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An Experimental Characterization of Downwards Gas-Liquid Annular Flow by Laser-Induced Fluorescence: Flow Regimes and Film Statistics

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Abstract

Downwards co-current gas-liquid annular flows were studied experimentally and characterized. An advanced optical laser-based measurement technique, namely Planar Laser-Induced Fluorescence (PLIF), was used for the visualization of the annular flow over a range of liquid Reynolds numbers $Re_L = 306 - 1532$ and gas Reynolds numbers $Re_G = 0 - 84600$. Four distinct flow regimes, namely the ‘dual-wave’, ‘thick ripple’, ‘disturbance wave’ and ‘regular wave’ regimes, have been identified based on qualitative information and a consequent quantitative analysis that provided information on the film thickness, interface and wave statistics, and gas entrainment into the liquid film. The mean film thickness data are generally in good agreement with previous studies. Evidence suggests that the turbulent gas phase affects strongly the shape of the interface, and that the mechanism for gas entrainment into the liquid film is strongly reliant on the existence of large-amplitude waves.

Keywords: Annular flow, entrainment, laser-induced fluorescence, flow regimes, film statistics

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1. Introduction

Two phase gas-liquid flow can exhibit a variety of flow regimes, which are dependent on the flow-rates of the two phases, the choice of fluids (gas and liquid), the sizes of the fluid domains, and also on the orientation of the flow. In the case of a vertically orientated pipe, the two-phase flow can be generally described as bubbly, slug or annular (Barnea et al., 1982; Usui, 1989). In the latter case of *annular* flow, which is observed at higher flow-rates, the gas phase occupies the central core of the pipe cross-section and the liquid phase flows as a film in contact with the inner surface of the pipe, in an annular region between the gas and the pipe wall. In addition, gas bubbles are entrained in the liquid film and liquid droplets are entrained in the gas core. In the special case of *downwards* annular flow, a film flow of liquid on the inner surface of the pipe wall can be formed even at zero gas velocities (the so-called ‘falling film’ case).

Understanding the behaviour of two-phase interfacial film flows is not only important from a fundamental perspective (e.g., for predicting heat transfer in such flows (Mathie et al., 2013)), but also from a practical point of view. In particular, annular flows are central to a large number of applications, covering a wide range of important industrial processes, systems and plants. Upwards flows are commonly found in the oil-and-gas industry (e.g., raisers, transfer pipelines, gas-liquid oil wells), whereas downwards annular flows are present in important industrial process units, such as condensers and evaporators, distillation towers, and chemical reactors. The industrial relevance of annular flows can be seen, for example, in the work of Nakoryakov et al. (1976), who reported that surface instabilities (i.e., wave activity) can in-

crease mass transfer during the absorption of carbon dioxide by water by up to 170%. An accurate identification of the particular flow regime exhibited in relevant process units, systems, transportation lines or plants is crucial for their correct design and operation. Both upwards and (less so) downwards annular flows have been studied by a number of investigators; in this paper we focus on a vertically downwards co-current gas-liquid annular flow.

The basis of our understanding of downwards co-current annular flows was laid in the 1960s and 1970s (Telles and Dukler, 1970; Chu and Dukler, 1974, 1975; Webb and Hewitt, 1975). In these pioneering early studies, annular flows were characterized based on geometrical or topological features of the gas-liquid flow, by identifying and grouping these features into flow regimes. Specifically, based on topological features, Webb and Hewitt (1975) identified four different flow regimes for downwards annular flow (DAF): (i) the ‘ripple’ regime, where the interface is occupied by a pattern of ripples, (ii) the ‘regular’ wave regime in which disturbance waves traverse a film substrate that is covered with ripples, (iii) the ‘dual-wave’ regime, where two different disturbance wave types coexist, and (iv) the ‘thick ripple’ regime in which the wave activity decreases with increasing liquid Reynolds number.

In fact, annular flow is often described in terms of the class of interfacial waves that cover the liquid substrate, which is a thin liquid region that is present between the waves. Two main classes of waves are typically identified: (i) ‘ripple’ waves, which are short-lived waves that cover the substrate and are associated with small spatial amplitudes that decay quickly after their inception, and (ii) ‘disturbance’ waves, which carry a significant portion of the flowing liquid and have amplitudes many times (typically a factor of

51 ~ 5) the mean film thickness and high propagation speeds (Chu and Dukler,
 52 1974, 1975). It is important to emphasise, however, that the wave type
 53 nomenclature remains unsettled. One can find references in the literature to
 54 ‘capillary’ (i.e., ripple) waves and ‘roll’ (i.e., disturbance) waves in various
 55 studies (Asali et al., 1985; Yu et al., 2006). Furthermore, the definitions
 56 of the two main wave types are not without some ambiguity. As a result,
 57 a clear, objective, and generally accepted distinction between the different
 58 wave types is still lacking, which, consequently, can lead to discrepancies in
 59 the definitions and distinctions between the different flow regimes.

60 Disturbance waves are considered to be crucial interfacial phenomena in
 61 annular flows, not least because they carry a significant fraction of the total
 62 liquid flow-rate. These waves are ring-like structures that are circumferen-
 63 tially coherent around the pipe (Zhao et al., 2013), however, the thickness
 64 of a given wave need not necessarily be uniform around the circumference.
 65 Alekseenko et al. (2012) linked the circumferential non-uniformity to the fre-
 66 quency, amplitude and circumferential size of ripple waves that are formed by
 67 the presence of the disturbance waves. The existence of disturbance waves is
 68 also important for the entrainment of the liquid phase into the gas core (Az-
 69 zopardi, 1997). The mechanism of entrainment, as described by Woodmansee
 70 and Hanratty (1969), is based on the acceleration of ripples on the crests of
 71 disturbance waves. This leads to the lifting and consequent atomization of
 72 the liquid into the gas core in the form of droplets, thus determining the
 73 liquid fraction that is carried by the gaseous phase. Three additional mech-
 74 anisms of liquid entrainment were identified by Ishii and Grolmes (1975):
 75 (i) undercutting of the liquid film by the gas flow, (ii) bursting of gas bub-

bles present in the liquid film, and (iii) impingement (direct impact) of the liquid drops entrained in the gas phase on the surface of the liquid film.

Extensive statistical analyses of the dynamics of DAF have shown that the liquid film thickness fluctuates in a stochastic way. The first comprehensive statistical examination of DAF was presented by Chu and Dukler (1974, 1975). Webb and Hewitt (1975) reported further that a critical gas flow-rate exists, and hence also a gas superficial velocity (defined as the volumetric flow-rate of the gas divided by the full internal cross-sectional area of the empty pipe) and gas Reynolds number (Re_G), below which the frequency of appearance of disturbance waves is independent of the gas flow-rate. An increase in the gas superficial velocity above this critical value leads to an increase in the wave frequency. Increasing the liquid superficial velocity (liquid volumetric flow-rate divided by the full internal cross-sectional area of the pipe) was also found to increase the frequency wave frequency. The excellent study by Karapantsios et al. (1989) focused on falling films (i.e., with zero gas velocity) and described them in terms of various statistical moments of a film thickness (i.e., mean, RMS, skewness and kurtosis), probability density functions (PDF) and autocorrelation functions (ACF) of the interfacial film shape (thickness variation). Additionally, the waves were characterized via the evaluation of the power spectral density (PSD) and PDF of the wave peaks. The main findings of this effort were that: (i) the mean thickness data deviate from the Nusselt relation for a liquid Reynolds number of about $Re_L \geq 400$; (ii) for $Re_L \leq 1\,250$ the disturbance waves dominate the flow and they increase in amplitude, but not frequency of appearance, with increasing Re_L ; and (iii) for $Re_L \geq 1\,250$ the amplitude and frequency of appearance of

the disturbance waves is constant, however, the substrate thickens gradually with increasing Re_L . More recently, similar observations to those of Webb and Hewitt (1975), concerning the effect of the liquid superficial velocity on the frequency wave frequency, were reported by Karimi and Kawaji (2000).

In addition to the aforementioned previous work on wave characterization, other experimental studies focused mainly on the global (or integral) characterization of these flows (Koskie et al., 1989; Azzopardi, 1997), for example by measuring pressure gradients or phase fraction/hold-up (e.g., with pinch valves), or employed intrusive or point measurement methods such as hot wire probes, fibre-optics or wire-meshes (e.g., Zhao et al. (2013)). Yet, annular flows comprise highly localized, small-amplitude and high-speed flow disturbances, and so the characterization of these flows presents a major challenge from the point of view of experimental design for the provision of accurate, detailed data for understanding and model development and validation. Although invaluable in providing insight into these flows, these techniques are intrusive, and introduce a degree of uncertainty into the final measurement. Non-intrusive measurement techniques, such as flush-mounted conductance probes or acoustic methods, have also been used, but these can only provide temporal measurements at one point (Clark, 2002). These methods are also associated with complications, such as a reduced spatial resolution due to their finite size, and an increasingly insensitive signal response at large thicknesses due to signal nonlinearity in the case of the conductance probes, leading to an inability to capture accurately short-wavelength and/or large-amplitude (including the all-important disturbance) waves.

Typically, conductivity probes have been used extensively to measure a

liquid film thickness in annular flows (Wolf et al., 2001; Belt et al., 2010). Recently, non-intrusive visualization techniques, such as Laser-Induce Fluorescence (LIF), are gradually being introduced into the efforts to measure and characterize multiphase flows (Liu et al., 2006; Schubring et al., 2010; Morgan et al., 2013). These techniques can allow both the detailed qualitative investigation of these flows (i.e., imaging), and the reliable and simultaneous evaluation of quantitative flow parameters, such as film thickness and bubble size. The present study applies a detailed, spatiotemporally resolved, non-intrusive optical technique, specifically two-dimensional (2-D) planar LIF (PLIF), to the measurement of downwards co-current gas-liquid annular flows (DAFs). The measurements using these methods provide qualitative and quantitative data based on which the downwards annular flows can be accurately characterized and mapped, and based on which existing models can be verified and on which new models can be proposed. Attention is focused on the development of the various regimes that accompany this flow. The distinction between these regimes is determined based on a quantitative analysis of the waves in terms of the film thickness, interface and wave statistics, and gas entrainment into the liquid film. Our results indicate that the interfacial shape is dependent upon the level of turbulence in the gas phase, and that the presence of large-amplitude waves exerts a strong influence on the rate of gas entrainment.

2. Experimental Methods

This section presents the experimental flow facility used (Figure 1), the investigated experimental conditions, measurement procedures and post-processing analysis methods. Specifically, Section 2.1 contains a description of the experimental flow facility and outlines the flow conditions investigated, Section 2.2 contains details of the design of the visualization test section and describes the equipment used for the optical flow measurements, and finally, Section 2.3 contains a description of the processing methods performed on the raw PLIF images and details on the consequent data extraction analysis.

2.1. Flow facility

The experimental campaign was carried out in the ‘Downwards Annular Flow Laser Observation Facility’ (DAFLOF) in the Department of Chemical Engineering of Imperial College London. The DAFLOF flow facility was constructed specifically in order to allow the *quantitative* visual observation and measurement of flow phenomena occurring in downwards co-current gas-liquid annular flows. A schematic illustration of the DAFLOF facility is shown in Figure 1(a). The test section consists of a 3 m long and $D = 32.4 \pm 0.4$ mm nominal bore (inner diameter, ID) pipe, made from fluorinated ethylene propylene (FEP). The ID of the pipe was selected to match one of the two sections investigated experimentally by Webb and Hewitt (1975), for the purposes of direct comparison with that work.

The test gas and liquid were air and water. The air was taken from the laboratory compressed air supply at 5 – 6 bar and was filtered prior to use by a filter/pressure regulator combination (Norgren Olympian B15-F3).

171 The outlet of the test section was open to the atmosphere, while the gas
172 pressure at the inlet to the test section was monitored by a pressure gauge.
173 In our experiments the deviations of the inlet pressure relative to atmospheric
174 conditions in the laboratory over the range of the investigated flow conditions
175 were no greater than +5%, in the worst case.

176 The water was circulated in a closed-loop circuit by two centrifugal pumps
177 (Wilo Gold RS60). Both air and water flow-rates were measured by tur-
178 bine flow-meters fitted with a digital display (Fluid Well F110-X LCD). The
179 manufacturer-stated accuracy of the flow turbines is $\pm 0.5\%$ of full-scale,
180 while their repeatability is $\pm 0.1\%$ of full-scale. The water was injected as a
181 film at the top of the pipe test section by a specially designed injector featur-
182 ing bevels. The bevels formed a narrow channel through which the liquid was
183 injected in an axial direction down the inner wall of the pipe. This design
184 was chosen specifically because it was proven to form annular liquid films
185 with much improved azimuthal uniformity compared to equivalent porous
186 injector designs. A detailed description of the liquid injector can be found in
187 Zhao et al. (2013).

188 The experimental conditions were chosen so as to achieve operation in
189 the annular flow regime (Webb and Hewitt, 1975; Kendoush and Al-Khatib,
190 1994). The investigated superficial gas (air) and liquid (water) velocities,
191 which are both defined over the entire cross-sectional flow area in the pipe A
192 $= \pi D^2/4$, were selected to be in the ranges $U_G = Q_G/A = 0 - 34 \text{ m}\cdot\text{s}^{-1}$ and
193 $U_L = Q_L/A = 0.034 - 0.182 \text{ m}\cdot\text{s}^{-1}$, where Q_G and Q_L denote the gas and liq-
194 uid volumetric flow-rates, respectively. The investigated experimental matrix
195 can be found in Table 1. In addition, the corresponding Reynolds number

ranges were $Re_G = 0 - 84\,600$ (for the gas core) and $Re_L = 306 - 1\,532$ (for the liquid films). The gas Reynolds number, Re_G , is defined according to Numrich (1995), based on the pipe diameter D and the gas superficial velocity U_G , i.e., as $Re_G = U_G D / \nu_G$, where ν_G is the gas kinematic viscosity. The liquid Reynolds number, Re_L , is based on the bulk-mean film velocity $\langle u_f \rangle = Q_L / A_f$ (average volumetric flow-rate Q_L divided by the mean liquid film flow area $A_f = \pi D \langle \delta \rangle$) and the mean film thickness $\langle \delta \rangle$, i.e., $Re_L = \langle u_f \rangle \langle \delta \rangle / \nu_L = (Q_L / \pi D \langle \delta \rangle) \langle \delta \rangle / \nu_L = (Q_L / \pi D^2) D / \nu_L$. It is noted that Re_L used here, based on the bulk-mean film velocity and mean film thickness, is four times smaller than that based on the liquid superficial velocity U_L and the full pipe diameter D , whereas Re_G based on the superficial and bulk velocities are similar due to the relatively small extent of the overall pipe diameter taken up by the thin liquid films. The values $1.57 \times 10^{-5} \text{ m}^2\text{s}^{-1}$ and $9.02 \times 10^{-7} \text{ m}^2\text{s}^{-1}$ have been used for the kinematic viscosities of air and water, respectively. These values correspond to the mean ambient temperature and the pressure in the laboratory over the experimental campaign, i.e., 22°C and 1 bar. It is noted that the temperature varied during the experiments by $\pm 3^\circ\text{C}$. Together with the aforementioned 0 – 5% inlet pressure variations, the uncertainties in the reported flow-rates from the turbine flow-meters and the uncertainty associated with our measurement of the tube bore D with Vernier calipers, the total uncertainties in the reported values of Re_G and Re_L at the inlet lie in the range 3 – 10% for the gas and 3 – 12% for the liquid.

2.2. Measurement techniques

The laser visualization section was located 2.35 m downstream of the injector, or $74 D$, in order to allow the flow to develop. The development lengths in

annular flow were reported in a number of studies (Webb and Hewitt, 1975; Takahama and Kato, 1980; Wolf et al., 2001). In terms of the mean film thickness, Takahama and Kato (1980) found that the length necessary for the flow to become fully developed depends on Re_L . For relatively low and high liquid Reynolds numbers, $Re_L = 400$ and 1000 , the development length was found to be $32D$ and $12D$ from the inlet, respectively. Nevertheless, although the mean film thickness is expected to be fully developed by this distance from the inlet in the present study, it is also known from Webb and Hewitt (1975) that DAFs continue to evolve: (i) for at least $160D$ from the injector in terms of the probability distribution of wave thickness amplitude, and (ii) for at least $470D$ in terms of the mean exhibited wave frequency. Such lengths (i.e., more than $470D$, or 15 m) were considered prohibitively long for the present investigation, and so it is noted that the frequency data are representative of the development region of these flows at a distance of $74D$ from the inlet. Moreover, Webb and Hewitt (1975) report that by a length of $150D$ from the inlet the mean wave frequency approaches its long-length, fully-developed asymptotic value to within 1% (high Re_G) and 10% (falling films), depending on the flow condition. Hence, according to the DAF data provided by Webb and Hewitt (1975), the long-length, fully-developed asymptotic values of the frequency data are expected to be between 1% (at high Re_G) and 30% (for falling films) higher than the data provided here.

Considerable effort was made when manufacturing the visualization section to match the refractive indices of the liquid and the pipe wall solid in the line of sight of the optical measurement. The purpose of this matching was to minimize any distortion that would arise in the images from the (round)

246 tube test section. A similar refractive-index matching approach was used by
 247 Zadrazil et al. (2012). The visualization section was enclosed in a Perspex
 248 box with square sides, and the volume between the main FEP tube and the
 249 internal walls of the Perspex box was filled with deionized water (see Fig-
 250 ure 1(c)) to match the elected liquid phase inside the pipe. The refractive
 251 indices of FEP and water are 1.34 and 1.33 at ambient conditions, respec-
 252 tively. The slight mismatch was corrected by image anti-distortion correction
 253 post-processing algorithms.

254 The flow was measured with a PIV system supplied by LaVision. A
 255 photograph of the experimental arrangement is shown in Figure 1(b). The
 256 system employs a double-pulsed frequency-doubled Nd:YAG laser, with an
 257 excitation wavelength at 532 nm. The flow was seeded with a fluorescent
 258 dye, namely Rhodamine-B, at a concentration of $0.05 \text{ mg}\cdot\text{L}^{-1}$, and was illu-
 259 minated by a laser sheet from dedicated optics with a divergence angle of
 260 20° . The sheet optics arrangement was capable of producing a sheet of light
 261 with a thickness of $\tau = 0.1 \text{ mm}$ at the working distance of approximately
 262 0.5 m. The scattered light was recorded by a camera positioned at 90° to
 263 the laser light sheet, as illustrated in Figure 1(c). A mid-speed monochro-
 264 matic CMOS camera (VC-Imager Pro HS 500, LaVision) with a 1280×1024
 265 pixel resolution and fitted with high-pass optical filters was used for this pur-
 266 pose. The area visualized for the PLIF imaging using the CMOS cameras
 267 was $29 \times 23 \text{ mm}$, corresponding to a spatial pixel resolution of $22 \mu\text{m}$. During
 268 each measurement a set of 500 images was taken at a frequency of 100 Hz.

269 2.3. Data analysis

270 The raw PLIF images were firstly converted into binarized black and white
271 equivalents by a custom MATLAB code developed in-house that used a
272 thresholding approach (see Figure 2(a) for a typical result). The algorithm
273 used for the raw image binarization consists of the following steps:

274 (i) An estimation of the light intensity threshold value for the gas-liquid
275 interface identification; this estimation is based on an *a priori* manual cal-
276 ibration procedure during which the interface is manually located at each
277 axial pixel position and correlated to the radial light intensity profile at that
278 axial position on the image, over a sufficiently large number of images and
279 light intensity conditions. The threshold values were chosen as a compromise
280 between small values that were more sensitive in identifying the interface and
281 larger ones that were less sensitive to the noise. Note that a different cali-
282 bration was undertaken separately for each flow condition.

283 (ii) Binarization of the raw images using the gas-liquid interface light in-
284 tensity threshold calibrations generated in (i), such that 1 280 specific thresh-
285 old values were used per image (one for each axial pixel position).

286 (iii) Removal of the entrained liquid and out-of-plane features from the
287 binarized images: the liquid is considered entrained when it is entirely sep-
288 arated from the liquid film. The result of the binarization is an image with
289 black and white regions representing the gas and liquid phases, respectively.

290 Based on the binarized PLIF images, the mean film thickness $\langle \delta \rangle =$
291 $\frac{1}{n} \sum_{i=1}^n \delta_i$, film roughness (or, standard deviation) $\sigma_\delta = \sqrt{\frac{1}{n-1} \sum_{i=1}^n (\delta_i - \langle \delta \rangle)^2}$,
292 probability density function (PDF) and higher statistical moments (i.e., skew-
293 ness $S_\delta = \frac{1}{n} \sum_{i=1}^n [(\delta_i - \langle \delta \rangle)/\sigma_\delta]^3$ and kurtosis $K_\delta = \frac{1}{n} \sum_{i=1}^n [(\delta_i - \langle \delta \rangle)/\sigma_\delta]^4$)

of the film thickness δ were evaluated. These quantities were calculated from 500 instantaneous images and 20 local film thickness measurements δ_i per image, such that the statistical analysis of the film thickness was performed on $n = 10\,000$ film thickness data points δ_i . The number of samples taken per image was based on the evaluated characteristic length scales for the flow conditions investigated (see Section 3.4) which are in the range of $1.3 \times 10^{-3} - 9.2 \times 10^{-3}$ m, corresponding to 3 – 22 samples per image. A further analysis of the influence of the number of samples per image on the calculated values of the mean and standard deviation of the film thickness showed independent results from 1 sample/image up to 40 samples/image, thus it was decided to pick a value of 20 samples/image for all investigated flows, as a good compromise between statistical convergence (owing to the increased number of samples used in the calculation of the moments) and statistical independence between the samples. The total uncertainty in the measurement of the instantaneous and local film thickness δ_i was ± 0.04 mm, which corresponds to 3 pixels. This uncertainty was estimated from the manual inspection of a large number of film thickness levels from the available sets of image sequences generated (comparing both raw and corresponding processed, i.e., binarized, images).

The sequences of binarized PLIF images (i.e., 500 instantaneous images per measurement flow condition) were used for the construction of long temporal film thickness time-traces. These were assembled by applying an adaptive two-pass cross-correlation algorithm to the consecutive binarized instantaneous images. A reference displacement was obtained during the first pass, in which only film displacements that had a normalized cross-correlation in-

319 intensity peak greater than 0.5 were considered. Displacement information in
 320 those image pairs that did not fulfil this requirement was set to the mean
 321 film displacement of adjacent valid image pairs. The reference displacement
 322 information from the first pass was then used in a second cross-correlation
 323 pass (i.e., the binarized images were shifted accordingly to the reference dis-
 324 placement), with the cross-correlation intensity peak threshold set to 0.7.
 325 The application of the adaptive two-pass cross-correlation method to the
 326 construction of the temporal film thickness time-traces greatly increases the
 327 precision of the image stitching processing, in a similar way to that achieved
 328 by multi-pass PIV algorithms when these are employed for the accurate cal-
 329 culation of velocity vector fields. Typical examples of the cross-correlation
 330 intensity as a function of the displacement and of a resulting temporal film
 331 thickness time-trace can be seen in Figure 2(c) and (d), respectively. The
 332 film thickness time-traces were used for the calculation of the interfacial wave
 333 frequency f_W , wave peak thickness δ_W , and characteristic time and length
 334 scales of the liquid film interface.

335 Two measures of the interfacial wave frequency f_W were obtained. The
 336 first is based on a direct counting of waves with a thickness $2 - 5 \times$ the mean
 337 film thickness $\langle \delta \rangle$, as they appear at the temporal film thickness time-traces.
 338 A wave was defined as a disturbance of the liquid film interface that appeared
 339 above the mentioned threshold, thus crossing the threshold level twice. In
 340 addition, a second wave frequency measure was obtained by carrying out a
 341 power spectral density (PSD) analysis on the temporal film thickness time-
 342 traces. The PSD was evaluated by using standard MATLAB functions.

343 A bubble size analysis was performed by a direct measurement of the

344 longitudinal (axial) $d_{a,i}$ and transverse (radial) $d_{r,i}$ cross-sections on a number
 345 of bubbles as they appeared on the PLIF images. For each set of conditions
 346 either all 500 consecutive images or 300 consecutive bubbles were evaluated.
 347 The average of the two lengths was then used as a mean bubble chord length
 348 $\langle d \rangle = \frac{1}{n} \sum_{i=1}^n (d_{a,i} + d_{r,i})/2$. The error in the measurement, which is based
 349 on the precision of the manual location of the bubble gas-liquid interface, of
 350 the raw axial $d_{a,i}$ and radial $d_{r,i}$ chord lengths is ± 0.04 mm. The ratio of the
 351 measured radial to axial chord lengths $\frac{1}{n} \sum_{i=1}^n d_{r,i}/d_{a,i}$ is on average 1.08.

352 Finally, the bubble frequency f_B was also calculated, as the number of
 353 bubbles identified per unit time. It should be noted that the reported bubble
 354 frequency herein is restricted to the 2-D observations of the liquid film at one
 355 side of the DAF in the pipe and therefore f_B does not reveal the information
 356 of the overall bubble frequency in the entire annular liquid film.

3. Results and Discussion

The PLIF results will be used in this section to provide insight into the flow structure of downwards co-current gas-liquid annular flows, or simply ‘downwards annular flows’ (DAFs). Flow regimes along with phenomenological observations based on a qualitative analysis of the instantaneous PLIF images are presented first, in Section 3.1. Following this, Section 3.2 presents results on the film thickness statistics (e.g., mean film thickness, film roughness and higher statistical moments such as the skewness and kurtosis). Wave statistics (e.g., wave amplitude and wave frequency) are presented in Section 3.3. The characteristic length and time scales of the interfacial structures in the investigated DAFs are reported in Section 3.4. Finally, Section 3.5 presents results for bubble size and frequency.

3.1. Flow regime map and phenomenological observations

A flow map featuring the various DAF regimes and drawn in terms of the gas and liquid phase Reynolds numbers (Re_G and Re_L) is shown in Figure 3, together with corresponding representative instantaneous images of each flow regime. The colour (or, greyscale level) of the instantaneous images corresponds to the local intensity of the emitted fluorescent light. The flow regimes were identified based on a synthesis of qualitative information from the instantaneous PLIF images, and specifically by taking into account the following: (i) substrate thickness δ_S , (ii) maximum wave thickness δ_W , (iii) substrate length λ_S , (iv) wave length λ_W , (v) gas entrainment events into the liquid film, and (vi) liquid entrainment events into the gas core. The definitions of these thickness and length scale variables are illustrated

in Figure 3(b). One should recall that the annular flow is usually described in terms of interfacial waves that cover the substrate, a thinner liquid region that is present between the waves. Therefore, the wave length corresponds to the length of a specific wave, and the substrate length to the length of the relatively quiescent region that is present between individual waves. Liquid entrainment was estimated from the occurrence of bag and ligament break-up events of liquid entrainment into the gas core (Azzopardi, 1997), whereas gas entrainment events were determined from direct observations of air bubbles penetrating into the liquid films (e.g., see Figure 3(c)).

Four distinct flow regimes have been observed in the current study. The ‘ripple’ regime was not observed here, due to flow-rate control limitations, and so the flow regime boundary for the ripple regime drawn in Figure 3(a) is taken from Webb and Hewitt (1975). The ‘dual-wave’ regime, in which negligible mixing between the two phases was observed, was characterized by large-amplitude waves (with $\delta_S < \delta_W$) separated by periods of relative inactivity in which the substrate is longer than the waves ($\lambda_S > \lambda_W$), and was identified over the range of conditions with $Re_G = 0 - 21\,100$ and $Re_L = 306 - 613$. No events that would indicate gas entrainment into the liquid film were observed in the dual-wave regime, however, a few liquid phase entrainment events into the gas core were identified.

The ‘thick ripple’ regime was observed for falling films ($Re_G = 0$) over the range $Re_L = 919 - 1\,226$, and for DAFs with $Re_G = 21\,100$ and $Re_L = 919$. In this regime large waves occurred sporadically, with the difference between the thickness of the substrate δ_S and the wave front thickness δ_W , and also between the substrate length λ_S and the wave length λ_W being minimal, or

406 $\delta_S \sim \delta_W$ and $\lambda_S \sim \lambda_W$. The characteristic alternation of waves and substrate
407 had a sinusoidal character (i.e., it was difficult to locate the beginning and
408 an end of a wave). Additionally, the gas phase was sporadically entrained
409 into the liquid film as bubbles, while rare instances of liquid entrainment into
410 the gas core via ligament break-up were observed.

411 A new DAF regime has been observed here for the falling films at the high-
412 est investigated liquid Reynolds numbers Re_L , and also at low gas Reynolds
413 numbers $Re_G = 21\,100$. The new regime, termed ‘disturbance wave’ regime,
414 is associated with $\delta_S \ll \delta_W$ and $\lambda_S > \lambda_W$, that is, very large-amplitude (i.e.,
415 disturbance) waves rising clearly above the substrate, with the waves oc-
416 ccurring more frequently than in the dual-wave or thick ripple regimes. In
417 addition, this regime displays high rates of entrainment of both the gas into
418 the liquid and the liquid into the gas.

419 Finally, the ‘regular’ wave regime was observed for all investigated Re_L
420 at the higher gas flow-rates and Reynolds numbers, $Re_G \geq 42\,300$. In this
421 flow regime, contrary to the previously mentioned regimes, the gas-liquid
422 interface is more strongly affected by the high levels of shear exerted by the
423 gas flow. The flow in the ‘regular’ wave regime is observed to be associated
424 with a more agitated interface, a considerably higher fraction of liquid being
425 entrained into the gas, as well as gas into the liquid film in the form of bubbles.
426 In addition, dry-out areas (i.e., regions of the pipe wall that become directly
427 exposed to the gas) can be sporadically observed. The overall flow regime
428 map in Figure 3 is in good general agreement with the one reported in the
429 work of Webb and Hewitt (1975).

430 Ishii and Grolmes (1975) and Azzopardi (1983) described five differ-

ent mechanisms of liquid entrainment into the gas phase for annular flows, namely ‘disturbance wave’, ‘wave undercut’, ‘ligament break-up’, ‘bubble burst’ and ‘liquid impingement’. The instantaneous images depicting these entrainment events, obtained by the direct observation from our investigated annular flows, are shown in Figure 4. It was observed that the drops often originate from large waves (e.g., disturbance waves), see Figure 4(a), which is in agreement with the findings from the experiments of Azzopardi and Whalley (1980). During the disturbance wave entrainment event, the crest of the large waves is either sheared off by the turbulent gas flow which results in a formation of small drops or is disrupted and eroded by ripples having high velocity. Figure 4(b) shows an liquid entrainment event when the liquid film is being undercut by the gas flow. This mechanism is sometimes referred as to ‘bag break-up’ in *upwards* annular flows (Azzopardi, 1983). A thin ligament is sometimes formed on a crest of both disturbance waves and ripples, see Figure 4(c). During the ligament break-up, a number of small liquid drops is formed. Figure 4(d)–(e) shows a bubble burst event taking place at different stages. A thin liquid filament is formed when a bubble reaches the gas-liquid interface. The droplets are formed due to the rupture of the filament and consequent dynamics of the surrounding liquid (i.e. filling the crater left by the bubble). The bubble burst mechanism gives rise to considerable amount of liquid being ejected into the gas phase.

According to James et al. (1980), the re-deposition of drops can be divided into two classes: (i) those that land by diffusion-like processes within the gas core, and (ii) those that arrive by direct impact (i.e., their motion is unaffected by the gas flow). Our qualitative observations show, see Figure 4(f)

that the advancing disturbance waves produce drops that are immediately deposited onto the substrate at the close proximity of the wave itself. The qualitative observations of the re-deposition processes indicate that the drops created by the ligament break-up and disturbance wave mechanisms are responsible for the diffusion-like re-deposition. Contrary, the direct impact re-deposition of the drops can be associated with the wave undercut, liquid impingement and bubble burst entrainment mechanisms.

3.2. Film thickness statistics

The temporal film thickness time-traces of falling films (i.e., $Re_G = 0$) that were constructed by using a two-pass cross-correlation between successive image pairs (see Section 2.3) are shown in Figure 5. These traces reveal that the falling film characteristics (i.e., film thickness, or interface morphology) are strongly dependent on Re_L . For relatively small liquid Reynolds numbers Re_L the large waves are well separated by a long film substrate. With increasing Re_L the distinction between the waves and the film substrate diminishes. At relatively large Re_L the amplitude and frequency of large waves increases significantly. Similar observations were made during the qualitative analysis of the instantaneous raw PLIF images (see Section 3.1). Furthermore, the following observations can be made: (i) the amplitude of large waves as well as the small waves is highly variable and intermittent, (ii) the large waves are often traversed by (and thus appear covered by) one or more smaller waves (i.e., ripples), (iii) at low Re_L and zero Re_G the substrate can be described as flat, whereas at high Re_L and non-zero Re_G the substrate is fully covered by small-amplitude waves (ripples), and (iv) the complexity of the waves increases with increasing Re_L and Re_G . Figure 6 shows equivalent results to

those contained in Figure 5, but over a range of Re_G from 21 100 to 84 600, and with Re_L fixed to a value of 306. From Figure 6 similar conclusions can be drawn to those above concerning the falling films.

Figure 7 shows the mean film thickness $\langle \delta \rangle$ for the falling films (i.e., with $Re_G = 0$) investigated in this work, along with: (i) a comparison with results from several previous studies (Webb and Hewitt, 1975; Takahama and Kato, 1980; Karapantsios et al., 1989; Karapantsios and Karabelas, 1995), and (ii) theoretical predictions of the mean film thickness from laminar Nusselt flow and turbulent (Mudawwar and El-Masri, 1986) falling films. It should be noted that data from several techniques for the measurement of film thickness are compared in Figure 7. We used directly PLIF images for measuring the film thickness contrary to parallel-wire conductance probes (Karapantsios et al., 1989; Karapantsios and Karabelas, 1995), platinum foil conductance probes (Webb and Hewitt, 1975) and needle contact and electric capacity probes (Takahama and Kato, 1980).

Our data are in good agreement with the published results. In addition, at low Re_L the data are well-described by the Nusselt flow predictions, though with increasing Re_L the theory tends to increasingly underpredict both our and the other previously published experimental data on $\langle \delta \rangle$. This arises due to the fact that Nusselt flow assumes a flat gas-liquid interface (i.e., without considering any interfacial instabilities) and a parabolic velocity profile, whereas an increasing number of wave disturbances (ripples) appear at the interface and the velocity profile also becomes modified with increasing Re_L . Our results at larger liquid Reynolds numbers, $Re_L \geq 800$, follow closely the experimental data from previous work at similar conditions (i.e., Re_L), as

well as the theoretical prediction for $\langle\delta\rangle$ in turbulent falling films. A wide range of Reynolds numbers for transition from laminar to turbulent flow can be found in the literature, from a low values of $Re_L = 250$ to higher values of $Re_L = 800$ (Fulford, 1964; Mudawwar and El-Masri, 1986). The transition from laminar to turbulent flow is indicated in Figure 7 by a dotted line and is situated to the intersect between Nusselt and turbulent predictions of the mean film thickness, $Re_L = 540$.

The mean film thicknesses $\langle\delta\rangle$ of the DAFs with non-zero Re_G are shown in Figure 8(a) and (b), as a function of the liquid and gas Reynolds numbers, respectively. The colours of the data points on these plots indicate different flow regimes, as identified in Section 3.1 and Figure 3. Generally, the thickest waves, in-the-mean, occur in the disturbance wave regime, and the thinnest in the regular wave regime. In general, the mean film thickness $\langle\delta\rangle$ increases with increasing Re_L , and decreases with increasing Re_G . A deviation from this trend can be seen for the thick ripple regime at $Re_G = 21\,100$ and $Re_L = 613$ (refer also to Figure 3). This can be due to either an acceleration of the flow within the liquid film due to the shearing forces from the gas flow, or by the onset of liquid entrainment into the gas core. It is interesting to point out that the transition from laminar to turbulent flow also occurs at these Re . We also note that for the highest investigated gas Reynolds number of $Re_G = 84\,600$ the mean film thickness $\langle\delta\rangle$ decreases slightly with increasing liquid Reynolds number Re_L . The slope of the trendlines $\langle\delta\rangle = f(Re_L)$ decreases gradually with increasing Re_G , which can be attributed to increased liquid entrainment into the gas core as can be seen from the qualitative observations, see Section 3.1. The effect of the gas Re_G on $\langle\delta\rangle$

531 can be seen in Figure 8(b). The liquid films appear progressively thinner
532 with increasing Re_G in a fashion that is approximately linear.

533 The absolute (σ_δ) and relative ($\sigma_\delta/\langle\delta\rangle$) liquid film fluctuation (or, ab-
534 solute and relative film roughnesses) as a function of liquid Reynolds number
535 Re_L are shown in Figure 9(a) and (b), respectively. The greatest interfacial
536 fluctuations (i.e., σ_δ) occur in the disturbance wave regime, and the highest
537 wave fluctuation intensities (relative to the mean, i.e., $\sigma_\delta/\langle\delta\rangle$) is observed
538 in the disturbance wave and regular wave regimes (recall, however, that the
539 mean film thickness $\langle\delta\rangle$ is minimal in this case; see Figure 8). The absolute
540 film roughness σ_δ , which is the standard deviation of the liquid film thick-
541 ness variation, shows two distinctive trends. (i) In the case of falling films
542 σ_δ decreases initially with increasing Re_L (at low Re_L), reaches a minimum
543 around $Re_L \approx 613 - 919$, and then increases again. A similar trend can
544 be seen for the lowest investigated gas Reynolds number of $Re_G = 21\,100$,
545 where a global minimum is observed around $Re_L \approx 919$. (ii) At the higher
546 gas Reynolds numbers, above $Re_G \geq 42\,300$, the absolute film roughness σ_δ
547 increases slightly but gradually with increasing liquid Reynolds number over
548 the entire investigated range from $Re_L = 306$ up to $1\,532$.

549 The relative film roughness, defined as $\sigma_\delta/\langle\delta\rangle$, is shown in Figure 9(b).
550 The different flow regimes exhibit their own distinctive behaviour with re-
551 spect to this variable. The relative film roughness attains values of $\sigma_\delta/\langle\delta\rangle =$
552 $0.3 - 0.4$ for the dual-wave regime. The smallest relative film roughnesses are
553 observed for the thick ripple regime, with $\sigma_\delta/\langle\delta\rangle = 0.2 - 0.3$. The transition
554 from the thick ripple to the disturbance wave regime is accompanied by a
555 sudden increase of the relative film roughness to $\sigma_\delta/\langle\delta\rangle = 0.6 - 0.9$. In fact,

556 this sudden transition in $\sigma_\delta/\langle\delta\rangle$ can be interpreted and used as a strong indi-
 557 cator of this flow regime transition. Finally, $\sigma_\delta/\langle\delta\rangle$ increases with increasing
 558 Re_L for the thick ripple regime within the range $\sigma_\delta/\langle\delta\rangle = 0.5 - 0.9$.

559 Figure 10 shows the measured probability distribution functions (PDFs)
 560 of the liquid film thickness $P(\delta)$ for: (a) $Re_L = 306 - 1\,532$ at a constant
 561 gas Reynolds number of $Re_G = 0$ (falling films), and (b) $Re_G = 0 - 84\,600$
 562 for a constant liquid Reynolds number of $Re_L = 306$. All the film thick-
 563 ness PDFs are asymmetric (+vely skewed) with a tail towards higher film
 564 thickness values. Increasing the liquid Reynolds number Re_L results in a
 565 broadening of the distributions towards thicker liquid film values. On the
 566 contrary, an increase in the gas Reynolds number Re_G leads to a narrowing
 567 of the distribution as the liquid films become progressively thinner.

568 Higher order statistical moments of the film thickness, specifically the
 569 third moment (skewness, S_δ) and fourth moment (kurtosis, K_δ), are a mea-
 570 sure of the asymmetry and flatness of the distributions shown above in Fig-
 571 ure 10 (as a reference point, $S_\delta = 0$ and $K_\delta = 3$ for a Gaussian distribution).
 572 Figure 11(a) and (b) shows the dependence of the skewness and kurtosis of
 573 $P(\delta)$ on the liquid Reynolds number Re_L . The skewness values occupy the
 574 range $S_\delta \approx 0.7 - 3$, and the kurtosis values the range $K_\delta \approx 4 - 24$.

575 A closer examination of Figure 10 reveals that falling films exhibit a non-
 576 monotonic behaviour, in that (starting from low Re_L) a gradual increase in
 577 Re_L leads first to more symmetric PDFs with reduced excursions away from
 578 the mean thickness $\langle\delta\rangle$, reaching a minimum around $Re_L \approx 613 - 919$, and
 579 then increasing again at the higher Re_L . This is in alignment with the ob-
 580 servations concerning the film roughness σ_δ in Figure 9. DAFs in general

show a very complex behaviour with respect to these two variables, S_δ and K_δ , reflecting the complex nature of these flows and the coupled, nonlinear processes between the liquid and gas phases that take place at the interface. In particular, the disturbance wave regime is associated with relatively high skewness S_δ and kurtosis K_δ , though not necessarily the highest values obtained in this work. Note, however, that in these two variables the central moments are normalized; in the case of S_δ the third moment is normalized by σ_δ^3 , and in the case of K_δ the fourth moment is normalized by σ_δ^4 .

3.3. Wave occurrence statistics

The interfacial wave frequency f_W was obtained from the temporal film thickness time-traces, such as those shown in Figures 5 and 6, by using two different methods: (i) by the direct counting of waves that had a thickness greater than $n = 2 - 4 \times$ the mean film thickness $\langle \delta \rangle$ ($f_{n\langle \delta \rangle}$), and (ii) by using a power spectral density (PSD) analysis (f_{PSD}). The wave frequencies $f_{n\langle \delta \rangle}$ obtained by the ‘wave counting technique’ can be seen in Figure 12.

As expected, the frequency values $f_{n\langle \delta \rangle}$ are dependent on the wave amplitude threshold factor n that is chosen to identify the large waves, with the frequency decreasing in general as factor n is increased. When including relatively smaller amplitude waves in the counting procedure, i.e., with amplitudes $> 2\langle \delta \rangle$ (Figure 12(a)), the wave frequency increases with increasing gas Reynolds number Re_G . For the falling films and the lowest gas Reynolds number flows with $Re_G = 21\,100$, the wave frequency $f_{2\langle \delta \rangle}$ decreases slightly with increasing Re_L up to $Re_L = 919$, reaches a minimum and then increases up to a maximum of ~ 8 Hz that is attained in the disturbance wave flows. The wave frequency $f_{2\langle \delta \rangle}$ for the higher gas Reynolds numbers $Re_G \geq 42\,300$,

when the flow is in the regular flow regime, is in the range 10 – 16 Hz. Clearly, this wave regime is associated with the greatest wave activity for relatively small-amplitude waves, with amplitudes $> 2\langle\delta\rangle$ (and also, $> 3\langle\delta\rangle$).

Interestingly, waves with amplitudes larger than $3\langle\delta\rangle$ (Figure 12(b)) were not observed in the thick ripple regime, which is indicated everywhere in Figure 12 by the green colour. Additionally, the frequency of the $3\langle\delta\rangle$ waves is smaller when compared to $2\langle\delta\rangle$ waves with a maximum of $f_{3\langle\delta\rangle} = 9$ Hz, while waves with very high amplitudes, i.e., $> 4\langle\delta\rangle$ (Figure 12(c)), appear only in the regular wave and the disturbance wave regimes; being the dominant wave type in the latter. These waves appear rarely, with a maximum frequency of 4 Hz for the $4\langle\delta\rangle$ case. The results in Figure 12 indicate that the disturbance waves generally (with some notable exceptions) become increasingly frequent when the gas Reynolds number Re_G is increased, which can be explained by the corresponding enhancement in the shearing force of the gas that sweeps the liquid off the pipe surface. This results in a thin substrate and relatively thick waves, but also with high gas entrainment, which was indeed confirmed by inspection of the sequences of instantaneous PLIF images.

We proceed now to the interfacial wave frequency measures that were obtained by the PSD analysis, f_{PSD} . The PSDs were constructed from temporal film thickness time-traces such as those in Figures 5 and 6. Results are shown in Figure 13 for: (a) falling films with a zero gas Reynolds number, $Re_G = 0$, and (b) DAFs with a constant liquid Reynolds number of $Re_L = 306$. The falling films in Figure 13(a) have peak signal powers at frequencies in the range 4 – 9 Hz, with the value of this peak frequency being mostly independent of Re_L . In addition, Figure 13(b) shows PSDs for DAFs with

different Re_G at constant $Re_L = 306$. The peak power frequency here is higher, and increases from 8 to 25 Hz with increasing Re_G from 0 to 84 600. The peak value of the power spectrum decreases with increasing Re_G .

The maximum power frequency f_{PSD} extracted from each PSD in Figure 13 is shown in Figure 14 as a function of: (a) liquid Reynolds number Re_L , and (b) gas Reynolds number Re_G . It is observed that the functional relationship of the peak power frequency with Re_G is stronger than that with Re_L , suggesting the the gas phase has a strong role to play in influencing the interfacial structures and dynamics. Specifically, the peak frequency f_{PSD} increases strongly with Re_G . Further, we may expect that the peak frequency values and trends can be attributed to the change of flow regimes. The highest frequencies are exhibited in the regular flow regime, and the lowest in the thick ripple and disturbance wave regimes.

Azzopardi (1997) attributed the PSD peak frequency to the frequency of disturbance waves. When Figures 12 and 14 are compared, it is concluded that there are significant differences between the two wave frequency measures. In fact, the PSD peak frequency corresponds more closely to the more frequent, but smaller amplitude waves (i.e., ripples) with an amplitude smaller than $2\langle\delta\rangle$, rather than the larger amplitude (e.g., disturbance) waves. It is interesting to observe that these small-amplitude waves carry the highest amount of energy in the shape of the liquid film interface, contrary to the large-amplitude disturbance waves as was previously assumed. Yet, this can be readily understood in terms of the regularity with which the two different types of waves occur, with the latter being relatively rare events, less able to affect the overall spectral energy contents of the interfacial signature signals.

656 This finding has significant implications in guiding the efforts to characterize
657 and to understand such flows, and is entirely consistent with a similar recent
658 finding in *upwards* annular flows (Zhao et al., 2013).

659 Figure 15 shows the power at the PSD peak frequency (i.e., the maximum
660 value of the PSD) as a function of: (a) liquid Reynolds number Re_L , and
661 (b) gas Reynolds number Re_G . Also, shown in Figure 15(c) is the high-
662 frequency (short wave length) decay power of the PSDs in Figure 13. The
663 PSD power values increase strongly with Re_L , reaching very high values for
664 the disturbance wave flows. In addition, the peak values are generally low for
665 regular wave flows, which are observed at the higher Re_G . It is interesting
666 to note that the identification of the disturbance wave regime can be more
667 readily done from the PSD peak value at f_{PSD} , rather than f_{PSD} itself, due to
668 the contamination of the latter variable from the smaller waves, as discussed
669 previously. The smaller waves are expected to be disproportionally associated
670 with higher frequencies, and as such it is expected that they will also affect
671 the energy decay at higher frequencies, which is considered in Figure 15(c).
672 Also shown here is the $-5/3$ power law decay of the Kolmogorov spectrum
673 for homogeneous and isotropic turbulence.

674 The PSD slopes at high frequencies shows a decay power exponent that is
675 clearly less steep in the regular wave flows. This observation ties in well with
676 the expectation that the regular wave regime, which is dominated by smaller
677 waves, will have a higher energy content at higher frequencies (i.e., shorter
678 wave lengths). The power exponent displays an almost linear link to the
679 gas Reynolds number Re_G , which suggests again the important role of the
680 turbulent gas phase in introducing energy at progressively higher frequencies

681 onto the interfacial structure as Re_G is increased.

682 Finally, we consider results for the mean and probability functions (PF)
683 of the wave amplitudes δ_W , defined as the height of the peaks of the waves
684 that are larger than the mean film thickness $\langle\delta\rangle$. The mean wave ampli-
685 tudes $\langle\delta_W\rangle$ are shown in Figure 16(a) as a function of the liquid Reynolds
686 number Re_G for various constant gas Reynolds numbers Re_G . The mean
687 wave amplitude $\langle\delta_W\rangle$ increases with increasing Re_L for a constant Re_G and
688 decreases with increasing Re_G for a constant Re_L . In the case of the falling
689 films and for the lowest investigated gas Reynolds number $Re_G = 21\,100$
690 a sharp increase of mean wave amplitude can be observed for the highest
691 liquid Reynolds numbers, $Re_L \geq 1\,226$. This increase corresponds to the
692 experimental conditions where the disturbance wave regime was observed.
693 Therefore, it can be concluded that the disturbance wave flows exhibit the
694 greatest wave amplitudes, and also that the regular flow regime is associated
695 with the smallest amplitudes. Further, Figure 16(b) shows the mean wave
696 amplitude normalized by the absolute roughness of the liquid film from Fig-
697 ure 9(a). Two bands of data can be discerned in this plot. The regular wave
698 and disturbance wave data collapse to a value of $\langle\delta_W\rangle/\sigma_\delta \sim 3$, while the thick
699 ripple and dual-wave regime flow data attain higher values, within the range
700 4 – 7, with a maximum appearing at intermediate Re_L .

701 More detailed information on the wave amplitudes is contained in the
702 probability functions of the wave amplitudes $P(\delta_W)$, which can be found in
703 Figure 17(a) and (b) for constant gas and liquid Reynolds numbers (i.e.,
704 $Re_G = 0$ and $Re_L = 306$) respectively. With increasing Re_L the probability
705 of the appearance of large waves increases and that of the appearance of

small waves decrease. In particular, the largest investigated liquid Reynolds number, $Re_L = 1\,532$, is characterized by a high probability of (disturbance) waves that are larger than 2 mm. Increasing the gas Reynolds number has an opposite effect on the wave occurrence probability, as shown in Figure 17(b), leading to a shift of the probability towards increasingly thinner waves. This finding is consistent with the expectation that higher interfacial shear stresses and increased liquid entrainment will both lead to lower amplitude liquid waves. Additionally, the probability functions $P(\delta_W)$ have shorter high wave amplitude tails and are narrower, i.e., a lower skewness and kurtosis.

3.4. Interface length and time scales

The autocorrelation functions (ACFs) evaluated from the interfacial time-traces for a constant gas Reynolds number $Re_G = 0$ (i.e., falling films) and a constant liquid Reynolds number $Re_L = 306$ are shown in Figure 18(a) and (b), respectively. In all cases the ACFs drop to near-zero values after a spatial distance Δl of a few cm. This is an indicator of the longitudinal (axial) length scale of the interfacial waves. The ACFs of falling films (Figure 18(a)) for the lowest and the two highest investigated liquid Reynolds numbers Re_L appear to have a similar shape, dropping to negative values. This is different from the shape of the ACFs at intermediate liquid $Re_L = 613 - 919$ (thick ripple regime), where no global minimum can be identified. Similar observations were made by Karapantsios et al. (1989). On the other hand, a gradual change in the shape of the ACFs of the DAFs with finite gas flow as a function of gas Reynolds number Re_G as can be seen in Figure 18(b)), where the position and the magnitude of the global minimum shifts towards shorter lengths Δl and smaller values with increasing Re_G .

731 An integration of the ACFs in Figure 18 allows the evaluation of length
732 and time scales associated with gas-liquid interface, a procedure similar to
733 that used for the evaluation of Taylor scales in turbulent (velocity) signals.
734 These are found to cover the ranges $0.5 \times 10^{-2} - 25 \times 10^{-2}$ s and $1.3 \times 10^{-3} - 9.2 \times 10^{-3}$ m
735 for the time and length scales, respectively. Both time and length scales were
736 found to decrease with increasing Re_G .

737 3.5. Bubble statistics

738 The mean bubble chord length $\langle d \rangle$ as a function of Re_L is shown in Fig-
739 ure 19(a). No gas entrainment into the liquid film was observed for falling
740 film at $Re_L = 306$. For falling films at $613 \leq Re_L \leq 919$ the level of gas
741 entrainment into the liquid film is negligible (see also Figure 20), however,
742 the mean size of the bubble chord lengths $\langle d \rangle$ is relatively large, being of
743 the order of ~ 0.8 mm. The large bubble chord lengths might be explained
744 by the lower shear stresses within the liquid films at low Re_L that may be
745 insufficient to affect a bubble after it has been entrained. Note that the liq-
746 uid film is most likely laminar where the large bubbles were observed (see
747 Fig. 7). With increasing Re_L the shearing forces within the liquid become
748 sufficiently high, enough to break the large bubbles into smaller ones. This
749 would be consistent with the sudden decrease in $\langle d \rangle$ demonstrated in the
750 falling films at $Re_L \geq 1\,226$, with $\langle d \rangle \sim 0.4 - 0.5$ mm, as well as the sudden
751 corresponding increase in the bubble frequency f_B in Figure 20. With the
752 exception of the falling films at the two lowest Re_L (≤ 919), all other flow
753 conditions including all DAFs at finite Re_G show an increasing mean bubble
754 chord length $\langle d \rangle$ with increasing Re_L .

755 The difference between the definition of a bubble chord length and a bub-

ble diameter is that the former length does not have to be at the centreline of the bubble. The size of the bubble (diameter μ_B) relative to the laser sheet thickness τ , together with the relative position of an individual bubble, define whether a chord length or a bubble diameter is being measured. When the bubble is much larger than the laser sheet thickness ($\mu_B/\tau \gg 1$), the outcome of the measurement is one of chord length (at some position where the laser sheet crosses the bubble). Contrary, when the bubble is much smaller than the laser sheet thickness ($\mu_B/\tau \ll 1$), the outcome of the measurement is the entire bubble diameter. In the former case (i.e., large bubbles) the mean chord length $\langle d \rangle$ can be used to estimate the mean bubble diameter μ_B by multiplying by a correction factor of $4/\pi$, whereas in the latter case the correction factor is equal to 1 (Hu et al., 2006, 2007; Morgan et al., 2013). The correction factor of $4/\pi$ arises from simple geometrical arguments that assume that over long times individual spherical bubbles appear uniformly spatially distributed around the laser sheet and an infinitely thin sheet. In any intermediate bubble-size case, the mean bubble diameter can be determined from the weighted ratio,

$$\frac{\langle d \rangle}{\mu_B} = \frac{\frac{\pi}{4} \left(\frac{\mu_B}{\tau} \right) + 1}{\left(\frac{\mu_B}{\tau} \right) + 1},$$

the inverse of which can be used as a correction factor to estimate μ_B from the direct measurement of $\langle d \rangle$, e.g., reported in Figure 19(a).

In our experiments the ratio μ_B/τ is found to be in the range of (approximately) 1 – 9 (from Figure 19(a)), such that the ratio above amounts to 0.8 – 0.9. Therefore, we can say that our mean chord lengths, as reported

778 in Figure 19(a), underestimate the true mean bubble diameters by approxi-
779 mately 10% for the smaller diameter bubbles and 20% for the larger ones.

780 The mean bubble chord length $\langle d \rangle$ normalized by the absolute film rough-
781 ness σ_δ is shown in Figure 19(b). It is interesting to note that the normalized
782 bubble chord length $\langle d \rangle / \sigma_\delta$ as a function of Re_L collapses onto a relationship
783 which qualitatively is very similar to that exhibited by the mean wave am-
784 plitude, normalized by the same parameter in Figure 16, i.e., $\langle \delta_w \rangle / \sigma_\delta$. This
785 suggests stingily that the mechanism for gas entrainment into the liquid film
786 may be strongly reliant on the existence of large-amplitude waves.

787 The bubble frequency f_B was calculated as the number of bubbles per
788 unit time as they appear within the visualized area (i.e., 2-D plane of one
789 side of the pipe). the highest bubble frequency f_B is observed in the regular
790 wave regime, followed by the disturbance wave regime. Specifically, at the
791 high gas Reynolds numbers associated with the regular wave regime (i.e.,
792 $Re_G \geq 42\,300$) the bubble frequency f_B is mostly independent of the liquid
793 Reynolds number Re_L and is found to be in the range of 500 – 700 Hz. At
794 smaller $Re_G \leq 21\,100$ the bubble frequency f_B shows an increasing trend for
795 increasing Re_L , with near-zero values for $Re_L \leq 919$; an indication that gas
796 entrainment into the liquid film is negligible at these conditions.

797 4. Summary and Conclusions

798 A non-intrusive laser-based flow measurement technique, namely PLIF, was
799 used for the detailed characterization of downwards annular flows across a
800 range of liquid (water) and gas (air) flow-rates (Reynolds numbers). Based
801 on qualitative observation of a large number of image sequences and a de-
802 tailed and extensive quantitative analysis, four distinctive flow regimes were
803 identified. These were described in terms of different statistical moments
804 of the film thickness (i.e., mean, standard deviation, skewness and kurtosis),
805 wave frequency (obtained by wave counting and PSD), wave amplitude
806 and gas entrainment into the liquid phase (mean bubble chord length and
807 frequency).

808 The dual-wave regime was observed for $Re_L \leq 613$ and $Re_G \leq 21\,100$.
809 The mean film thickness in this regime was, generally, well described by
810 Nusselt flow, suggesting that the liquid film is laminar. The temporal film
811 thickness time-traces revealed that the interface comprised a long substrate
812 (which was found to be flat) separating individual waves. Both ripples and
813 disturbance waves were observed, however, the frequency of the disturbance
814 waves was found to be low, i.e., below 1 Hz.

815 The thick ripple regime was characterized by an absence of disturbance
816 waves and the sole presence of small-amplitude ripples. This regime can be
817 found at $Re_L = 919 - 1\,226$ and $Re_G \leq 21\,100$. The substrate is fully covered
818 by ripples, which have a near-sinusoidal shape. The mean film thickness
819 deviates from the Nusselt relation for laminar flow and follows the curve for
820 turbulent flow as described by Mudawwar and El-Masri (1986). Additionally,
821 large bubbles with a mean bubble chord length of $\langle d \rangle \sim 0.5 - 0.8$ mm can be

822 sporadically observed.

823 A disturbance wave regime was also uncovered at relatively high liquid
824 and low gas Reynolds numbers, $Re_L \geq 1\,532$ and $Re_G \leq 21\,100$. As the
825 name suggests, the gas-liquid interface is occupied by large-amplitude waves
826 $> 4 - 5 \times \langle \delta \rangle$, where the thickness of individual waves can be up to 5 mm. The
827 disturbance waves themselves were best identified by direct wave counting,
828 as opposed to considerations of the interface PSDs, since the latter contain
829 significant information from smaller amplitude waves.

830 Finally, a regular wave regime was observed at high gas Reynolds numbers
831 $Re_G \geq 42\,300$ and all investigated Re_L . This flow regime is characterized
832 by thin liquid films, high frequency small- and large-amplitude waves and
833 significant gas entrainment into the liquid film.

834 Our results indicate that the turbulent gas core plays a significant role
835 in introducing energy at progressively higher frequencies onto the interfacial
836 structure as Re_G is increased, and also that the mechanism for gas entrain-
837 ment into the liquid film is strongly reliant on the existence of large-amplitude
838 waves. This information is important in helping to understand and charac-
839 terize these complex flows. In addition, the detailed quantitative information
840 in this paper can assist in the development and validation of advanced two-
841 phase models for downwards annular flow.

842 **Acknowledgement**

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845 EP/K003976/1).

846 Figure captions

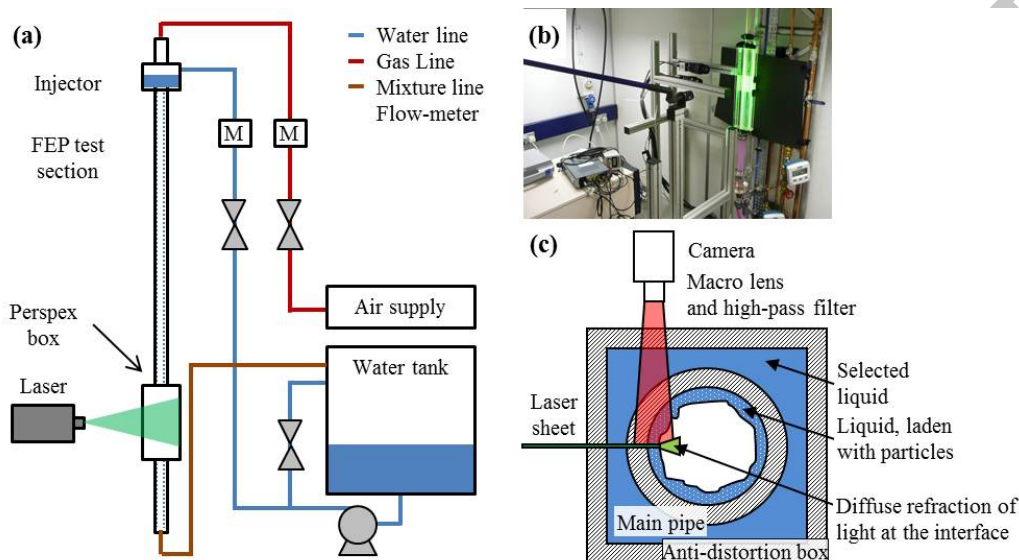


Figure 1: (a) Schematic layout of DAFLOF, (b) photograph of the visualization section with the laser sheet in position, and (c) simplified schematic illustration of the experimental visualization setup.

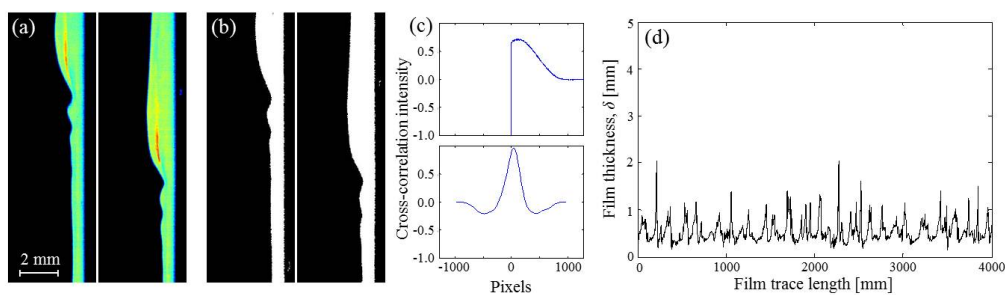


Figure 2: Examples of: (a) raw instantaneous PLIF images, (b) binarized equivalents of (a), (c) two-pass cross-correlation functions of (b), and (d) film thickness traces that were constructed from the stitching of 500 consecutive instantaneous images, after using the cross-correlation information in (c).

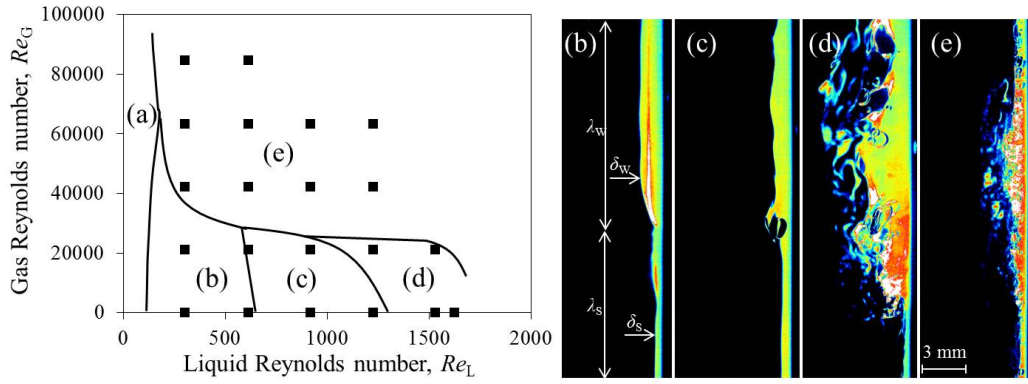


Figure 3: Flow regime map constructed from a series of PLIF images and instantaneous images of wave fronts in various conditions. (a) ‘Ripple’ regime, (b) ‘dual-wave’ regime (‘falling film’ with $Re_L = 306$), (c) ‘thick ripple’ regime (falling film with $Re_L = 919$), (d) ‘disturbance wave’ regime ($Re_G = 21\,100$ and $Re_L = 1\,226$), and (e) ‘regular wave’ regime ($Re_G = 42\,300$ and $Re_L = 306$). The names of the flow regimes (except the newly uncovered ‘disturbance wave’ regime) are used according to Webb and Hewitt (1975).

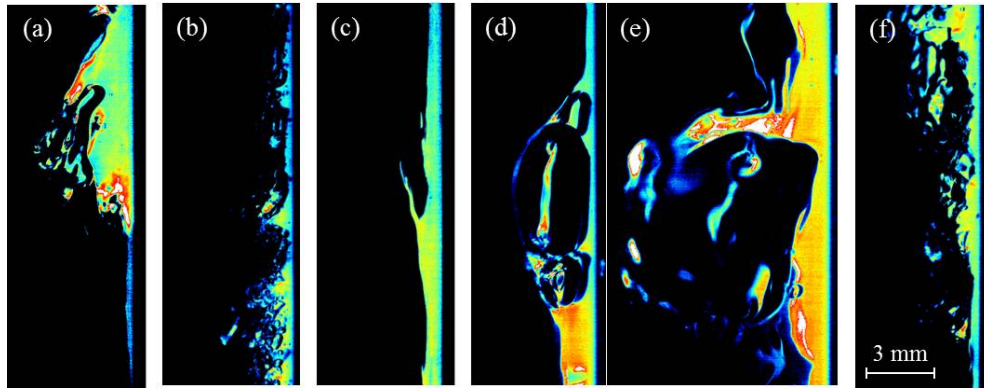


Figure 4: Instantaneous PLIF images of events that lead to entrainment of liquid into the gas phase. (a) ‘disturbance wave’, (b) ‘wave undercut’, (c) ‘ligament break-up’, (d)–(e) different stages of a ‘bubble burst’ and (f) ‘liquid impingement’. The selected images are from annular flows at: (a)–(c), (f) $Re_L = 1\,226$; $Re_G = 21\,100$ and (d)–(e) $Re_L = 1\,532$; $Re_G = 0$.

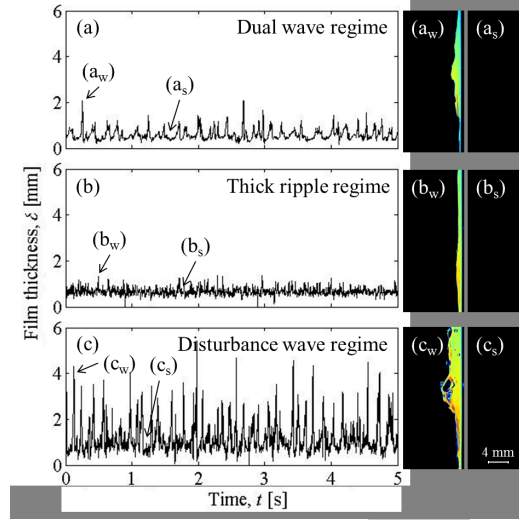


Figure 5: Temporal film thickness time-traces and instantaneous raw PLIF images of wave fronts (x_w) and substrates (x_s) for falling films ($Re_G = 0$) and liquid Reynolds numbers Re_L of: (a) 306, (b) 613, and (c) 1532.

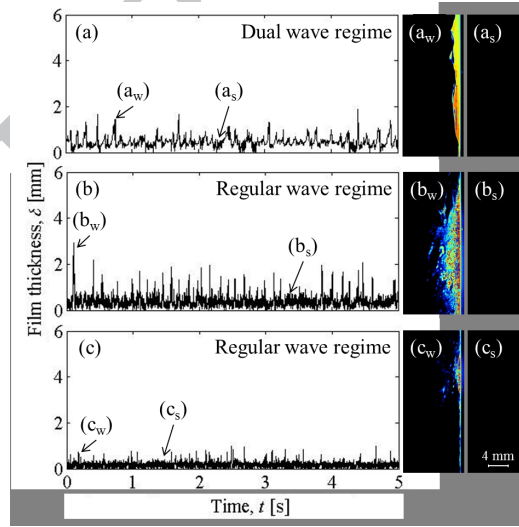


Figure 6: Temporal film thickness time-traces and instantaneous raw PLIF images of wave fronts (x_w) and substrates (x_s) for a liquid Reynolds number $Re_L = 306$ and gas Reynolds numbers Re_G of: (a) 21 100, (b) 42 300, and (c) 84 600.

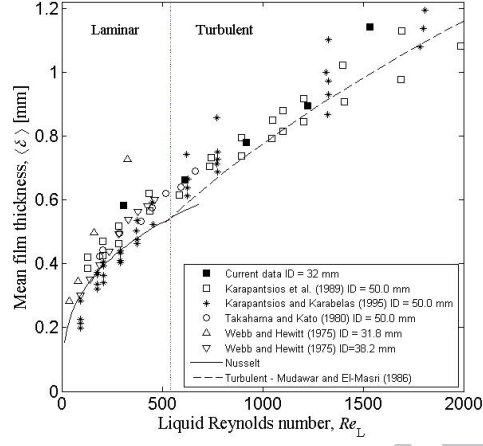


Figure 7: Comparison of the mean film thickness $\langle \delta \rangle$ with data from other investigators.

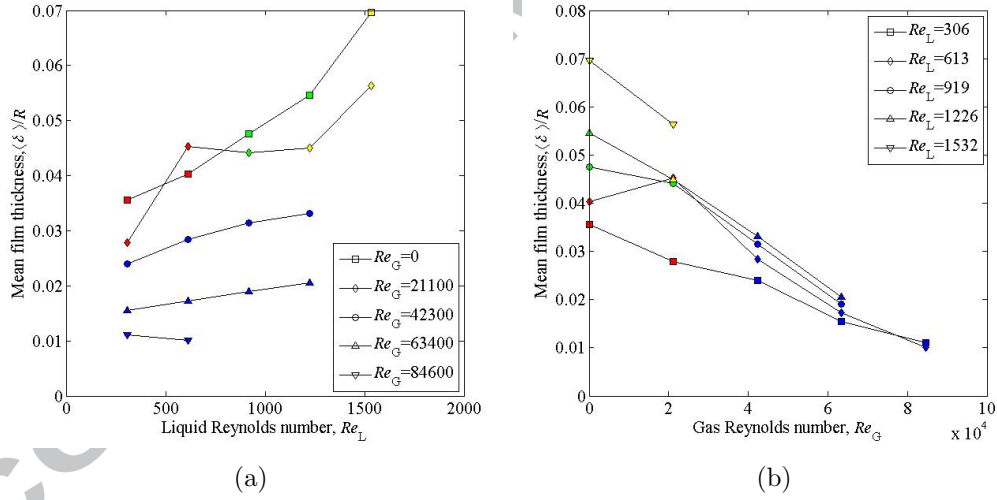


Figure 8: Mean film thickness $\langle \delta \rangle$ as a function of: (a) liquid Reynolds number Re_L , and (b) gas Reynolds number Re_G . The colours indicate different flow regimes identified in Section 3.1 and Figure 3: dual-wave regime (red), thick ripple regime (green), disturbance wave regime (yellow), and regular wave regime (blue).

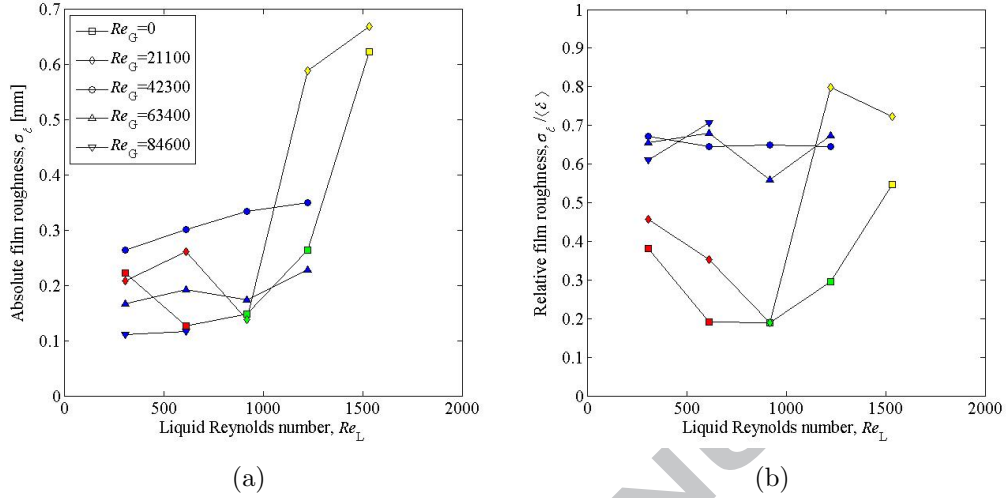


Figure 9: (a) Absolute roughness σ_δ , and (b) relative roughness $\sigma_\delta / \langle \delta \rangle$ of the liquid film as a function of liquid Reynolds number Re_L for various gas Reynolds numbers Re_G . The colours indicate different flow regimes identified in Section 3.1 and Figure 3: dual-wave regime (red), thick ripple regime (green), disturbance wave regime (yellow), and regular wave regime (blue). The legend in Figure 9(a) applies to both figure subplots.

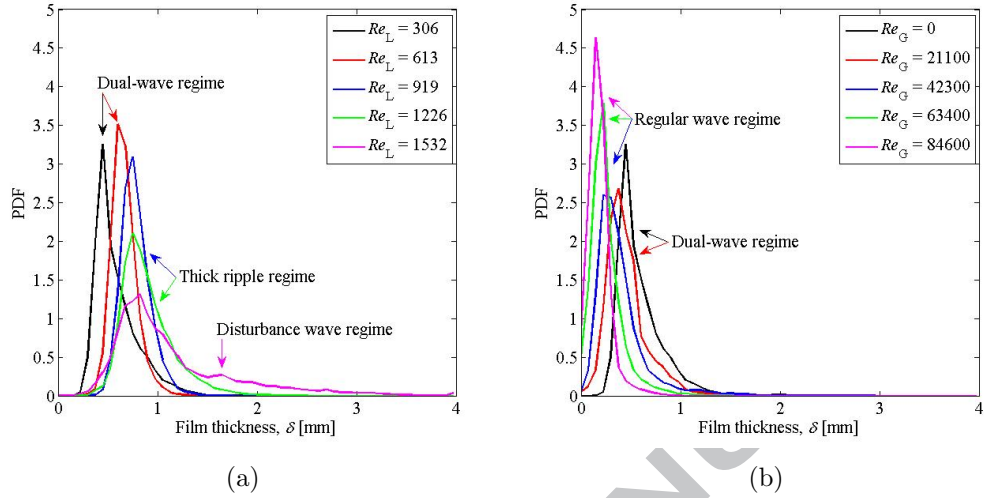


Figure 10: Probability density functions (PDFs) of film thickness $P(\delta)$ for: (a) constant gas Reynolds number of $Re_G = 0$ (i.e., falling films), and (b) constant liquid Reynolds number of $Re_L = 306$.

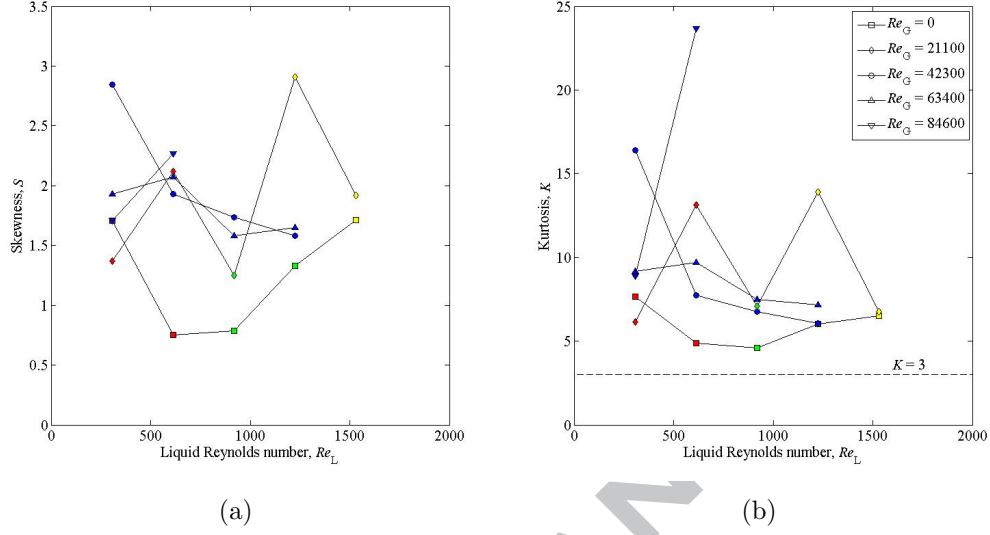


Figure 11: (a) Skewness S_δ , and (b) kurtosis K_δ of the probability density functions of film thickness $P(\delta)$ as a function of liquid Reynolds number Re_L . The colours indicate different flow regimes identified in Section 3.1 and Figure 3: dual-wave regime (red), thick ripple regime (green), disturbance wave regime (yellow), and regular wave regime (blue). The legend in Figure 11(b) applies to both figure subplots.

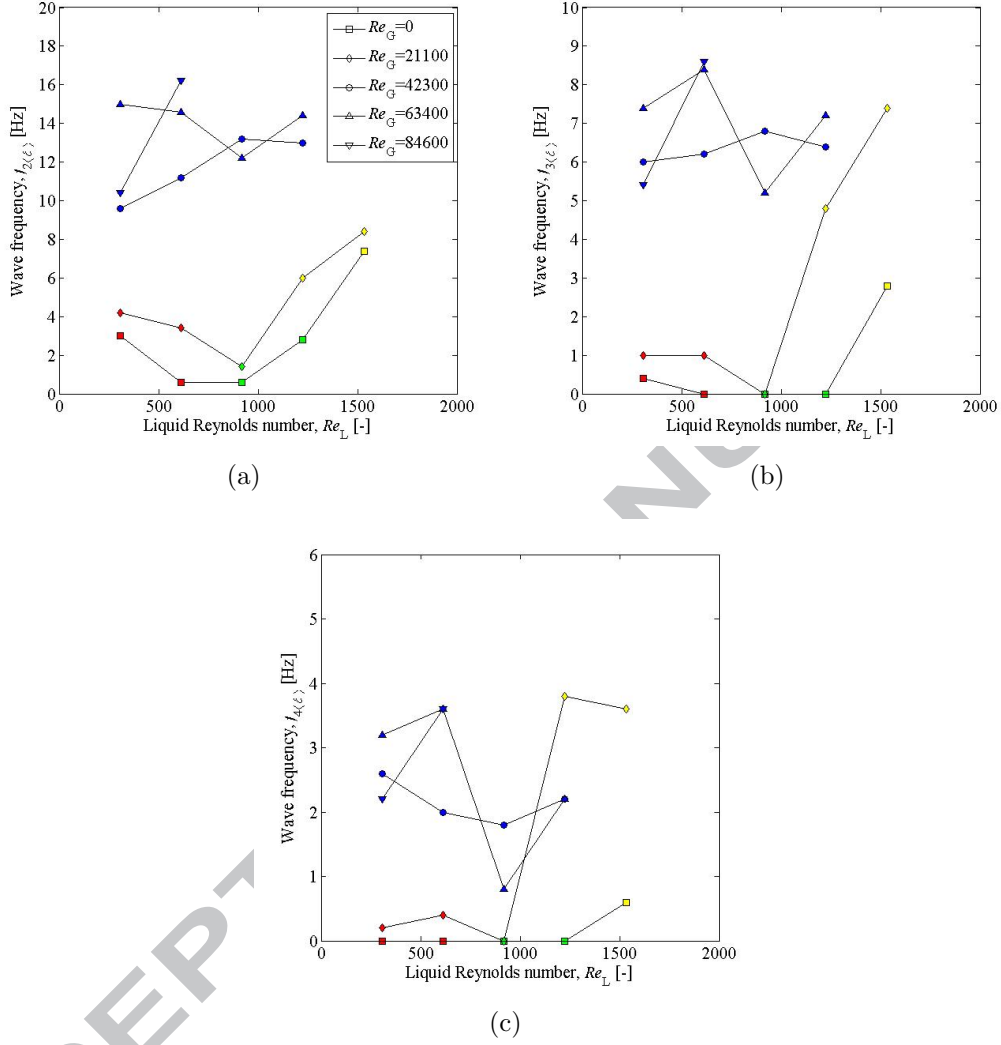


Figure 12: Frequency of waves f_W with amplitudes greater than: (a) $2\langle\delta\rangle$, (b) $3\langle\delta\rangle$ and (c) $4\langle\delta\rangle$. The colours indicate different flow regimes identified in Section 3.1 and Figure 3: dual-wave regime (red), thick ripple regime (green), disturbance wave regime (yellow), and regular wave regime (blue). The legend in Figure 12(a) applies to all figure subplots.

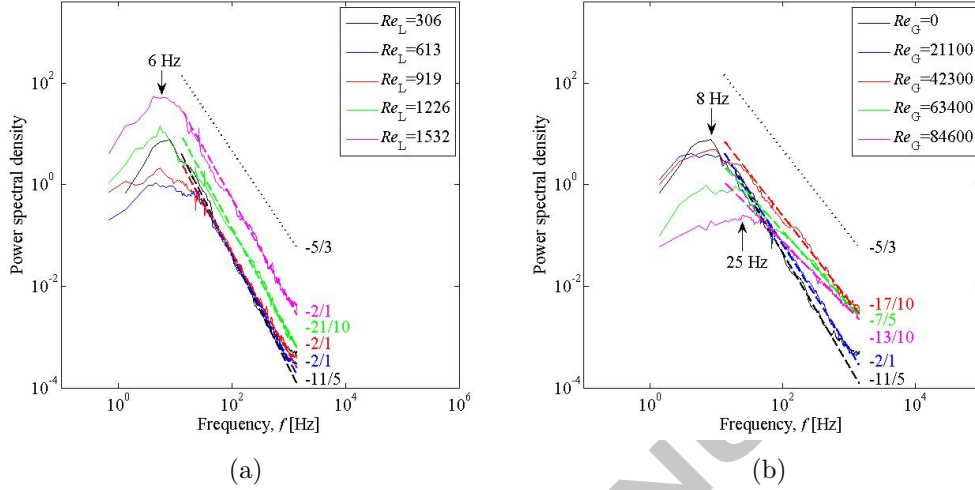


Figure 13: Power spectral densities (PSDs) for: (a) constant gas Reynolds number $Re_G = 0$ (i.e., falling films), and (b) constant liquid Reynolds number $Re_L = 306$.

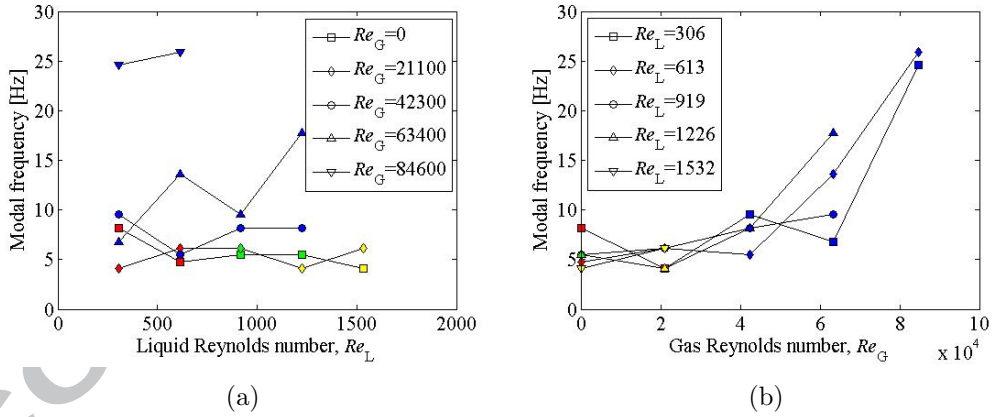


Figure 14: Peak power frequency f_{PSD} as a function of: (a) liquid Reynolds number Re_L , and (b) gas Reynolds number Re_G . The colours indicate different flow regimes identified in Section 3.1 and Figure 3: dual-wave regime (red), thick ripple regime (green), disturbance wave regime (yellow), and regular wave regime (blue).

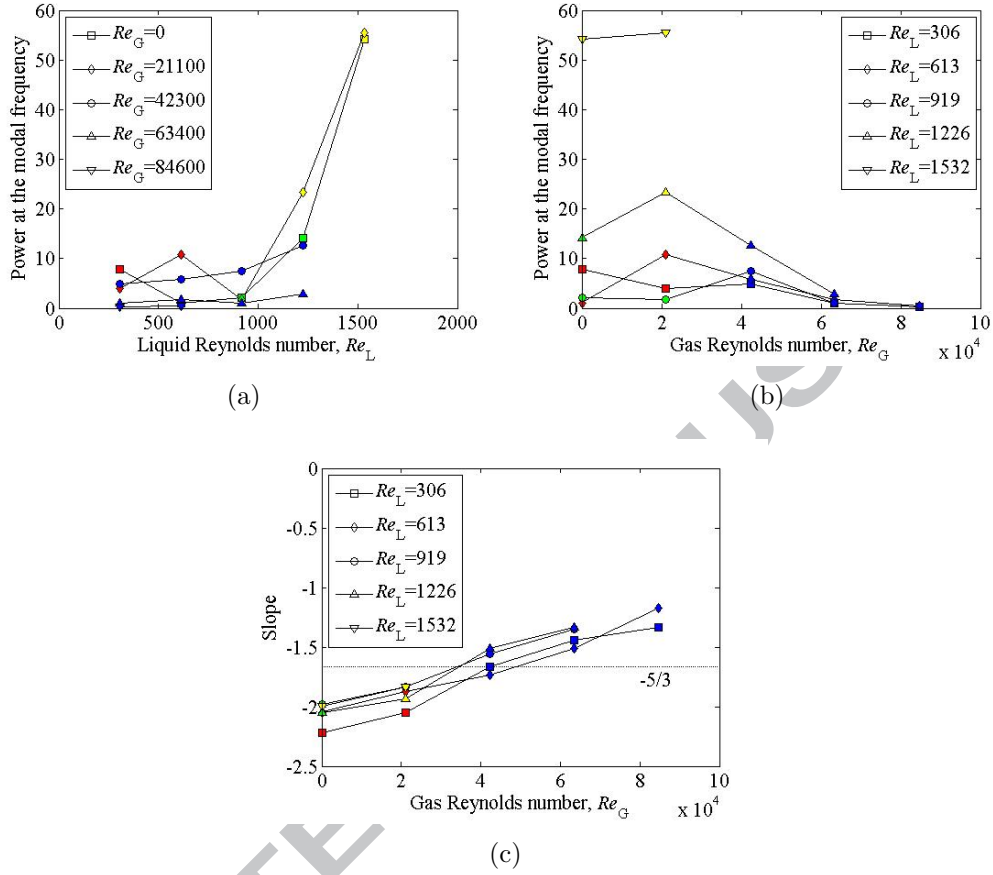


Figure 15: Power density at the PSD peak frequency as a function of: (a) liquid Reynolds number Re_L , and (b) gas Reynolds number Re_G . (c) Power law decay of the PSD slopes at high frequencies as a function of gas Reynolds number Re_G . The colours indicate different flow regimes identified in Section 3.1 and Figure 3: dual-wave regime (red), thick ripple regime (green), disturbance wave regime (yellow), and regular wave regime (blue).

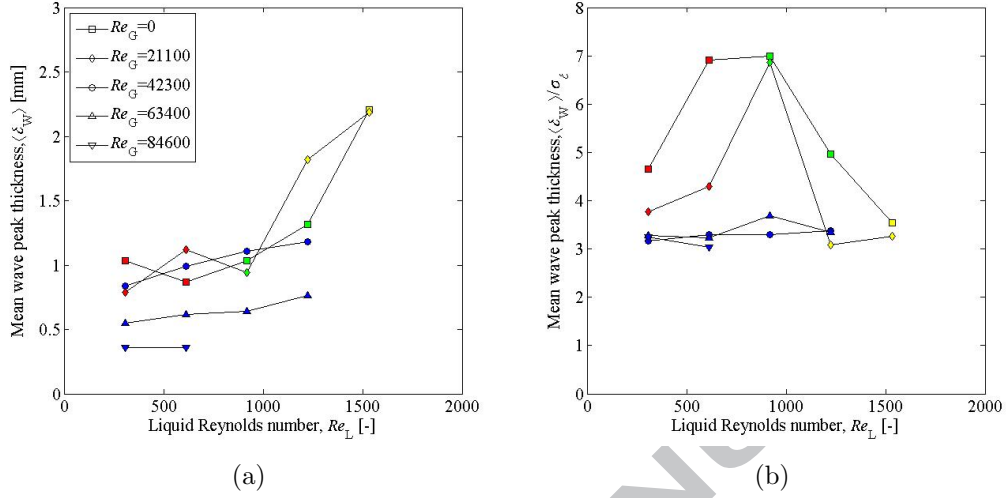


Figure 16: (a) Mean wave amplitude $\langle \delta_W \rangle$, and (b) mean wave amplitude normalized by the absolute wave roughness $\langle \delta_W \rangle / \sigma_\delta$ as a function of liquid Reynolds number Re_L . The colours indicate different flow regimes identified in Section 3.1 and Figure 3: dual-wave regime (red), thick ripple regime (green), disturbance wave regime (yellow), and regular wave regime (blue). The legend in Figure 16(a) applies to both figure subplots.

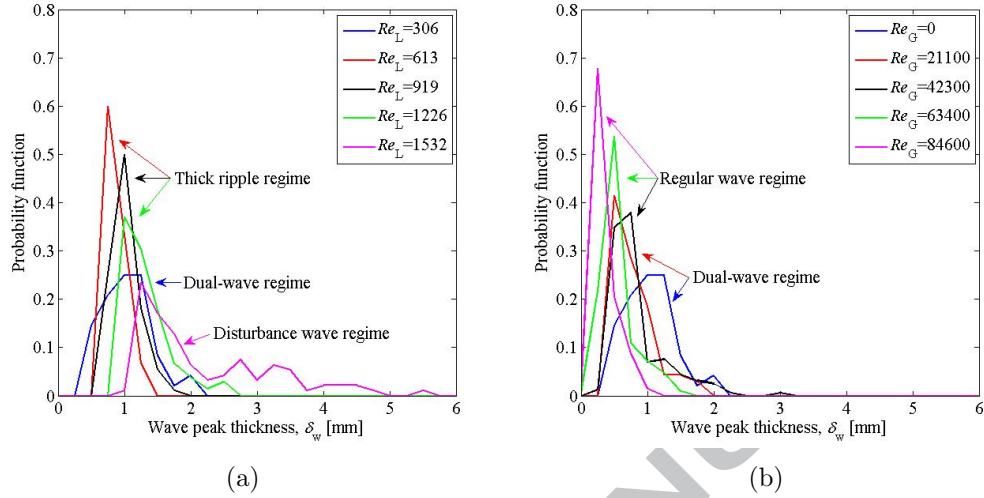


Figure 17: Probability functions (PFs) of wave amplitude $P(\delta_W)$ for: (a) constant gas Reynolds number $Re_G = 0$ (i.e., falling film), and (b) constant liquid Reynolds number $Re_L = 306$.

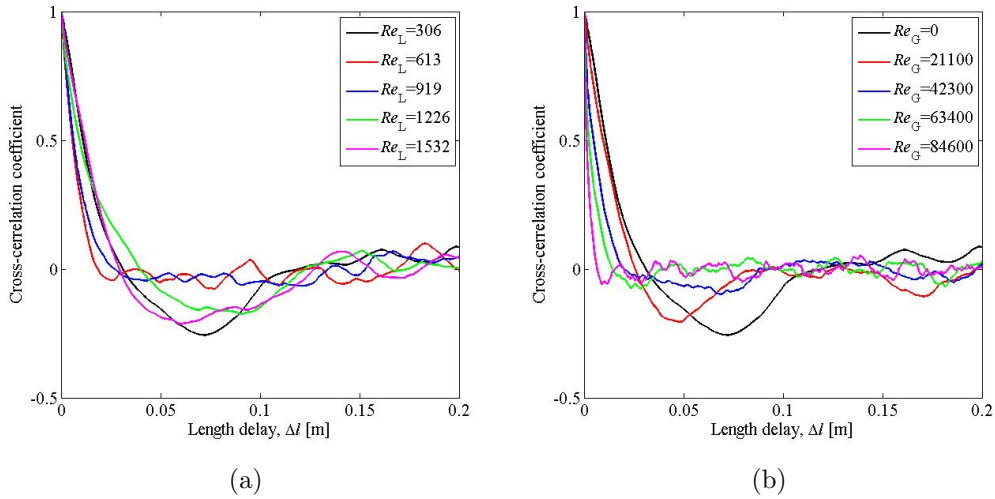


Figure 18: Autocorrelation functions (ACFs) of the film thickness for: (a) constant gas Reynolds number $Re_G = 0$, and (b) constant liquid Reynolds number $Re_L = 306$.

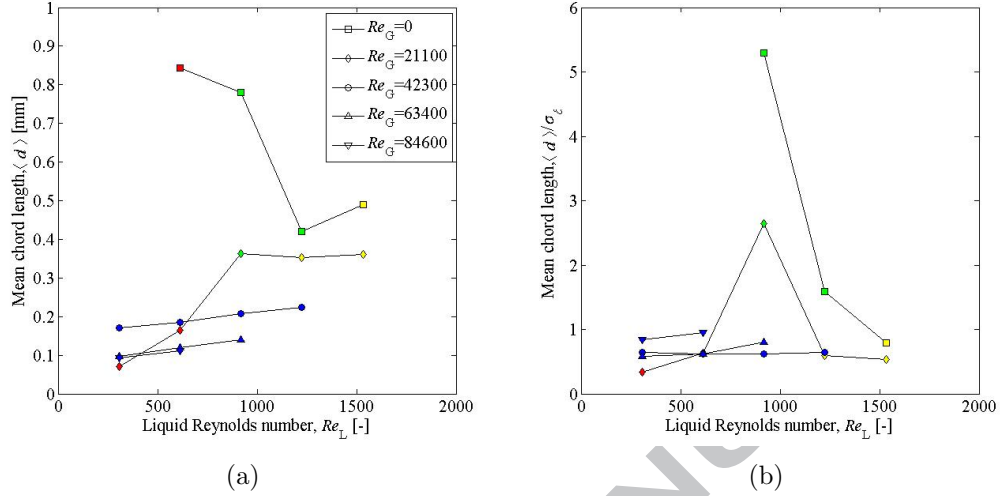


Figure 19: (a) Mean bubble chord length $\langle d \rangle$, and (b) mean bubble chord length normalized by the absolute film roughness $\langle d \rangle / \sigma_\delta$ as a function of liquid Reynolds number Re_L . The colours indicate different flow regimes identified in Section 3.1 and Figure 3: dual-wave regime (red), thick ripple regime (green), disturbance wave regime (yellow), and regular wave regime (blue). The legend in Figure 19(a) applies to both figure subplots.

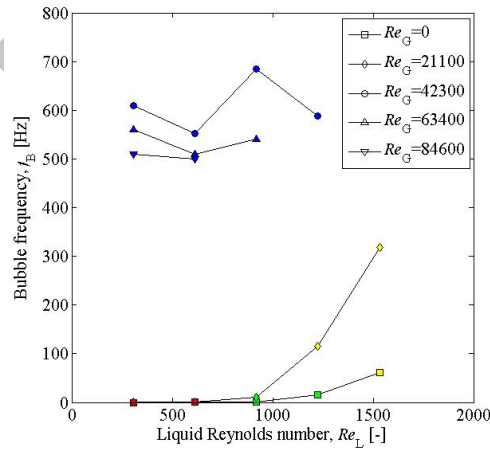


Figure 20: Bubble frequency f_B as a function of liquid Reynolds number Re_L . The colours indicate different flow regimes identified in Section 3.1 and Figure 3: dual-wave regime (red), thick ripple regime (green), disturbance wave regime (yellow), and regular wave regime (blue).

847 **Table captions**

Table 1: Experimental matrix of the investigated flow conditions. Here, Re_L^* corresponds to the liquid Reynolds number based on the liquid superficial velocity U_L and the full pipe diameter D .

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849 **References**

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$Q_L \cdot 10^{-5}$ $\text{m}^3 \cdot \text{s}^{-1}$	$Q_G \cdot 10^{-3}$ $\text{m}^3 \cdot \text{s}^{-1}$	$U_L \cdot 10^{-2}$ $\text{m} \cdot \text{s}^{-1}$	U_G $\text{m} \cdot \text{s}^{-1}$	Re_L -	Re_L^* -	Re_G -
2.8	0.0	3.4	0.0	306	1 224	0
5.6	0.0	6.7	0.0	613	2 452	0
8.4	0.0	10.1	0.0	919	3 676	0
11.1	0.0	13.5	0.0	1226	4 904	0
13.9	0.0	16.8	0.0	1532	6 128	0
2.8	8.3	3.4	8.5	306	1 224	21 100
5.6	8.3	6.7	8.5	613	2 452	21 100
8.4	8.3	10.1	8.5	919	3 676	21 100
11.1	8.3	13.5	8.5	1226	4 904	21 100
13.9	8.3	16.8	8.5	1532	6 128	21 100
2.8	16.7	3.4	17.0	306	1 224	42 300
5.6	16.7	6.7	17.0	613	2 452	42 300
8.4	16.7	10.1	17.0	919	3 676	42 300
11.1	16.7	13.5	17.0	1226	4 904	42 300
2.8	25.0	3.4	25.2	306	1 224	63 400
5.6	25.0	6.7	25.2	613	2 452	63 400
8.4	25.0	10.1	25.2	919	3 676	63 400
11.1	25.0	13.5	25.2	1226	4 904	63 400
2.8	33.3	3.4	34.0	306	1 224	84 600
5.6	33.3	6.7	34.0	613	2 452	84 600

An Experimental Characterization of Downwards Gas– Liquid Annular Flow by Laser-Induced Fluorescence: Flow Regimes and Film Statistics

Ivan Zadrazil, Omar K. Matar and Christos N. Markides

- Downwards annular flow was characterized experimentally over a range of conditions
- Spatially resolved LIF measurements were used to visualize the flows quantitatively
- Flow behaviour and gas entrainment into the liquid film were observed and analyzed
- Statistical data on liquid film thickness, wave events and gas entrainment are reported
- Four flow regimes were identified, including a new ‘disturbance’ wave regime