Supplementary information

Inflow boundary conditions determine T-mixer efficiency

Tobias Schikarski,^a Holger Trzenschiok,^a Wolfgang Peukert,^a and Marc Avila^b

^a Institute of Particle Technology, Department of Chemical and Biological Engineering, Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen, Germany; E-mail: <u>tobias.schikarski@fau.de</u> ^b Center of Applied Space Technology and Microgravity, Universität Bremen, Am Fallturm, 28359 Bremen, Germany

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S1 Transition to turbulence

In section §3, we argue that the natural and random occurrence of turbulent puffs beyond $Re \gtrsim 1460$ influences mixing in the outlet channel and results in a large deviation in mixing time t_m (see Fig. 1b in paper). To support this statement, the standard deviation of mixing time, normalized by the mean $\operatorname{std}(t_m)/t_m$, is



Fig. S1: Standard deviation of the experimentally obtained mixing time, scaled with the mean $\operatorname{std}(t_m)/t_m$, as function of Reynolds number Re. The gray-shaded area depicts the Reynolds number range where turbulent puffs are expected to occur, whereas the red dashed line marks the natural onset of transition, which in our experiments is at $Re \approx 1400$.

shown as a function of Reynolds number in Fig. S1. Clearly, the deviation in measured mixing time increases strongly in the regime where turbulent puffs can occur. Besides, large deviations are also observed in the intermediate regime, where engulfing and symmetric flow structures compete. This emphasizes the general sensitivity of mixing on the inflow in the T-mixer.

S2 The intensity of turbulence in a T-mixer

The collision of inlet streams is used in common mixing devices to trigger strong turbulent motions, hence mixing, at relatively low inflow Reynolds numbers. The accurate computation of the turbulent statistics in our simulation method allows to estimate the turbulent intensity in the mixing channel and compare it to canonical flows, e.g. pipe flow. To do so, the definition of the Taylor microscale for homogeneous isotropic turbulence, $\lambda = \sqrt{10\nu k/\epsilon}$, where k and ϵ are the turbulent kinetic energy and the dissipation, respectively, is assumed to hold. Then, the turbulent Reynolds number $Re_{\lambda} = \sqrt{k\lambda}/\nu$ can be calculated by extracting the values of k and ϵ from Figure 10. Taking the maximum values of k and ϵ (at around x = (1, 2)), we obtain $Re_{\lambda} \approx 50$ and $Re_{\lambda} \approx 125$ for Re = 650 and Re = 4000, respectively. Comparing these values to pipe flow (S.C.C. Bailey *et al.*¹ we are not aware of any study in square duct flow providing similar data), the turbulence level in a T-mixer at Re = 4000 is comparable to $Re \approx 60000$ in cylindrical pipe flow.

A more direct comparison to square-duct flow can be done by comparing the pressure loss Δp of our



Fig. S2: Darcy friction factor, as defined in Eq. (3), as function of Reynolds number Re. Symbols represent the data obtained from simulation in the T-mixer. The solid and the dashed line denote the laminar and turbulent friction factor of square duct-flow, see Eqs. (4) and (5) respectively.

T-mixer to that of a square duct of the same length by using the Darcy friction factor

$$f_d = 2\frac{\Delta p d_h}{L\rho u_0^2},\tag{3}$$

where d_h is hydraulic diameter (of the outlet in case of the T-mixer), u_0 is the mean velocity and L, here 15.5d, is the length between inlet and outlet. In square duct flow, the Darcy friction factor is

$$f_d = \frac{57}{Re},\tag{4}$$

for fully developed laminar flow, whereas in the fully turbulent regime it is commonly estimated with an empirical correlation proposed by Jones⁹

$$f_d^{-1/2} = 2\log_{10}(2.25Ref^{1/2}) - 0.8.$$
(5)

Fig. S2 shows that at low *Re* the friction factor of the T-mixer is close the laminar value of square-duct flow. However, as *Re* increases, the friction factor gradually departs from it because of the subsequent hydrodynamic instabilities, which result in turbulent flow at the junction. As the turbulence develops fully, the friction factor of the T-mixer appears to evolve nearly parallel to that of square-duct flow, but it is about 5 times larger in magnitude.

S3 Turbulent statistics of square duct flow

In our simulations of the flow in a T-mixer at large Re, we apply fully developed turbulent flow at the inlet boundaries. To generate a fully developed turbulent inflow, we simulated the fluid flow at the desired



Fig. S3: Turbulent statistics (black line) obtained at Re = 4000 are compared to reference data (blue circles¹² and red triangles⁶) obtained at Re = 4410. (a) and (b) show the mean $\langle (\bullet) \rangle$ and root mean square velocity $\langle (\bullet)'(\bullet)' \rangle^{1/2}$ components at position z = 0.35, respectively. The top and bottom row depict the streamwise v and cross-stream u velocity component, respectively. Note that the orientation of coordinates of the T-mixer inlet are used as well as only the half of the channel width is shown.

Reynolds number in a periodic square duct with stream-wise length of L = 30d (see Fig. 6 in paper) and mapped the stored data for two selected cross sections to the two inlet boundaries of the T-mixer. To avoid correlated results, the data of the two cross sections is used with $100d/u_0$ delay.

Fig. S3 shows a comparison of the first and second order turbulent statistics obtained with the our DNS to reference data^{6,12} at a fixed height, here z = 0.35. We averaged our data in time and in the (homogeneous) streamwise direction, which is denoted by the operator $\langle \rangle$. Good agreement between our simulations and the reference simulations in the mean velocity components $\langle u \rangle$ and $\langle v \rangle$ (see Fig. S3(a)) was obtained. Slight deviations apart from the wall in cross-stream velocity component $\langle v \rangle$ are attributed to the too short average time interval. Similar, the normal Reynolds stresses $\langle u'u' \rangle^{1/2}$ and $\langle v'v' \rangle^{1/2}$, where $u' = u - \langle u \rangle$ describes the velocity fluctuations, agree very well to the reference data (see Fig.S3(b)).

S4 Pressure drop measurements



Fig. S4: (a) Sketch of the experimental setup. (M), (TC) and (PR) denote the stepping motor, temperaturecontrolled barrels and the position of the pressure recording, respectively. A zoom of the T-mixer is shown in (b). The grey shaded T-mixer depicts the domain used in the simulations.

Fig. S4 shows a sketch of the full experimental setup, which consists of two inlets of square cross section of hydraulic diameter d = 1mm, which discharge in a mixing channel of 1mm height, 2mm width and

11.5mm length. The inlets are 18mm long, whereas in the simulations the inlets are just 3mm long because we impose fully developed flow as boundary condition. The geometry of the simulations is shown gray shaded in Fig. S4. Pressure transmitters model A09 (Sensor-Technik Wiedemann GmbH, Germany) with a range up to 0.5bar relative pressure were used to measure the pressure difference between the fluid in the T-mixer and the environment. To avoid disturbing the inflow close to the junction, we did not install pressure probes at the locations of the simulated geometry (i.e. 3mm before the junction). Instead, the pressure probes were installed before the connections to the inlets of the T-mixer device (PR). To enable comparison with the simulations, a second device consisting of a square duct of 15mm length was manufactured (the dashed line in Fig. S4(b) depicts the position its outlet). With this second device, experiments where performed to measured the pressure loss. This was then subtracted from the total loss measured in the experiments with the actual T-mixer, thereby allowing a direct comparison to the simulations.

Before each experimental run, a baseline pressure signal was recorded for 5 seconds. Then, after the pistons started moving and the flow structures developed, the pressure signal reached a plateau, indicating operation in steady-state conditions. For each experiment, the average pressure of the baseline was sub-tracted to the average steady-state pressure. Finally, we note that at low Reynolds numbers (up to Re 400) the resulting pressure loss is of the order of the measurement uncertainty. Although deviations are expected, the agreement with numerical simulations is excellent, see Fig. 11(a) in § 4 (paper).



S5 Villermaux-Dushman Reaction

Fig. S5: Experimentally obtained mixing efficiencies (segregation index X_s [-] and mixing time t_m [s]) as function of the Reynolds number. The inset shows the concentration dependence of $a = t_m/X_s$ as function of Reynolds number Re.

S5.1 Background

Chemical reactions, used as molecular probes for mixing, are powerful tools to gain insights into the mixing process. In the literature, several competitive consecutive and parallel chemical reactions have been proposed, where the distribution of quantifiable chemical species depicts the mixing history.^{10, 11, 13} Fournier *et al.*⁵ proposed the parallel use of the neutralization of boric acid

$$H_2BO_3^- + H^+ \rightleftharpoons H_3BO_3. \tag{6}$$

with the Dushman-reaction³

$$5I^{-} + IO_{3}^{-} + 6H^{+} \rightleftharpoons 3I_{2} + 3H_{2}O, \tag{7}$$

namely the Villermaux–Dushman reaction,² to quantify the mixing efficiency of mixing devices. In this reaction system, sulfuric acid is added in a stoichiometric deficit. If mixing is nearly perfect, then sulfuric acid is entirely consumed due to the very fast kinetics, whereas if mixing is poor the by-product iodine I_2 emerges from the Dushman reaction.

While the neutralization of boric acid in Eq. 6 is quasi-instantaneous, the kinetics of the Dushman reaction is much slower and in the range of the mixing process.⁸ The iodine formed in the redox-reaction will further react in a quasi-instantaneous reaction with iodide ions and form an equilibrium with triiodide-ions,⁷ which reads

$$I^- + I_2 \rightleftharpoons I_3^-. \tag{8}$$

The concentration of the formed triiodide ions I_3^- was measured here with a Cary 100 Scan UV/Vis spectrometer (Varian Deutschland GmbH) at a wavelength of 353 nm and quantified by applying the Beer–Lambert law

$$[I_3^-] = \frac{\text{OD}}{\epsilon_{353} \cdot l} \qquad \text{valid if } 0.1 < \text{OD} < 2.5, \tag{9}$$

where OD, ϵ_{353} and *l* denote the optical density (absorbance), the extinction coefficient of triiodide at wavelength 353nm and the optical path length within the measurement cell, respectively. Note that the UV/VIS spectrometer was not connected to the mixing setup. Thus the samples collected in a beaker were measured offline. In doing so, at least 1.5ml of the mixture was collected for each experimental trial, while 1ml of this collected amount was needed for optical density measurements itself.

S5.2 Notes on mixing time

The mixing efficiency is quantified in the Villermaux-Dushman characterization by the segregation index

$$X_s = \frac{Y}{Y_{st}},\tag{10}$$

which is the fraction of H^+ ions used to form iodine I_2 , i.e.

$$Y = \frac{[I_2] + [I_3^-]}{[H^+]_0} \quad \text{and} \quad Y_{st} = \frac{6[IO_3^-]_0}{[IO_3^-]_0 + [H_2BO_3^-]_0}, \tag{11}$$

where the subscript $_0$ denotes the initial concentration. The weakness of this approach lies in the dependence of the segregation index X_s on the initial concentrations. In our experiments, the mixing efficiency changes enormously as Re increases from 100 to 4000, and this requires the application of different concentrations sets to stay in the linear regime of the detector of the spectrometer (see Eq. 9). However, using different concentrations sets results in a discontinuous transition in the segregation index X_s as the Reynolds number increases, see Fig. S5. Two different concentrations sets are applied in this study (see Tab. S1), which are changed at Re = 650.

Commenge *et al.*² solved this problem by providing a single empirical a master curve capturing multiple concentration sets. In particular, their master curve converts the measured optical density (effectively the segregation index) into a mixing time

$$t_m = 0.33(\text{OD})[H^+]_0^{-4.55}[I^-]_0^{-1.5}[IO_3^-]_0^{-5.8}[NaOH]_0^{-2}[H_3BO_3]_0^{-2}.$$
(12)

Their approach relies on the interaction of exchange with the mean (IEM) model, which incorporates the influence of the mixing time t_m on the Villermaux-Dushman reaction and the actual reaction system including the kinetics.^{2,4} By varying the mixing time in the model, different segregation indices X_s were obtained and compared to the measured ones. Doing this for different mixers, Reynolds numbers and concentration sets, large datasets were gathered and collapsed together on one master curve by normalizing with the product of reactant concentrations as introduced above in Eq. 12. Two of the concentration sets given in their work enabled the seamless identification of mixing times between 0.1 and 0.001s in our work, whereby the actual concentrations are given in Tab. S1.

In practice, within one concentration set the mixing time t_m can be obtained by just multiplying a constant factor to the segregation index X_s as

$$t_m \approx a \cdot X_s,\tag{13}$$

where the constant a depends solely on the concentration set as depicted in Fig. S5. Thus, the herein used mixing time t_m is nothing else than a normalized segregation index.

S5.3 Experimental protocols

Impurities brought into the reaction system can drastically change the kinetics of chemical reactions. This could occur either by unwanted alternative reactions or by merely changing the ionic strength. To detect such problems, out T-mixer was carefully cleaned every day before and after the experimental runs. Each experiment was repeated at least three times and at different days using solutions freshly prepared each day.

Reactants		concentrations mol l^{-1}	
		Re = (100, 650)	Re = (700, 4000)
acid	H_2SO_4	0.015	0.025
	NaI	0.	032
haaa	KIO ₃	0.006	
base	NaOH	0.09	
_	H_3BO_3	0	.09

Tab. S1: Base concentrations and acid concentrations used for the Villermaux–Dushman reaction.

In all experiments, ultrapure water, produced with a Purelab[®] Ultra from Elga LabWater (Veolia Water Solutions & Technologies, France), was used. For the Villermaux–Dushman characterization, the chemicals used were all of high purity, either analytical grade (p.a. or Reag. PhEur) or of 99.8% purity. Sulfuric acid 96% (p.a.), potassium iodate (p.a.) (Carl Roth GmbH, Germany), sodium iodite (Reag. Ph Eur) (Merk KgaA, Germany), sodium hydroxide (reagent grade)(Sigma-Aldrich Chemie GmbH, Germany) and boric acid (99,8%) (Alfa Aesar, Germany) were used. The solutions were prepared as described by Guichardon *et al.*⁷ to prevent thermodynamic triiodide formation. Additionally, before the solutions were prepared, the ultrapure water was stripped with nitrogen gas to avoid the oxidation of iodide to iodine.⁸ The employed chemicals do not influence the material parameter of the aqueous solution, e.g. viscosity and density, and thus enable to compare the results qualitatively between simulation and experiment.

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