Investigation of emulsification in static mixers by optical measurement techniques using refractive index matching

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Abstract

This study compares a two-phase flow through a foam-like porous structure and a Sulzer SMXTM static mixing element. Refractive index matching between two immiscible fluids and the internal structures enables optical measurements to be performed. The droplet size and position within the internal structures were observed by laser-induced fluorescence. The results show that at low flowrates, droplets follow preferred paths, whereas at higher flowrates, they are more homogeneously distributed within the structures. The droplet size distribution was found to be well represented by the Sauter mean diameter. Measuring along the axis of the two static mixers, we found that droplets disintegrate more quickly in the foam-like porous structure. As both geometries have the same porosity and hydraulic diameter, we conclude that the change in the free cross section is also an important parameter. We observed that in the geometry with large changes in the free cross section, the droplets are smaller.

Keywords: flow chemistry, process intensification, two-phase flow, porous structure, refractive index matching, laser-induced fluorescence (LIF)

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1. Introduction

Contact between two fluids and a solid is required for heterogeneous catalysis with a two-phase educt (e.g., hydrogenation, oxidation, and hydroxylation). An extensive review of the reactions that are frequently encountered in the pharmaceutical and fine-chemicals industries is provided in Mills and Chaudhari (1997). These reactions are conventionally conducted in batch vessels, that are used as multipurpose plants. However, many alternative reactors are available (Stitt, 2002). A continuous tubular reactor with a porous structure is frequently proposed as an alternative (e.g., Stankiewicz and Moulijn, 2004). It enables new chemical routes by providing good heat / mass-transfer and improved safety. In addition, the continuous operation mode allows heat integration and therefore reduces energy consumption. The internal structures in tubular reactors can be used as a catalyst support and as static mixing elements. In multiphase reactions, these structures can also serve to produce small droplets. In this study, we investigate a two-phase flow through a foam-like porous structure and a Sulzer SMXTM static mixing element.

Studies of multiphase flow through porous structures can be macroscopic (system scale) or microscopic (pore scale). Many studies on multiphase flows in porous structures have considered macroscopic quantities such as the pressure drop, hold-up, mass transfer, residence time distribution, or interfacial area density (Edouard et al., 2008; Losey et al., 2001; Molga and Westerterp, 2012). These macroscopic quantities are strongly affected by the microscopic composition of the two-phase flow. Therefore, to understand the transport

25 processes on a macroscopic scale, the microscopic effects need to be investi-
26 gated. In many microscopic studies of two-phase flows in porous structures,
27 the droplet size was larger than the pore size (Ovdat and Berkowitz, 2006;
28 Darnault et al., 1998; Ng et al., 1978; Krummel et al., 2013; Montemagno and
29 Gray, 1995; Kong et al., 2011; Stöhr et al., 2003). Such situations are encoun-
30 tered in oil recovery, for example. In this case, a single-phase flow can occur
31 at the pore scale even though a two-phase flow occurs at the macroscopic
32 scale.

33 In a tubular reactor, it could be beneficial for the droplets to be smaller
34 than the pores as this would lead to more homogeneous reaction conditions
35 within the reactor. Droplets smaller than the pores are also encountered in
36 emulsification processes using static mixing elements. Owing to the limited
37 access to measurement equipment, the droplet size is frequently measured at
38 the exit of a static mixing element (Al Taweel and Walker, 1983; Fradette et
39 al., 2007). Alternatively, it is measured offline. By microencapsulation via
40 an interface polymerization reaction, the droplet size of an emulsion can be
41 preserved for later analysis by optical microscopy or laser diffraction (Legrand
42 et al., 2001; Lemenand et al., 2003; Theron and Le Sauze, 2011).

43 In the present study, we investigate a two-phase flow inside a porous
44 structure by optical measurements. This is enabled by matching the refrac-
45 tive index (RI) of the two liquids and the solid. Below, we review optical
46 measurement techniques for multiphase flows.

47 *1.1. Optical measurement techniques*

48 Many tomographic methods including X-ray radiography, neutron trans-
49 mission tomography, nuclear magnetic resonance imaging, electrical capaci-

50 tance tomography, and optical methods have been applied for visualizing the
 51 phase distribution (Chaouki et al., 1997). In the present study, we use an
 52 optical measurement technique with RI matching. Below, we review optical
 53 measurement techniques with a focus on studies that employed RI match-
 54 ing. We start with two-phase flows in ducts, continue to single-phase flows
 55 in porous structures, and end with two-phase flows in porous structures.

56 *1.1.1. Two-phase flow in ducts*

57 For phenomenological studies such as column flooding (Stemmet et al.,
 58 2005) or phase inversion (Piela et al., 2006), a standard camera can be used
 59 without RI matching. For quantitative measurements by laser-induced fluo-
 60 rescence (LIF) and particle image velocimetry (PIV) a system consisting of
 61 an illuminating laser and a camera is needed. When used for investigating
 62 multiphase flows, the main challenge faced with these optical measurement
 63 techniques is the refraction of light at phase boundaries. Some authors have
 64 applied optical measurement techniques without RI matching (Unadkat et
 65 al., 2009; Lindken et al., 1999; Fujiwara et al., 2004). In this case, a laser-
 66 based measurement technique is frequently combined with shadowgraphy.
 67 The limitation of the shadowgraphy method is that it is only applicable to
 68 two-phase flows in a bubbly flow regime with low hold-up of the dispersed
 69 phase. This limitation can be overcome by matching the RI of the disperse
 70 phase and the continuous phase.

71 RI matching was applied by Svensson and Rasmuson (2006) for two-phase
 72 flow in stirred tanks. RI-matched two-phase flow through ducts was inves-
 73 tigated by LIF (Liu et al., 2005) and PIV (Conan et al., 2007). Augier et
 74 al. (2003) applied simultaneous PIV and LIF measurements in RI matched

liquid/liquid flow through a duct. The images of the particles and fluorescent dye were separated based on their gray values. This allowed simultaneous PIV and LIF with only one camera. Similarly, Morgan et al. (2013) investigated RI-matched liquid/liquid flow in a duct. They simultaneously measured the droplet size by LIF and the velocity field by PIV/PTV.

1.1.2. *Single-phase flow in porous structures*

RI matching between a solid and a liquid allows optical measurements inside porous structures. Budwig (1994) reviewed RI matching techniques for a single-phase flow. Wiederseiner et al. (2011) recently reviewed RI matching for particle suspensions. These two reports provide an overview of fluid recipes corresponding solid materials for RI matching. Whereas many studies have focused on RI matching between one fluid and one solid, few have focused on using RI matching for two-phase flow in porous structures.

1.1.3. *Two-phase flow in porous structures*

Ng et al. (1978) investigated liquid/liquid two-phase flow through a porous structure by RI matching of the wetting liquid and the solid. The motion of single droplets was observed by viewing the setup from two perpendicular directions. Kong et al. (2011) investigated gas/liquid two-phase flow inside a porous structure by RI matching of the continuous liquid phase and the solid. A moving laser sheet allowed an incremental 3D scan of the gas phase, in turn allowing a 3D reconstruction of the distribution of the gas phase.

Few studies have employed RI matching between two fluids and a solid. Burdett et al. (1981) was the first to match the RI of two fluids and a solid for optical measurements. Silicone rubber with $n = 1.422$ was used owing

99 to its low RI. This allowed the use of methylcyclohexane and a 67.9% w/w
 100 glycerine/water mixture. The mean hold-up along a line was estimated using
 101 a light absorption technique. Montemagno and Gray (1995) were the first to
 102 apply RI matching for investigating two-phase pore scale flow. Two aqueous
 103 and two nonaqueous liquids were used in a random packing of fused silica
 104 ($n = 1.46$). Tracer dyes were added to both phases. The aqueous phase was
 105 doped with a dye that congregates along the fluid-fluid interface. Plane illu-
 106 mination by a coherent laser enabled identification of the two phases. A 3D
 107 scan of the two fluids in the porous structures was obtained by using precision
 108 translators for the optical measurement equipment. Alternatively, Stöhr et
 109 al. (2003) measured the 3D field by the translation of the porous structure.
 110 They used silicone oil and a zinc chloride solution as RI matching fluids for
 111 fused silica. Two optical filters were mounted on a PC-controlled filter wheel
 112 to allow separate LIF measurements of the phases with one camera. Ov-
 113 dat and Berkowitz (2006) obtained similar measurements from polymethyl
 114 methacrylate (PMMA). The phases were distinguished using a fluorescent
 115 dye in the disperse phase. Finally, Krummel et al. (2013) used two fluids
 116 that closely match the RI of glass beads to measure the 3D phase distri-
 117 bution. This was achieved using a confocal microscope that (slice-by-slice)
 118 observed the fluorescence of a tracer in the wetting continuous liquid phase.

119 In comparison to the abovementioned studies that used particles with
 120 random packing, the porous structure investigated in our study has a defined
 121 geometry that is manufactured by rapid prototyping. This enables optical
 122 measurement techniques for two-phase flow in designed porous structures.

123 1.2. Definitions

124 Nondimensional numbers are defined with continuous phase properties,
125 hydraulic diameter, and interstitial velocity. The hydraulic Reynolds number
126 is defined as follows:

$$Re_h = \frac{u \cdot d_h}{\nu_b}. \quad (1)$$

127 The interstitial velocity u was calculated from the total volumetric flowrate
128 $\dot{V}_a + \dot{V}_b$. The hydraulic Weber number is defined as follows:

$$We_h = \frac{\rho_b u^2 d_h}{\sigma} \quad (2)$$

129 The hydraulic diameter is defined with the volume of fluid (V_f) and the
130 interface area between solid and fluid (A_{sf}) according to Carman (1937):

$$d_h = \frac{4V_f}{A_{sf}} \quad (3)$$

131 The choice of the characteristic diameter in flow through porous struc-
132 tures is somewhat arbitrary. As discussed in Häfeli et al. (2014), suitable
133 choices of the characteristic diameter could be the cell diameter (Dukhan
134 et al., 2014), ligament diameter (Zenklusen et al., 2014), particle diameter
135 (Jolls et al., 1966), pore diameter (Häfeli et al., 2013), or the diameter of
136 particles with the same surface area per solid volume (Horneber et al., 2012).
137 The choice of the characteristic diameter strongly influences the Reynolds
138 number. In our geometry, for example, the cell diameter is 2.8 times larger
139 than the pore diameter. Therefore, choosing the cell diameter as the char-
140 acteristic length leads to Reynolds numbers that are 2.8 times higher than if
141 the pore diameter were chosen instead. This explains why it is generally dif-
142 ficult to compare studies by different authors. Therefore, the characteristic

143 diameter to be chosen depends on the dominant physics. Some porous struc-
 144 tures are built up as a network of cylinders. In such a case, the results are
 145 frequently compared to studies on flow around single cylinders (Zenklusen et
 146 al., 2014). It is therefore helpful to use the cylinder diameter as the charac-
 147 teristic diameter. In studies of flow through packed beds of spheres, it can
 148 be more convenient to take the sphere diameter as the characteristic length
 149 scale. However, if the goal is to compare flow through porous structures of
 150 arbitrary geometry, the characteristic length can be defined based on the
 151 hydraulic diameter.

152 The volumetric transport fraction of the disperse phase is defined as fol-
 153 lows:

$$\dot{\epsilon} = \frac{\dot{V}_a}{\dot{V}_a + \dot{V}_b} \quad (4)$$

154 where $\dot{V}_a$ denotes the volumetric flowrate of the disperse phase (anisole), and
 155 $\dot{V}_b$ the flowrate of the continuous phase (brine).

156 2. Experimental

157 2.1. Solid material and geometry

158 Two geometries are investigated. The first is the foam-like porous struc-
 159 ture introduced by Hutter (2010). This structure is designed as the negative
 160 of tetrahedrally arranged overlapping spheres. Fig. 1 shows an auxiliary view
 161 of one periodic unit of the porous structure (left) and a top view on the mea-
 162 surement plane (right). The large void spaces inside the structure are referred
 163 to as *cells* in the following text. The openings between two adjacent cells
 164 are called *pores*. A cell is connected to twelve other cells. A fluid primarily
 165 enters a cell from its three upstream neighbors, and it primarily leaves the

Figure 1: One periodic unit of the foam-like porous structure (Häfeli et al., 2014). Left: auxiliary view, right: cut at the measurement plane.

Table 1: Characteristic dimensions of the foam-like porous structure.

Empty pipe diameter D [mm]	20
Length of periodic unit [mm]	19.25
Cell diameter [mm]	7.94
Cell distance [mm]	7.86
Rounding radius [mm]	0.34
Pore diameter [mm]	2.88
Bulk Porosity [%]	78.7
Porosity of manufactured structure [%]	70.4
Hydraulic diameter [mm]	6.23

166 cell to enter the three downstream neighbors. The remaining six neighbors
 167 are perpendicular to the main flow axis, and therefore there is very little flow
 168 to these cells. The measurements are conducted in a plane through the axis
 169 of the tube. Of the twelve pores per cell, only two are in the measurement
 170 plane. Table 1 lists the dimensions of the foam-like porous structure. Owing
 171 to the boundary effects, the porosity of the manufactured structure is smaller
 172 than the bulk porosity.

173 The second geometry under investigation is a Sulzer SMXTM static mixer.
 174 It is made up of ligaments that are inclined at 45° with respect to the axial
 175 direction (Fig. 2). It has the same hydraulic diameter and same porosity as
 176 the foam-like porous structure. Table 2 lists its dimensions. The measure-
 177 ment plane was chosen to be 2 mm off-center. By doing so, the measurement
 178 plane is exactly between the two ligaments.

179 A major difference between the foam-like porous structure and the SMX

Figure 2: Four periodic units of a Sulzer SMXTM static mixer. Left: auxiliary view, right: cut at the measurement plane.

Table 2: Characteristic dimensions of Sulzer SMXTM.

Empty pipe diameter D [mm]	20
Length of periodic unit [mm]	13.5
l_1 [mm]	27
l_2 [mm]	6.75
l_3 [mm]	2.8
l_4 [mm]	4
Bulk Porosity [%]	70.78
Porosity of manufactured structure [%]	70.67
Hydraulic diameter [mm]	6.14

Figure 3: Relative free cross-sectional area along one periodic unit of the two geometries. Left: cut through Sulzer SMXTM showing the free cross section (black) and the solid in the area of the cut (yellow). Right: cut through foam-like porous structure.

180 is the free cross-sectional area, i.e., the area in the tube that is not occupied
 181 by the porous structure. Fig. 3 shows the free cross-sectional area divided by
 182 the cross section of the empty tube as a function of the axial coordinate. On
 183 average, both geometries have a free cross-section of approximately 70.5 %
 184 (70.7 % and 70.3 %). However, the foam-like structure shows a wide range of
 185 variation (from 53 % to 92 %). In comparison, the free cross section in the
 186 SMX shows only a small range of variation (from 64 % to 75 %).

187 The investigated structures are made of Somos[®] WaterShed XC 11122
 188 (hereafter called WaterShed). This is a transparent material that was devel-
 189 oped for manufacturing with stereolithography. During the experiments, it
 190 was observed that the RI of the solid changes slowly owing to the absorption
 191 of liquid. To prevent RI changes, WaterShed was cleaned every day with
 192 distilled water and then flushed with dry compressed air to allow drying and

193 desorption.

194 When WaterShed was used for single phase flow experiments with anisole,
195 the structure had to be coated (Butscher et al., 2012; Häfeli et al., 2014).
196 Without varnish coating, anisole would dissolve WaterShed within a short
197 time. However, in two-phase flow experiments, it is not necessary to coat
198 the structure. As anisole was used as the disperse phase, it was not in direct
199 contact with the solid. Therefore, the uncoated structure of WaterShed was
200 stable.

201 *2.2. RI matching liquids*

202 Two fluids had already been used in single-phase flow experiments (Butscher
203 et al., 2012; Häfeli et al., 2014). The organic liquid anisole is used as a pure
204 substance. The second fluid is an aqueous mixture of sodium iodide and zinc
205 iodide. The used fluid contained 1000 g of water, 1400 g of sodium iodide,
206 and 680 g of zinc iodide.

207 More details such as the temperature and composition dependence of the
208 RI are provided in Häfeli et al. (2014). There, the concentration of zinc iodide
209 was slightly higher because the RI of WaterShed was measured as 1.519 by
210 an Abbe refractometer. For the experiments presented here, we decided to
211 match the RI to $n = 1.515$ in accordance with the manufacturers datasheet
212 because we found that the measurements of the RI of the solid could have
213 some uncertainty owing to the use of a contact liquid that is absorbed by
214 WaterShed. The interfacial tension between the two liquids was measured
215 with the Wilhelmy plate method. The interfacial tension was estimated to
216 be 9.6 mN m^{-1} .

217 The selection criteria for the two fluids was that they be immiscible and

218 that they have the same RI as WaterShed. In addition, a fluorescent dye
 219 should be stable in the disperse phase, and PIV particles should be dispersible
 220 in the continuous phase. For the selected system, Rhodamine B can be used
 221 as a fluorescent dye. We found that it dissolves in anisole and is stable.
 222 Different seeding particles for PIV were evaluated. The density of glass
 223 beads and polyamide particles was found to be too low. These particles float
 224 on the brine and collect at the interface between brine and anisole in the
 225 two-phase separator. We therefore chose particles of higher density; in this
 226 regard, we found 2 μm aluminum oxide particles with a density of 3.95 g cm^{-3}
 227 to be suitable.

228 In Häfeli et al. (2014), we discussed the discoloration of brine that occurs
 229 owing to the oxidation of zinc iodide. As an improvement over the previous
 230 setup, we used compressed nitrogen instead of compressed air to flush the
 231 system. This prevented the oxidation of brine. Therefore, the brine remained
 232 transparent. However, the simultaneous use of two fluids caused some new
 233 problems as they are not completely inert. In the two-phase separator, we
 234 observed the formation of a slurry. It must have formed from anisole as it is
 235 present only in that phase. Its density is slightly higher than that of anisole,
 236 and it collects near the liquid-liquid interface in the two-phase separator. The
 237 suction line of the pump for anisole was placed such that only pure anisole
 238 is sucked from the two-phase separator.

239 *2.3. Experimental setup*

240 The setup was configured for simultaneous PIV and LIF measurements.
 241 Fig. 4 shows the arrangement of the laser, measurement section and cameras.
 242 A pulsed Nd-YAG laser with a wavelength of 532 nm was used (Litron Nano

Figure 4: Illustration of the measurement equipment. A green laser illuminates a plane in the porous structure. The laser light is reflected by PIV particles (green) and excites the fluorescent tracer dye (orange). The light from the two sources is separated by a dichroic mirror and is recorded by two cameras equipped with filters.

243 L 135-15). The laser has a pulse energy of 120 mJ and a frequency of 15 Hz.
 244 A set of cylindrical lenses expanded the laser beam to a sheet of 100 μm
 245 thickness. The light is then reflected by PIV particles in the continuous
 246 phase, and it excites the fluorescent dye in the disperse phase. The reflected
 247 light from PIV particles has the same wavelength as the laser (532 nm). The
 248 light emitted by the fluorescent dye has a higher wavelength ($> 570\text{ nm}$).
 249 This allows us to separate the two images by using a dichroic mirror. In
 250 addition, we mounted filters on the two camera lenses.

251 Because the brine was relatively expensive, a closed system was built for
 252 them (Fig. 5). The liquid-liquid separator is a simple glass vessel with a
 253 volume of 2.5 L. The heavier brine is collected at the bottom of the ves-
 254 sel whereas the lighter anisole is collected at the top. Two gear pumps

Figure 5: Experimental setup. Fluids are stored in a liquid-liquid separator ①. From there they are sucked by two gear pumps ②. The flowrate is controlled by mass-flow controllers ③. After tempering ④, the brine enters a calming section ⑤. The anisole is injected via a nozzle immediately in front of the porous structure ⑥. Several positions are equipped with temperature (T), pressure (P), and flowrate (F) indicators (I), controllers (C), and alarms (A).

(VGS200 from Verder Catag) are used to convey the liquids. The flowrate is controlled using a mass-flow controller, which works based on the Coriolis principle (Bronckhorst M55-AGD-22-O-S). The mass-flow controllers measure the flowrate with an accuracy of 0.2%. In addition, they measure the density and temperature. A thermostat is used to remove heat from the pumps and to bring the system to a constant temperature of 20 °C. The brine then enters an empty pipe with an inner diameter of 20 mm and a length of 100 mm. After this section and right in front of the porous structure, the anisole is injected through a nozzle with a diameter of 1 mm. After the measurement section, the fluids flow back to the two-phase separator.

The choice of materials that are compatible with anisole and brine is somewhat limited. The piping and fittings were made out of stainless steel and polyamide. The plant further comprised some seals. In a first attempt, we used nitrile butadiene rubber (NBR) sealing rings. These are chemically resistant to both fluids; however, they swell up in anisole. This caused some problems when dismantling the plant, and the sealing rings had to be replaced from time to time. Ultimately, we found it to be worthwhile to invest in more expensive perfluoroelastomer (FFKM) sealing rings. These proved to be chemically resistant to both fluids.

2.4. Droplet detection

The procedure to evaluate the LIF images starts with background subtraction and masking of the solid region. The image is then converted to a black/white image (b/w). To find the right threshold, the gray-value histogram of an image is analyzed. An intensity peak is found at a gray value corresponding to the background. The peak of the histogram plus two times

280 its standard deviation is then set as the initial threshold. After this step, all
 281 droplets that contain less than 65 pixels are rejected. This corresponds to
 282 droplets with a diameter of 88 μm . Dark spots inside the droplets are then
 283 closed. A disadvantage of this step is that if there are brine droplets inside
 284 anisole droplets, this area is falsely attributed as anisole. This has to be
 285 kept in mind when images with many droplets in droplets are analyzed. The
 286 number of pixels in a droplet multiplied by the area of one pixel gives the
 287 cross-sectional area of the droplet. This is used to define the characteristic
 288 diameter of the droplet. The characteristic diameter of a droplet was defined
 289 as the diameter of a circle with equal cross section. The effect of observing
 290 a sectional plane of the droplet instead of the real droplet diameter will be
 291 discussed in [Section 3.2](#).

292 Some droplets are so close to each other that they are recognized as a
 293 single droplet. We found that the center of a droplet is brighter than its
 294 border. This causes droplets to be smaller when the b/w threshold is higher.
 295 We therefore developed a procedure that is based on incrementally increasing
 296 the b/w threshold. When two droplets stick together in the b/w image with
 297 a low threshold, they will be separated at a certain higher threshold. If
 298 the two new droplets are more round than the original, we define them as
 299 two droplets; however, if they are less round, we assume that the original
 300 was the real droplet. The reason for this distinction is that the laser sheet
 301 is not completely homogeneous but contains some darker stripes. Fig. 6
 302 shows the change in b/w images for droplets that are very near (top) and
 303 a droplet that is inhomogeneously illuminated (bottom). In the images on
 304 top, three droplets are very close to each other. In a b/w image with low

Figure 6: Influence of changing threshold for sticking bubbles (top) and bubbles in a laser sheet with stripes (bottom). The left shows a grayscale image, the middle shows a b/w image with a low threshold, and the right shows a b/w image with a higher threshold.

305 threshold (top,center), the three droplets are recognized as a single droplet.
 306 However, in an image with higher threshold (top,right), the three droplets
 307 are separated. The bottom of Fig. 6 shows a droplet that is in a region of
 308 inhomogeneous laser illumination. With a low b/w threshold, the droplet
 309 is recognized correctly. However, at higher threshold, the droplet appears
 310 as three non-round regions. Such droplets are identified correctly using the
 311 above-described procedure.

312 This post-processing is quite simple compared to post-processing proce-
 313 dures in other measurement techniques. In shadowgraphy, for example, one
 314 observes overlapping bubbles and bubbles that are out-of-focus. Lelouvetel
 315 et al. (2011) distinguished focused bubbles from unfocused ones by filtering
 316 the gradient of the gray value at the boundary of the bubble. Bröder and
 317 Sommerfeld (2007) used a Sobel filter to identify the bubble contours and

318 then used the gray value gradient to identify unfocused bubbles. Honkanen
 319 et al. (2005) show an algorithm that identifies strongly overlapping bubbles
 320 by fitting ellipses on the clustered arcs of the perimeter of bubbles. As we
 321 applied plane illumination, we did not observe unfocused droplets, and we
 322 could use a relatively simple detection method.

323 3. Error sources

324 3.1. *Effect of dichroic mirror*

325 A dichroic mirror was used to separate the PIV and the LIF image. It was
 326 found that the light reflected by the mirror is blurred. Fig. 7 shows an image
 327 (of droplets inside a cell) obtained without a dichroic mirror. On the other
 hand, Fig. 8 shows an image of the light reflected by the dichroic mirror. In

Figure 7: LIF image obtained without a
dichroic mirror.

Figure 8: LIF image obtained with a
dichroic mirror.

328

329 a direct comparison of measurements obtained with and without the mirror,
 330 we found that the droplet size is 9.3% larger when a mirror is used. It could

Figure 9: Measured droplet size on image as a function of its position relative to the laser sheet. A small droplet $d = 0.6$ mm and a large droplet $d = 1.2$ mm are shown.

therefore be argued that it is better to not use a mirror. However, we found that the evaluation of the images is more difficult when no mirror is used because the droplet images can show many fine stripes from inhomogeneous laser illumination. This makes it difficult to correctly identify the droplets. Therefore, the dichroic mirror is used for LIF measurements when no PIV measurements were performed simultaneously.

3.2. Error due to plane-illumination

As we apply plane-illumination, we see only a slice of the droplet. The measured droplet diameter is therefore smaller than the real diameter. In the following, we estimate the error due to plane-illumination. For the calculation, we assume spherical droplets. Fig. 9 shows the measured droplet size as a function of the droplet position relative to the laser sheet. The laser sheet is located between 0 mm and 0.1 mm on the x-axis. A droplet with a diameter of 0.6 mm is not visible when its center is at a position $x < -0.3$ mm.

345 When the center of the droplet is in the laser sheet, the largest diameter of
 346 the droplet is illuminated and the correct droplet size is measured. When the
 347 center is between -0.3 mm and 0 mm, a slice of the droplet is illuminated. It
 348 is assumed that the largest illuminated diameter of that slice is observed in
 349 the image. In this case, the observed diameter as a function of the position
 350 is between 0 mm and the actual size when the droplet travels from -0.3 mm
 351 to 0 mm on the x-axis.

352 Averaged over all positions, the measured droplet size is slightly lesser
 353 than the actual droplet size. The relative error depends on the droplet size
 354 and the thickness of the laser sheet. In our experiments, the laser sheet had
 355 a thickness of $100\text{ }\mu\text{m}$. Based on the calculation above, a droplet with a
 356 diameter of 0.6 mm is measured as being 0.49 mm across. A droplet with a
 357 diameter of 1.2 mm is measured as being 0.96 mm across. The ratio between
 358 the measured and the real droplet size is 81.5% for the small droplet and
 359 80.1% for the large droplet. If the laser sheet were infinitesimally thin, the
 360 measured diameter would be $\frac{\pi}{4} = 78.5\%$ of the real diameter. The maximal
 361 error due to plane illumination is therefore -21.5% . One could conclude
 362 that a thick laser sheet is beneficial for the measurement of the droplet size.
 363 However, if the laser sheet is too thick, we observe overlapping droplet images.
 364 This makes it difficult to identify single droplets in the images. A thin laser
 365 sheet is therefore preferred.

4. Results and Discussion

4.1. Local droplet concentration

Using the procedure described in Section [refdroplet detection](#), the LIF images were used to identify the regions of the disperse phase, i.e. the regions of anisole. After postprocessing, an image contains zeroes where there is brine and ones where there is anisole. Averaging 2000 images results in the probability that the corresponding location is occupied by anisole. This is referred to as local hold-up. Fig. 10 shows the local distribution of the hold-up in the Sulzer SMXTM static mixer. The fluid streams upwards and the ligaments in the measurement plane are aligned from bottom left to top right. It is observed that at low Reynolds numbers, the droplets are inhomogeneously distributed. The highest local hold-up is observed below the ligaments. The accumulation of droplets below the ligament can be described by buoyancy. As the droplets have a lower density than the continuous phase, they possess a higher velocity and collect below the ligaments of the static mixer. At higher Reynolds numbers ($Re_h = 135$), droplets are distributed more homogeneously. However, there remain some regions of higher droplet concentration. Apart from the area below the ligament, there are two areas with higher droplet concentration. At Reynolds numbers of $Re_h = 180$ and $Re_h = 269$, droplets can be found at any position within the static mixer. At these Reynolds numbers, only the small zone behind the ligament near the wall exhibits higher droplet concentrations, possibly because droplets are guided there by other ligaments.

An additional observation is that there seem to be lines where very few droplets are observed. These lines appear due to the refraction of light at

Figure 10: Average distribution of hold-up in SMX static mixer at volumetric transport fraction of 4 %. The boundary of the tube as well as the in-plane ligaments are indicated by solid lines. Out-of-plane ligaments are indicated by dotted lines.

the ligaments that are between the measurement plane and the camera. As the RI of the solid and the fluid is matched, refraction should not occur. However, the match is not perfect, and some refraction always occurs. This is especially the case when a light ray crosses a phase interphase with an incident angle close to 90° . According to Snell's law, the relation between the angle of incidence (θ_1) and the angle of refraction (θ_2) is given as follows:

$$\frac{\sin(\theta_1)}{\sin(\theta_2)} = \frac{n_2}{n_1} \quad (5)$$

If the RI of the solid and the fluid show a mismatch in the third digit, e.g., $n_2 = 1.515$ and $n_1 = 1.514$, an incident light beam with an angle of 90° leaves the interface with an angle of 87.9° . In contrast, an incident light beam with an angle of 10° is refracted with an angle of 9.99° . This shows that even though refraction was generally not a problem, it can still disturb experiments in cases where the light passes a phase interface with a high angle to the normal.

In the foam-like porous structure, droplets were not observed to accumulate due to buoyancy. Also in the foam-like porous structure, droplets were observed to follow preferred paths at low flowrates. Fig. 11 shows the local hold-up **in the cell already shown in Fig. 7**. For orientation, the velocity field inside the porous structure is shown (Häfeli et al., 2014). The velocity field was measured by PIV in single-phase flow. Despite all differences, it is expected that the mean velocity field in two-phase flow shares some characteristics with single-phase flow. For example, it is observed in single-phase flow that the fluid entering the cell from the bottom left leaves the measurement plane and exits the cell via two pores on the bottom right of the cell (not visible in the measurement plane). Similarly, the fluid enters through

Figure 11: Velocity field (Häfeli et al., 2014) and average distribution of hold-up in foam-like porous structure. The volumetric transport fraction in these experiments was 4%.

two pores on the top left of the cell and leaves the cell through the visible pore at the top right. It is expected that in a first approximation, this feature of the flow through the porous structure remains the same in two-phase flow.

At a Reynolds number of 137 it can be seen that droplets are mainly found in the bulk flow. At higher flowrates, the distribution of droplets is more homogeneous. Again, the limitation of the RI matching technique for the measurement of local hold-up distributions is visible. At the right of the pore, there is a band of low hold-up because the illuminating laser crosses a phase interface at a very high angle to the interface normal. As discussed above, this leads to strong refraction even for accurately matched RIs. A similar effect occurs on the path of light between the measurement plane and the camera. It crosses a phase interface perpendicularly and is refracted. This produces a circle of low hold-up in the center of the measurement plane. This effect is less pronounced at low flowrates because droplets are larger and can still be correctly identified even when a stripe within the droplet is insufficiently illuminated.

4.2. Droplet size distribution

The droplet sizes are estimated from 2000 images. The histogram of the number distribution is obtained by classifying the size in bins of 0.05 mm. For the modeling of mass transfer in the chemical reactor, we are interested in the area rather than the number of droplets. Therefore, we transform the number distribution into an area distribution. Fig. 12 shows the number and area distributions of measurements upstream and inside the foam-like porous structure. In the measurements upstream the porous structure we observe the droplets as they were produced by the nozzle. At this position

Figure 12: Histogram of droplet sizes for an experiment with 10 % volumetric transport fraction and $Re_h = 241$; size distribution in front of porous structure and inside porous structure (201 mm from the entrance).

the droplets are larger and have a wider size distribution than inside the porous structure. Comparing the area with the number distribution it is observed that the mode of the area distribution is at a larger diameter than the mode of the number distribution because larger droplets have a larger surface area.

A meaningful characteristic of the droplet size distribution is the maximum stable droplet diameter d_{max} . Instead of estimating d_{max} , droplet sizes are frequently related to d_{90} , a value that is measured with more confidence

448 (Theron and Le Sauze, 2011). d_{90} , d_{50} and d_{10} are the largest diameters that
 449 represent 90 %, 50 %, and 10 % of the volume of the droplets, respectively.
 450 Instead of using d_{max} or d_{90} to characterize the droplet size distribution,
 451 models in the literature usually report the Sauter mean diameter (Lemenand
 452 et al., 2003):

$$d_{32} = \frac{\sum_i n_i d_i^3}{\sum_i n_i d_i^2} \quad (6)$$

453 where n_i is the number of droplets of size d_i . The Sauter mean diameter is the
 454 diameter of monodisperse droplets that have the same area to volume ratio
 455 as the measured droplet collective. This is motivated by a study by Sprow
 456 (1967), who found that in a stirred tank emulsification, numerous average
 457 diameters are directly proportional to the maximum drop size. Fig. 13 shows
 458 that in our experiments, both d_{90} and d_{10} are linearly correlated to the Sauter
 459 mean diameter. It is also observed that the ratios d_{90}/d_{32} and d_{10}/d_{32} are
 460 similar for the Sulzer SMXTM static mixer and the foam-like structure. We
 461 conclude that the droplet size distribution can be well characterized by the
 462 Sauter mean diameter.

463 4.3. *Droplet size along the structures*

464 Fig. 12 already shows that in front of the porous structure, the droplets
 465 are larger and have a wider size distribution. Inside the porous structure, the
 466 droplets are smaller and show a narrower size distribution. More quantita-
 467 tive insight into the break-up mechanism is gained by measuring the droplet
 468 size along the porous structure. Fig. 14 shows the evolution of d_{32} in the
 469 foam-like porous structure and in the SMXTM. Measurements were made
 470 for volumetric transport fractions of $\dot{\epsilon} = 4\%$ and $\dot{\epsilon} = 10\%$. We first dis-
 471 cuss the results for $\dot{\epsilon} = 10\%$. The initial droplet size (produced by a nozzle

Figure 13: Characteristic diameters d_{90} , d_{10} and d_{32} in comparison.

Figure 14: Break-up of droplets as they travel through porous structures at varying volumetric transport fraction. Brine flowrate was constant, resulting in Reynolds numbers between 224 and 241.

472 of 1 mm diameter) is 1.20 mm in the experiment with the foam-like porous
 473 structure and 1.49 mm in the experiment with the Sulzer SMXTM. The dif-
 474 ference can be caused by several factors. One is that the measurement plane
 475 for the SMX was 2 mm off center owing to the requirements of the structure.
 476 Another reason is a difference in pressure drop between the two structures.
 477 This results in different pressures at the nozzle, which eventually affects the
 478 operation of the nozzle. Another effect could be a coupling between pressure
 479 variations in the porous structure and at the nozzle. Notwithstanding these,
 480 differences the droplet size at the entrance varies by only 24 % between the
 481 foam-like structure and the SMX. In the foam-like porous structure, it is ob-
 482 served that within the first 50 mm, the droplet size is reduced from 1.20 mm
 483 to 0.74 mm. Further downstream, the droplets approach a diameter of ap-
 484 proximately 0.55 mm. In the SMX, the reduction of droplet size is slower,
 485 and the droplets approach a final size of approximately 0.95 mm.

486 In the experiments with $\dot{\epsilon} = 4\%$, the observations are similar. The
 487 initial droplet size was 1.45 mm in the experiment with the foam-like porous
 488 structure and 1.80 mm with the SMX. This corresponds to a difference of
 489 24 %, and the same reasons as for the experiments with $\dot{\epsilon} = 10\%$ apply. The
 490 difference in initial droplet size between the experiments with $\dot{\epsilon} = 4\%$ and
 491 $\dot{\epsilon} = 10\%$ can be explained by the production of the droplets by the nozzle.
 492 At a lower flowrate ($\dot{\epsilon} = 4\%$), the nozzle produces smaller droplets. Toward
 493 the end of the structures, the difference in droplet size for the experiments
 494 with $\dot{\epsilon} = 4\%$ and $\dot{\epsilon} = 10\%$ diminishes.

Figure 15: Droplet size as a function of interstitial velocity and volumetric transport fraction for Sulzer SMXTM static mixer and foam-like porous structure.

4.4. Effect of flowrate and volumetric transport fraction

Fig. 15 shows the droplet size as a function of the volumetric transport fraction and interstitial velocity. These measurements are conducted at ≈ 200 mm from the entrance of the two geometries. Again, we observe that the droplets in the foam-like structure are smaller than in the SMX. The interstitial velocity appears to have a strong effect on the droplet size. In comparison, changing the volumetric transport fraction from $\dot{\epsilon} = 4\%$ to $\dot{\epsilon} = 10\%$ appears to have a minor effect on the droplet size. A clear trend cannot be identified. At high flowrates, larger droplets are observed at higher

504 volumetric transport fractions. This is according to expectations as a higher
 505 droplet density generally leads to a higher droplet coalescence rate. However,
 506 at low flowrates, the droplets are smaller for higher volumetric transport frac-
 507 tions. This observation is made in the foam-like porous structure as well as in
 508 the SMX because of variations in the inlet droplet size. As discussed above,
 509 the droplets are produced by a nozzle with an inner diameter of 1 mm. The
 510 size of the droplets produced by this nozzle depends on the flowrate through
 511 the nozzle. It is possible that some effect of the volumetric transport fraction
 512 stems from this. According to Fig. 14, small changes in the droplet size still
 513 occur after 200 mm.

514 4.5. Comparison to literature

515 Middleman (1974) proposed a correlation assuming Kolmogoroff's theory
 516 of turbulence, and here, it is written according to hydraulic numbers:

$$\frac{d_{32}}{d_h} = C_1 We_h^{-0.6} f_h^{-0.4} \quad (7)$$

517 In the turbulent pipe flow, the friction factor f_h is a weak function of the
 518 Reynolds number. With the approximation

$$f_h \propto Re_h^{-0.25} \quad (8)$$

519 the droplet size was given as a function of the Weber and Reynolds numbers:

$$\frac{d_{32}}{d_h} = C_1 We_h^{-0.6} Re_h^{0.1} \quad (9)$$

520 Some authors estimated the value of the exponents by varying dimensionless
 521 numbers (e.g. Legrand et al., 2001). When operating in the turbulent flow
 522 regime, the exponent of the Weber number reported in the literature ranges

523 from -0.5 to -0.859 ; but most authors report a value close to -0.6 (Theron
524 and Le Sauze, 2011).

525 At intermediate Reynolds numbers, the droplet size is still a function of
526 the Reynolds and Weber number; however, the exponents change. Theoretically,
527 experimental data could be used to fit the exponents of a correlation.
528 However, to do so, the nondimensional numbers should be varied independently.
529 This can be achieved by varying the viscosity and interface tension.
530 In our case, the challenge is that the RI has to be constant. Therefore, we
531 only varied the flowrate and volumetric transport fraction and compared our
532 experiments to correlations available in the literature. A study of emulsification
533 in a Sulzer SMXTM static mixer at similar Reynolds numbers was
534 conducted by Legrand et al. (2001). They used three fluid pairs to investigate
535 the droplet size at Reynolds numbers between 4 and 320. Their correlation
536 in terms of hydraulic numbers is as follows:

$$\frac{d_{32}}{d_h} = C_2 We_h^{-0.2} Re_h^{-0.16} \quad (10)$$

537 It described their experimental values with a mean error of approximately
538 10 %. We found that our experiments could also be well described by an
539 expression with these exponents. Fig. 16 shows the droplet size as a function
540 of $We_h^{-0.2} Re_h^{-0.16}$. It appears that the curves of the two static mixers show
541 a different slope. The data of the SMX is correlated by $C_2 = 0.47$ and
542 shows an error of 13 %. The data of the foam-like structure is correlated by
543 $C_2 = 0.34$ and shows an error of 4 %.

544 The two investigated structures show different emulsification performances
545 even though they have the same hydraulic diameter and the same porosity.
546 The differences in droplet size therefore arise from differences in the geome-

Figure 16: Correlation of experimental data in terms of the hydraulic Weber and Reynolds numbers.

try. A substantial difference between the structures is their free cross section. The average free cross section of the two structures is equal as they have the same porosity. However, the foam-like structure shows strong variations, as discussed in Section 2.1. Therefore, the fluid is repeatedly accelerated and decelerated. In our experiments, it was observed that in the structure that shows large changes in the free cross section, the droplets produced are smaller.

5. Conclusions

We introduced a system of two immiscible fluids and a solid (manufactured by rapid prototyping), all three of which have the same RI. This system permits optical measurements in two-phase flow through designed porous structures. We used this to investigate liquid-liquid two-phase flow through a foam-like porous structure and a Sulzer SMXTM static mixing element.

Using LIF, a fluorescent tracer in the disperse phase was observed. This enabled us to observe the droplet size and position inside a porous structure. We observed that at low flowrates, droplets follow preferred paths, whereas at higher flowrates, they are more homogeneously distributed within the structures.

The droplet size distribution was found to be well represented by the Sauter mean diameter d_{32} . By measuring along the porous structure, we found that the droplet size is quickly reduced in the foam-like porous structure. In comparison, the droplets in the SMX disintegrate more slowly, and the final droplet size is larger. As both geometries have the same porosity and hydraulic diameter, we conclude that the change in the free cross section

571 is also an important parameter. We observed that in the geometry with large
572 changes in the free cross section, the droplets are smaller.

573 Further, we investigated how the droplet size is affected by changes in
574 flowrate and volumetric transport fraction. We observed that the droplets
575 are smaller at higher flowrates. The effect of the volumetric transport frac-
576 tion was found to be undetermined. Owing to droplet coalescence, larger
577 droplets are generally expected when the volumetric transport fraction is
578 higher. However, this was only observed at high Reynolds numbers. We
579 conclude that the measured droplet size is affected by the size produced by
580 the nozzle. Small changes in droplet size are observed even after 200 mm of
581 the structures.

582 Finally, droplet sizes were correlated as a function of dimensionless num-
583 bers. It was found that the droplet size correlates well to $(We_h^{-0.2} Re_h^{-0.16})$,
584 as observed by Legrand et al. (2001) for similar Reynolds numbers.

585 **Nomenclature**

586 *Roman letters*

587	A_{sf}	Interface area between solid and fluid [m ²]
	D	Empty pipe diameter [m]
	d	Droplet diameter of an individual droplet [m]
	d_{32}	Sauter mean droplet diameter [m]
	d_{10}	Largest diameter representing 10 % of volume [m]
	d_{90}	Largest diameter representing 90 % of volume [m]
	d_{max}	Largest droplet diameter [m]
	d_h	Hydraulic diameter of a porous structure [m]
	f_h	Friction factor [m/s]
	l_1, l_2, l_3, l_4	Dimensions of Sulzer SMX TM [m]
	n	Refractive index [-]
	Re_h	Reynolds number based on hydraulic diameter [-]
	u	Interstitial velocity [m s ⁻¹]
	V_f	Volume of fluid [m ³]
	$\dot{V}$	Volumetric flowrate [m ³ s ⁻¹]
	We_h	Weber number based on hydraulic diameter [-]
	x_1	Axial coordinate [m]
	x_2	In-plane radial coordinate [m]

588 *Greek letters*

589	$\dot{\varepsilon}$	Volumetric transport fraction [-]
	ν_b	Kinematic viscosity of continuous phase [$\text{m}^3 \text{s}^{-1}$]
	ρ_b	Density of continuous phase [kg m^{-3}]
	σ	Interface tension [N m^{-1}]
	θ_1, θ_2	Angle of incidence and refraction [$^\circ$]

590 *Abbreviations*

591	b/w	Black / White
	LDA	Laser Doppler anemometry
	LIF	Laser induced fluorescence
	PIV	Particle image velocimetry
	PTV	Particle tracking velocimetry
	RI	Refractive index

592 *Sub- and superscripts*

593	a	Anisole (disperse phase)
	b	Brine (continuous phase)

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