

Lagrangian μ -Particle Tracking Velocimetry Analysis of Microscale Flow over Structured Surfaces

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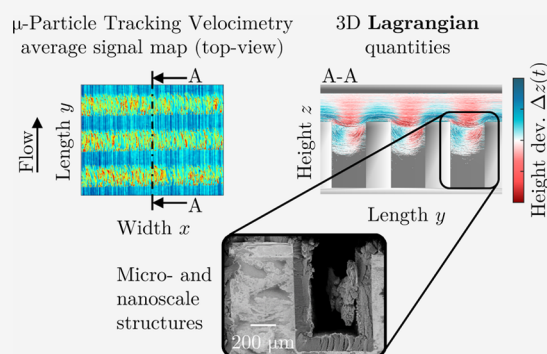
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ABSTRACT: This work aims to provide an experimental basis for the improvement of (bio)reactors by studying flow over micro- and nano-structured surfaces, with a focus on microscale mass transport toward and away from the surface. μ -Particle tracking velocimetry is employed in a microreactor to experimentally obtain Lagrangian trajectories of flow-following tracer particles. The method also resolves three-dimensional, time-dependent velocity fields. The derived mean velocity profiles agree well with the theory of flow over porous walls. Beyond averaged Eulerian velocities, the Lagrangian framework captures near-surface interactions. The Lagrangian analysis of different surfaces indicates that rectangular structured microgrooves increase the wall contact of the tracer particles, especially when the flow is oriented perpendicular to the microgrooves. Particularly, this approach enables resolution of trapping phenomena and localized recirculation dynamics. Nanostructuring by growing a carbon nanotube forest increases surface area and modifies local fluid dynamics, thereby enhancing contact with (bio)catalysts that are potentially functionalized on the nanotubes in future applications.



INTRODUCTION

The coupling of a free-fluid flow with porous media represents a fundamental phenomenon in many industrially relevant processes such as catalytic reactors and biochemical systems.^{1,2} In such systems, transport phenomena strongly influence the effective reaction rate in porous catalytic structures.³ Surfaces dominate the fluid flow characteristics through various physical quantities, such as wettability, surface roughness, and surface patterns.⁴ These surface effects govern mass transport toward and away from the interface and can therefore impact reaction performance by changing velocity gradients.^{3,5}

The relevance of interfacial fluid dynamics becomes even more pronounced for biochemical systems such as biofilms or enzyme-based reactors.^{6,7} In recent years, such systems have gained importance in sustainable process engineering due to their high selectivity, mild operating conditions, and renewable resources.⁸ In these systems, not only the reaction performance but also the formation and stability of the catalytic layer are influenced by surface properties and local flow conditions.^{6,9,10} To obtain a high yield within these reactors, it is desirable that the fluid is well mixed and that fluid containing reactants reaches the active media continuously.

Static mixers with catalytically active walls enhance mixing and reaction rates in gas flows¹¹ and are also commonly used in liquid–liquid reactions.^{12,13} Besides the usual T- and Y-type micromixer, other geometries like split-and-recombine mixers and various mixing principles have been employed for different applications, as summarized by Cai et al.¹⁴ Many effective microstructured mixers have a rather complex three-dimensional (3D) structuring. However, biochemical systems are sensitive to fluid dynamic stresses. High shear may influence the adhesion behavior of biofilms and intricate 3D geometry might favor clogging.^{9,15} For this reason, such shear-sensitive systems with biofilms¹⁶ or enzyme layers are typically operated under laminar flow in overflow configurations, often over electrode surfaces. Such operating conditions enable control of reaction parameters and residence time.^{16–19} In such scenarios, the influence of the porous medium at the reactor bottom on

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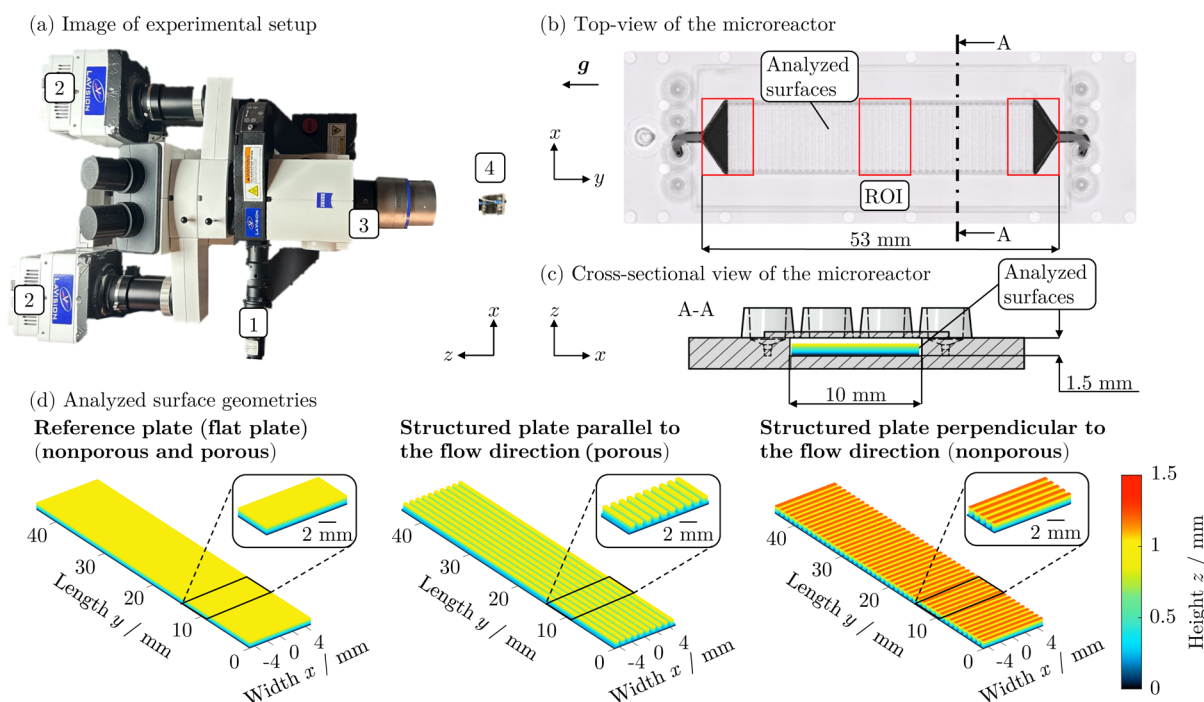


Figure 1. (a) Experimental setup used for the μ -PTV measurements. A Nd:YLF laser (1) and two high-speed cameras (2) are coupled through a microscope (3) to image the microreactor (4). (b) Top view of the microreactor with inserted plate. Red regions show the ROI for inlet, central, and outlet regions. The main flow direction is along the y -direction against g . (c) Cross-sectional view of the microreactor. (d) Analyzed surface geometries: a reference geometry (flat plate) and structured substrates with periodic continuous microgrooves, oriented parallel and perpendicular to the main flow direction. Nonporous surfaces correspond to fused silica, whereas porous surfaces are obtained by coating the reference geometry and the structured substrate with parallel microgrooves with MWCNTs.

external flow has been analyzed²⁰ as porous interfaces allow momentum exchange between the external and the internal flow.²¹ However, translating these fluid dynamic observations into design principles for catalytic structures remains challenging. Consequently, the engineering of catalytic structures to optimize transport processes often still relies on empirical trial-and-error approaches.^{22,23}

Recent advances in the development of tailorable porous materials provide new opportunities for structural improvement.²⁴ Carbon nanotubes (CNTs) are tubular nanostructures with diameters of several nanometers and can grow on substrates in densely packed, vertically aligned, forest-like shapes.^{25,26} This material offers considerable potential for industrial applications due to their exceptional thermal and mechanical properties.²⁷ Reported specific surface areas of multi-walled CNT (MWCNT) forests typically range from 150 to 350 m² g⁻¹, depending on structural parameters such as CNT diameter, length, bulk density, and alignment.²⁸ Such high surface areas make them suitable for catalytic reactor applications.^{29,30} Several studies show the potential for CNT-supported catalysts to enhance the efficiency of processes.^{29,31–33} Chemical vapor deposition (CVD) has emerged as a preferred growth method for CNTs.³⁰ Substrate-supported CVD can adjust the morphology of CNT forests, and the use of structured substrates can tailor the forest geometry to enhance transport processes.³⁰ For the study of such phenomena, Beavers and Joseph introduced experiments on the flow over porous surfaces in 1967.¹ Several studies have investigated flow over porous media using experimental, analytic, and numerical approaches.^{2,20,21} However, most of these studies are limited to two-dimensional flow data, while experimentally resolved 3D measurements remain scarce,

although such data are indispensable to understand microscale transport phenomena.³⁴ This is especially relevant for flows involving rough and porous surfaces, as well as wettability effects, where uncertain boundary conditions limit 3D numerical simulations. Since surface interactions, recirculation zones, and vertical velocity components can influence transport and residence times in all spatial directions, experimentally resolved 3D flow data are required for a comprehensive analysis.

Micron-resolution particle tracking velocimetry (μ -PTV) is a nonintrusive technique to track individual fluorescent particles within a fluid flow.¹⁰ In contrast to micron-resolution particle image velocimetry (μ -PIV), which uses particle pattern image correlation to derive Eulerian velocity fields, given at fixed spatial locations,¹⁰ μ -PTV enables a Lagrangian analysis of individual particle trajectories. By resolving these individual particle trajectories, a Lagrangian approach captures the time-dependent transport dynamics.³⁵ In this study, μ -PTV is applied to structured and porous CNT forest surfaces to resolve the three-dimensional particle trajectories and quantify microscale transport dynamics. These findings advance the understanding of engineered porous surfaces and support the development of novel reactor concepts within the collaborative research center 1615 SMART Reactors, which is funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation). The study is organized as follows: the experimental setup, the fabrication of the customized surfaces with and without nanostructures, and the data analysis are described. Further, the results are discussed beginning with an analysis of the studied structures with regard to the visualized flow, the Eulerian velocity field, and the average velocity profile. Subsequently, the Lagrangian trajectories are used to

evaluate the transport of the tracers close to the wall for different surface geometries. The findings are summarized and put into context in the final section.

■ EXPERIMENTAL SECTION

The experimental setup employed in this study for μ -PTV measurements is introduced first. The different surface geometries used for the flow experiments, as well as their fabrication processes, are described subsequently. The μ -PTV technique is applied to obtain 3D particle trajectories within the used microreactor. The corresponding data processing procedure is outlined thereafter.

Experimental Setup

The experimental setup is shown in Figure 1a and comprises an optical system (μ -PTV system) and a microreactor. The optical system is part of the research equipment MUST, which stands for mobile multianalysis unit for flow investigations in the context of power-to-x technologies and hydrogen applications, and is funded by the DFG. The whole μ -PTV system is acquired from LaVision GmbH (Göttingen, Germany). In this system, a Nd:YLF laser (Photonics Industries International Inc., USA, 1) is optically coupled to two high-speed cameras (Phantom VEO 640L, Vision Research, USA, 2) and directed through a stereomicroscope (SteREO Discovery.V12, Carl Zeiss AG, Germany, 3). The laser is a dual-cavity, high-speed system providing a pulse energy of 30 mJ per cavity at a maximum repetition rate of 10 kHz and an emission wavelength of 527 nm. The output intensity is adjusted with the operating current between 18 and 41 A in combination with an integrated attenuator to excite fluorescent polystyrene particles (PS-FluoRot-Fi219, microParticles GmbH, Germany) with a diameter of $d_p = (7.76 \pm 0.12) \mu\text{m}$. The maximum excitation of the particles at 530 nm is close to the emission wavelength of the Nd:YLF laser, while the fluorescence maximum of the particles is at 607 nm. A bandpass ($\lambda \geq 545 \text{ nm}$) filter in the microscope prevents the laser excitation light from passing to the cameras. The laser pulses are synchronized with the image acquisition frequency of the cameras at 100 Hz using an electronically controlled shutter. Each of the two high-speed cameras has a resolution of 2560×1600 pixels. They are arranged so that their optical axes are aligned with the direction of the incident laser beam. The microscope operates at magnifications in the range from 1.28 $\times$ to 16 $\times$. In the present experiments, total magnifications of 1.8 $\times$ and 2.4 $\times$ are used with a maximum numerical aperture of 0.03. The depth of field (DOF), which is inversely proportional to the microscope magnification, differs between the two configurations. For magnifications of 2.4 $\times$ and 1.8 $\times$, the resulting DOFs are 571 and 902 μm , respectively.

For flow experiments, a custom-designed microreactor fabricated from Topas cyclic olefin copolymers (COC) polymer (microfluidic ChipShop GmbH, Germany) is positioned in front of the microscope, as shown in Figure 1a labeled as (4). The microreactor features a defined geometry, as illustrated in Figure 1b,c. It has a width of $W = 10 \text{ mm}$ and a length of $L = 53 \text{ mm}$, with a channel height of $H = 1.5 \text{ mm}$ in the baseline reactor configuration. The width corresponds to the x -direction, the length to the y -direction, and the height to the z -direction used for the flow analysis. The inlet and outlet geometries are triangular, as shown in the top view of the microreactor in Figure 1b. The reactor is sealed at the top with 100 μm -thick Topas COC foil, which both closes the reactor and provides optical access. The material exhibits a refractive index of 1.53 at 589 nm and 25 $^\circ\text{C}$.³⁶ Measurements are conducted on different surface geometries at three distinct regions of interest (ROIs), namely, the inlet, central, and outlet sections, as depicted in Figure 1b.

Three specific surface geometries are investigated, as illustrated in Figure 1d. As a reference case, a flat, unstructured plate is examined. To enhance mass transport from and to the wall, the plate geometry is structured. The surface structuring consists of periodic continuous so-called microgrooves oriented both parallel and perpendicular to the main flow direction. The surface geometry of the fabricated samples is characterized using a profilometer (Keyence Corporation, Japan) to

evaluate the actual structural geometry. The unstructured plate corresponds to the intended design geometry. For the microgrooves aligned with the flow direction, widths of $(453 \pm 10) \mu\text{m}$ were measured for the grooves, while the plateaus exhibited widths of $(517 \pm 8.2) \mu\text{m}$. In contrast, the microgrooves oriented perpendicular to the main flow direction showed microgroove widths of $(407 \pm 10) \mu\text{m}$ and plateau widths of $(559 \pm 7.5) \mu\text{m}$. Possible reasons for these deviations and the different widths of microgrooves and plateaus are related to the fabrication process of the substrates, which is discussed in the section describing the substrate manufacturing process. The total height of the structured substrates is 1.0 mm and 1.2 mm for the parallel and perpendicular geometries, respectively. The actual geometry of the substrates is taken into account in the following analyses. The different structured plates are incorporated into the microreactor from the bottom for experimental studies. Thus, their incorporation reduces the effective (smallest) flow channel height to $H_{f,\min} = 500 \mu\text{m}$ for the reference geometry and for the structured plate with microgrooves aligned parallel to the main flow direction and to a channel height to $H_{f,\min} = 300 \mu\text{m}$ for the structured plate with microgrooves oriented perpendicular to the main flow direction (see Figure 1c). Further fluid-filled regions exist within the microgrooves, increasing the total fluid-filled height to $H_f = 1.35 \text{ mm}$.

Throughout this work, four different plates are analyzed, comprising porous and nonporous cases with varying wettability. The nonporous case corresponds to a fused silica plate. Porous cases refer to surfaces whose geometries are coated with MWCNTs. The fabrication process is described later in this section. The following enumeration summarizes the studied cases.

- (i) Reference plate as (nonporous) fused silica plate.
- (ii) Reference plate with (porous) hydrophobic MWCNTs
- (iii) Structured plate with parallel microgrooves with (porous) hydrophilic MWCNTs.
- (iv) Structured plate with perpendicular microgrooves as (nonporous) fused silica plate.

The nonporous reference plate is used for the validation of the experimental setup and analysis procedure, which is discussed in Figure S1 in the Supporting Information. In addition, a hydrophobic MWCNT-coated reference geometry is compared with the structured (parallel to flow direction) hydrophilic MWCNT-coated surface in Figure 3. Furthermore, a nonporous structured plate with microgrooves aligned perpendicular to the main flow direction is studied. The detailed analysis focuses on cases (i), (iii), and (iv).

Operating Conditions

Bidistilled water at a temperature of $T = (20 \pm 1) ^\circ\text{C}$ is fed with a syringe pump (KD Scientific Inc., USA) into the microreactor. The material properties of bidistilled water are a density of $\rho_L = (998.2 \pm 0.2) \text{ kg m}^{-3}$ and a viscosity of $\eta = (1.00 \pm 0.02) \text{ mPa s}$.³⁷ According to the demands in SMART reactors, a volumetric flow rate of 0.08 mL min^{-1} is selected, corresponding to a mean axial flow velocity magnitude of $\bar{v} = 0.27$ to 0.44 mm s^{-1} , dependent on the effective flow height $H_{f,\min}$. The operating conditions are summarized in Table 1 for all cases. For evaluating the operating conditions, the porous plate with parallel microgrooves is considered equivalent to the reference plate, as the CNT coating fills the grooves, making the surface comparable in terms of the following fundamental fluid dynamics considerations. For the rectangular cross-section, the hydraulic diameter is defined as $D_h = 2WH_{f,\min} / (W + H_{f,\min})$. This diameter is used as the characteristic length scale in the definition of the Reynolds number

$$Re = \frac{D_h \rho_L \bar{v}}{\eta} \quad (1)$$

yielding a Reynolds number of $Re \approx 0.25$, which lies within the laminar regime and can be classified as Stokes flow ($Re \ll 1$). In Stokes flow, inertial forces are smaller than viscous forces.^{38,39} The operating conditions are chosen to be representative for biochemical processes, which require long residence times for high yields and Reynolds numbers of $Re < 1$.^{8,19,40}

The microreactor is operated counter to the direction of gravitational acceleration. Prior to each measurement, gas bubbles are flushed out by temporarily applying an elevated flow velocity. The presence of gas bubbles alters the effective flow cross-sectional area and affects both the pressure drop and the velocity profile as they block the free flow. In addition, their presence can induce local accelerations, recirculation zones, and elevated shear gradients, which also have been shown to enhance mixing.⁴¹ Once a bubble-free state is achieved, the measurements are initiated. To quantify the influence of gravitational forces on the analysis, the Bond number Bo

$$Bo = \frac{(\rho_L - \rho_G)gD_h^2}{\sigma} \quad (2)$$

is evaluated.³⁸ Here, $\rho_G = 1.204 \text{ kg m}^{-3}$ is the gas density, $\sigma = 72.74 \text{ mN m}^{-1}$ is the surface tension between air and water at 20 °C, and $g = 9.81 \text{ m s}^{-2}$ is the gravitational acceleration.³⁷ According to eq 2, the resulting values of the Bond number are $Bo \approx 0.05$ to 0.1. Since $Bo \ll 1$, gravitational effects are negligible under the present operating conditions.³⁸

For the μ -PTV measurements, tracer particles are employed. The bidistilled water is seeded with fluorescent monodisperse polystyrene particles. The resulting particle seeding density is 0.032 ± 0.044 particle per pixel for the different measurements. To assess whether the particles can be considered flow-following, two Stokes numbers are evaluated, namely the Stokes number for gravitational settling St_g and the (inertial) Stokes number St .^{42,43} The former Stokes number is based on the buoyancy-induced settling velocity, which quantifies the extent to which gravitational effects cause the particle motion to deviate from the fluid motion.⁴⁴ The settling velocity v_s can be calculated according to

$$v_s = \frac{(\rho_p - \rho_L)gd_p^2}{18\eta} \quad (3)$$

where $\rho_p = 1050 \text{ kg m}^{-3}$ and d_p denotes the density and diameter of the tracer particles, respectively. The validity of this expression requires the particle Reynolds number Re_p

$$Re_p = \frac{d_p \rho_L v_s}{\eta} \quad (4)$$

to remain well below unity. Evaluating eq 3 yields a settling velocity of $v_s = 1.7 \text{ } \mu\text{m s}^{-1}$, which in turn confirms $Re_p \ll 1$ and thus the applicability of eq 3.⁴⁵ The relevance of gravitational settling relative to the flow-induced particle motion is quantified by

$$St_g = \frac{v_s}{\bar{v}} \quad (5)$$

A value of $St_g = 3.8 \cdot 10^{-3}$ to $6.4 \cdot 10^{-3} \ll 1$ confirms that settling effects are negligible relative to the flow-induced particle displacement.^{42,46} The (inertial) Stokes number St is defined as the ratio of the relaxation time of the tracer particle τ_p to the characteristic time of the flow τ_F .^{43,47} The particle relaxation time is given by

$$\tau_p = \frac{d_p^2 \rho_p}{18\rho_L \nu} \quad (6)$$

where ν is the kinematic viscosity of the liquid. Furthermore, the characteristic time scale of the flow is defined as

$$\tau_F = \frac{D_h}{\bar{v}} \quad (7)$$

where D_h represents the hydraulic diameter, taken here as the characteristic length scale.^{43,47} The resulting definition of the Stokes number is expressed as

$$St = \frac{\tau_p}{\tau_F} = \frac{d_p^2 \rho_p \bar{v}}{18\rho_L \nu D_h} = \frac{d_p^2 \rho_p}{18\rho_L D_h^2} Re \quad (8)$$

Evaluation of eq 8 results in Stokes numbers of $St \approx 2.7 \cdot 10^{-6}$ to $9.8 \cdot 10^{-7}$. The dimensionless numbers discussed previously are also included in Table 1. The condition $St \ll 1$ indicates that the particles

Table 1. Operating Conditions and Dimensionless Numbers for the Reference and Structured Plates Studied

parameter	reference plate (nonporous)	struc. plate parallel microgrooves (porous)	struc. plate perp. microgrooves (nonporous)
mean axial velocity $\bar{v}$ /mm s ⁻¹	0.27	0.27	0.44
hydraulic diameter D_h /μm	952	952	583
Reynolds number Re /–	0.25	0.25	0.26
Bond number Bo /–	0.1	0.1	0.05
gravitational Stokes number St_g /–	$6.4 \cdot 10^{-3}$	$6.4 \cdot 10^{-3}$	$3.8 \cdot 10^{-3}$
Stokes number St /–	$9.8 \cdot 10^{-7}$	$9.8 \cdot 10^{-7}$	$2.7 \cdot 10^{-6}$

are considered flow-following.⁴⁸ In addition, for $St < 0.2$, velocity slip errors are expected to be less than 2%.^{49,50} Prior to the measurements, the particles are dispersed by mixing them with the bidistilled water and repeatedly pumping the suspension back and forth through the system such that the particles are homogeneously distributed. Following the characterization of the tracer particles, the fabrication of the surface geometries studied in this work is described below.

Fabrication of Analyzed Surface Geometries

In this work, both nonporous and porous cases are considered. All cases are based on fused silica plates. The fused silica substrates are fabricated using selective laser-induced etching with the help of the 3D printer from Lightfab (Lightfab GmbH, Germany). This two-step process enables the subtractive 3D microstructuring of transparent materials.⁵¹ In a first step, the fused silica bulk material is locally modified by multiphoton absorption using a highly focused femtosecond laser (emission wavelength of 1030 nm, pulse duration of 0.5 ps, spot size of around 2 μm) without affecting the surrounding areas. In a second step, the modified regions can be etched in an ultrasonic bath at 82 °C in 8 mol L⁻¹ potassium hydroxide significantly faster than the unmodified regions, with selectivities of over 1000 being achievable.⁵¹ Here, the selectivity is defined as the ratio of the etch rate of laser-modified regions to that of unmodified regions. Due to the etching of unmodified regions, so-called overetching occurs, resulting in a deviation between designed and manufactured dimensions. As a consequence, the microgrooves exhibited larger widths than the plateaus due to the additional material removal during fabrication. These deviations were quantified in this study using a profilometer and were found to range from approximately 32 to 78 μm for the substrates with microgrooves aligned parallel and perpendicular to the flow direction, respectively.

In addition to nonporous fused silica plates, porous MWCNT forests are studied. Therefore, two fused silica structures (reference plate and structured plate with parallel microgrooves) are further coated with a MWCNT forest synthesized on the substrates via a CVD process, following the procedure reported by Kröning et al.⁵² A horizontal tube furnace (Carbolite Gero GmbH & Co. KG, Germany) is used for the synthesis. The furnace has a quartz glass tube of 110 mm inner diameter and 1100 mm length (Glasatelier Sillart BV, Belgium) and comprises three independently controlled heating zones. As the carbon source and catalyst precursor, a solution containing 5 wt % ferrocene (Merck KGaA, Germany, purity = 98%) dissolved in toluene (Th. Geyer GmbH & Co. KG, Germany) is employed. The precursor solution is introduced into the reactor via an evaporation unit positioned upstream of the tube furnace. Prior to

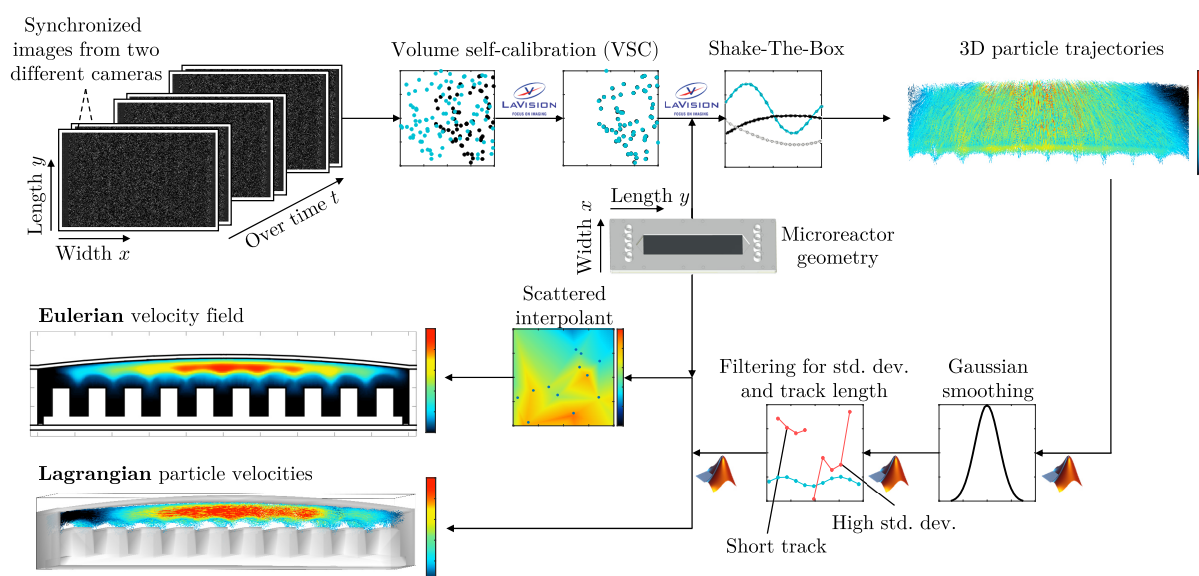


Figure 2. Workflow of the Lagrangian data analysis applied to the μ -PTV measurements. Following stereoscopic image acquisition with two synchronized high-speed cameras of the MUST facility, a volume self-calibration (VSC) is performed. Three-dimensional particle trajectories are reconstructed in DaVis using the Shake-The-Box algorithm while incorporating the reactor geometry. The resulting tracks are postprocessed in MATLAB, including Gaussian smoothing, selective filtering based on standard deviation of the particle velocities and track length, as well as a geometry-based validity check. After these steps, the final Lagrangian particle trajectories and velocities are obtained and can be analyzed. An Eulerian velocity field is generated by interpolating the scattered Lagrangian data onto a structured spatial grid.

injection, the reactor is purged with argon (Westfalen AG, Germany, purity = 99.999 vol %) at a flow rate of 0.4 L min⁻¹. The heating zones are positioned sequentially along the gas flow direction, with 200 °C in the inlet zone and 790 °C in the subsequent downstream zones. After thermal equilibrium is reached, the evaporation unit is heated to 200 °C, and the argon flow is reduced to 0.2 L min⁻¹. Hydrogen (Westfalen AG, Germany, purity = 99.999 vol %) is used as a reducing agent at a flow rate of 55 mL min⁻¹. The synthesis is initiated by feeding the precursor solution into the system at a rate of 20 mL h⁻¹ using a syringe pump (Kisker Biotech GmbH & Co. KG, Germany). After 3 h, corresponding to a total injected precursor volume of 60 mL, the precursor feed and hydrogen supply are terminated. The reactor is subsequently cooled to room temperature under an increased argon flow rate of 0.4 L min⁻¹. Upon reaching ambient conditions, the substrate coated with the MWCNT forest is retrieved.

Since the flow experiments are conducted with bidistilled water, the surface wettability plays a crucial role.⁴ Based on Young's equation, wetting phenomena can be described quantitatively by placing a liquid droplet on a solid surface and measuring the contact angle.⁵³ Wettability does not limit experiments with fused silica, which exhibits a contact angle of 0° at 25 °C,⁵⁴ indicating complete wetting. In contrast, MWCNT forests display superhydrophobic behavior after synthesis, with contact angles exceeding 150°.⁵⁵ Although individual CNTs exhibit a contact angle of approximately 85°,⁵⁵ the superhydrophobicity of the forest structure arises from the Cassie–Baxter wetting state, in which air pockets trapped within the porous network substantially increase the apparent contact angle.⁵⁶ However, this state is metastable: external influences can induce a transition from the Cassie–Baxter state to the Wenzel state by overcoming an associated energy barrier. In the Wenzel state, the air pockets are filled with liquid, leading to a decrease in the contact angle and resulting in hydrophilic wetting behavior, which allows droplets to penetrate the MWCNT network.⁵⁶ A targeted transition between these wetting states can be achieved through controlled functionalization of the MWCNT forest. This involves introducing polar functional groups into the forest to increase the polarity of individual CNTs. In this work, treatment with oxygen plasma enables grafting of oxygen-containing functional groups onto the tips of CNTs, resulting in hydrophilic wetting behavior.⁵⁷ For plasma generation, a low-pressure

plasma system (SmartPlasma 10, plasma technology GmbH, Germany) is employed. The MWCNT plates are placed inside the plasma chamber, which is subsequently evacuated. For the plasma treatment, an oxygen partial pressure of 0.3 mbar (Westfalen AG, Germany, purity = 99.999 vol %) is maintained. Functionalization is carried out at 300 W for 3 min. The hydrophilized samples are removed at ambient pressure. μ -PTV is used to analyze the flow over different surface geometries. The measurements are then processed using a Lagrangian procedure, as described below.

Particle Tracking Velocimetry Analysis Procedure

In the present study, a Lagrangian and an Eulerian perspective are used. For the study of fluid–surface interactions, transport phenomena, and residence times, a Lagrangian perspective is advantageous, as these phenomena cannot be fully captured within an Eulerian framework. However, the Eulerian approach remains valuable for describing global flow structures and time-averaged behavior. In general, the velocity field $\mathbf{v}(\mathbf{x}, t)$ in Eulerian description is defined at fixed points in space as

$$\mathbf{v}(\mathbf{x}, t) = (u, v, w) \quad (9)$$

where (u, v, w) represents the Cartesian velocity components in the x -, y -, and z -directions, respectively, and $\mathbf{x}$ is the spatial coordinate, and t is the time. The Eulerian velocity at position $\mathbf{x}'$ can be derived from the Lagrangian particle velocity using

$$\mathbf{v}(\mathbf{x}'(\mathbf{x}_0, t), t) = \frac{\partial \mathbf{x}'(\mathbf{x}_0, t)}{\partial t} \approx \frac{\Delta \mathbf{x}'(\mathbf{x}_0, t)}{\Delta t} \quad (10)$$

Here, $\mathbf{x}'(\mathbf{x}_0, t)$ denotes the position of the particle at time t , with an initial position of $\mathbf{x}_0$. The approximation in eq 10 is valid, if the time difference Δt is small compared to the temporal change of the flow and the spatial difference $\Delta \mathbf{x}'$ is small compared to the spatial change of the flow.

The μ -PTV measurements are evaluated using an elaborate Lagrangian analysis workflow to obtain Eulerian velocity fields and Lagrangian velocities, as shown in Figure 2. After image acquisition, the recordings are processed in DaVis 11.2 software (LaVision GmbH, Germany) for particle reconstruction and tracking. The resulting trajectory data are postprocessed in MATLAB using the integrated Image Processing Toolbox (MathWorks Inc., USA).

Table 2. Metrics for Spatial Calibration and VSC (1) as well as the Number of Tracks, Trajectories, and Reconstructed Process Volumes with Average Position Uncertainties in the Three Spatial Directions by Shake-The-Box (2), as Obtained from DaVis

	parameter	reference plate (nonporous)	struc. plate parallel microgrooves (porous)	struc. plate perp. microgrooves (nonporous)
(1)	magnification/–	2.4×	2.4×	1.8×
	DOF/ μm	571	571	902
	increment/ μm	300	300	500
	scale factor/pixel mm^{-1}	239.64	241.59	179.52
	fit error/pixel	0.015	0.011	0.014
(2)	temporal resolution $\Delta t_M/\text{s}$	0.01	0.01	0.01
	time steps/–	3,120	2,900	3,916
	volume $L_x \times L_y \times L_z/\text{mm}^3$	$10.79 \times 6.74 \times 0.75$	$10.37 \times 6.67 \times 1.50$	$10.00 \times 9.01 \times 1.35$
	av. uncertainty in $x/\mu\text{m}$	0.110	0.211	0.154
	av. uncertainty in $y/\mu\text{m}$	0.114	0.206	0.164
	av. uncertainty in $z/\mu\text{m}$	0.448	1.005	0.860
	number of particles/–	11,000	34,000	30,000
	number of trajectories/–	73,000	376,000	316,000

The two cameras coupled to the stereomicroscope look from slightly different angles onto the microreactor, which causes the two images to appear slightly shifted along the reactor width (x -direction). This configuration results in two different lines of sight and thus enables the reconstruction of positions in three-dimensional space by stereoscopic triangulation. To calibrate the investigated volume in front of the microscope, a calibration target (Model MP 50 $\times$ 38 0.5 mm, LaVision GmbH, Germany) is used. It has a size of 50 $\times$ 38 mm², featuring a grid of dots with diameters of 167 μm , which are arranged at a spacing of 500 μm . Three-dimensional calibration is achieved by adjusting the distance between the microscope and the target to span the entire DOF. For this purpose, the microscope is moved in 300 or 500 μm increments (depending on the used magnification), resulting in three calibration planes in z -direction, with the metrics for spatial calibration summarized in Table 2. Multiple planes are required when using a two-dimensional calibration target to account for optical distortions and the finite thickness of the measurement volume. A subsequent volume self-calibration (VSC) is performed in DaVis to correct mapping function errors based on the actual particle recordings.^{58,59} The VSC is an iterative process that projects the positions of triangulated particles back onto the original image. The resulting disparity vectors are used to calibrate the perspective down to subpixel accuracy. The mean spatial resolution is 180 pixel mm^{-1} for the magnification of 1.8 $\times$, and about 240 pixel mm^{-1} for the magnification of 2.4 $\times$, with a maximum fitting error of 0.015 pixel. The calibration metrics of the VSC are summarized in Table 2. The resulting enhancement in reconstruction accuracy, particularly along the height (z -direction), is of particular importance, as only two cameras are employed in the present configuration.⁶⁰ For the subsequent particle reconstruction, an optical transfer function is calculated for each camera at different subvolumes within the investigated volume.

Tracer particle tracking is conducted using the Shake-The-Box algorithm implemented in DaVis.^{61,62} In the recorded images, tracer particles appear as nearly Gaussian intensity peaks with a typical size of about 3 $\times$ 3 pixels. Since this study investigates rough and structured surfaces, such as MWCNT forests and microstructured surfaces, tracer particles become trapped on the surface and obstruct the flow analysis. Thus, a background image is generated by averaging all recorded images prior to the Shake-The-Box analysis. As stationary particles remain at fixed positions throughout the image sequence, they appear in the resulting average image. This background image is subtracted from each individual frame, thereby removing stationary particles from the image sequence before particle tracking and preventing them from being identified by the Shake-The-Box algorithm. Particle trajectories are recorded over approximately 2900 to 3900 consecutive time steps with a temporal resolution of $\Delta t_M = 0.01$ s, corresponding to an acquisition frequency of 100 Hz. Using this data, the reconstructed process volumes ($L_x \times L_y \times L_z$) after applying the Shake-The-Box method are summarized in Table 2.

L_x , L_y , and L_z represent the lengths of the reconstructed volume in the x -, y -, and z -directions, respectively. In the analysis, only every tenth time step is evaluated, resulting in an effective temporal resolution of $\Delta t_A = 0.1$ s. All temporal references hereafter refer to this temporal resolution. To derive a positional uncertainty, the DaVis software applies a polynomial least-squares fit along each particle trajectory, with the uncertainty corresponding to the confidence intervals of the fitted coefficients.^{62,63} The average uncertainties of all tracks in each of the three spatial dimensions are reported in Table 2. However, it should be kept in mind that further sources of uncertainty may exist as discussed in ref 64.

Irregular plate surfaces resulting from the surface roughness can promote particle trapping and adhesion. These immobilized particles may still be identified by the tracking algorithm and, consequently, generate artifacts. Moreover, since the stereoscopic reconstruction relies on only two camera perspectives, vibrations or camera misalignment can introduce triangulation errors. These errors may result in ambiguous particle matching, resulting in ghost particles,⁶⁵ especially on the edge of the z -limits of the microreactor. These artifacts affect the velocity reconstruction, resulting in elevated unphysical local velocities. Thus, the reactor geometry is included as an STL file in the analysis in DaVis to restrict it to physically possible flow regions in the Shake-The-Box method.

The resulting particle trajectories are further processed in MATLAB, as shown in Figure 2. The particle tracks are smoothed using a Gaussian filter to reduce measurement noise using the built-in MATLAB function `smoothdata`. Furthermore, trajectories shorter than 150 time steps were excluded from the analysis. Trajectories exhibiting a standard deviation higher than 10 in velocity with regard to their neighbors are removed,⁶⁴ as such behavior typically indicates tracking errors, mismatches, or ghost particle artifacts. Particle velocities along the reconstructed trajectories were determined using a fourth-order central difference scheme. In an additional processing step for the derivation of the Eulerian velocity fields, particles located outside the physically admissible measurement domain were excluded. This is based on the known geometry of the microreactor by placing zero-velocity boundary conditions at the reactor walls, the fused silica surfaces, and outside the flow domain. For each measurement case, between 73,000 and 376,000 trajectories are processed. These numbers, along with the corresponding number of particles, are summarized in Table 2. On the basis of the processed trajectories, two complementary perspectives are distinguished: the Lagrangian and the Eulerian approach. In the Lagrangian framework, quantities such as changes in position or velocity are evaluated along the reconstructed three-dimensional particle trajectories, as illustrated in Figure 2.

In contrast, the Eulerian velocity field is obtained by interpolating the irregularly distributed Lagrangian particle tracks onto a structured spatial grid using MATLAB's built-in `scatteredInterpolant` function for each time step. The resulting temporally resolved velocity

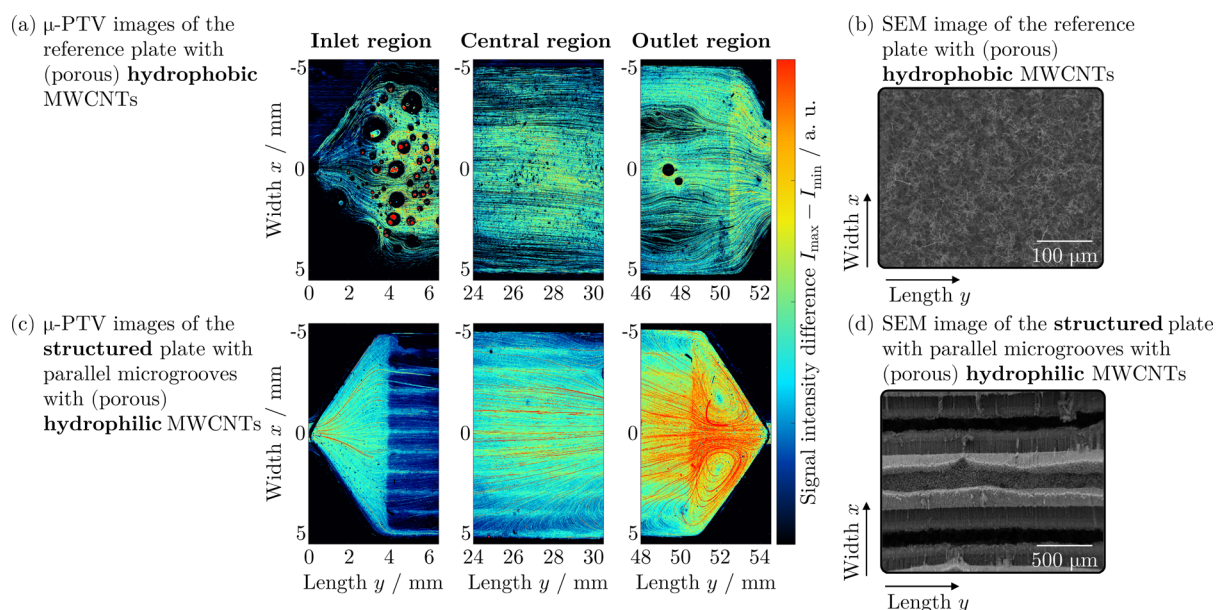


Figure 3. Time-averaged μ -PTV images of the flow over different MWCNT forests shown as signal intensity maps, where the color scale represents the difference between the maximum and minimum recorded signal intensities. Images are acquired in the inlet, central, and outlet regions of the microreactor. (a) As-synthesized (porous) hydrophobic MWCNT forest on the reference plate, exhibiting a gas bubble accumulation and flow disturbances. (c) Structured plate with parallel microgrooves and a (porous) hydrophilic MWCNTs, showing improved wetting and reduced bubble presence. Corresponding top-view SEM images prior to flow experiments are presented in (b) and (d) for the configurations shown in (a) and (c), respectively.

fields are analyzed here by applying spatial averaging in the flow direction as well as temporal averaging. An example of the averaged velocity field is shown in Figure 2. However, it must be highlighted that the temporally resolved velocity fields are also obtained with the presented measurement and analysis techniques but are not shown here.

RESULTS AND DISCUSSION

First, characteristic flow structures within the microreactor are evaluated by investigating the flow over MWCNT forests with varying wettabilities using a two-dimensional visual approach based on the pathlines of the tracer particles. Second, a three-dimensional Eulerian analysis is performed for micro- and nanostructured MWCNT forests. The analysis is subsequently extended to a Lagrangian framework to resolve time-resolved fluid–surface interactions. Tracking individual tracer trajectories provides direct insight into near-wall transport processes and fluid–surface contact.

Two-Dimensional Visual Characterization of the Flow

Figure 3a,c presents μ -PTV images from one camera, shown as signal intensity maps. Two different MWCNT forests with distinct surface properties in terms of geometry and wettability are compared. The color scale represents the difference between the highest and lowest signal intensities, time-averaged over the measurement duration of approximately 30 s. The tracer particles are illuminated by a laser and exhibit a high signal intensity in the images. Thus, regions of elevated signal intensity represent the sum of several particle trajectories and indicate dominant flow pathways within the microreactor. This approach provides insight into the global flow structures based on the time-averaged behavior. From the observed preferred pathline patterns, distinct flow phenomena can be identified, which can be attributed to the combined effects of reactor geometry, surface structure, and gas bubbles. Thus, the

signal intensity maps are displayed at different streamwise positions within the microreactor (ROIs in Figure 1b).

The MWCNT forest synthesized on the reference plate, which exhibits a superhydrophobic surface (hydrophobic MWCNT forest), is considered. A top-view scanning electron microscopy (SEM) image of this surface is shown in Figure 3b. All SEM images presented in this work are captured using a Zeiss Supra 55 VP (Carl Zeiss AG, Germany) at an acceleration voltage of 2 kV. The MWCNTs are vertically aligned, forming a rough and heterogeneous structure with unevenly distributed clusters. This porous morphology exhibits superhydrophobic behavior.⁵⁶ Considering Figure 3a, in the inlet region, the superhydrophobic nature of the surface is evident, as the MWCNT forest is not wetted and remains partially covered by gas bubbles. Although a sophisticated bubble-removal procedure is employed, some gas bubbles remain stable on the surface and can be seen as particle-free zones (low signal intensity in Figure 3a). These bubbles significantly disturb the incoming flow and locally alter the velocity field. In addition, the surface is in a Cassie–Baxter state, where air is entrapped between the individual MWCNTs. Consequently, both the gas bubbles and the entrapped air pockets reduce the liquid–solid contact area and result in a liquid–air–solid interphase.

In contrast, the flow field shown in Figure 3c exhibits a different behavior. This difference can be attributed to the surface wettability and geometry. Here, the plate features parallel microgrooves coated with a hydrophilic MWCNT forest (microstructured hydrophilic MWCNT forest). The hydrophilic behavior is achieved by introducing polar functional groups into the MWCNT forest. In addition, the microstructured surface is shown in the SEM image in Figure 3d. The image demonstrates that CNTs grow perpendicular to the local surface.²⁵ Consequently, the underlying microgroove structure is replicated within the MWCNT forest, resulting in a

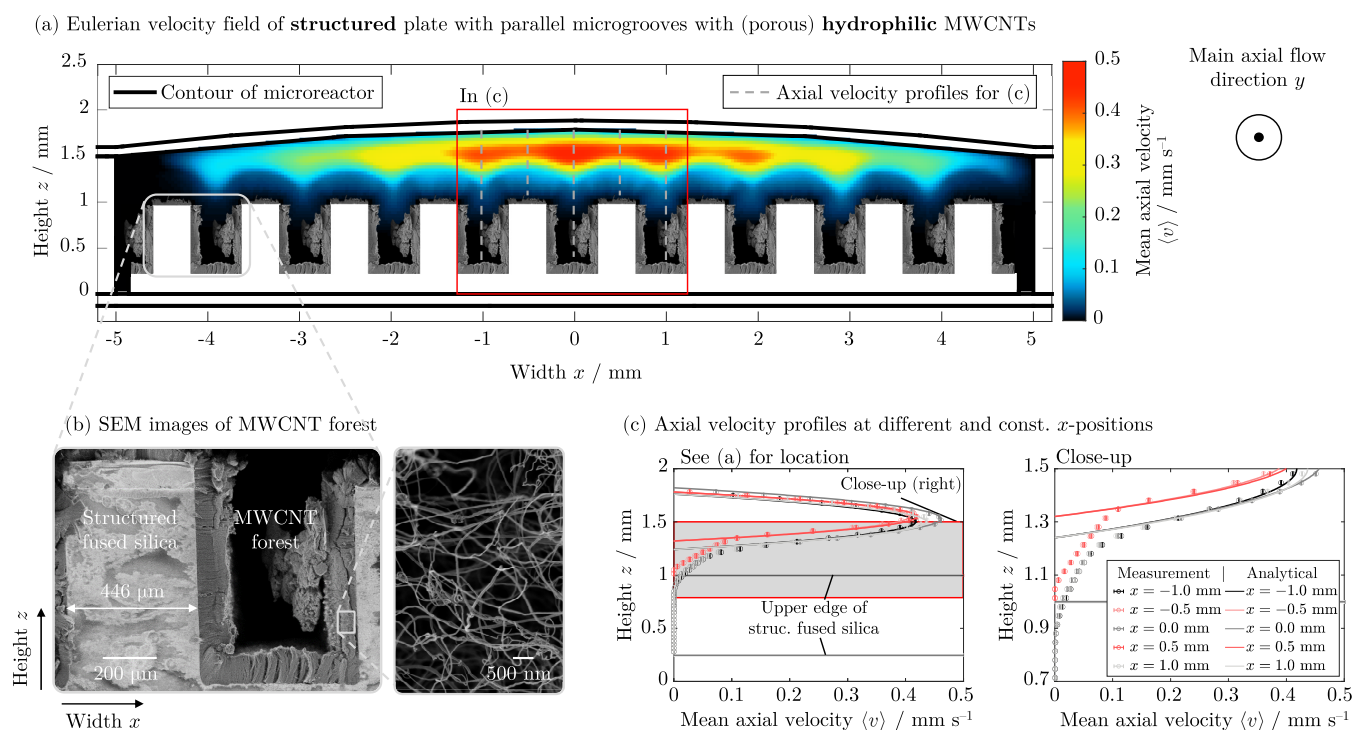


Figure 4. Temporally and spatially averaged axial velocity $\langle v \rangle$ for flow over the microstructured MWCNT forest in the central region of the microreactor in (a). The MWCNT forest is grown on top of a structured fused silica substrate. (b) SEM image of the cross-sectional area corresponding to (a). The close-up shows a higher-magnification view highlighting individual CNTs. (c) One-dimensional axial velocity profiles are compared with the analytical solution for Poiseuille flow in a rectangular channel of assumed infinite width (see eq 11). The profiles are extracted from the averaged velocity field shown in (a). A magnified view highlights the near-wall region. The error bars indicate the standard error of the temporally and spatially averaged velocities.

micro- and nanostructured surface topology. Thus, this structure integrates multiple levels of porosity. Such hierarchical systems have attracted rapidly growing attention due to their large accessible surface area and potential for enhanced transport processes.⁶⁶ The CNTs reduce the original characteristic microgroove spacing with a width of (453 ± 10) μ m for the fused silica plate.

The effect of this surface topology on the flow is visible in Figure 3c, which shows a homogeneous expanding flow in the inlet region. Within the triangular section of the reactor, the flow is uniformly distributed across the entire reactor width. Upon reaching the porous surface at $y = 4$ mm, fluid–surface interaction becomes apparent. Pronounced channeling effects are observed along the microgrooves, with the incoming flow following the geometric pathways. However, in the region between $x = -5$ and -3 mm along the width of the reactor, deviations from the regular channeling pattern are visible. In this region, one microgroove appears to be partially obstructed, possibly due to irregular CNT growth. The flow field is altered in this region. From a transport perspective, this phenomenon may enhance convective transport in open pathways, whereas partially blocked regions may exhibit increased residence times and diffusion-dominated transport. These details cannot be detected from the time-averaged flow analysis and therefore require a Lagrangian perspective.

The outlet region demonstrates a change in the flow field. Symmetrically arranged vortical structures appear around the outlet. Gravitational effects can be excluded based on the Bond number of $Bo \approx 0.1$ (see Table 1), indicating that surface tension dominates buoyancy forces. This interpretation is further supported by the absence of comparable vortical

structures in the hydrophobic configuration. Since fluid penetration into the hydrophobic MWCNT forest is not expected, fluid–surface interaction is low in that case. Conversely, the microstructured hydrophilic MWCNT forest allows liquid penetration, potentially enhancing fluid–surface interaction between the flow and the porous structure. Therefore, it is possible that detached CNT clusters accumulate near the outlet, leading to partial clogging and an associated increase in local pressure drop. The removal of MWCNTs is confirmed by SEM images through a comparison of the surface before and after flow, which is shown in the Supporting Information (see Figure S2). In particular, regions directly exposed to the flow exhibit a removal of MWCNTs. Possible reasons for the limited mechanical robustness include MWCNT clusters sticking out into the flow, which are subjected to increased hydrodynamic forces due to their exposed surface area. In addition, structural defects within the MWCNTs may promote removal and weak bonds at the MWCNT–substrate interphase can lead to their detachment from the surface.⁶⁷ The debonding energy of individual MWCNTs strongly depends on the interaction between substrate, catalyst particles, and the MWCNTs.^{67,68} Therefore, further investigations are required to identify the removal mechanisms under flow conditions.

Thus, the observed backflow may therefore result from a cluster accumulation in the outlet and could vary among experiments. The resulting backflow is not limited to the outlet but extends upstream into the central region of the reactor. This effect is visible between approximately $x = 3$ and 5 mm along the width of the reactor, where nearly vertical pathlines indicate the presence of recirculating flow structures. Such

recirculation zones may locally modify the residence time and mass transport to the surface, potentially leading to spatial variations in reaction conditions. These fluid dynamic inhomogeneities could influence catalytic performance, although their exact impact cannot be conclusively assessed without detailed concentration measurements of the particular reaction considered.

In the central region, isolated gas bubbles are visible, particularly in the range of $x = -4$ to -3 mm at streamwise positions of approximately $y = 25$ and 30 mm. The upstream bubble in particular exerts an influence on the surrounding flow field, locally deflecting pathlines and perturbing the velocity distribution. For (bio)chemical reactions occurring at the structured surfaces, such bubble-covered regions are effectively inactive, as the gas phase prevents direct liquid-surface contact and thus inhibits mass transport toward and away from the reactive interface.

However, the presence of gas bubbles is limited, especially in comparison with the hydrophobic configuration, where trapped gas bubbles prevent reproducible flow conditions. Furthermore, incomplete liquid-surface contact restricts the direct interaction between the fluid phase and the MWCNT forest, which limits the analysis of the near-surface transport processes. For this reason, the subsequent analysis focuses on the microstructured hydrophilic MWCNT forest, where fluid penetration into the porous layer is expected. To minimize the influence of inlet and outlet effects, the following evaluation is further restricted to the central region of the microreactor. Although this two-dimensional visual analysis already provides insight into characteristic flow features and the global fluid dynamics of the microreactor, three-dimensional and time-resolved data enhance the level of detail and physical understanding.

Three-Dimensional Eulerian Characterization of the Flow Field

The μ -PTV images are processed according to the analysis workflow shown in Figure 2 to get Eulerian velocity fields. The experimental setup and analysis procedure are validated using measurements with the reference plate (nonporous flat fused silica) and precisely known boundary conditions. The validation results are presented in the Supporting Information in Figure S1. Using the validated workflow, the Eulerian velocity field for the flow over the microstructured hydrophilic MWCNT forests is obtained as presented in Figure 4a (corresponding to the central region in Figure 3c). In the following, this surface is termed the microstructured MWCNT forest. The Eulerian velocity field is temporally and spatially averaged for the axial velocity component. Temporal averaging is performed over 290 time steps, and spatial averaging is performed over 5.1 mm in the axial direction y . The velocity field is presented in the x - z plane, displaying the mean axial velocity $\langle v \rangle$. The MWCNT forest is approximated in the microreactor geometry with a uniform height of approximately $150 \mu\text{m}$, which is visualized in Figure 4a. However, the growth of the MWCNT forest is not homogeneous and exhibits irregularities, as shown in the SEM images in Figures 3d and 4b. These images reveal that individual MWCNT clusters extend up to $300 \mu\text{m}$ into the microgroove and toward the top foil. As previously discussed in Figure 3c, such structures locally intrude into the flow domain, partially obstructing pathways for the fluid and tracer particles. Owing to the heterogeneous growth of the MWCNT forests, the SEM

images represent only a limited section of the surface morphology. It is possible that CNT clusters fully block individual channels, leading to even stronger local modifications of the flow behavior.

Considering Figure 4a, the top foil of the microreactor exhibits a bulging deformation of approximately $300 \mu\text{m}$. This deformation was already observed during the measurement process and could be quantitatively characterized due to the three-dimensional nature of the acquired data. Consistent with previous reports of pressure-induced membrane deformation in liquid-cell systems,⁶⁹ the experimentally determined bulging is incorporated into the geometric model used for data evaluation and approximated by a quadratic function. However, for future reactor designs, the foil should be replaced with more mechanically stable substrates.

Distinct flow features can be identified above the nanostructured porous surface. As shown in Figure 4a, the highest velocities occur in the free central region of the channel. A region of reduced velocity is observed directly above the MWCNT forest. This behavior can be further interpreted using Poiseuille's theory. Under the assumptions of steady, fully developed laminar flow (here $Re < 1$), and no-slip boundary conditions on the walls, an analytical solution for the axial velocity profile $v(z)$ can be derived from the Navier–Stokes equations. For a rectangular channel with an aspect ratio $WH_f^{-1} \gg 1$ (here $WH_f^{-1} \approx 7$ to 20 with $H_f \approx (500$ to $1500) \mu\text{m}$), the axial velocity profile according to pressure-driven channel flow is given by

$$v(z) = v_{\max} \left(\frac{4z}{H_f} \left(1 - \frac{z}{H_f} \right) \right) \text{ for } 0 \leq z \leq H_f \quad (11)$$

which exhibits a parabolic form across the channel height. Despite the bulging, the channel can still be approximated as rectangular, as the deformation is small relative to the channel width and does not significantly affect the aspect ratio. This one-dimensional axial velocity profile can be compared with the measured reactor velocity field.

For this purpose, Figure 4c presents measured one-dimensional axial velocity profiles evaluated at five distinct locations satisfying $-1 \text{ mm} \leq x \leq 1 \text{ mm}$ together with the analytical solution following eq 11. The parameters are adjusted to match the measured velocity profile. While the upper boundary of the reactor is an impermeable wall, the lower boundary consists of a porous MWCNT forest. This asymmetry is also reflected in the axial velocity profiles. At the upper wall and the center of the channel, the measured velocity profile agrees well with the axial profile predicted by eq 11. Conversely, a deviation is observed toward the surface with the MWCNT forest. Such deviations have been reported in both numerical and experimental studies and are commonly attributed to the formation of a boundary or transition layer on the surface between a free-flow region and a porous medium.^{20,21} This explanation is consistent with the structure of the porous MWCNT forest, which consists of individual nanotubes, as illustrated in Figure 4b. Due to this nanostructure, the porous interface enables momentum exchange between the external flow and the flow within the porous layer, resulting in a slip condition at the surface. The observation of the development of such a layer therefore provides additional validation of the applied measurement technique. Since a measurable influence of the porous layer on the bulk flow is evident, it can be concluded that the fluid

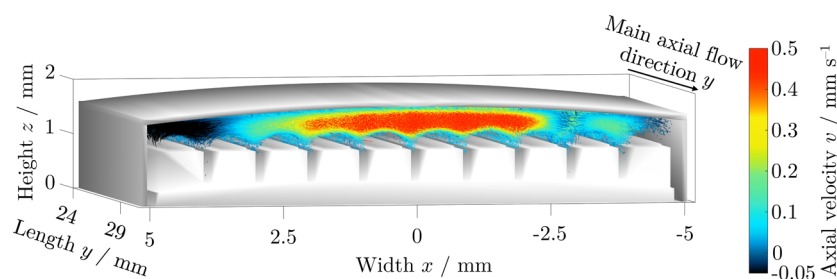


Figure 5. Lagrangian particle trajectories are measured by μ -PTV for the flow over the microstructured MWCNT forest in the central region. The axial velocity component v is represented by the color scale. The visualization is presented from a rear perspective, corresponding to the downstream side of the ROI. The reactor geometry is included for reference. For clarity, the figure shows only every second trajectory.

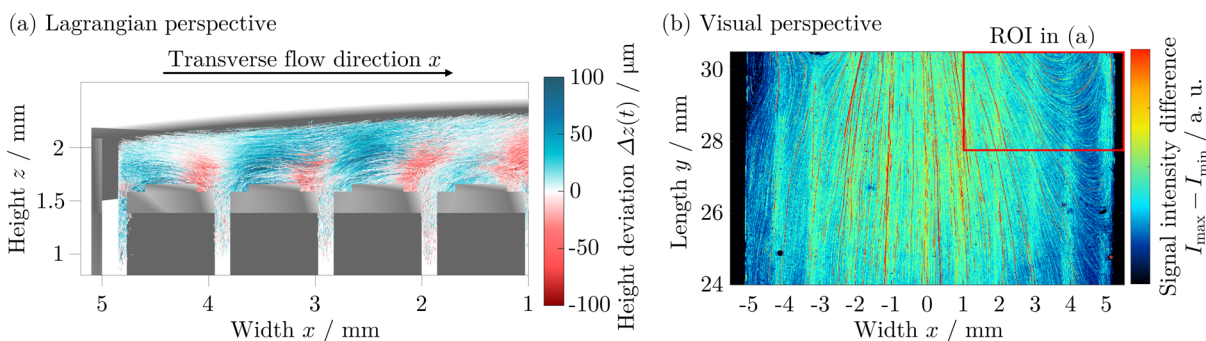


Figure 6. 3D particle trajectories for the flow over the microstructured MWCNT forest shown in (a). The trajectories are colored according to the height deviation calculated from eq 12 along each individual trajectory. The reactor geometry is included for reference. The overall flow direction is out of the paper plane toward the reader. The visualization is shown from a rear perspective, corresponding to the downstream side of the ROI. The ROI in (a) is visualized on the signal intensity map in (b). Thus, the flow over the microgrooves is subjected to transverse flow at the axial position $x \approx 4$ mm and $y \approx 29$ mm.

penetrates into the microstructured MWCNT forest, resulting in a fluid dynamic interaction between the fluid and the MWCNT forest. The forest can reshape under the influence of flow. As a result, a structure can emerge with a characteristic size between the microstructure of the fused silica plate and the nanostructure of the MWCNT forest, which can, for example, include additional flow channels through the forest. Such phenomena and the penetration are relevant for future applications involving (bio)catalytically functionalized nanotubes. The structures within different characteristic size ranges affect transport processes, while the increased accessible surface area can improve catalytic performance. To enable a more detailed analysis, structural parameters such as the porosity and thickness of the porous layers can be varied.

However, experimental constraints, such as the finite DOF, set a limit on acquiring further height information, and the bulging of the top foil contributes to this limitation. Moreover, resolving the flow field even closer to the surface remains a challenge in the present method with the examined surfaces. Because of the heterogeneous and rough morphology of the CNT layer, tracer particles become trapped within the structure. Surface roughness could be reduced by optimized CVD parameters or post-treatment steps, such as mechanical ablation. Thus, the surface roughness could be reduced to obtain smoother porous geometries, thereby enabling flow resolution closer to the porous surface or increased to produce an even larger fluid–surface interaction radius. Furthermore, velocities directly at the surface are small, such that particle displacements during the measurement interval might fall below a detection limit. Nevertheless, the measurement technique used enables a three-dimensional, time-resolved

analysis of the phenomena at the microscale. To the best of our knowledge, experimental data have only been reported on slightly larger scales, without temporal resolution, and in a Lagrangian fashion.²⁰

Lagrangian Characterization of the Particle Trajectories

To complement the Eulerian results, the analysis is extended to a Lagrangian framework, which enables a time-resolved investigation of the particle trajectories. The individual Lagrangian particle trajectories for the microstructured MWCNT forest are thoroughly analyzed in the following. As illustrated in Figure 2, the particle trajectories serve as the basis for this analysis. The 3D Lagrangian velocities are presented in Figure 5.

The phenomena already visible in the top view shown in Figure 3c become more evident when analyzed from a Lagrangian perspective. In particular, the backflow region can also be identified between $x = 3$ and 5 mm, extending throughout the channel height. The Lagrangian analysis enables a more detailed quantification of the interaction between the porous wall and the fluid flow. Thus, the height deviation $\Delta z(t)$ is defined along an individual trajectory of a particle. This criterion serves as an indicator of fluid–surface interaction. It is computed as

$$\Delta z(t) = z(t) - z(t_0) \quad (12)$$

relative to the particle position $z(t_0)$ at time t_0 . Hence, the height deviation quantifies the upward or downward deviation of a particle over the course of its trajectory, providing an indication of vertical transport dynamics and potential penetration into the porous region. Figure 6a illustrates the

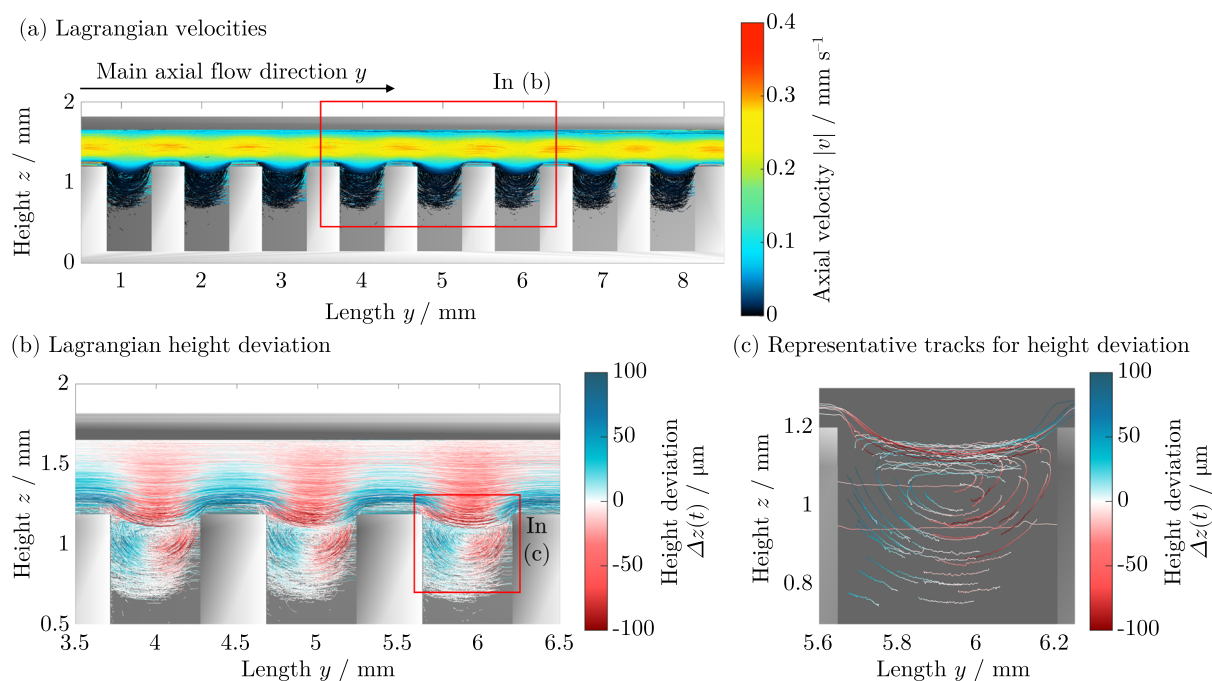


Figure 7. 3D particle trajectories for the flow over a structured fused silica plate with microgrooves perpendicular to the main flow direction in the central region of the reactor during a measurement duration of 40 s. (a) Full ROI in the y - z plane is shown with the main flow direction along the y -axis. The trajectories are colored according to the absolute axial velocity. Only every second Lagrangian trajectory is shown in (a). (b) Representative section of the microgrooves is illustrated. Here, the trajectories are colored according to the height deviation calculated from eq 12 along each individual trajectory. (c) Trajectories longer than 300 time steps ($t > 30$ s) are depicted, with the height deviation used as the color scale. The reactor geometry is included for reference in all cases.

particle trajectories colored using eq 12. The view into the reactor is oriented downstream.

Of particular interest is the region characterized by the beginning of recirculation at $x \approx 4$ mm and $y \approx 29$ mm, as discussed in Figure 3c. This specific ROI considered here is illustrated in Figure 6b. In this area, the flow over the microgrooves is in the transverse direction, whereas the main bulk flow proceeds primarily in the positive y -direction. This lateral flow over the microgrooves of the MWCNT forest is also reflected in the variations in height deviation. The results clearly indicate that the fluid penetrates the microgrooves, as evidenced by the alternating upward and downward particle motions. Consequently, an interaction emerges between the porous surface and the surrounding fluid. Although the structured plate used for the analysis features microgrooves oriented parallel to the main flow direction, the cross-flow region reveals an enhanced interaction between the fluid and the surface.

To study this effect in a controlled way, a fused silica plate is analyzed according to the geometry shown in Figure 1d with microgrooves perpendicular to the main flow direction. This plate does not have a porous or rough surface. Thus, fluid-surface interactions can be examined more systematically, leaving aside the additional effects of the MWCNT forest. Figure 7 represents the Lagrangian analysis for the flow over the (nonporous) structured plate with microgrooves oriented perpendicular to the main flow direction (plate with perpendicular microgrooves) in the central region of the reactor. The main flow direction is along the positive y -axis at a volumetric flow rate of 0.08 mL min^{-1} , which is used for all cases.

Figure 7a shows the absolute axial velocities $|v|$ within the Lagrangian framework. The highest velocities occur in the free central region of the channel. Approaching the surface, the axial velocity gradually decreases. A clear interaction between the fluid and the structured surface becomes visible through wavy particle trajectories induced by the microgrooves. The influence of the surface extends up to a height of approximately $z = 1.5$ mm. Inside the microgrooves, particles are trapped and exhibit low axial velocities of $|v| \lesssim 0.01 \text{ mm s}^{-1}$. To further examine this phenomenon, the height deviation along each trajectory is analyzed based on eq 12. As shown in Figure 7b, the particles exhibit alternating upward and downward motions, consistent with the wavy trajectories. A comparable behavior is also observed for the microstructured MWCNT forest under transverse flow conditions (see Figure 6a). The amplitude of the height deviation for the plate with perpendicular microgrooves is of the order of $|z(t)| \approx 100 \text{ } \mu\text{m}$ within each microgroove. As noted previously, particles are trapped within the microgrooves. Once trapped, the particles undergo a rotational motion within the microgrooves, resulting in local recirculation zones in which the particle partially travels opposite to the main flow direction. The flow structures occurring within the cavities, referred to as microgrooves in the present work, have been investigated in both experimental and analytical studies.^{70–73} The vortices due to flow separation measured in the rectangular microgrooves of the present study exhibit characteristics consistent with those described in the literature.⁷⁰ Further evidence of this behavior is provided by individual long trajectories (exceeding 300 time steps), as shown in Figure 7c. Some particles enter the microgrooves without being trapped, while others remain temporarily confined. Trapped particles can eventually exit the micro-

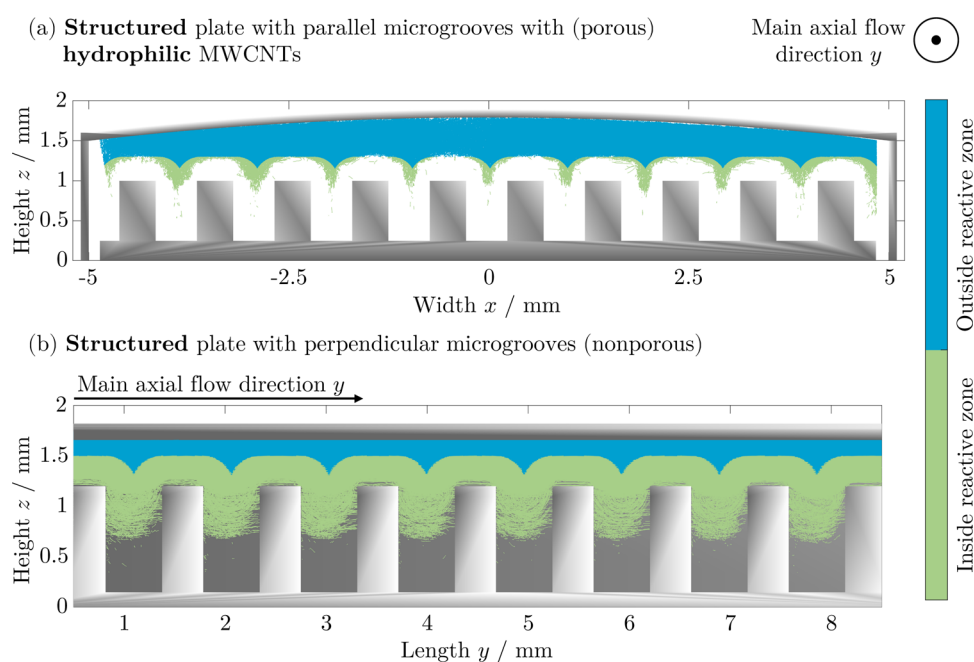


Figure 8. 3D particle trajectories for two flow configurations, with the main flow direction along the y -axis. The axial direction y refers to the starting point within the ROI. (a) Structured plate with parallel microgrooves coated with (porous) hydrophilic MWCNTs in the x - z plane. (b) Structured plate with perpendicular microgrooves in the y - z plane. Trajectories are color-coded: particles that approach closer than $300\ \mu\text{m}$ to the surface (reactive zone) are shown in green, while those remaining outside this zone are shown in blue. The reactor geometry is included for reference, and every second track is shown for both measurements.

grooves and rejoin the main flow. By analyzing Lagrangian velocities, the number of circulation cycles a particle undergoes within one or several microgrooves, as well as its residence time within these structures, can be quantified, further highlighting the importance of the Lagrangian perspective. This approach enables explicit resolution of trapping phenomena, repeated surface contacts, and localized recirculation dynamics. Such effects are often smoothed out or underestimated in averaged Eulerian velocity fields, derived, e.g., by μ -PIV or steady-state flow simulations.

This Lagrangian analysis can be extended further. To this end, a region is defined that extends $300\ \mu\text{m}$ from the surface. Trajectories that are inside this so-called reactive zone are marked in green, as illustrated in Figure 8. The extent of the reaction zone into the reactor is chosen to be $300\ \mu\text{m}$ based on the characteristic growth height of the MWCNT forests (see Figure 3d). The two cases discussed above are compared, namely the structured plate with parallel microgrooves coated with (porous) hydrophilic MWCNTs in Figure 8a and the structured plate with perpendicular microgrooves (nonporous) in Figure 8b. To ensure maximum comparability and minimize parasitic effects, the analysis for both cases is restricted to the central region of the microreactor, away from the inlet and outlet. In future applications within SMART reactors, these MWCNT forests will be functionalized with (bio)catalysts and are therefore intended to actively interact with the fluid.

Considering Figure 8, the interaction of particles with the reaction zone is considerably higher for the plate with perpendicular microgrooves. This difference results from the surface orientation of the microgrooves: parallel microgrooves limit surface interaction, whereas perpendicular microgrooves promote trapping and enhance the fluid–surface contact. To quantify fluid–surface interactions, trajectory-based classifica-

tion is applied. The resulting statistical metrics are summarized in Table 3 for both cases.

Table 3. Statistical Classification of Particle Trajectories Relative to the Reactive Zone, Including Stay Outside, Stay Inside, Enter, Leave, and Reentering Categories Analyzed within the Central Region of the Microreactor (See Figure 8)

analyzed geometry	stay outside reac. zone	stay inside reac. zone	enter reac. zone	leave reac. zone	reentering reac. zone
struc. plate parallel microgrooves (porous)	84%	9.9%	1.4%	1.5%	2.9%
struc. plate perp. microgrooves (nonporous)	20%	56%	0.5%	0.4%	23%

Trajectories that remain outside the reactive zone throughout the entire observation period pass above the surface without a significant wall interaction. These constitute the largest fraction in the case of the microstructured MWCNT forest (84%), which is consistent with laminar flow conditions characterized by stable pathlines and limited transverse mixing. Under such conditions, particle trajectories largely followed the main flow direction. Deviations occur when interactions with the surface take place. Thus, for the plate with perpendicular microgrooves, the fraction of particles outside the reactive zone remains the largest but is significantly reduced to 20% compared to the parallel microstructured MWCNT forest, as trajectories are trapped within the microgrooves. A similar trend is observed for trajectories that remain entirely within the reactive zone and, therefore, exhibit an increased likelihood of surface contact and potential

reaction. While only 9.9% of trajectories fall into this category for the flow over the parallel microstructured MWCNT forest, the corresponding fraction for the fused silica plate with perpendicular grooves amounts to 56%. This behavior can be attributed to structural differences between the surfaces. The porous MWCNT network features only channels oriented parallel to the main flow direction, so particle deviation and surface interaction occur mainly due to a heterogeneous surface and cross-flow phenomena. In contrast, the plate with perpendicular microgrooves promotes particle trapping and recirculation, thereby increasing the probability of surface contact and the potential reaction.

Of particular interest are the trajectories that enter or leave the reactive zone during their motion. Entry into the zone may indicate enhanced transport of reactants toward the surface, whereas exit from the zone can be interpreted as the removal of fluid parcels or reaction products from the near-surface region. For the microstructured MWCNT forest, 1.4% of trajectories are classified as entering the reactive zone and 1.5% as leaving it (see Table 3). In comparison, the plate with perpendicular microgrooves exhibits similar low fractions of 0.5% and 0.4% for entering and exiting trajectories, respectively. A small fraction of trajectories repeatedly enter and leave the reactive zone, which are grouped into a single category, denoted as reentering. This fraction is significant for the plate with perpendicular microgrooves, accounting for 23% of trajectories, compared with only 2.9% for the plate with parallel microgrooves. These results indicate an enhanced fluid–surface interaction for the surface with perpendicular microgrooves.

Overall, the analysis indicates that under current flow conditions, a plate with microgrooves aligned parallel to the main flow direction is inferior to a plate with microgrooves oriented perpendicular to the flow in terms of the fluid–surface interaction. For future investigations of reactive flows, surface structures with perpendicular microgrooves should be considered. These findings underscore the advantages of the Lagrangian framework over the Eulerian approach, as such insights are only accessible through time-resolved particle velocities and trajectory analysis. In summary, the Lagrangian perspective enables a quantitative characterization of fluid–surface interactions. The presented analysis approach therefore provides new opportunities to investigate and optimize the flow behavior within hierarchical porous structures.

CONCLUSION AND OUTLOOK

This work studies the flow over microstructured plates and micro- and nanostructured substrates coated with porous MWCNT forests. To assess their impact on microscale transport processes toward and away from the surface, μ -PTV is used in a custom-built microreactor. The flow over the surfaces under consideration is visualized using fluorescent tracer particles. The measured flow field is analyzed using Eulerian and Lagrangian perspectives.

On the basis of Eulerian velocity fields, a time-averaged analysis reveals that gas bubbles significantly influence the flow field, particularly in the case of hydrophobic MWCNT forests. Thus, the hydrophilicity of the MWCNT forest is essential to ensure surface wetting, thereby promoting reproducible flow conditions. In addition, the three-dimensional Eulerian velocity fields further show a characteristic velocity decay toward the porous interface, which is in good agreement with findings reported in the literature. A Lagrangian analysis is applied to

quantify particle transport on the micrometer scale near the surface. Importantly, structured rectangular microgrooves enhance wall contact of the passive tracer when the flow is oriented perpendicular to the grooves, resulting in a 46%-point increase in the fraction of particles staying within the reactive zone. For the analysis of fluid–surface interactions, a Lagrangian perspective is advantageous as such effects cannot be evaluated in an Eulerian framework.

The used technique enables a three-dimensional, time-resolved flow analysis at the microscale. While previous studies resolved concentration fields using μ -laser-induced fluorescence (μ -LIF),¹² the present work extends this approach to three-dimensional, time-resolved trajectory, transport, and velocity measurements. Future studies may use the μ -PTV data as a foundation for the validation of numerical approaches, enabling detailed analyses of near-surface transport phenomena. The flow characterization presented provides a robust basis for reactive studies in microreactors. For that purpose, coating the MWCNT forests with a catalyst, which can be biobased or a conventional heterogeneous catalyst, would allow the study of reaction yields under characterized fluid dynamic conditions. In this context, concentration field measurements at reactive surfaces using μ -LIF should be performed and correlated with the flow-field measurements. Such coupled analysis enables a comprehensive understanding of the interaction between flow, mass transport, and surface reactions for the application in SMART reactors.

ASSOCIATED CONTENT

Data Availability Statement

The particle trajectory data obtained from the Shake-The-Box analysis performed with DaVis 11.2 (LaVision GmbH, Germany), along with the MATLAB (MathWorks Inc., USA) generated data sets and selected processing scripts, are openly available in TORE at [10.15480/882.16568](https://doi.org/10.15480/882.16568).⁷⁴ The scripts are based on ref 75 and further developed for the present study. Raw camera recordings can be made available from the corresponding author upon reasonable request.

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.iecr.6c01159>.

Validation of the experimental setup and the analysis procedure; SEM images of flow-induced MWCNT removal (PDF)

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Notes

The authors declare no competing financial interest.

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ABBREVIATIONS

3D	three-dimensional
μ -LIF	μ -laser-induced fluorescence
μ -PIV	μ -particle image velocimetry
μ -PTV	μ -particle tracking velocimetry
COC	cyclic olefin copolymer
CNT	carbon nanotube
CVD	chemical vapor deposition
DOF	depth of field
DFG	Deutsche Forschungsgemeinschaft (German Research Foundation)
MWCNT	multi-walled carbon nanotube
ROI	region of interest
SEM	scanning electron microscope
VSC	volume self-calibration

NOMENCLATURE

Roman Symbols

D_h	hydraulic diameter/ μm
d_p	tracer particle diameter/ μm
g	gravitational acceleration/ m s^{-2}
H	height of microreactor/ mm
H_f	flow channel height of microreactor/ μm
$H_{f,\min}$	smallest flow channel height of microreactor/ μm
$I_{\max}$	maximum signal intensity/—
$I_{\min}$	minimum signal intensity/—
L	length of microreactor/ mm
L_x	length of processed volume in x / mm
L_y	length of processed volume in y / mm
L_z	length of processed volume in z / μm
T	temperature/ $^{\circ}\text{C}$
t	time/ s
Δt_M	temporal resolution for measurement/ s
Δt_A	temporal resolution for analysis/ s
t_0	initial time of each trajectory/ s
u	velocity in x -direction/ mm s^{-1}
v	axial velocity in y -direction/ mm s^{-1}
$\vec{v}$	velocity vector/ mm s^{-1}
$\bar{v}$	mean axial velocity in y -direction/ mm s^{-1}
$v_{\max}$	maximal axial velocity in y -direction/ mm s^{-1}
v_s	settling velocity of particles/ $\mu\text{m s}^{-1}$
W	width of the microreactor/ mm
w	velocity in z -direction/ mm s^{-1}
x	width/ mm
x	spatial position/ mm
x'	particle position/ m
x_0	initial position of a particle/ m
y	length/ mm
z	height/ mm

Greek Symbols

η	viscosity/ Pa s
ν	kinematic viscosity/ $\text{m}^2 \text{s}^{-1}$
ρ_G	gas density/ kg m^{-3}
ρ_L	fluid density/ kg m^{-3}
ρ_p	tracer particle density/ kg m^{-3}
λ	wavelength/ nm
σ	surface tension/ N m^{-1}
τ_F	characteristic time scale of the flow/ s
τ_p	particle relaxation time/ s

Dimensionless Numbers

Bo	Bond number
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Re Reynolds number
 Re_p particle Reynolds number
 St Stokes number
 St_g gravitational Stokes number

REFERENCES

- (1) Beavers, G. S.; Joseph, D. D. Boundary conditions at a naturally permeable wall. *J. Fluid Mech.* **1967**, *30*, 197–207.
- (2) Naqvi, S. B.; Bottaro, A. Interfacial conditions between a free-fluid region and a porous medium. *Int. J. Multiphase Flow* **2021**, *141*, No. 103585.
- (3) Freund, H.-J.; Meijer, G.; Scheffler, M.; Schlögl, R.; Wolf, M. CO oxidation as a prototypical reaction for heterogeneous processes. *Angew. Chem., Int. Ed.* **2011**, *50*, 10064–10094.
- (4) Cao, B.-Y.; Sun, J.; Chen, M.; Guo, Z.-Y. Molecular momentum transport at fluid-solid interfaces in MEMS/NEMS: a review. *Int. J. Mol. Sci.* **2009**, *10*, 4638–4706.
- (5) Weckhuysen, B. M. Chemical imaging of spatial heterogeneities in catalytic solids at different length and time scales. *Angew. Chem., Int. Ed.* **2009**, *48*, 4910–4943.
- (6) Kim, J.; Kim, H.-S.; Han, S.; Lee, J.-Y.; Oh, J.-E.; Chung, S.; Park, H.-D. Hydrodynamic effects on bacterial biofilm development in a microfluidic environment. *Lab Chip* **2013**, *13*, 1846–1849.
- (7) He, B.; Doubrovinski, K.; Polyakov, O.; Wieschaus, E. Apical constriction drives tissue-scale hydrodynamic flow to mediate cell elongation. *Nature* **2014**, *508*, 392–396.
- (8) Chook, K. Y.; Aroua, M. K.; Gew, L. T. Enzyme biocatalysis for sustainability applications in reactors: A systematic review. *Ind. Eng. Chem. Res.* **2023**, *62*, 10800–10812.
- (9) Purevdorj, B.; Costerton, J. W.; Stoodley, P. Influence of hydrodynamics and cell signaling on the structure and behavior of *Pseudomonas aeruginosa* biofilms. *Appl. Environ. Microbiol.* **2002**, *68*, 4457–4464.
- (10) Wang, H.; Hong, L.; Chamorro, L. P. Micro-scale particle tracking: from conventional to data-driven methods. *Micromachines* **2024**, *15*, No. 629.
- (11) Khinast, J. G.; Bauer, A.; Bolz, D.; Panarello, A. Mass-transfer enhancement by static mixers in a wall-coated catalytic reactor. *Chem. Eng. Sci.* **2003**, *58*, 1063–1070.
- (12) Hoffmann, M.; Schlüter, M.; Rübiger, N. Experimental investigation of liquid–liquid mixing in T-shaped micro-mixers using μ -LIF and μ -PIV. *Chem. Eng. Sci.* **2006**, *61*, 2968–2976.
- (13) Frey, T.; Kexel, F.; Dittmer, K. R.; Bohne, S.; Hoffmann, M.; Trieu, H. K.; Schlüter, M. A novel approach for visualizing mixing phenomena of reactive liquid-liquid flows in Milli-and micro-channels. *Front. Chem. Eng.* **2022**, *4*, No. 874019.
- (14) Cai, S.; Jin, Y.; Lin, Y.; He, Y.; Zhang, P.; Ge, Z.; Yang, W. Micromixing within microfluidic devices: Fundamentals, design, and fabrication. *Biomicrofluidics* **2023**, *17*, No. 061503.
- (15) Menzel, F.; Conradi, B.; Rodenacker, K.; Gorbushina, A. A.; Schwibbert, K. Flow chamber system for the statistical evaluation of bacterial colonization on materials. *Materials* **2016**, *9*, No. 770.
- (16) Klein, E. M.; Heintz, H.; Wurst, R.; Lapp, C. J.; Edel, M.; Hähl, H.; Jacobs, K.; Gescher, J. Tailored enzyme expression modifies *Shewanella oneidensis* biofilms and increases current density. *New Biotechnol.* **2025**, *89*, 1–10.
- (17) Kirschning, A.; Altwicker, C.; Dräger, G.; Harders, J.; Hoffmann, N.; Hoffmann, U.; Schönfeld, H.; Solodenko, W.; Kunz, U. PASSflow Syntheses Using Functionalized Monolithic Polymer/Glass Composites in Flow-Through Microreactors. *Angew. Chem., Int. Ed.* **2001**, *40*, 3995–3998.
- (18) Asanomi, Y.; Yamaguchi, H.; Miyazaki, M.; Maeda, H. Enzyme-immobilized microfluidic process reactors. *Molecules* **2011**, *16*, 6041–6059.
- (19) Wen, K.; Gorbushina, A. A.; Schwibbert, K.; Bell, J. Microfluidic platform with precisely controlled hydrodynamic parameters and integrated features for generation of microvortices to accurately form and monitor biofilms in flow. *ACS Biomater. Sci. Eng.* **2024**, *10*, 4626–4634.
- (20) Wu, Z.; Mirbod, P. Experimental analysis of the flow near the boundary of random porous media. *Phys. Fluids* **2018**, *30*, No. 047103.
- (21) Kang, J.; Wang, M. Brinkman double-layer model for flow at a free-porous interface. *Int. J. Mech. Sci.* **2024**, *263*, No. 108770.
- (22) Coasne, B. Multiscale adsorption and transport in hierarchical porous materials. *New J. Chem.* **2016**, *40*, 4078–4094.
- (23) Friend, C. M.; Xu, B. Heterogeneous catalysis: a central science for a sustainable future. *Acc. Chem. Res.* **2017**, *50*, 517–521.
- (24) Huber, P. Soft matter in hard confinement: phase transition thermodynamics, structure, texture, diffusion and flow in nanoporous media. *J. Phys.:Condens. Matter* **2015**, *27*, No. 103102.
- (25) Shah, K. A.; Tali, B. A. Synthesis of carbon nanotubes by catalytic chemical vapour deposition: A review on carbon sources, catalysts and substrates. *Mater. Sci. Semicond. Process.* **2016**, *41*, 67–82.
- (26) De Jong, F. J. The Filtration of Nanoparticles in Nanoporous Media; PhD dissertation, Technische Universität Hamburg-Harburg: Hamburg, Germany, 2016.
- (27) Rao, R.; Pint, C. L.; Islam, A. E.; Weatherup, R. S.; Hofmann, S.; Meshot, E. R.; Wu, F.; Zhou, C.; Dee, N.; Amama, P. B.; Carpena-Núñez, J.; Shi, W.; Plata, D. L.; Penev, E. S.; Yakobson, B. I.; Balbuena, P. B.; Bichara, C.; Futaba, D. N.; Noda, S.; Shin, H.; Kim, K. S.; Simard, B.; Mirri, F.; Pasquali, M.; Fornasiero, F.; Kauppinen, E. I.; Arnold, M.; Cola, B. A.; Nikolaev, P.; Arepalli, S.; Cheng, H.-M.; Zakharov, D. N.; Stach, E. A.; Zhang, J.; Wei, F.; Terrones, M.; Geohagan, D. B.; Maruyama, B.; Maruyama, S.; Li, Y.; Adams, W. W.; Hart, A. J. Carbon nanotubes and related nanomaterials: critical advances and challenges for synthesis toward mainstream commercial applications. *ACS Nano* **2018**, *12*, 11756–11784.
- (28) Chen, G.; Davis, R. C.; Futaba, D. N.; Sakurai, S.; Kobashi, K.; Yumura, M.; Hata, K. A sweet spot for highly efficient growth of vertically aligned single-walled carbon nanotube forests enabling their unique structures and properties. *Nanoscale* **2016**, *8*, 162–171.
- (29) Sherbi, M.; Wesner, A.; Wisniewski, V. K.; Bukowski, A.; Velichkova, H.; Fiedler, B.; Albert, J. Superior CNT-supported bimetallic RuCu catalyst for the highly selective hydrogenolysis of glycerol to 1, 2-propanediol. *Catal. Sci. Technol.* **2021**, *11*, 6649–6653.
- (30) Moyer-Vanderburgh, K.; Park, S. J.; Fornasiero, F. Growth of carbon nanotube forests on flexible metal substrates: Advances, challenges, and applications. *Carbon* **2023**, *206*, 402–421.
- (31) Luo, Z.; Wang, X.; Yang, D.; Zhang, S.; Zhao, T.; Qin, L.; Yu, Z.-Z. Photothermal hierarchical carbon nanotube/reduced graphene oxide microspherical aerogels with radially orientated microchannels for efficient cleanup of crude oil spills. *J. Colloid Interface Sci.* **2020**, *570*, 61–71.
- (32) Bychko, I.; Bazylevska, A.; Khavrus, V.; Tang, J.; Strizhak, P. Ni@CNT nanocomposites as novel highly effective carbocatalysts for Hydrogenation/Dehydrogenation reactions. *J. Catal.* **2023**, *427*, No. 115108.
- (33) Vanoye, L.; Guichet, B.; Rivera-Cárcamo, C.; Contreras, R. C.; de Bellefon, C.; Meille, V.; Serp, P.; Philippe, R.; Favre-Régouillon, A. Process intensification of the catalytic hydrogenation of squalene using a Pd/CNT catalyst combining nanoparticles and single atoms in a continuous flow reactor. *Chem. Eng. J.* **2022**, *441*, No. 135951.
- (34) Yu, C.-H.; Yoon, J.-H.; Kim, H.-B. Development and validation of stereoscopic micro-PTV using match probability. *J. Mech. Sci. Technol.* **2009**, *23*, 845–855.
- (35) Llamas, C. G.; Spille, C.; Kastens, S.; Paz, D. G.; Schlüter, M.; von Kameke, A. Potential of Lagrangian analysis methods in the study of chemical reactors. *Chem. Ing. Technol.* **2020**, *92*, 540–553.
- (36) Polymers, T. A. Topas COC datasheets 2026 https://topas.com/wp-content/uploads/2023/04/TDS-S013S-04_e_US.pdf (accessed Feb 20, 2026).
- (37) Stephan, P.; Kabelac, S.; Kind, M.; Mewes, D.; Schaber, K.; Wetzel, T. VDI-Wärmeatlas: Fachlicher Träger VDI-Gesellschaft Verfahrenstechnik und Chemieingenieurwesen; Springer-Verlag, 2019.

- (38) Shui, L.; Eijkel, J. C.; Van den Berg, A. Multiphase flow in microfluidic systems—Control and applications of droplets and interfaces. *Adv. Colloid Interface Sci.* **2007**, *133*, 35–49.
- (39) Cengel, Y. A.; Cimbala, J. M. *Fluid Mechanics: Fundamentals and Applications*, 3rd ed.; McGraw-Hill Education: New York, 2013.
- (40) Tišma, M.; Zelić, B.; Vasić-Rački, Đ.; Žnidaršič-Plazl, P.; Plazl, I. Modelling of laccase-catalyzed L-DOPA oxidation in a microreactor. *Chem. Eng. J.* **2009**, *149*, 383–388.
- (41) Bratsun, D.; Mizev, A.; Siraev, R.; Krasnyakova, E. Control of mixing in a continuous-flow microreactor with an integrated bubble. *J. Flow Chem.* **2025**, *15*, 59–66.
- (42) Lucci, F.; Castro, N. D.; Rostami, A. A.; Oldham, M. J.; Hoeng, J.; Pithawalla, Y. B.; Kuczaj, A. K. Characterization and modeling of aerosol deposition in Vitrocell exposure systems—exposure well chamber deposition efficiency. *J. Aerosol Sci.* **2018**, *123*, 141–160.
- (43) Ouellette, N. T.; O'malley, P.; Gollub, J. P. Transport of finite-sized particles in chaotic flow. *Phys. Rev. Lett.* **2008**, *101*, No. 174504.
- (44) Parker, J. T.; DeBardinis, J.; Mäkiharju, S. A. Enhanced laboratory x-ray particle tracking velocimetry with newly developed tungsten-coated O (50 μm) tracers. *Exp. Fluids* **2022**, *63*, 184.
- (45) King, R. P. *Introduction to Practical Fluid Flow*; Elsevier: 2002.
- (46) Bazant, M. Z.; Bush, J. W. A guideline to limit indoor airborne transmission of COVID-19. *Proc. Natl. Acad. Sci. U.S.A.* **2021**, *118*, No. e2018995118.
- (47) Urizarna-Carasa, J.; Ruprecht, D.; von Kameke, A.; Padberg-Gehle, K. Relevance of the Basset history term for Lagrangian particle dynamics. *Chaos* **2025**, *35*, No. 073122.
- (48) Raffel, M.; Willert, C. E.; Scarano, F.; Kähler, C. J.; Wereley, S. T.; Kompenhans, J. *Particle Image Velocimetry: A Practical Guide*; Springer: 2018.
- (49) Samimy, M.; Lele, S. Motion of particles with inertia in a compressible free shear layer. *Phys. Fluids A* **1991**, *3*, 1915–1923.
- (50) Martins, F. J. W. A.; Kirchmann, J.; Kronenburg, A.; Beyrau, F. Quantification and mitigation of PIV bias errors caused by intermittent particle seeding and particle lag by means of large eddy simulations. *Meas. Sci. Technol.* **2021**, *32*, 104006.
- (51) Gottmann, J.; Hermans, M.; Repiev, N.; Ortmann, J. Selective laser-induced etching of 3D precision quartz glass components for microfluidic applications—up-scaling of complexity and speed. *Micromachines* **2017**, *8*, No. 110.
- (52) Kröning, K.; Krause, B.; Pötschke, P.; Fiedler, B. Nanocomposites with p-and n-type conductivity controlled by type and content of nanotubes in thermosets for thermoelectric applications. *Nanomaterials* **2020**, *10*, No. 1144.
- (53) Butt, H.-J.; Kappl, M. *Surface and Interfacial Forces*; John Wiley & Sons: 2018.
- (54) Senn, B.; Pigram, P.; Liesegang, J. Surface electrical resistivity and wettability study of fused silica. *Surf. Interface Anal.* **1999**, *27*, 835–839.
- (55) Liu, H.; Zhai, J.; Jiang, L. Wetting and anti-wetting on aligned carbon nanotube films. *Soft Matter* **2006**, *2*, 811.
- (56) Han, Z.; Tay, B.; Tan, C.; Shakerzadeh, M.; Ostrikov, K. Electrowetting Control of Cassie-to-Wenzel Transitions in Superhydrophobic Carbon Nanotube-Based Nanocomposites. *ACS Nano* **2009**, *3*, 3031–3036.
- (57) Van Hooijdonk, E.; Bittencourt, C.; Snyders, R.; Colomer, J.-F. Functionalization of vertically aligned carbon nanotubes. *Beilstein J. Nanotechnol.* **2013**, *4*, 129–152.
- (58) Wieneke, B. Volume self-calibration for 3D particle image velocimetry. *Exp. Fluids* **2008**, *45*, 549–556.
- (59) Wieneke, B. Improvements for volume self-calibration. *Meas. Sci. Technol.* **2018**, *29*, No. 084002.
- (60) Schanz, D.; Schröder, A.; Gesemann, S.; Michaelis, D.; Wieneke, B. PIV13; 10th International Symposium on Particle Image Velocimetry, Delft: The Netherlands, 2013.
- (61) Schanz, D.; Gesemann, S.; Schröder, A. Shake-The-Box: Lagrangian particle tracking at high particle image densities. *Exp. Fluids* **2016**, *57*, 70.
- (62) Schröder, A.; Schanz, D. 3D Lagrangian particle tracking in fluid mechanics. *Annu. Rev. Fluid Mech.* **2023**, *55*, 511–540.
- (63) Janke, T.; Michaelis, D. 14th international symposium on particle image velocimetry. 2021; Vol. 1.
- (64) Kameke, A.; Uphoff, R.; Steuwe, E.; Nissen, J.; Hoffmann, M.; Schlüter, M.; Kexel, F. Experimental analysis of time resolved three-dimensional velocity and vorticity fields behind single rising bubbles using Lagrangian particle tracking velocimetry. *Chem. Eng. Sci.* **2026**, *326*, No. 123403.
- (65) Aguirre-Pablo, A. A.; Aljedaani, A. B.; Xiong, J.; Idoughi, R.; Heidrich, W.; Thoroddsen, S. T. Single-camera 3D PTV using particle intensities and structured light. *Exp. Fluids* **2019**, *60*, 25.
- (66) Pérez-Ramírez, J.; Christensen, C. H.; Egeblad, K.; Christensen, C. H.; Groen, J. C. Hierarchical zeolites: enhanced utilisation of microporous crystals in catalysis by advances in materials design. *Chem. Soc. Rev.* **2008**, *37*, 2530–2542.
- (67) Lahiri, I.; Lahiri, D.; Jin, S.; Agarwal, A.; Choi, W. Carbon nanotubes: how strong is their bond with the substrate? *ACS Nano* **2011**, *5*, 780–787.
- (68) Ageev, O. A.; Blinov, Y. F.; Il'ina, M. V.; Il'in, O. I.; Smirnov, V. A.; Tsukanova, O. G. Study of adhesion of vertically aligned carbon nanotubes to a substrate by atomic-force microscopy. *Phys. Solid State* **2016**, *58*, 309–314.
- (69) Bottauscio, N.; Ramadan, M.; Lin, O.; Lyu, Z.; Kim, C.; Tidemand-Lichtenberg, S.; Lajer, J.; Chen, Q.; Jensen, E. C.; Mølhav, K. S. Trapping Nanoparticles in Nanofluidic Traps Using a Monolithic Liquid Phase-TEM Chip. *Microsc. Microanal.* **2025**, *31*, 1834–1835.
- (70) Taneda, S. Visualization of separating Stokes flows. *J. Phys. Soc. Jpn.* **1979**, *46*, 1935–1942.
- (71) Moffatt, H. K. Viscous and resistive eddies near a sharp corner. *J. Fluid Mech.* **1964**, *18*, 1–18.
- (72) Shen, C.; Floryan, J. M. Low Reynolds Number Flow Over Cavities. *Phys. Fluids* **1985**, *28*, 3191–3202.
- (73) Pan, F.; Acrivos, A. Steady Flows in Rectangular Cavities. *J. Fluid Mech.* **1967**, *28*, 643–655.
- (74) Merbach, T.; Nissen, J.; Riebesehl, F.; Rennpfert, L.; Steuwe, E.; Hoffmann, M.; von Kameke, A. Data supplement for publication: Lagrangian μ -Particle Tracking Velocimetry Analysis of Microscale Flow over Structured Surfaces, 2026 DOI: 10.15480/882.16568.
- (75) von Kameke, A. WakeMix – Lagrangian particle tracking data and bubble shapes for rising bubble experiments (data paper1). 2026, DOI: 10.15480/882.16724.