyst (0.3 wt % Pt/Al_2O_3)).

bed before and after each experiment day shows that the catalyst bed did not undergo any flow-induced changes during the experiment.

A reference experiment (yellow triangle symbols) was performed at the beginning of each experimental day to detect a potential deactivation of the catalyst. Due to the strict shutdown and start-up protocol, no significant catalyst deactivation was detected during experiments. This reference experiment is also used to estimate the reproducibility of liquid holdup determination as this operating point was run daily. The average liquid holdup of the reference experiment was 0.345 with a standard deviation of 7.2%. The standard deviation corresponds to a uncertainty of ±0.025 in holdup determination. The standard operating point was daily maintained for 60 min, to quantify the temporal stability of the measured liquid holdup. An absolute deviation below 0.04 was determined. Together with the stable hydrogen production rate, neither catalyst deactivation nor measurement drift influenced the determined holdup profiles.

In addition, the reference experiment was conducted on one experimental day prior to the reactor shutdown and demonstrated a high degree of reproducibility for the liquid holdup determination on the same experimental day.

To conclude, the reproducibility tests show that γ-ray densitometry is suitable to estimate the liquid holdup accurately and noninvasively under the demanding conditions of H12-BT dehydrogenation.

3.3. Experimental Determination of the Axial Liquid Holdup Profile

In this section the influence of the catalyst temperature and the DoDH on the liquid holdup is analyzed. The presented results provide the first spatially resolved experimental insight into the

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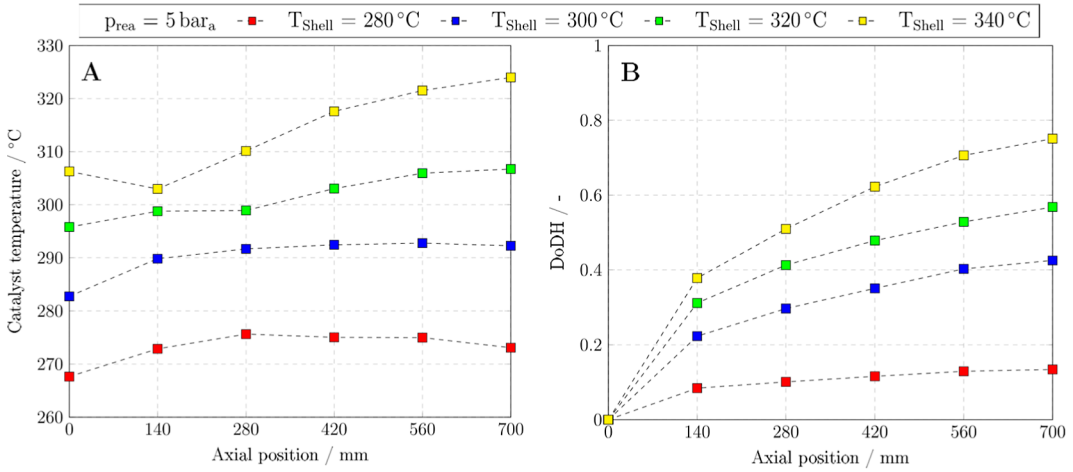


Figure 6. (A) Mean catalyst temperatures at different shell temperatures for (p_{rea} = 5 bar_a, $\dot{V}_{in}$ = 15 $mLmin^{-1}$, 347.4 g of catalyst (0.3 wt % Pt/Al_2O_3)); (B) measured DoDH at different shell temperatures (p_{rea} = 5 bar_a, $\dot{V}_{in}$ = 15 $mLmin^{-1}$, 347.4 g of catalyst (0.3 wt % Pt/Al_2O_3)).

axial liquid holdup profile of a multiphasic fixed-bed LOHC dehydrogenation reactor under reactive conditions. The discussion focuses on experiments conducted at a reaction pressure of 5 bar_a and a liquid feed rate of 15 mL min^{-1}, corresponding to a LHSV of 1.3 h^{-1}. The results for all other operating conditions are available via zenodo.[55]

Figure 6A shows the axial catalyst temperature profile for varying shell temperatures. The catalyst temperature is the relevant parameter for the estimation of the liquid holdup and the LOHC evaporation. The catalyst temperature increases as expected with higher shell temperature. However, an increase of 20 °C in shell temperature does not correspond to an increase of 20 °C of catalyst temperature as the hydrogen release rate also increases and this consumes more heat. The axially decreasing hydrogen release rate leads also to an axial increase in catalyst temperature as less heat is consumed by the dehydrogenation with increasing reactor height. Furthermore, the ratio of hydrogen to H*x*-BT should influence the liquid holdup and is thus shown in Figure 6B as the axial DoDH profile. The DoDH increases continuously along the reactor axis, reflecting progressive hydrogen release. The observed profiles for temperature and DoDH are consistent with the findings of Willer et al.,[6] confirming the reproducibility of the reaction behavior under the applied operating conditions. Only a slight increase in the axial DoDH is observed above 140 mm for a shell temperature of 280 °C due to the slow kinetics at this low temperature.

Figure 7 shows the determined axial liquid holdup profile. Increasing the catalyst temperature enhances the intrinsic reaction rate, resulting in a higher DoDH with respect to a higher ratio of hydrogen to H*x*-BT. Consequently, this leads to a lower liquid holdup as more hydrogen gas is in the system. Additionally, the higher hydrogen release decreases the H*x*-BT partial pressure triggering H*x*-BT evaporation, which reduces the amount of liquid H*x*-BT intensifying the reduction in liquid holdup. The liquid holdup decreases steeply for a shell temperature of 320 and 340 °C, resulting in almost complete evaporation at an axial position of 280 mm. Remarkably, the axial holdup profile is fundamentally different for a shell temperature below 300 °C as at such lower temperatures the axial decrease of the liquid holdup is significantly less pronounced. At lower shell temperatures, the axial increase in catalyst temperature and DoDH is less pronounced,

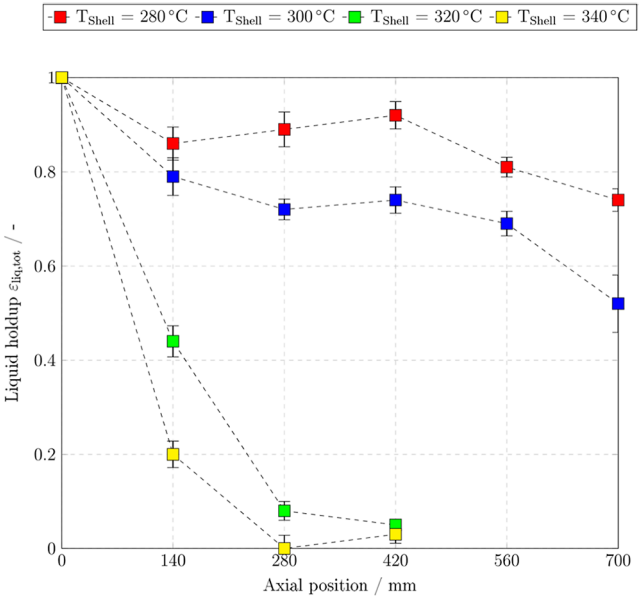


Figure 7. Dependency of the axial liquid holdup profile on shell temperature. As a liquid holdup of almost 0 was determined at an axial position of 420 mm for a shell temperature of 320 and 340 °C, the determination at 560 and 700 mm was omitted because no significant liquid holdup can be expected under these conditions (p_{rea} = 5 bar_a, $\dot{V}_{in}$ = 15 $mLmin^{-1}$, 347.4 g of catalyst (0.3 wt % Pt/Al_2O_3)).

resulting in weaker axial gradients in hydrogen generation and H*x*-BT evaporation. Consequently, the resulting change in gas and liquid superficial velocities is reduced, which leads to a smaller decrease in liquid holdup.

Complete evaporation of H*x*-BT results in a liquid holdup of 0. The approximate axial position of complete H*x*-BT evaporation can be estimated using eq 1. Complete H*x*-BT evaporation is predicted, e.g., for a catalyst temperature of 303 °C and a DoDH of 0.38. These values are predicted for a shell temperature of 340 °C at an axial position of 140 mm. However, experimental results show a liquid holdup of around 0.2, indicating still a significant amount of liquid at this axial position. Similar effects can also be observed at other conditions and reactor positions. A schematic representation

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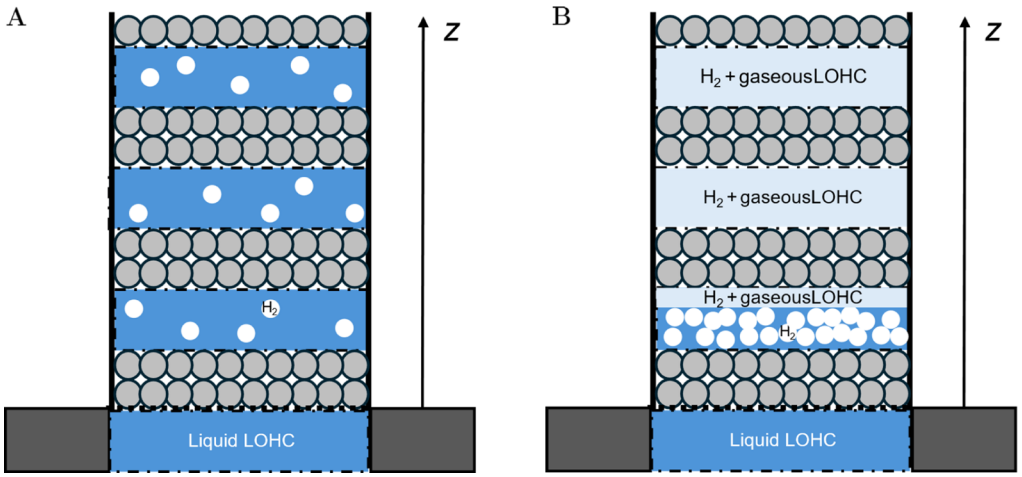


Figure 8. Visual representation of different hydrodynamic regimes along the reaction length axis at different shell temperatures. (A) Shell temperature of 280 °C. Liquid LOHC compounds (dark blue) enter the reaction zone. In contact with the catalyst (gray), small amounts of hydrogen (white) are released from the LOHC system forming a two-phase flow. Due to slow hydrogen release and comparably low catalyst temperatures, no LOHC evaporation takes place. (B) Shell temperature of 340 °C. Liquid LOHC compounds (dark blue) enter the reaction zone. In contact with the catalyst (gray), hydrogen (white) is released rapidly from the LOHC system forming a two-phase flow. Due to fast hydrogen release and comparably high catalyst temperatures, LOHC evaporation is fast, leading to complete evaporation after a short path length.

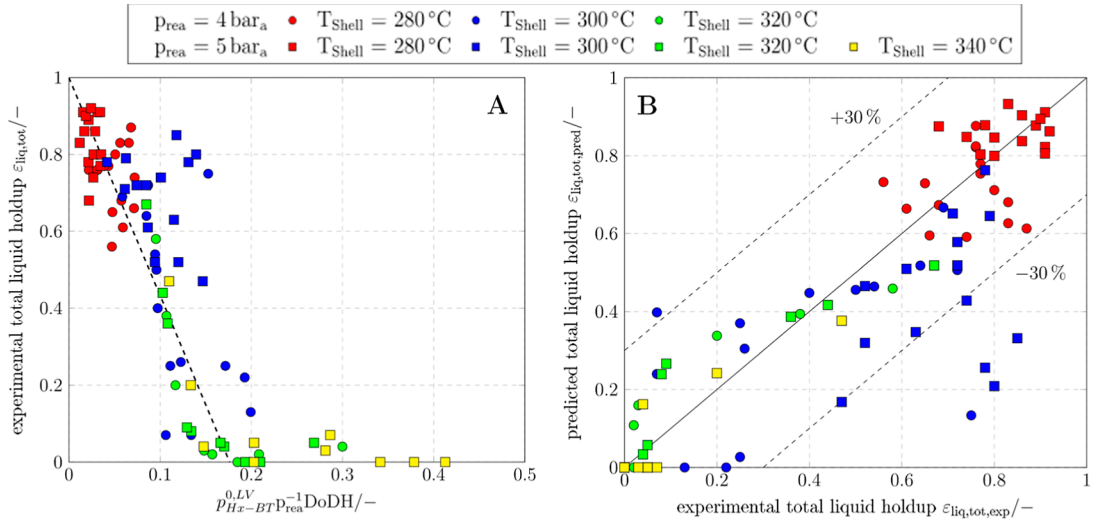


Figure 9. (A) Dependency of experimental liquid holdup on lumped parameter x; (B) Parity Plot comparing the experimental liquid holdup with the predicted liquid holdup estimated by the proposed correlation. Dashed lines indicate an absolute deviation of ±30%. Range of validity: T_{Shell} = 280 to 340 °C, p_{rea} = 4 bar_a to 5 bar_a, · $\dot{V}_{in}$ = 5 mLmin^{-1} to 25 mL min^{-1}.

of the hydrodynamic regimes at the extreme conditions of the shell temperatures 280 and 340 °C is given in Figure 8.

A plausible explanation for this effect is the LOHC condensation inside the catalyst pores. The pressure in the pores exceeds the pressure in the bulk due to capillary effects, preventing complete evaporation of Hx-BT in the catalyst pores. Please note that the interparticle volume of the used catalytic fixed bed (178.9 cm^3) is approximately equal to the catalyst pore volume (163.3 cm^3). However, since the radiation beam penetrates both porous particles and the interparticle void space, it is not possible to distinguish between internal and external liquid holdup.

The resulting capillary pressure can be estimated according to the Young−Laplace equation. A mean pore diameter of 30 nm, a surface tension of benzyltoluene of approximately 7.0 mN m^{-1} at 300 °C, and a contact angle below 15° on alumina is assumed for the estimation of the capillary pressure. A capillary pressure of 9 bar is estimated for the given parameters. So, the total pressure in the pores is the sum of capillary pressure (9 bar) and bulk pressure (5 bar) resulting in a total pressure of 14 bar. This higher pressure results in suppression of complete evaporation of Hx-BT, explaining the elevated liquid holdup that is determined experimentally. Thus, a major part of the experimentally determined liquid holdup is in fact an internal liquid holdup in the porous catalyst.

3.4. Liquid Holdup Correlation

In this section, a first empirical correlation for the radial-averaged liquid holdup in a vertical LOHC dehydrogenation reactor is proposed. The developed correlation can serve as a basis for the optimized design of future dehydrogenation reactors. Within the investigated operating parameters (T_{Shell} = 280 to 340 °C, p_{rea} = 4 bar_a to 5 bar_a, LHSV = 0.6 h^{-1} to 3.2 h^{-1}), the liquid holdup was found to be nearly linearly dependent on the product of (i) the ratio of LOHC vapor

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pressure to reaction pressure and (ii) DoDH, which can be seen in Figure 9A. This combined parameter

$$x = \left(\frac{P^{LV}_{0,Hx\text{-}BT}}{p_{rea}} \right) \times \text{DoDH} \tag{14}$$

can be interpreted as a lumped description of the two dominant phenomena governing liquid holdup under reactive conditions:

1 Thermodynamic driving force for evaporation, represented by the ratio of vapor pressure of the pseudo LOHC component to process pressure $P^{LV}_{0,Hx\text{-}BT} p^{-1}_{rea}$, which reflects the propensity of the LOHC to evaporate.
2 Gas generation by reaction, represented by the DoDH, which determines the volumetric fraction of hydrogen formed and thus directly affects phase distribution and local hydrodynamics.

Complete evaporation, i.e., 0% liquid holdup, was found for x exceeding 0.1763. Because the liquid holdup must be 0 in the case of complete evaporation, the correlation for the predicted total liquid holdup $\epsilon_{liq,tot,pred}$ must be defined as a stepwise defined function

$$\epsilon_{liq,tot,pred} = \begin{cases} -5.6726 \times x + 1, & 0 < x \leq 0.1762 \\ 0, & x > 0.1762 \end{cases} \tag{15}$$

The proposed correlation differs fundamentally from classical liquid holdup correlations for both trickle-bed and bubble columns, which commonly express liquid holdup as a function of externally imposed superficial liquid and gas velocities, fluid properties, and dimensionless numbers such as Reynolds, Froude, or Etveös. In those systems, gas and liquid velocities are independent boundary conditions controlling hydrodynamics.

In the LOHC dehydrogenation reactor, however, the gas phase is generated internally via the chemical reaction, as expressed by the dimensionless DoDH. Simultaneously, the flow of the liquid phase is reduced by evaporation driven by the hydrogen release expressed by the dimensionless DoDH and the ratio of vapor pressure and system pressure, another dimensionless parameter. Although the empirical correlation is specific for the studied LOHC system, reactor geometry, and operating conditions, the proposed dimensionless structure suggests potential applicability to other gas-generating liquid-phase reactions.

Due to its simplicity, this linear approach yields a root-mean-square error (RMSE) of 0.19. These deviations are substantial and clearly reflect the complexity of the underlying multiphase reactive system. Nevertheless, given the superposition of reaction kinetics, phase equilibrium, capillary condensation, and two-phase hydrodynamics, the ability of a single linear descriptor to capture the dominant trend is noteworthy. Given the proposed correlation, 91.9% of all predicted values deviate by less than ±30% absolute, see Figure 9B. The developed correlation relies purely on bulk properties, and capillary effects are considered via the fitting parameters. The discrepancy between interparticle (internal) and intraparticle (external) holdup is most pronounced at 300 °C shell temperature and could be responsible for the deviations between the developed correlation and the experimental results. However, the experimentally determined liquid holdup is unexpectedly high for a shell temperature of 300 °C and thus deviations may also be caused by experimental errors. One source of error can be a slight overestimation of the DoDH caused by our sampling procedure (which followed Kadar et al.[8]), which is more pronounced under multiphasic conditions. Overestimating the DoDH results in an unreasonably high value for the parameter x, which could explain some of the observed deviations.

4. CONCLUSION

The present study provides the first axially resolved in situ determination of the liquid holdup in a fixed-bed LOHC dehydrogenation reactor under fully reactive conditions. Using γ-ray densitometry, the cross-sectional averaged total liquid holdup was quantified under technically relevant operating conditions. The applied experimental protocol demonstrated good reproducibility under the investigated conditions. Repeated measurements at a defined standard operating point over multiple experimental days show only minor deviations within a low uncertainty. This confirms both the stability of the experimental setup and the robustness of the densitometric method under reactive, high-temperature conditions.

A strong dependence of liquid holdup on catalyst temperature and the amount of hydrogen released was observed. Increasing temperature accelerates hydrogen release and promotes evaporation of the LOHC, resulting in a pronounced axial decline in liquid holdup. At elevated temperatures above 320 °C shell temperature, liquid holdup approaches negligible values within the first half of the catalyst bed, indicating complete gas phase dehydrogenation in the second half of the reactor.

The axial position of complete evaporation can also be estimated via vapor−liquid equilibrium calculations, and complete evaporation is predicted significantly faster than experimentally determined. This behavior is consistently explained by capillary effects within the porous catalyst structure, which leads to condensation of Hx-BT within the pores of the catalyst due to the higher pressure inside the pores.

A first empirical correlation for the local liquid holdup was derived as a function of vapor pressure, system pressure, and DoDH. This correlation provides a pragmatic engineering approximation for predicting axial liquid holdup profiles during the course of reactor design studies. Future work should aim to separate internal and external liquid contributions experimentally. This would allow us to link measured liquid holdup profiles directly to local heat-transfer coefficients and catalyst deactivation behavior.

To further increase the accuracy and general applicability of the proposed correlation, the experimental database should be extended by incorporating a broader range of operating conditions, flow configurations, or LOHC systems.

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Notes

Declaration of generative: AI and AI-assisted technologies in the manuscript preparation process. During the preparation of this work the authors used ChatGPT in order to edit the language. After using this tool/service, the authors reviewed and edited the content as needed and take full responsibility for the content of the published article.

The authors declare the following competing financial interest(s): The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Peter Wasserscheid is founder and minority shareholder of the company Hydrogenious LOHC technologies (www.hydrogenious.net) that offers commercially hydrogen storage systems based on the LOHC technology. There is no conflict of interest to declare with regard to the specific scientific results reported in this paper.

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■ NOMENCLATURE

$\dot{N}$	counting rate of detector
$\dot{V}$	volume flow (mL min^{-1}; NL min^{-1})
μ	attenuation coefficient (m^{-1})
μ	mean value of Gaussian fitting curve
ρ	liquid density (kg m^{-3})
Al_2O_3	alumina oxide
H_2	hydrogen
ϵ	liquid holdup (-)
$\epsilon_{liq,dyn}$	dynamic liquid holdup (-)
$\epsilon_{liq,ext}$	external liquid holdup (-)
$\epsilon_{liq,int}$	internal liquid holdup (-)
$\epsilon_{liq,stat}$	static liquid holdup (-)
$\epsilon_{liq,tot}$	total liquid holdup (-)
A_i,	area species (pA s)
e_ϵ	deviation of calculated liquid holdup (-)
E_{liq}	maximum radiation attenuation (-)
I_0	emitted intensity by radiation source (-)
I_x	attenuated intensity (-)
I_{gas}	attenuated intensity of gas reference measurement (-)
I_{liq}	attenuated intensity of liquid reference measurement (-)
I_{tpf}	attenuated intensity of two-phase flow with unknown phase distribution (-)
P_0^{LV}	vapor pressure (Pa)
Cs	Cesium
d	diameter (mm)
d	length of penetrated material (m)
DoDH	degree of dehydrogenation (-)
FID	flame ionization detector
GC	gas chromatography
H0-BT	benzyltoluene
H6-BT	partially hydrogenated benzyltoluene
H12-BT	perhydro benzyltoluene
H*x*-BT	mixture of H0-, H6- and H12-BT with unspecified hydrogen content
H18-DBT	perhydro dibenzyltoluene
H0-DBT	dibenzyltoluene
K	density correction factor
l	length (m)
LHSV	liquid hourly space velocity (h^{-1})
LOHC	liquid organic hydrogen carrier
MCA	multichannel analyzer
p	pressure (Pa)
PHS	pulse height spectra
Pt	platinum
RMSE	root-mean-square error
T	temperature (°C)
VLE	vapor−liquid equilibrium

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