



Sedimentation behavior of suspensions in milliflow reactors

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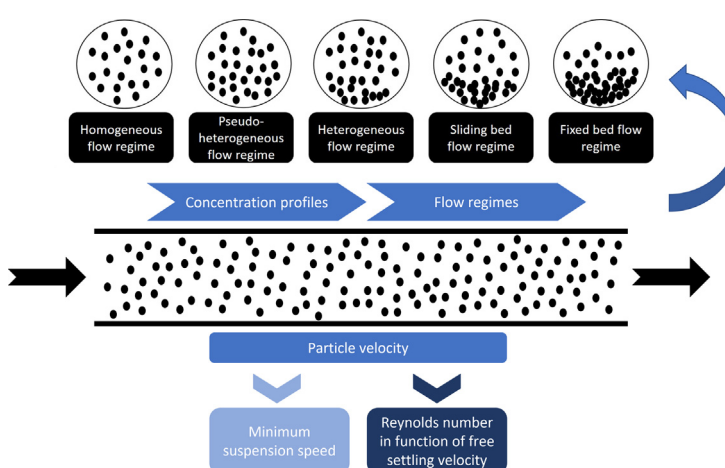
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HIGHLIGHTS

- Concentration and velocity profiles of solids to determine the flow regimes.
- Determination of the minimum suspension speed for different liquid and solid properties.
- Prediction of the Reynolds number for suspensions via the free settling velocity.

GRAPHICAL ABSTRACT



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ABSTRACT

The increasing industrial interest in performing heterogeneous solid–liquid reactions in tubular reactors requires control over sedimentation and accurate data on the solid velocity in order to optimize residence time distribution. In this research, concentration profiles, velocity profiles and the minimum suspension speed of non-interacting solids in milliflow reactors are determined via image analysis and particle image velocimetry. First, the five different flow regimes are identified. Then, the minimum suspension speed, ensuring a homogeneous/pseudo-heterogeneous flow regime is observed to be between 0.7 and 1 m/s depending on liquid and solid properties. Reducing the liquid viscosity and density difference, decreases the minimum suspension speed. Based on the free settling velocity, the Reynolds number to avoid sedimenting of particles can be calculated. The identification of minimum suspension speed and Reynolds number were found to be the determining factors enabling broader application of heterogeneous solid–liquid reactions in milliflow reactors, in academic and industrial environments.

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1. Introduction

Traditionally, most pharmaceutical and fine chemical reactions are performed in batch reactors. The research conducted by Roberge (2005) estimates that 59 % of the reactions in the fine/pharmaceutical industry would benefit from a continuous process (Roberge et al., 2005). One of the major hurdles in flow chemistry is the handling of multiphase systems, specifically a solid–liquid suspension velocity to avoid sedimentation in milliflow reactors is unknown (Wu & Kuhn, 2014). In general, two different forces determine the settling velocity of a particle in a quiescent fluid, namely the gravitational force and the drag force. At a certain point, both forces will be in equilibrium when a particle is sedimenting. The particle then has zero acceleration and the constant vertical velocity of the particle at this point is defined as the terminal velocity or the free settling velocity. Perry and Green (1984) have proposed an equation to calculate the free settling velocity, V_t , as is shown in Eq. (1).

$$V_t = \left(\frac{4gd_p(\rho_s - \rho_l)}{3C_D\rho_l} \right)^{1/2} \quad (1)$$

With g (m/s^2) the gravitational constant, d_p (m) the particle diameter, ρ_s (kg/m^3) the solid density, ρ_l (kg/m^3) the liquid density and C_D (-) the drag coefficient which is dependent on the particle Reynolds number, Re_p . The particle Reynolds number can be calculated with Eq. (2) and the hydrodynamic regimes for settling particles with their corresponding expression of C_D are shown in Table 1.

$$Re_p = \frac{\rho_l V_t d_p}{\mu} \quad (2)$$

With ρ_l (kg/m^3) the liquid density, V_t (m/s) the free settling velocity, d_p (m) the particle diameter and μ (kg/m s) the liquid dynamic viscosity.

In addition to the gravitational and drag force, literature also mentions influences of the Saffman force, the Magnus force, lift force, virtual mass, lubrication,... in flow reactors (Farajpour et al., 2019b; Shang et al., 2013; Solsona et al., 2020; Wang & Fan, 2013). These forces influence the settling velocity of particles and the particle concentration in the radial direction in milliflow reactors. Five different flow regimes can occur with a heterogeneous solid–liquid feed: homogeneous, pseudo-heterogeneous, heterogeneous, heterogeneous with sliding bed and heterogeneous with fixed bed (Faddick, 1970; Peker & Helvaci, 2008). These regimes, with their corresponding particle velocity and volumetric concentration profiles, are visualized in Fig. 1.

Flow regimes are widely discussed, modelled and described in literature. However, the experimental sources either determine the concentration profiles or the minimum settling velocity. They are able to discriminate between heterogeneous and sliding bed regime, but cannot determine all the different flow regimes. When the particles are evenly distributed over the tube cross section a homogeneous flow regime is established. The homogeneous flow regime is only possible if the flow rate is high enough to create turbulence or if chaotic advection exists, resulting in vortices which

prevent settling of particles. If the suspension flow rate, and thus turbulence intensity decreases, accumulation of particles in the lower half of the tubing occurs, resulting in a pseudo-heterogeneous and eventually heterogeneous flow regime. The corresponding velocity profile also becomes less symmetric. By further decreasing the flow rate, particles will sediment at the bottom of the tube, creating a heterogeneous regime with sliding bed. The whole layer moves, which is visible in the velocity profile, but only the top particles of the layer are entrained by the fluid, thus having the highest velocity. In the heterogeneous regime with fixed bed, the particles at the bottom of the sedimented layer are not moving, while the particles at the top of that layer tend to roll over the layer (Doron & Barnea, 1996). The occurrence of a fixed bed can lead to clogging of a milliflow reactor (Christopher E. Brennen, 2005; Polansky, 2014; Reynier et al., 2011). A heterogeneous regime with sliding or fixed bed indicates that there is a large difference in the residence time between the particles and the liquid due to the sedimentation of particles. This difference can have a negative influence on reaction rate and product yield because these parameters depend on the concentration of the reagent and/or product.

To determine the minimum suspension velocity in milliflow reactors, it is important to measure particle velocity profiles with high precision. Several techniques are available to measure or to calculate particle velocities inside a milliflow reactor. The main used methods are laser Doppler velocimetry (LDV), pulsed ultrasonic Doppler velocimetry (PUDV), particle image velocimetry (PIV) and computational fluid dynamics (CFD) (Doran, 2012; Dramicanin, 2018; Eesa & Barigou, 2009; Gao et al., 2013; Ozalp et al., 2013; Posada et al., 2014; Sad Chemloul et al., 2009; Tarbell et al., 1986; Tu et al., 2008). The principle, advantages and disadvantages of the techniques are summarized in Table 2.

This study investigates, via PIV and image analysis, particle concentration and particle velocity profiles and determines the flow regimes of heterogeneous solid–liquid mixtures in milliflow reactors, yielding macroscopic and microscopic behavior of solids in tubular reactors. With the use of PIV, it is possible to either determine the particle velocity and to do image analysis. Via concentration and velocity profiles, all flow regimes are determined and compared with other particle sizes. Finally, a criterion is proposed to ensure a homogeneous or pseudo-heterogeneous flow regime and corresponding suspension velocities for different solvents are discussed. The influence of particle concentration, particle density, particle size and liquid density and liquid viscosity is investigated.

2. Material & methods

2.1. Setup

The experimental setup is shown in Fig. 2, it consisted of a beaker (1L) with an overhead stirrer, stirring between 800 and 1,000 rpm to obtain a homogeneous suspension. Next, a centrifugal pump (M510S-V, TCS micropumps), was used to recycle the heterogeneous mixture. The maximum flow rate of water was 1,200 ml/min. The tubing was PFA tubing (Polyfluor) with OD of ¼" and ID of 4.35 mm and had a length of 1.25 m until the connector. To record high quality images of the suspension inside the tubing, a quartz glass tube with a length of 32 cm, OD of 6.2 mm and ID of 4 mm was mounted in front of the high speed camera equipped with a microscopic lens (Fastcam mini AX100, Photron – 12x zoom lens with x2 F mount adapter, Navitar) and 18 cm after the connection between PFA and glass tubing. CFD simulations showed steady state conditions at the recording place after the small constriction of the connection. The magnification of the microscopic lens was set to 3x. Videos were recorded with a

Table 1
Particle Reynolds number and drag coefficient in different hydrodynamic regimes (Paul et al., 2004).

Hydrodynamic regime	Particle Reynolds number	Drag coefficient
Laminar (Stokes' law)	$Re_p < 0.3$	$C_D = 24/Re_p$
Intermediate	$0.3 < Re_p < 1,000$	$C_D = 18.5/Re_p^{3/5}$
Turbulent (Newton's law)	$1,000 < Re_p < 34 \times 10^4$	$C_D = 0.445$

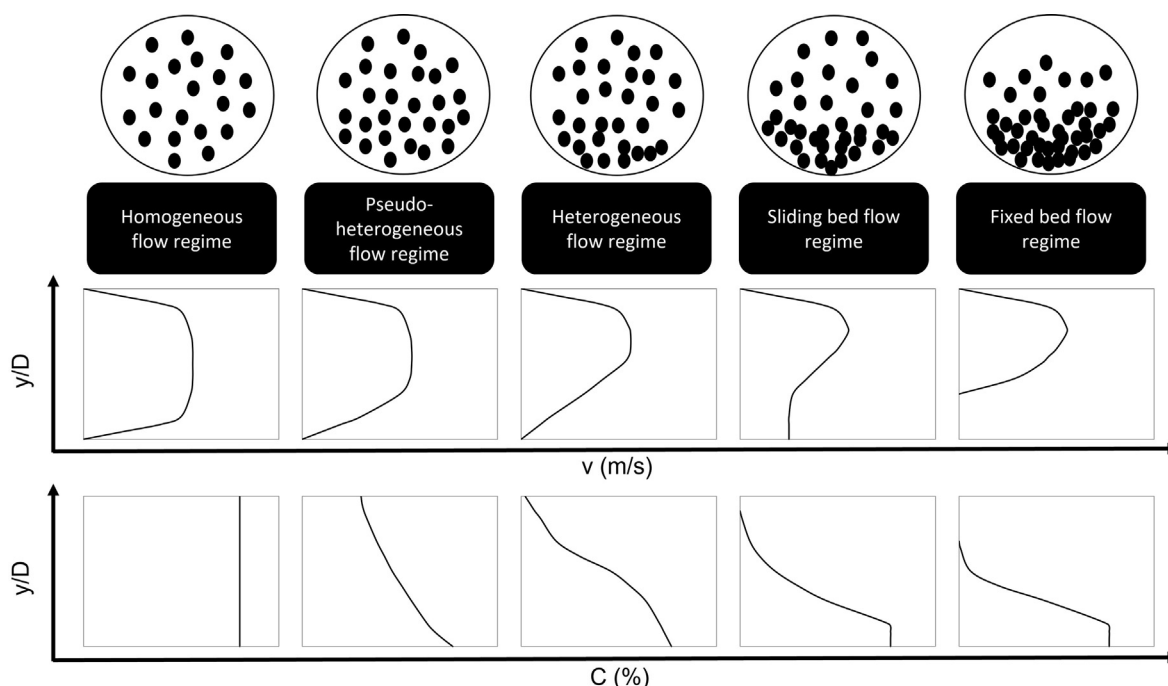


Fig. 1. Flow regimes with particle velocity, v , and volumetric concentration profiles, C , along the y axis position over tube diameter (y/D) in milliflow reactors (Miedema, 2016).

framerate of 5,000 fps, a resolution of 896×896 pixels² and a shutter speed of 1.04 μ s to determine the concentration and velocity profiles. To achieve sharp images and videos, a strong LED light (Highpower LED, CREE) with a maximum of 2180 lm at 1 A was installed behind the tubing. This LED light was controlled by a voltage controller with a constant current of 0.3 A and the LED was placed on a heat sink. At the end of the recirculation loop, a Coriolis flow meter (M15-AGD-22-0-S, Bronkhorst) was implemented to measure the flow rate of the suspension.

2.2. Suspension properties and experiments

Multiple parameters that have an influence on the concentration and velocity profile in milliflow reactors were investigated: particle size, density and concentration (vol%) and liquid density and dynamic viscosity. The particle size of the used products was analyzed and summarized in the [supplementary information](#) (section determination of particle size distribution). The viscosity of suspensions is independent of particle size and for low solid fractions it can be calculated with the Einstein equation, Eq. (3) (Konijn et al., 2014).

$$\mu_{\text{suspension}} = \mu_0 \left(1 + \frac{5}{2} \phi \right) \quad (3)$$

With μ_0 (kg/m s), the dynamic viscosity of the liquid and with ϕ , the volumetric particle concentration. The properties of the particles and liquids are summarized in [Table 3](#) and the experimental suspensions are shown in [Table 4](#).

The KBr-solution with a density of 1,260 kg/m³ was made by dissolving 378.3 g in 1 L water (3.18 mol/l) and ethylene glycol was diluted to 10 m% in water. Both liquid densities were measured by weighing with a pycnometer (0.0001 g accuracy) and double checked by measuring the density with the Coriolis mass flow meter. The particle concentration was varied between 0.4 vol% and 1.0 vol% since higher volumetric concentrations caused too much light reflection and scattering resulting in unus-

able images. In each experiment, the suspension flow rates were typically ranging from 350 to 1,000 ml/min for the suspensions with glass beads and from 200 to 1000 ml/min for suspensions with PMMA and PCM, which resulted in laminar and turbulent flow. All experiments started at the highest possible flow rate delivered by the centrifugal pump to evenly distribute all particles in the setup. Next, the flow rate under investigation was programmed and images are recorded after 2 min which ensures steady state conditions.

2.3. Determination of the velocity profile of the particles

To determine the particle velocity the PIV software PIVlab (Matlab software) developed by William Thielicke, was used (Thielicke & Sonntag, 2021). In the [supplementary information](#), table 2 gives a summary of all parameters which were used in PIVlab. For each experiment, 500 images were analyzed in PIVlab, and a velocity profile along the vertical direction of the tube was extracted from the mean velocity vectors.

2.4. Determination of concentration profile

The concentration profile was determined via the pixel intensity, ranging from 0 to 255, of the recorded images with Matlab (R2020a). First, a region of interest (ROI) was selected on a recorded image, where the top and bottom of the ROI was the wall of the tube. Next, for each row of the ROI, the mean pixel intensity of 1,000 images of one flow rate was calculated. The calculated pixel intensity for each row of the ROI was called (pixel intensity)_y where the y indicates the height of the ROI. The pixel intensities of the highest measured flow rate were chosen as standard, (pixel intensity)_{y,std}. At last, each flow rate was standardized with Eq. (4).

$$\text{Standardized pixel intensity} = \frac{(\text{pixel intensity})_y}{(\text{pixel intensity})_{y,\text{std}}} \quad (4)$$

Table 2

Summary of techniques to measure or calculate particle velocity profiles.

Name analysis method	Principle	Advantage	Disadvantage
Laser Doppler velocimetry	Measurement of the frequency shift (Doppler effect) of scattered laser beams which is proportional to particle velocity	- High velocities measurable - High measurement accuracy - Non-invasive method	- Limited resolution due to particle concentration - Fixed at one location - Need of optically clear surface - Suffers from Nyquist theorem
Pulsed ultrasonic Doppler velocimetry	Use of ultrasound and Doppler effect to measure particle velocity	- Opaque surfaces useable - Ease of operation - Non-invasive method	- Need of relatively large measurement volume - Suffers from Nyquist theorem
Particle image velocimetry	Measurement of particle displacement via image analysis	- High velocities measurable - Particle velocities in two or three dimensions - Non-invasive method	- Limited particle concentration - Does not take the particle fraction in interrogation area into account
Computational fluid dynamics	Numerical analysis for fluid flows. The Discrete Element Method is applied to solve the particle motion and particle-particle interactions, whereas CFD solves the Navier-Stokes equations to predict the fluid velocity	- Detailed information compared to experimental results - Couples experimental and analytical approaches	- Need of experimental input in complex cases - Calculations are very time consuming for slurries

3. Results and discussion

3.1. Determination of flow regimes based on concentration profiles

Concentration profiles can provide information about the sedimented layer thickness and about particle distribution in the tube. The five different flow regimes are determined via image and video analysis and derived from concentration and velocity profiles. Fig. 3 shows results of experiments with 1.0 vol% of medium sized glass beads (58 μm). The x-axis is the standardized pixel intensity and the y-axis indicates the normalized height in the tube with 1 the top and maximum diameter (4 mm).

Fig. 3 shows a lower standardized pixel intensity when particles are sedimented which is due to lower pixel intensity for darker pixels in the image. The highest flow rate, 914 ml/min, is set as standard value 1 and shows a homogeneous flow regime in the corresponding recorded image (Fig. 3a). For all lower flow rates, deviations from the homogeneous flow regime are observed. Accumulation of particles in the lower half of the tube and depletion of particles in the upper half of the tube is indicated by pixel intensities smaller and >1 , respectively. Fig. 3b and Fig. 3c show the particle distribution for flow rates of 590 and 481 ml/min. Particles are more concentrated towards the bottom of the tube and at 481 ml/min, a layer of particles is observed at the bottom of the tube. The flow regime between 856 and 590 ml/min is pseudo/heterogeneous and for 481 ml/min, the flow is heterogeneous with a sliding bed as confirmed via the video images. For the lowest measured flow rate, 384 ml/min, the image (Fig. 3d) shows a larger and den-

ser layer of particles at the bottom of the tube. In the concentration profile, the particle layer is also visible and has a thickness of 0.8 mm and since particles are not moving in the sedimented layer, heterogeneous flow regime with fixed bed is observed. The evolution of the flow regimes is explained by the decrease of the linear velocity, resulting in a decrease of the lift/drag force, causing a net downwards force when gravitational force is dominant.

In addition, the concentration profiles of 0.2 vol% large glass beads of 141 μm in water are shown in Fig. 4. For the recorded images of the highest measured flow rate, 1,021 ml/min (Fig. 4a), a pseudo-heterogeneous flow regime is visible. The particles are distributed over the whole tube but not uniformly. Fig. 4 also shows that the deviation on the standardized pixel intensity (1,021 ml/min) for all tested flow rates in the upper part of the tube is small, resulting in profiles which are almost the same as the highest flow rate, 1,021 ml/min. Although, the low particle concentration and increased particle size (141 μm) has a minor influence on the pixel intensity and leads to less pronounced results. The recorded image of 568 ml/min (Fig. 4b), shows an increased particle density towards the bottom of the tube, typically for a heterogeneous flow regime. At the two lowest flow rates, 433 and 358 ml/min, sedimentation of particles is visible (Fig. 4c and Fig. 4d), resulting in a sliding bed flow regime for 433 ml/min and fixed bed flow regime for 358 ml/min. In the concentration profile (Fig. 4), the particle layer is visible for both lowest flow rates, 433 and 358 ml/min. The sedimented particle layer thickness for 358 ml/min is approximately 0.6 mm. However, caution must be applied with the layer thickness since there are no suspended

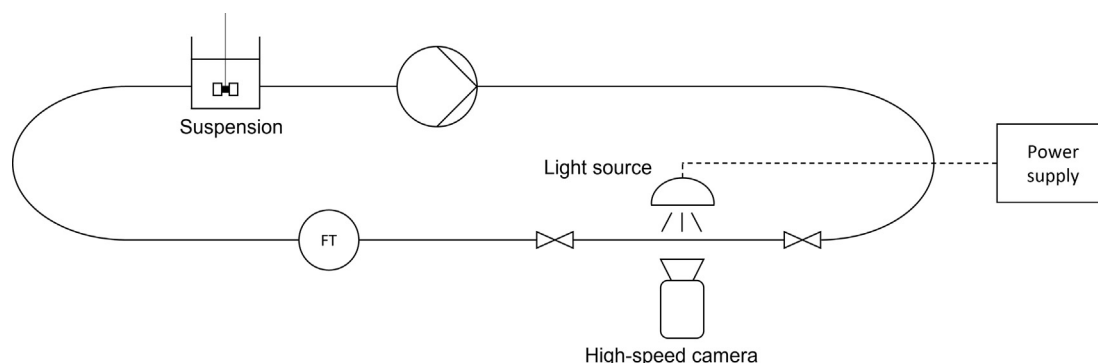
**Fig. 2.** Setup with flow indicator (FI), high-speed camera, feeding pump and light source with power supply.

Table 3

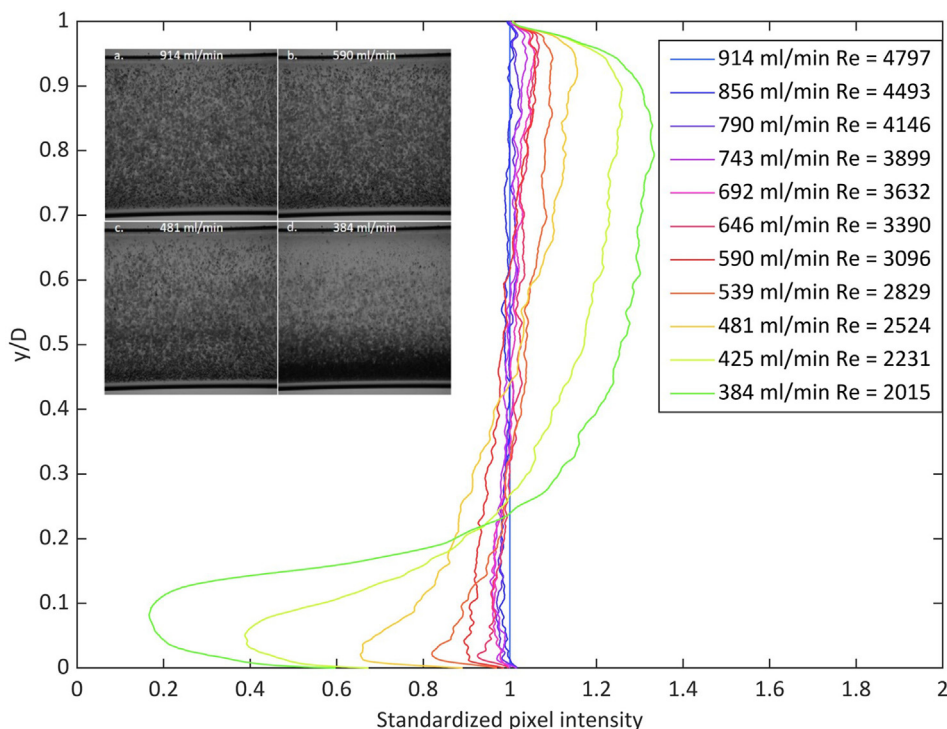
Product information.

Material	Company	Purity	Physical state	Density (kg/m ³) at 20 °C	Dynamic viscosity (kg/m s) at 20 °C
Water	/	Conductivity < 1.5 µS/cm	Liquid	999	1.0×10^{-3}
Ethanol	VWR Chemicals	90 %	Liquid	860	1.2×10^{-3}
Methanol	VWR Chemicals	>99.8 %	Liquid	790	0.61×10^{-3}
Isopropyl alcohol (IPA)	VWR Chemicals	>98 %	Liquid	786	2.2×10^{-3}
Ethylene glycol	VWR Chemicals	>98 %	Liquid	1,115	21×10^{-3}
Potassium bromide	UCB	3.18 mol/l	Aqueous solution	1,260	0.85×10^{-3}
Glass beads	Silibeads/	/	Solid	2,500	(Abdulagatov & Azizov, 2008)
Polymethylmethacrylaat (PMMA)	Cospheric Dynoadd P-560	/	Solid	1,200	/
Micronized paracetamol (PCM)	Research sample	83 %	Solid	1,263	/
Silicone antifoam emulsion	Serva	30 %	liquid	1,000	/

Table 4

Experimental information of the used suspensions.

Particle	Particle density (kg/m ³)	Liquid density (kg/m ³)	Particle size d ₅₀ (µm)	Volumetric concentration (vol%)
Glass beads	2,500	999 (Water)	54; 58; 72; 141	1.0; 0.8; 0.6; 0.4
		860 (Ethanol)	58	1.0; 0.8; 0.6; 0.4
		790 (Methanol)	58; 141	1.0; 0.8; 0.6; 0.4
		786 (isopropyl alcohol)	58	1.0; 0.8; 0.6; 0.4
		1,260 (KBr-solution)	58; 141	1.0; 0.8; 0.6; 0.4
		1,010 (Ethylene glycol – water mixture)	141	1.0
PMMA	1,200	999 (water + antifoam)	62	1.0; 0.8; 0.6; 0.4
PCM	1,263	999 (water + antifoam)	80	1.0; 0.8; 0.6; 0.4

**Fig. 3.** Concentration profiles and recorded images of suspension of 1.0 vol% of glass beads (58 µm) in water at different flow rates.

particles present in the upper part of the tube at 358 ml/min, indicating that the particles may have sedimented in earlier parts of the setup.

Comparison of these results (Figs. 3 and 4), shows that the flow regimes for 58 µm and 141 µm glass beads particles are similar for each flow rate. However, once gravitational forces are dominant,

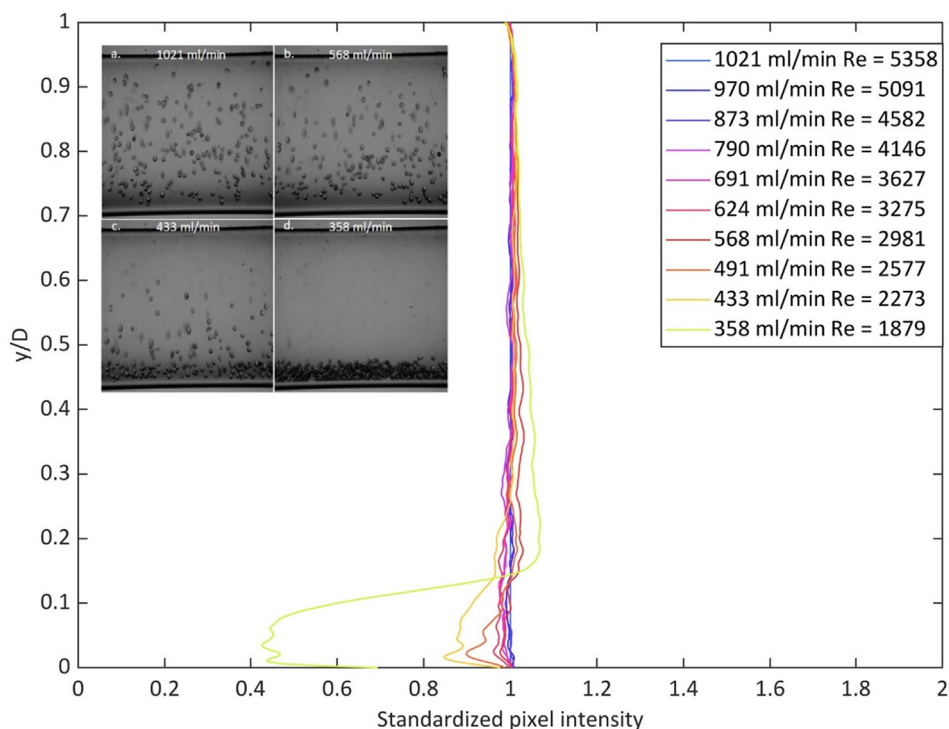


Fig. 4. Concentration profiles and recorded images of suspension of 0.2 vol% of glass beads (141 μm) in water at different flow rates with y/D indicating the height in the tube.

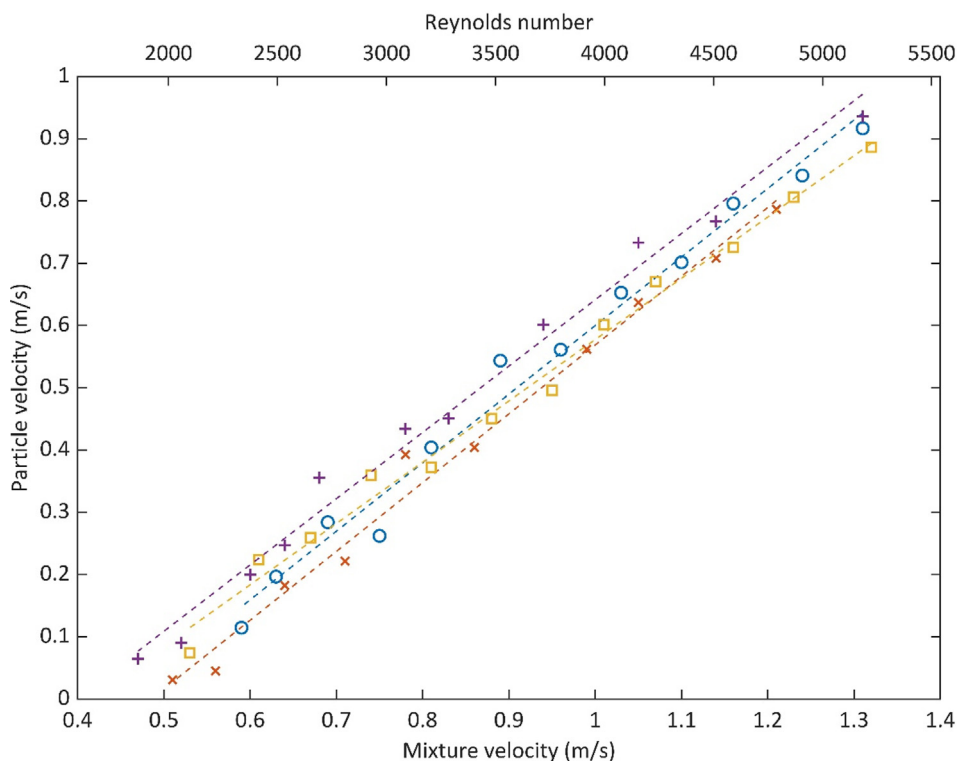


Fig. 5. Velocity of particles at the bottom of the tube at different mixture velocities for different particle sizes of glass beads 54 μm (○), 58 μm (x), 72 μm (□) and 141 μm (+). Linear trendlines are added as a visual guideline.

larger particles will sediment faster than smaller particles. In milliflow reactors, not only the drag force and gravitational force determines the net force balance on a particle. Farajpour et al. have

reported that the magnus force and thus lift force increases with increasing particle radius in microfluidic devices (Farajpour et al., 2019a), (Bluemink et al., 2010).

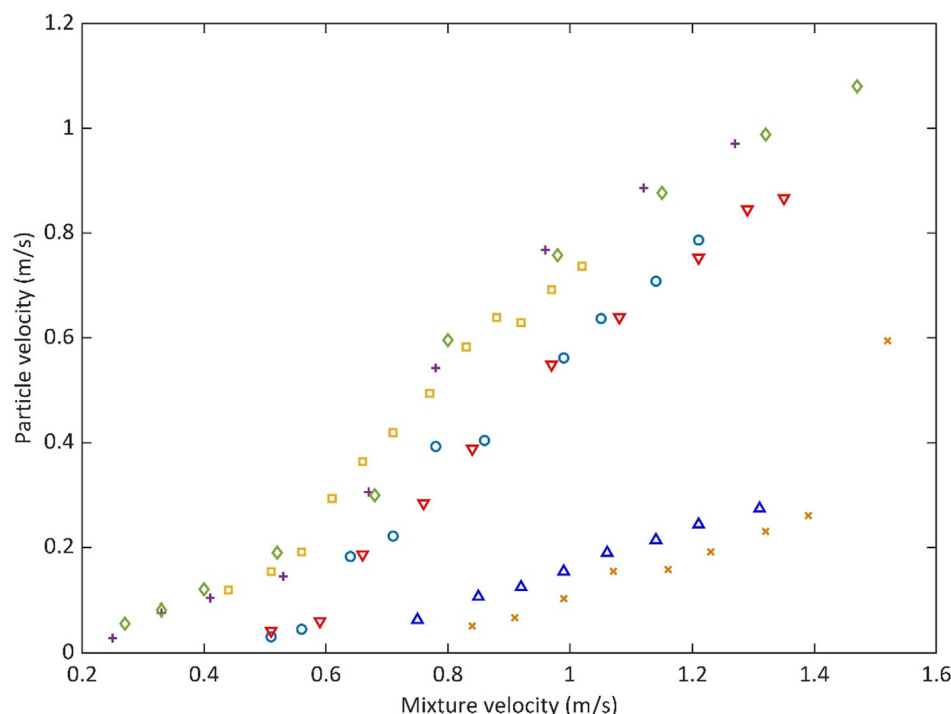


Fig. 6. Velocity of particles at the bottom of the tube at different mixture velocities for different particle densities, glass beads of 58 μm ($\circ$), PCM ($\diamond$) and PMMA (+) in water and for different liquid densities and viscosities, ethanol (x), IPA (Δ), methanol (∇) and KBr-solution ($\square$).

3.2. Determination of velocity profiles and minimum suspension velocity in milliflow reactors

Based on the velocity profiles of the particles, the particle velocity at the bottom of the tube can be examined. An example of a velocity profile of glass beads of 58 μm in water at two different flow rates is visible in the [supplementary information](#). In Fig. 5, the particle velocity at the bottom of the tube for different particle sizes of glass beads is plotted in function of the suspension velocity, measured with a flow meter. At low mixture velocities, sedimentation is visible. Particle velocity is measured 0.235 mm above the bottom of the tube, to avoid wall effects. Fig. 5 shows for each different particle size of glass beads, the same linear trend. Notice the highest velocity at the bottom of the tube of the largest (141 μm) particles which is counterintuitive since they have the highest sedimentation velocity. Wiedmeyer et al. also observed this in experiments with coiled tubing (Wiedmeyer et al., 2017). Due to the parabolic liquid velocity profile, larger particles experience a higher velocity gradient than smaller particles located at the same location, specifically at the solid boundary (wall).

Next, the influence of the particle density, liquid density and viscosity on the particle velocity at the bottom of the tube is researched in function of the mixture velocity (Fig. 6). Fig. 6 shows a linear trend for every suspension. However, results of ethanol and IPA are not comparable with the other results. In all mixture velocities with ethanol and IPA, sedimentation of particles is visible, resulting in low particle velocities at the bottom of the tube, even at high mixture velocities. Methanol (790 $\text{kg/m}^3 - 0.61 \times 10^{-3} \text{ Pa s}$) has a lower viscosity and same density in comparison to IPA (786 $\text{kg/m}^3 - 2.2 \times 10^{-3} \text{ Pa s}$) but no sedimentation is visible with methanol as liquid. Therefore, lowering of the viscosity leads to higher particle velocities at the bottom of the tube. Viscosity has an influence on the parabolic liquid profile, which is most pronounced close to the wall, and thus also an influence on particle velocity. Another parameter which is researched is the difference between solid and liquid density, ρ_{s-l} . For the results of PCM,

PMMA and glass beads, lowering of ρ_{s-l} results in higher particle velocities at the bottom of the tube. PCM and PMMA have the same particle velocities at the bottom of the tube due to the low ρ_{s-l} . Also in the KBr-solution, glass beads move faster than in water due to a lower ρ_{s-l} and a lower viscosity ($0.85 \times 10^{-3} \text{ Pa s}$) of the KBr-solution in comparison to water.

When using these results to design heterogeneous flow processes for reaction synthesis, next to avoiding blockage, limiting the residence time distribution of particles (and fluid) is very important. Therefore, the determination of the minimum suspension velocity is needed. The ratio of the maximum particle speed to the particle speed at the bottom of tube is a measure for the residence time distribution of the particles. In Fig. 7, this ratio is plotted versus the flow rate of the mixture, with corresponding Reynolds number, for different particle sizes of glass beads. To limit the residence time distribution of particles, we opted to allow a maximum ratio ($V_{\text{max}}/V_{\text{particles_bottom}}$) of 2. Consequently, the minimum suspension velocity is defined as the velocity at which the slowest particles move twice as slow as the fastest particles. In Fig. 7, all particle sizes show the same increasing trend of the ratio ($V_{\text{max}}/V_{\text{particles_bottom}}$) with decrease of the mixture velocity. The determined minimum suspension speed of glass beads and water lays between 0.88 and 0.98 m/s which corresponds with a Reynolds number between 3,480 and 3,880, indicating a turbulent flow regime.

Next, the minimum suspension speed ($V_{\text{max}}/V_{\text{particles_bottom}} = 2$), is determined for other particles and liquid densities and liquid viscosities, and results are visible in Fig. 8. All results show an increasing trend of the ratio when the mixture velocity decreases. It is visible, that lowering the particle density (PMMA or PCM) reduces the minimum suspension speed, now ranging from 0.68 to 0.71 m/s. The explanation for this result is the lower ρ_{s-l} . Also for the results with KBr-solution as liquid, the minimum suspension speed ranges from 0.68 and 0.71 m/s. KBr-solution has a lower viscosity and a lower ρ_{s-l} . If ρ_{s-l} and viscosity increases, which is the case for ethanol and IPA, it is visible that even at the highest tested mixture

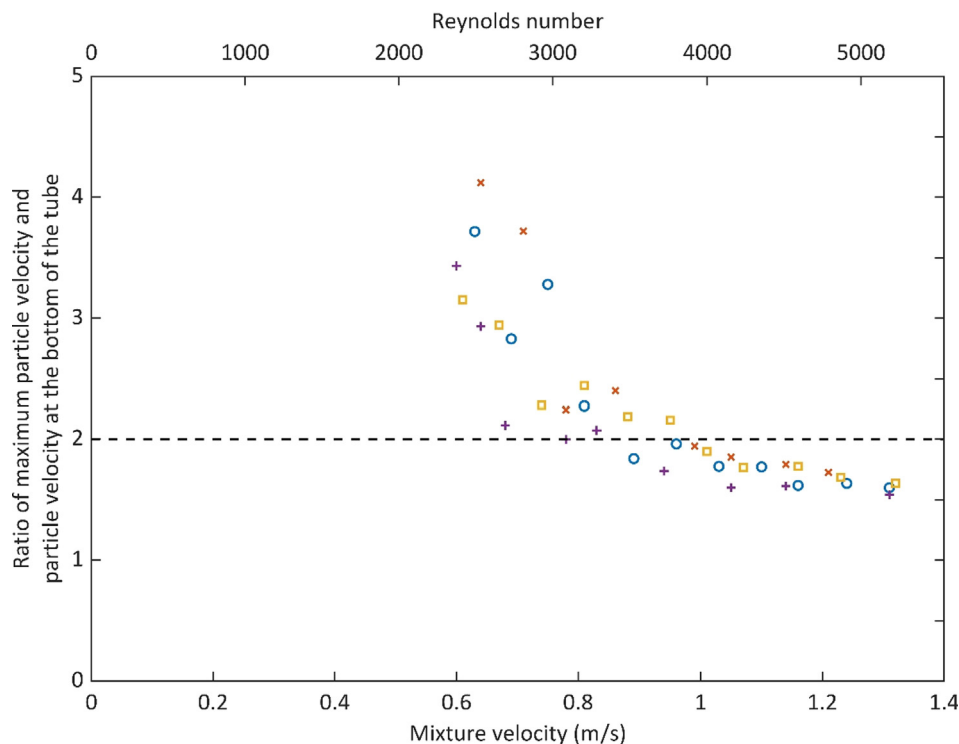


Fig. 7. Ratio of maximum particle velocity and particle velocity at the bottom of the tube at different mixture velocities for different particle sizes of glass beads 54 μm ($\circ$), 58 μm ($\times$), 72 μm ($\square$) and 141 μm ($+$).

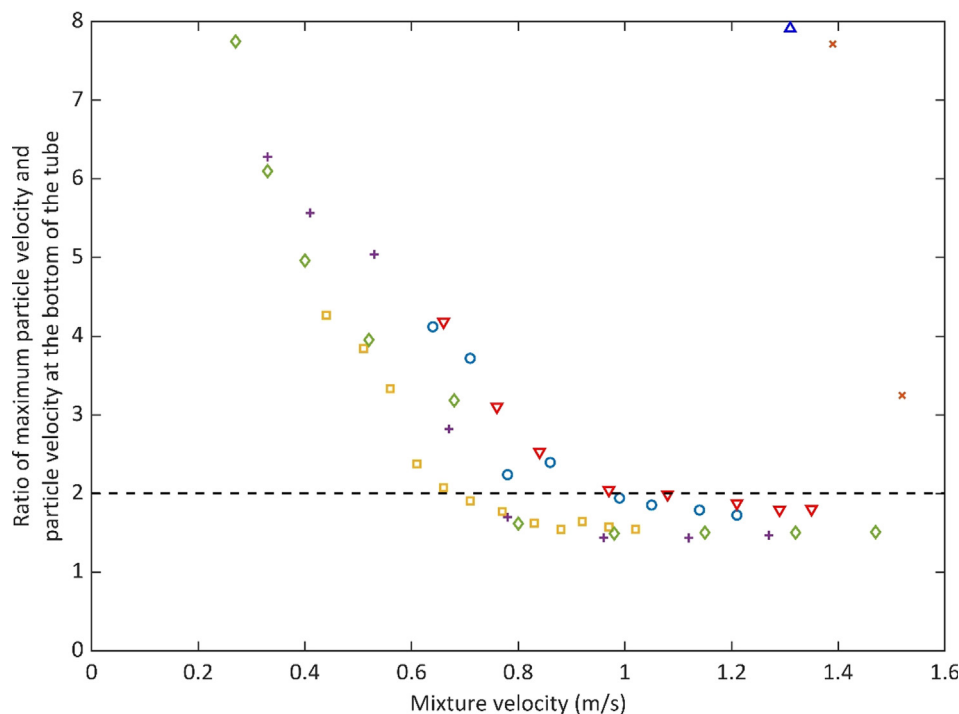


Fig. 8. Ratio of maximum particle velocity and particle velocity at the bottom of the tube at different mixture velocities for different particle densities, glass beads of 58 μm ($\circ$), PCM ($\diamond$) and PMMA ($+$) in water and for different liquid densities and viscosities with glass beads of 58 μm , ethanol ($\times$), IPA (Δ), methanol (∇) and KBr-solution ($\square$).

velocities, the ratio is higher than 2 and particles at the bottom of the tube are moving very slowly in comparison to the liquid. At last, the viscosity is researched with three different liquids with approximately the same density: methanol, ethanol and IPA. Results show that only methanol has ratios lower than 2 with all

measured mixture velocities and the results of methanol almost correspond with the results of water. The minimum suspension speed for methanol-glass beads mixtures is between 0.97 and 1.04 m/s. IPA and ethanol have a higher viscosity than methanol, resulting in higher ratios at the same mixture velocity. In general,

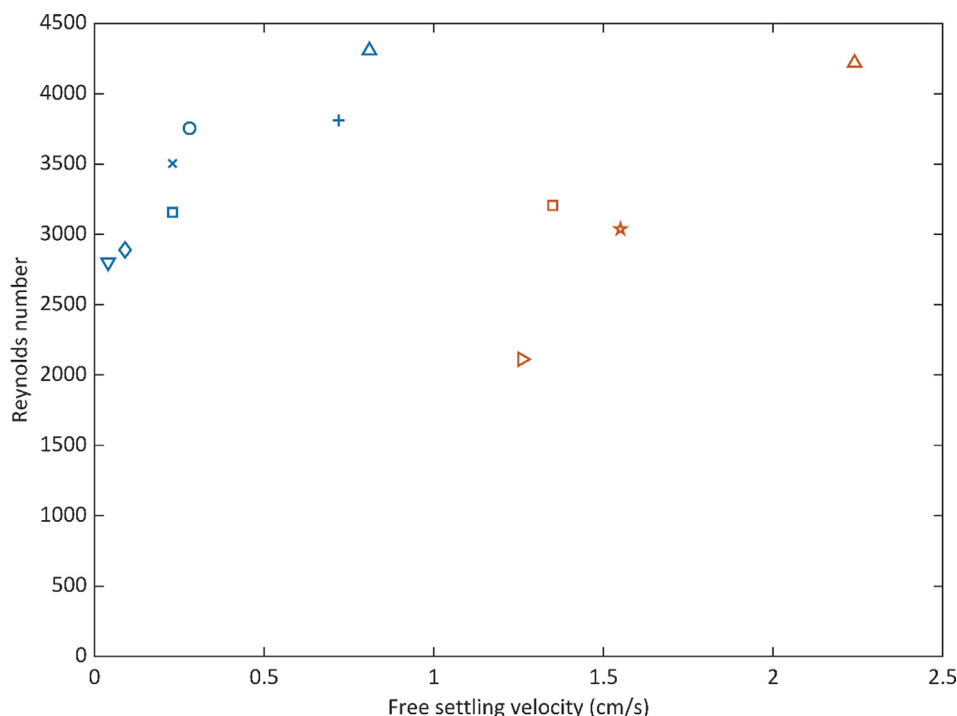


Fig. 9. Reynolds number in function of free settling velocity with two different ranges of particle sizes (bleu: 54 – 80 μm and orange: 141 μm) and different particle composition and liquids: glass beads water (○ and), KBr-solution (□), methanol (Δ) and ethylene glycol–water (▷), glass beads of 54 μm (x) 72 μm (+) in water, PMMA of 62 μm in water (▽) and paracetamol of 80 μm in water (◇).

it can be concluded that lowering of the viscosity and lowering of ρ_{s-l} is beneficial to reduce residence time distribution and sedimentation of particles in milliflow reactors. Finally, the Reynolds number of each experiment is determined and plotted in function of the calculated free settling velocity of the particles (Fig. 9).

Two different trends are visible, each with a different particle size (blue: 54 – 80 μm and orange: 141 μm). Therefore, the results indicate the importance of the particle size in the velocity and settling of particles. Larger particles have a higher free settling velocity, but do not need higher Reynolds numbers compared to smaller particles, in order to reach the minimum suspension speed ($V_{\text{max}}/V_{\text{particles_bottom}} = 2$). Within each size category, the needed Reynolds number to reach the minimum suspension speed increases linearly with higher free settling velocities. With these results, it is possible to predict the necessary Reynolds number in milliflow reactors by calculating the free settling velocity.

4. Conclusion

The determination of particle velocities is important to perform heterogeneous solid–liquid reactions in a milliflow reactor. This research investigates concentration and velocity profiles of particles, flow regimes of suspensions and it determines the minimum suspension velocity. First, via concentration profiles and image recording, all flow regimes (homogeneous, pseudo-heterogeneous, heterogeneous, sliding bed and fixed bed) are determined. Examining the velocity of the particles at the bottom of the tube shows that larger particles have higher velocities at the bottom of the tube than smaller particles. Larger particles experience larger particle velocities due to the parabolic profile of the liquid. Beside the influence of the particle size, lowering of the liquid viscosity or particle density, which also lowers the density difference (ρ_{s-l}), results in higher velocities at the bottom of the tube. Next, the minimum suspension speed is determined and is

defined as the suspension speed at which the ratio of the maximum particle velocity to the minimum particle velocity is 2. For suspensions with high ρ_{s-l} (1,500 kg/m^3), the minimum suspension speed lays between 0.88 and 0.98 m/s and with low ρ_{s-l} (200 kg/m^3), the minimum suspension speed decreases to 0.68 m/s . Alcohols have lower densities, resulting in higher density differences with the particles and thus resulting in a higher minimum suspension velocity which lays above 0.97 m/s for methanol as liquid. The corresponding Reynolds numbers are plotted in function of the calculated free settling velocity and results shows two different trends, each for a different particle size. With the knowledge of the free settling velocity, the needed Reynolds number can be calculated which ensures a limited residence time of particles in the tubular reactor. When applying the results of the minimum suspension velocity on a heterogeneous solid–liquid reaction, this can lead to a practical issue. The residence time for reactions in milliflow reactors generally varies from 1 min until 15 min. Therefore, the length of the milliflow reactors may be prohibitively long to reach the residence time with the corresponding minimum suspension speed, specifically for reactions with a catalyst which has a high ρ_{s-l} in most cases or with high liquid viscosities. A solution for this issue could be the use of pulsation in milliflow reactors. In literature, it is already proven that the use of pulsation is beneficial to reach a high degree of plug flow even at low flow rates. However, the influence of pulsation on liquid is known but for suspensions, it needs further investigation. In general, it can be concluded that via a high-speed camera and PIV, flow regimes, velocity profiles and minimum suspensions speeds can be determined with high accuracy.

CRediT authorship contribution statement

Joris Claes: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Validation, Visualization,

Writing – original draft, Writing – review & editing. **Bert Metten:** Conceptualization, Writing – review & editing. **Bert Brabants:** Conceptualization, Writing – review & editing. **Mumin Enis Leblebici:** Writing – review & editing. **Simon Kuhn:** Conceptualization, Writing – review & editing. **Leen C.J. Thomassen:** Conceptualization, Funding acquisition, Project administration, Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ces.2022.117794>.

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