

Development of new fluidic devices for processing droplets

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by

Ana Teresa Sanches Cerdeira

Supervised by

José Daniel Pacheco Araújo

João Mário Rodrigues Miranda

João Bernardo Lares Moreira de Campos



Department of Chemical and Biological Engineering
Faculty of Engineering, University of Porto, Portugal

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Ana Teresa Sanches Cerdeira

up201811565@up.pt

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Department of Chemical and Biological Engineering

Faculty of Engineering of the University of Porto

Rua Dr. Roberto Frias, s/n, 4200-465, Porto, Portugal

www.fe.up.pt

feup@fe.up.pt

The work was developed in Centro de Estudos de Fenómenos de Transporte (CEFT), and supervised by:

José Daniel Pacheco Araújo

daraujo@fe.up.pt

João Mário Rodrigues Miranda

jmiranda@fe.up.pt

João Bernardo Lares Moreira de Campos

jmc@fe.up.pt

Department of Chemical and Biological Engineering

Faculty of Engineering of the University of Porto

Rua Dr. Roberto Frias, s/n, 4200-465, Porto, Portugal

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Abstract

Two-phase flows involving droplets play a central role in a wide range of industrial and biomedical applications, including artificial blood substitutes, emulsification processes, multiphase chemical reactors, oil-water separation in petroleum extraction systems, wastewater treatment, and microfluidic technologies. Particularly at milli and microscales, the rapid development of fabrication techniques have enabled the design of complex structured devices incorporating contractions, expansions, T-junctions, and arrays of obstacles to manipulate multiphase flows. However, the behavior of droplets in such confined geometries remains governed by intricate interfacial dynamics involving deformation, coalescence, and breakup processes. A comprehensive understanding of these mechanisms is essential for the design and optimization of fluidic systems.

A literature review is first presented, addressing gas-liquid and liquid-liquid flows involving Newtonian and non-Newtonian fluids in microfluidic elements such as contractions, expansions, constrictions, obstacles, and T-junctions. The review highlights the main physical mechanisms governing droplet and bubble dynamics and identifies key parameters influencing interfacial phenomena in confined multiphase flows.

This PhD thesis investigates the hydrodynamic behavior, deformation, and breakup of droplets flowing through constricted geometries and structured channels at the milli and microscales using a combined experimental and numerical approach. Experimental investigations were conducted in 3D-printed millifluidic constricted channels to study droplet dynamics under a wide range of viscosity ratios ($10^{-2} < X < 10^2$) and Capillary numbers of the continuous phase ($10^{-4} < Ca_c < 10^{-1}$). Numerical simulations were performed with a Computational Fluid Dynamics (CFD) tool, more specifically, the commercial software package Ansys FLUENT[®] with a model that included the Volume of Fluid (VOF) methodology for interface tracking. These simulations allowed to analyze droplet deformation, velocity, and film thickness in both milli and microscale geometries. The numerical results showed strong agreement with experimental measurements of droplet velocity and length in constricted regions, validating the computational model and enabling a detailed analysis of the governing flow mechanisms.

Further experiments explored droplet deformation and breakup in 3D-printed millifluidic channels containing arrays of rectangular obstacles with different configurations, including transverse, longitudinal, and herringbone arrangements. The experiments considered viscosity ratios (X) between 10^{-2} and 10^2 , and Capillary numbers of the dispersed phase (Ca_d) between 10^{-4} and 10^0 . The results demonstrated that droplet size at the outlet is primarily controlled by obstacle configuration and channel porosity, while viscous stresses associated with increasing Ca_d promote stronger droplet deformation and breakup. Among the tested designs, the herringbone configuration proved to be the most effective in producing smaller droplets due to enhanced droplet-obstacle interactions.

Finally, a combined milli and microfluidic strategy was developed to produce biocompatible microparticles with controlled size suitable for biomedical applications. Monodisperse droplets generated in a millifluidic flow-focusing device with a herringbone array of obstacles were subsequently processed in a microchannel containing a transverse array of obstacles, leading to successive breakup events and significant size reduction. Based on fluid systems of biomedical relevance, including perfluorodecalin dispersed in sodium alginate solution and sodium alginate droplets dispersed in mineral oil, a final droplet size of approximately $10\text{ }\mu\text{m}$ was achieved.

Overall, this work demonstrates that passive geometrical fluidic elements such as constrictions and arrays of obstacles provide effective hydrodynamic mechanisms for controlling droplet deformation and breakup across multiple length scales. The results contribute to a deeper understanding of multiphase flow in confined geometries and provide design guidelines for milli and microfluidic systems aimed at droplet manipulation and the scalable production of microparticles for biomedical applications.

Keywords: droplet milli/microfluidics; 3D printing; CFD; constrictions; arrays of obstacles; droplet breakup; droplet deformation.

Resumo

Os escoamentos bifásicos que envolvem gotas desempenham um papel central numa vasta gama de aplicações químicas e biomédicas, incluindo substitutos artificiais do sangue, processos de emulsificação, reatores químicos multifásicos, separação óleo-água em sistemas de extração de petróleo, tratamento de águas residuais e tecnologias microfluídicas. Em particular para sistemas às escalas mili e micro, o rápido desenvolvimento das técnicas de fabricação tem impulsionado o desenvolvimento de dispositivos complexos que incorporam contrações, expansões, junções em T e matrizes de obstáculos com o objetivo de manipular escoamentos multifásicos. No entanto, o comportamento das gotas em geometrias confinadas é fortemente dependente de fenómenos complexos que controlam os processos de deformação, coalescência e fragmentação. Assim sendo, para projetar e otimizar sistemas fluídicos de forma eficiente, é essencial ter uma compreensão abrangente e detalhada deste tipo de processos.

Numa fase inicial, foi realizada uma revisão aprofundada da literatura, abordando escoamentos gás-líquido e líquido-líquido com fluidos Newtonianos e não Newtonianos em elementos microfluídicos, tais como contrações, expansões, constrições, obstáculos e junções em T. Esta revisão destaca os principais mecanismos físicos que controlam a dinâmica de gotas e bolhas e identifica os parâmetros fundamentais que influenciam os fenómenos interfaciais em escoamentos multifásicos confinados.

Esta tese de doutoramento recorre a abordagens experimentais e numéricas para investigar o comportamento hidrodinâmico, a deformação e a fragmentação de gotas em escoamento através de geometrias com constrições e canais com matrizes de obstáculos às escalas mili e micro. O trabalho experimental foi realizado em canais milifluídicos com constrições fabricados por impressão 3D, de forma a estudar a dinâmica das gotas para uma ampla gama de razões de viscosidade ($10^{-2} < X < 10^2$) e números de Capilaridade da fase contínua ($10^{-4} < Ca_c < 10^{-1}$). Foram também realizadas simulações numéricas com uma ferramenta de Dinâmica de Fluidos Computacional (CFD), mais especificamente, o pacote comercial Ansys FLUENT® com um modelo que incorpora a metodologia do Volume de Fluido (*Volume of Fluid*, VOF) para descrever a interface entre fases. Estas simulações permitiram analisar a

deformação da gota, a sua velocidade e a espessura do filme em geometrias às escalas mili e micro. Os resultados numéricos mostraram uma forte concordância com as medições experimentais da velocidade e do comprimento das gotas nas regiões com estreitamento, o que valida o modelo computacional e permitiu uma análise detalhada dos mecanismos de escoamento dominantes.

Adicionalmente, foram realizadas experiências que exploraram a deformação e fragmentação de gotas em canais milifluídicos, fabricados por impressão 3D, com matrizes de obstáculos retangulares de diferentes configurações, incluindo arranjos transversais, longitudinais e em “espinha de peixe”. Nestas experiências foram utilizadas razões de viscosidade (X) entre 10^{-2} e 10^2 e números de Capilaridade da fase dispersa (Ca_d) entre 10^{-4} e 10^0 . Os resultados demonstraram que o tamanho das gotas à saída é controlado essencialmente pela configuração dos obstáculos e pela porosidade do canal, enquanto as tensões viscosas associadas ao aumento do Ca_d promovem uma deformação e fragmentação mais intensas das gotas. Entre as configurações testadas, a configuração em “espinha de peixe” revelou-se a mais eficaz na produção de gotas de menor dimensão devido à intensificação das interações gota-obstáculo.

Por fim, foi desenvolvida uma estratégia combinada de mili e microfluídica para produzir micropartículas biocompatíveis com tamanho controlado e adequadas para aplicações biomédicas. As gotas foram geradas num dispositivo milifluídico de *flow-focusing* composto por uma matriz de obstáculos em “espinha de peixe”, e posteriormente foram processadas num microcanal com uma matriz transversal de obstáculos, o que conduziu a eventos sucessivos de fragmentação e a uma redução significativa do seu tamanho. Utilizando sistemas de fluidos com relevância biomédica, incluindo perfluorodecalina dispersa numa solução de alginato de sódio e gotas de alginato de sódio dispersas em óleo mineral, foi possível obter gotas com um tamanho final de cerca de $10\ \mu\text{m}$.

De forma global, este trabalho demonstra que a aplicação de elementos fluídicos geométricos passivos, tais como constrições e matrizes de obstáculos, constitui uma abordagem hidrodinamicamente eficaz para controlar a deformação e fragmentação de gotas às escalas mili e micro. Os resultados contribuem para uma compreensão mais aprofundada do escoamento multifásico em geometrias confinadas e fornecem diretrizes de projeto relevantes para sistemas mili e microfluídicos destinados à

manipulação de gotas e à possível produção de micropartículas para aplicações biomédicas.

Palavras-chave: mili/microfluídica de gotas; impressão 3D; CFD; constrições; matrizes de obstáculos; fragmentação de gotas; deformação de gotas

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Introduction

Introduction

1.1. Relevance and motivation

Two-phase flows involving immiscible fluids are ubiquitous in natural systems (e.g., milk emulsions [1], geysers and magma-gas flows in volcanoes [2]) and industrial processes. In particular, they have a decisive role in a broad range of applications, including energy production (e.g., oil-water separation in petroleum extraction [3] and gas-liquid flows in heat exchangers [4]), chemical processing (e.g., emulsification, liquid-liquid extraction and multiphase reactors [5]), biomedical technologies (e.g., droplet-based microfluidic drug delivery [6], lab-on-a-chip assays [7] and artificial blood substitutes [8, 9]), and environmental systems (e.g., wastewater treatment [10]). In such systems, the interaction between phases gives rise to complex flow structures whose characteristics strongly influence transport phenomena, operational efficiency, and system reliability [11, 12]. Understanding and controlling these interactions is therefore a central challenge in multiphase flow research.

Among the different two-phase flow patterns, dispersed flows – where one phase is transported as droplets within a continuous phase – are of particular importance given their prevalence [13, 14]. Depending on the simultaneous phenomena involved, droplets act as fundamental transport units that strongly influence mass and heat transfer rates, and reaction efficiency [5, 15, 16]. Droplets are governed by the interaction between hydrodynamic forces and interfacial tension, and so, their behavior is highly sensitive to fluid properties, flow conditions, and geometric confinement [17, 18]. Consequently, droplet deformation, motion, and breakup are key mechanisms that must be controlled to optimize a wide range of processes, including emulsification, encapsulation, extraction, mixing, and chemical reactions [6, 19, 20].

The increasing use of miniaturized and microstructured devices has further emphasized the importance of droplet-scale phenomena [21-23]. At milli and microscales, surface tension and viscous forces dominate over inertia, leading to flow behaviors that differ fundamentally from those observed in macroscale systems. Under these conditions, channel geometry becomes a primary tool for flow manipulation rather than being a merely passive conduit [24]. Small fluidic geometrical elements – such as contractions (Figure 1.1a), expansions (Figure 1.1b), constrictions (Figure 1.1c and Figure 1.1d), and obstacles (Figure 1.1e) – can induce strong local variations in droplets' flow, shape, stability and velocity [17, 25-29].

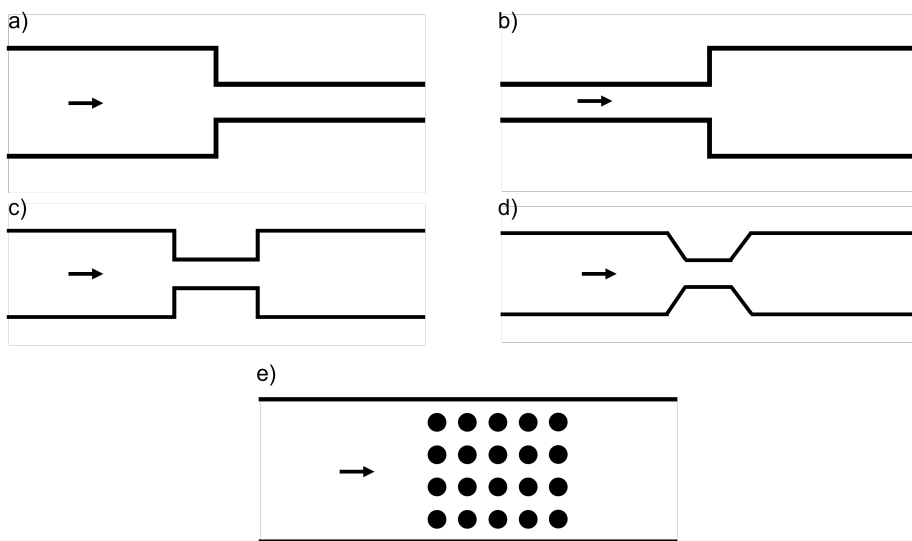


Figure 1.1. Schematic illustrative representation of several fluidic geometrical elements: a) contraction; b) expansion; c) abrupt constriction; d) gradual constriction; and e) array of obstacles.

Concurrently, advances in additive manufacturing – particularly stereolithography three-dimensional (3D) printing [30, 31] – have enabled high-precision rapid prototyping of complex millifluidic geometries. This technological progress opens new possibilities to systematically investigate how constrictions and different arrays of obstacles configurations influence droplet dynamics under controlled conditions.

Microfluidic devices commonly exploit fluidic geometrical elements to generate, manipulate, and control droplets. Droplet formation is typically achieved using T-junction or flow-focusing configurations [32-35], while constrictions [36-42] and obstacles [43-47] are employed to promote deformation, coalescence, or breakup. Sudden changes in channel geometry can promote distinct droplet response regimes, including trapping, squeezing, and breakup [48, 49]. Identifying the range of conditions where each regime occurs is crucial, since undesired breakup may compromise functionality in applications like drug delivery, while controlled breakup can be beneficial for enhancing mixing or mass transfer processes, and for producing microparticles [6, 50].

In this context, structured channels containing arrays of solid obstacles have emerged as an effective passive strategy for manipulating multiphase flows [24, 51]. As droplets traverse these arrays, repeated interactions with solid boundaries generate complex stress fields capable of enhancing deformation, inducing breakup, or regulating droplet trajectories without the need for external actuation. Arrays of obstacles are therefore widely used to intensify phase separation [52], improve mixing through chaotic advection [53-55], and enable precise fluidic control in lab-on-a-chip systems [43, 44, 46, 47, 56].

Despite their practical relevance, research addressing predictive understanding of droplet transport through complex arrays of obstacles [52, 57, 58] remains scarce, particularly when compared to the extensive literature on straight channels or single constrictions.

This gap in knowledge is especially relevant given the growing reliance on droplet-based microfluidics in chemical engineering, biotechnology, and materials science. Applications such as single-cell analysis, droplet-based polymerase chain reaction (PCR), high-throughput screening, and microscale reactors demand consistent control over droplet integrity and size distribution. In many scenarios, arrays of obstacles show

a considerable potential to transform polydisperse droplets into more uniform populations [29] or to intensify mixing between liquid phases [53-55].

The controlled production of microparticles with precise size and transport properties remains a key challenge in biomedical engineering, particularly for applications such as oxygen therapeutics, cell encapsulation, and blood-analogue fluids [19, 20, 59-65]. In these systems, particle size is critical to ensure adequate microvascular transport, controlled mass transfer, and biocompatibility. However, achieving uniform, scalable, and reproducible microparticle production without depending on complex external actuation or energy-intensive methods is still an important limitation.

1.2. Constrictions and arrays of obstacles

Among several geometrical fluidic elements (Figure 1.1), this PhD thesis focuses on abrupt constrictions and arrays of obstacles. These are passive geometrical elements that induce strong and localized modifications in the flow field. On one hand, constrictions generate variations in confinement, leading to pronounced changes in pressure, velocity, and interfacial stresses acting on droplets. These effects make them ideal for investigating fundamental mechanisms such as droplet deformation and breakup under controlled conditions. On the other hand, arrays of obstacles introduce repeated and spatially distributed perturbations to the flow. As droplets interact successively with multiple solid structures, complex stress distributions and flow circulation patterns develop, and breakup occurs, resulting in cumulative effects that cannot be achieved with isolated fluidic elements. Constrictions serve to understand the physics governing droplet behavior in confined environments, whereas arrays of obstacles represent a scalable strategy for practical applications, particularly for the production of microparticles with biomedical interest. Indeed, this work does not merely aim to analyze droplet behavior, but also to translate physical understanding into design principles for novel fluidic devices.

The problem addressed in this PhD thesis arises from a central challenge in droplet-based microfluidics: the need for precise, robust, and scalable control over droplet size, stability, shape, and dynamics. This work explores how constrictions and arrays of

obstacles can be used to control droplet deformation and induce breakup in a predictable and tunable manner.

The theoretical foundation underlying this work is based on the physics of multiphase flows in confined geometries, where the balance between viscous forces and interfacial tension governs the system behavior. A key aspect of this framework is the use of dimensional analysis, which is presented in detail in Chapter 2.

1.3. Numerical and experimental approach

To address the research problem, this thesis adopts a combined experimental and numerical methodology. The experimental approach is based on the fabrication and characterization of milli and microfluidic devices incorporating constrictions and arrays of obstacles. Advanced fabrication techniques, including stereolithography 3D printing and soft lithography, are employed to produce channels with well-defined geometries. Flow visualization and image analysis methods, including droplet tracking, are used to quantify droplet properties such as size and velocity, enabling a detailed characterization of droplet dynamics under a wide range of operating conditions.

Complementing the experiments, numerical simulations are performed using Computational Fluid Dynamics (CFD), with particular emphasis on the Volume of Fluid (VOF) method for interface tracking. These simulations provide access to detailed flow information, such as velocity fields and film thickness measurements. The numerical model is validated against experimental data, ensuring its reliability.

In summary, the focus on constrictions and arrays of obstacles reflects a deliberate strategy to bridge fundamental understanding and practical application in droplet microfluidics. By investigating how these fluidic geometrical elements influence droplet behavior, this thesis contributes to the development of passive, efficient, and scalable fluidic devices, particularly for microparticle production in biomedical contexts.

1.4. Objectives

The main scientific objective of this work is to achieve a more comprehensive understanding on the influence of fluidic geometrical elements – constrictions and arrays of obstacles – and surface properties (mainly the contact angle) on droplets' flow behavior. At both milli and microscales, an optimal combination of these geometric and physical parameters can significantly impact applications in medical devices, water treatment, and chemical production. Contributing to the knowledge about these multiphase flow systems has the potential to drive technological advancements, enabling the design of more efficient fluidic devices.

To accomplish this main goal, several experimental and numerical intermediate objectives were pursued:

- Fabrication of milli and microchannels using two different approaches: 1) stereolithography 3D printing to produce millichannels featuring a flow-focusing region followed by a constricted zone or an array of obstacles region; and 2) polydimethylsiloxane (PDMS) microchannels comprising an array of obstacles fabricated by soft lithography.
- Numerical validation: assess the applicability of the Ansys FLUENT® software using the VOF methodology to simulate droplet dynamics in three-dimensional constricted milli and microchannels. This validation is supported by systematic experiments in 3D-printed millichannels.
- Quantitative droplet characterization: analyze droplet size and velocity distributions within the designed geometrical elements using ImageJ®/Fiji® segmentation and particle tracking plugins, supplemented by Python scripts for comprehensive post-processing of experimental data.
- Droplet deformation and breakup analysis: investigate the effects of rectangular arrays of obstacles with different geometrical configurations on droplet deformation and breakup.
- Passive droplet control strategies: employ arrays of obstacles as an efficient passive approach to promote droplet breakup and produce droplets with uniform sizes.

- Production of biomedical-relevant droplets: generate size uniform microdroplets suitable for biomedical applications, including artificial oxygen carriers and blood-analogue fluids.

The outcomes of this study provide relevant insights that are very useful for the design and optimization of multiphase fluidic systems where precise droplet control is critical, such as in targeted drug delivery, biomedical diagnostics, environmental monitoring, and emulsification processes. By using intrinsic fluidic geometrical elements instead of external fields, these elements offer scalable and energy-efficient strategies for droplet manipulation, enabling the development of compact, adaptive, and high-performance milli and microfluidic devices.

1.5. Thesis outline

After the present introductory chapter, this PhD thesis is organized into four separate chapters, each based on a paper that has been published or is currently under review in scientific journals:

- Chapter 2: A. T. S. Cerdeira, J. B. L. M. Campos, J. M. Miranda, and J. D. P. Araújo, "Review on Microbubbles and Microdroplets Flowing through Microfluidic Geometrical Elements", published in *Micromachines*.
- Chapter 3: A. T. S. Cerdeira, J. B. L. M. Campos, J. M. Miranda, and J. D. P. Araújo, "Numerical and experimental analysis of droplet dynamics in constricted milli and microchannels", published in *Chemical Engineering and Processing - Process Intensification*.
- Chapter 4: A. T. S. Cerdeira, J. B. L. M. Campos, J. M. Miranda, and J. D. P. Araújo, "Droplet deformation and breakup in 3D-printed millichannels featuring arrays of rectangular obstacles with different configurations", published in *Chemical Engineering Science*.
- Chapter 5: A. T. S. Cerdeira, J. B. L. M. Campos, J. M. Miranda, and J. D. P. Araújo, "Production of microparticles with biomedical interest using array of obstacles devices", under review in *Chemical Engineering and Processing - Process Intensification*.

In Chapter 2, a review about the manipulation of bubbles and droplets in common microfluidic geometrical elements (i.e., constrictions, contractions, expansions, obstacles, and T-junctions) is presented. It addresses gas-liquid and liquid-liquid flows involving both Newtonian and non-Newtonian fluids. This review covers the relevant theoretical background of two-phase flows in these elements and summarizes key experimental and numerical studies along with their applications. Particular attention is devoted to the kinematics, fluid dynamics, and deformation behavior of bubbles and droplets, with emphasis on deformation, coalescence, and breakup mechanisms occurring as they pass through the aforementioned microfluidic elements. Overall, the review provides a structured and comprehensive survey of current advances in the study of two-phase flows in microfluidic geometrical elements. This review is further complemented in Appendix A with a state-of-the-art overview covering the past six years, including the most recent studies in this field.

Starting with the constrictions, Chapter 3 explores the three-dimensional flow of a single droplet through a constricted channel at milli and microscales using combined experimental and numerical approaches. This publication addresses a topic of high relevance for chemical and biomedical multiphase systems. Experiments were conducted in a novel 3D-printed constricted millichannel using different immiscible fluid pairs, marking one of the first applications of 3D printing to study droplet flow in such geometries. Complementary numerical simulations, based on the VOF methodology, replicated the droplet flow at both milli and microscales across a wide range of viscosity ratios (X) and Capillary numbers of the continuous phase (Ca_c). The values of droplet velocity and length within the constricted region were compared between experiments and simulations, showing strong agreement and validating the computational model. Additionally, numerical results provided a detailed analysis of velocity fields, droplet deformation, and film thickness. This kind of information is extremely helpful to clarify the effects of X and Ca_c , and offers valuable guidance for the optimization of these multiphase microfluidic systems. Appendix B provides supplementary material for Chapter 3.

In order to investigate the influence of more complex fluidic geometrical elements, Chapter 4 features droplet deformation and breakup in 3D-printed millichannels containing arrays of rectangular obstacles with different configurations. Experiments were conducted with two fluid pairs, systematically varying the flow rate ratio, the

Capillary number of the dispersed phase (Ca_d), and the array porosity to evaluate their combined effects on droplet dynamics. Quantitative flow visualization and image analysis were used to characterize droplet area and velocity at different millichannel regions. The novelty of this work lies in the direct comparison of distinct arrays of obstacles using high-precision 3D printing and robust quantitative analysis. Overall, the findings demonstrate that arrays of obstacles are effective passive strategies for promoting droplet breakup and size uniformity, providing modern design guidelines for droplet manipulation in millifluidic systems. Appendix C includes additional material related to Chapter 4.

Chapter 5 demonstrates that integrating arrays of obstacles within combined milli and microfluidic devices provides a robust and scalable platform for producing biocompatible microparticles with controlled size and transport properties. Two biomedically relevant fluid systems, perfluorodecalin (PFD) droplets in sodium alginate and sodium alginate droplets in mineral oil, were processed using a two-stage strategy: initial droplet generation in a 3D-printed millifluidic flow-focusing device, followed by secondary hydrodynamic refinement in a PDMS microchannel containing an array of rectangular obstacles. The synergy between alginate hydrogel chemistry, oxygen-carrying perfluorocarbon phases, and obstacle-based microfluidic design establishes a versatile framework for engineering functional microparticles, with potential for in-channel gelation, cell encapsulation, and scalable continuous manufacturing.

Finally, Chapter 6 summarizes the main contributions of this PhD thesis and outlines several directions for future research.

Nomenclature

Acronyms	Definition
3D	Three-dimensional
CFD	Computational Fluid Dynamics
PCR	Polymerase chain reaction
PDMS	Polydimethylsiloxane
PFD	Perfluorodecalin
VOF	Volume of Fluid

Latin	Definition	Units
Ca_c	Capillary number of the continuous phase	—
Ca_d	Capillary number of the dispersed phase	—
X	Viscosity ratio	—

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Review on Microbubbles and Microdroplets Flowing through Microfluidic Geometrical Elements

A. T. S. Cerdeira, J. B. L. M. Campos, J. M. Miranda, and J. D. P. Araújo, "Review on Microbubbles and Microdroplets Flowing through Microfluidic Geometrical Elements," *Micromachines*, vol. 11, no. 2, p. 201, 2020, doi: [10.3390/mi11020201](https://doi.org/10.3390/mi11020201)

*Review on Microbubbles and Microdroplets Flowing through Microfluidic Geometrical Elements***Abstract**

Two-phase flows are found in several industrial systems/applications, including boilers and condensers, which are used in power generation or refrigeration, steam generators, oil/gas extraction wells and refineries, flame stabilizers, safety valves, among many others. The structure of these flows is complex, and it is largely governed by the extent of interphase interactions. In the last two decades, due to a large development of microfabrication technologies, many microstructured devices involving several elements (constrictions, contractions, expansions, obstacles, or T-junctions) have been designed and manufactured. The pursuit for innovation in two-phase flows in these elements require an understanding and control of the behavior of bubble/droplet flow. The need to systematize the most relevant studies that involve these issues constitutes the motivation for this review. In the present work, literature addressing gas-liquid and liquid-liquid flows, with Newtonian and non-Newtonian fluids, and covering theoretical, experimental, and numerical approaches, is reviewed. Particular focus is given to the deformation, coalescence, and breakup mechanisms when bubbles and droplets pass through the aforementioned microfluidic elements.

2.1. Introduction

Flow patterns are defined as the different types of spatial distribution of immiscible phases that develop as they flow simultaneously in a channel. These patterns are essentially dependent on the channel geometry, fluid physical properties, and flow rates of each phase. Furthermore, within each flow pattern, the channel geometry has a large impact on the behavior of characteristic hydrodynamic features. This impact must be studied in detail, since the geometry is directly involved on the performance of practical applications, ranging from micro to macroscales.

Macroscale pipe networks comprise fluidic elements that normally process two-phase mixtures. Among the most common are expansions, contractions, and constrictions. These fluidic elements can be found in chemical reactors, gas-lift in oil wells, petrochemical plants, natural gas transportation in subsea pipelines, and power generation units [1]. As an example, the splitting of Taylor bubbles into two daughter bubbles, being caused by the passage through an expansion, is still scarcely studied experimentally [2], and there are only two published computational works of Taylor bubbles rising through expansions in pipe diameter [3, 4]. Taylor bubbles/droplets are long and wide bullet-shaped entities – occupying almost all the cross-section of a channel – that appear in one of the most frequent multiphase flow patterns, named slug flow. Despite the intrinsic complexity involved, the slug flow hydrodynamics in straight channels is a well-documented and well-studied subject [5], but the cases that involve other fluidic elements still need to be systematically studied. For the motion of droplets through a sudden/gradual expansion/contraction, several numerical and experimental works considered the flow through horizontal pipes, but they are mainly focused on pressure drop [6-9].

The heat and mass transfer can be significantly enhanced in a microsystem due to the high surface volume ratio. Currently, bubble/droplet-based microfluidics has been successfully applied in chemical and biological analysis [10, 11], the synthesis of advanced materials [12], sample pretreatment [13], and the encapsulation of cells [14]. Droplets are liquid entities that flow in an immiscible liquid continuous phase, and bubbles are gas entities also flowing in a liquid fluid. The use of droplets as microreactors presents a lot of advantages when compared with single-phase microfluidics, such as: confinement of reactants or prevention of longitudinal

dispersion, and cross contamination between samples [15]. The reduction of unwanted adhesion/absorption of the material confined in droplets at the microchannel walls, the possibility of varying, in each droplet the physicochemical conditions under which chemical or biological process develop, and the facilitated heat/mass transport due to the fast mixing promoted by droplets are other benefits [15, 16]. The use of bubbles might be interesting to produce contrast agents in medical applications [17], as a source of reactants in the gas phase to promote reactions in liquid or solid (i.e., microchannel wall) phases [18], and to study in vitro gas embolisms [19].

Microfluidic devices (length scales lower than one millimeter) comprise microfluidic elements, analogous to their macroscale counterparts, to transport and distribute fluids, to promote mixing, reaction, and mass transfer. In the case of multiphase flows, these microfluidic elements also include expansions, contractions, constrictions, obstacles, and T-junctions that can be used to promote the formation, breakup, or coalescence of microdroplets and microbubbles.

Taylor microbubbles/droplets have garnered significant attention in various fields, including enhanced oil recovery, surface cleaning, medical purposes, and chemical and biomedical (such as drug delivery) engineering applications [20, 21]. Good knowledge on two-phase flow hydrodynamics in millichannels and microchannels is important for the design, optimization, and control of structured microflow systems, since the physics governing bubble/droplet passing through microfluidic elements is really complex [22, 23].

The present review addresses several aspects of handling bubbles/droplets in some of the most common microfluidic elements and the theoretical, experimental, and numerical tools that are used to tackle this problem. The scope of this review is mainly oriented towards the physics, experimental, and modelling approaches, and the designs of microfluidic elements for the control of deformable bubbles and droplets. A summary of the main experimental and numerical studies, together with their applications, is provided after a general overview of the theoretical background supporting the study of two-phase flows in microfluidic geometrical elements. The kinematics, fluid dynamics, and deformation dynamics of the bubbles/droplets are also discussed. Particular emphasis is given to the analysis of coalescence and breakup mechanisms that bubbles and droplets experience when they pass through microfluidic

elements. With this review, the overall picture regarding two-phase flows in microfluidic geometrical elements is systematized.

2.2. Theoretical background

Firstly, it is important to provide an overview of the main concepts that are involved in the flow of bubbles and droplets through microfluidic elements. Subsection 2.2.1 enunciates a description of the squeezing process in these fluidic elements, together with a quantitative evaluation of the droplet deformation and the channel confinement. The main types of microfluidic elements reported in the literature, with different geometries and arrangements, are addressed in Subsection 2.2.2. A dimensional analysis and the main operation parameters (e.g., pressure and transit time) of the problem are also discussed (Subsection 2.2.3).

2.2.1. Squeezing process in microfluidic geometrical elements

The continuous phase transports shear stresses and deforms the dispersed phase when flowing around a dispersed phase. The deformation of the bubble/droplet depends on the flow conditions, physical properties of the dispersed phase, and geometry of the microfluidic element that is involved (Subsection 2.2.2).

During a squeezing process through a microfluidic element, the bubble/droplet can take different shapes, varying from a parachute to a dumbbell shape. In the particular case of Taylor bubbles, several interface shapes can be observed by tuning the Reynolds (Re) and Capillary (Ca) numbers [22]. The deformed bubble/droplet assumes normally a bullet form while considering a Newtonian bubble or droplet and a Newtonian continuous phase. The deformation of a Newtonian bubble or droplet in a viscoelastic continuous phase induces, in most of the cases, an ellipsoid shape [24].

The deformation of the dispersed phase is commonly quantified by the Deformation Factor, also known as the Elongation Factor. Additionally, the bubble/droplet deformation is associated to a change in the geometry of the channel, so, to describe the degree of deformation in relation to the size of the contraction, a confinement parameter, known as the Confinement Factor, can also be adopted. When the

dimensions of the channels are smaller than the diameter of the droplets/bubbles, the Deformation Factor is set as the ratio between the maximum length of the droplet/bubble during the deformation process and the un-deformed droplet/bubble diameter. The Confinement Factor is defined as the non-dimensional ratio between the diameter of the un-deformed droplet/bubble and the diameter of the constricted channel. Summaries of the most relevant analytical expressions that are available in the literature for evaluating the Deformation and Confinement Factors are presented in Table 2.1 and Table 2.2, respectively.

Table 2.1. Analytical expressions available in the literature for the Deformation Factor of a droplet/bubble.

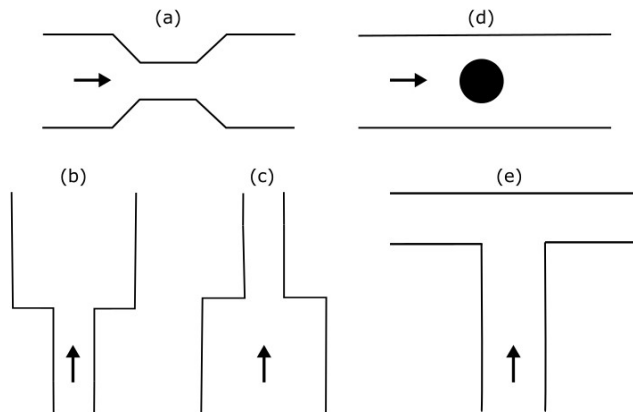
Deformation Factor	Comments	References
$\frac{L - B}{L + B}$	Taylor deformation factor: L : maximum length of the droplet/bubble during the deformation process; B : minimum length of the droplet/bubble during the deformation process.	[25]
$\frac{L}{D}$	L : maximum length of the droplet/bubble during the deformation process; D : un-deformed droplet/bubble diameter.	[26]
$\frac{P}{2\sqrt{\pi A}}$	P : imaged perimeter of the droplet/bubble; A : imaged area of the droplet/bubble.	[27]

Table 2.2. Analytical expressions that are available in the literature for the Confinement Factor of the microchannel.

Confinement Factor	Comments	References
$\frac{A_D}{A_H}$	A_D : cross-section of the un-deformed droplet/bubble; A_H : cross-section of the constricted channel.	[28]
$\frac{D-d}{D}$	D : diameter of the droplet/bubble; d : diameter of the constricted channel.	[29]
$\frac{r}{r_H}$	r : un-deformed droplet/bubble radius; r_H : hydraulic radius of the constricted channel, $r_H = \frac{W_c H}{W_c + H}$ (W_c is the width of the constricted channel, and H the height of the constricted channel).	[27]

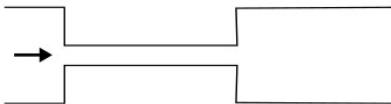
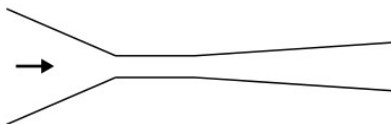
2.2.2. Microfluidic elements

Microfluidic elements can exist in many forms, and so, a wide variety of geometries and arrangements have already been designed (see Figure 2.1). The most common types are constrictions (Figure 2.1a), expansions (Figure 2.1b), and contractions (Figure 2.1c). Other cases of interest involve the study of the droplet/bubble dynamics through obstacles (e.g., cylinders, squares) in microchannels (Figure 2.1d), and in T-junction channels (Figure 2.1e).


Figure 2.1. Schematic representation of the most common microfluidic elements: (a) constriction; (b) expansion; (c) contraction; (d) obstacle; and (e) T-junction.

Within constrictions, the number of possible arrangements is high, and Table 2.3 summarizes the main types of constrictions that are frequently reported in the literature.

Table 2.3. Constriction channel geometry designs.

Constriction Geometry	Design Purpose	References
Straight 	Droplet breakup	[30]
Tapered/ conical 	Droplet breakup	[27]
Sinusoidal/ cosine 	Oil trapping, droplet breakup	[31]
Double- bend 	Provides deformation and rotation	[25]
Saw-tooth- shaped 	Directional transportation by periodic half-open capillary channels	[32]
Hyperbolic channel 	Capsule dynamics, encapsulate droplets	[33]
Shark teeth 	Provides rapid deformation	[34]

2.2.3. Operational and geometrical parameters

2.2.3.1. *Non-dimensional parameters*

When passing through a microfluidic element, a bubble/droplet deforms and changes the dimensions. The dimension of the deformed bubble/droplet, L_{de} , depends on: the physical properties of the continuous (denoted by the index c) and the dispersed (index d) phases, namely their density, ρ , and viscosity, μ , as well as on the surface tension between both of the phases, σ ; velocity of the continuous phase, U_c ; geometrical parameters describing the bubble/droplet, given by its length (before deformation), L ; and, widths of the channels in different regions, w_1 e w_2 . When working with microfluidic devices, the interfacial forces become more prominent than gravitational forces, which tend to be negligible. For this reason, the acceleration due to gravity does not influence the dimension of the deformed droplet and will not be considered in the following dimensional analysis.

Dimensional analysis, which was based on the Buckingham theorem, leads to the following relation for the bubble/droplet deformation:

$$\frac{L_{de}}{D} = f\left(\frac{\rho_d}{\rho_c}, \frac{\mu_d}{\mu_c}, \frac{U_c \mu_c}{\sigma}, \frac{w_1 \rho_c U_c}{\mu_c}, \frac{w_2}{w_1}, \frac{L}{w_1}\right) \quad (2.1)$$

This relation is equivalent to say that the deformation, L_{de}/D , is a function of the density ratio, ρ_d/ρ_c , viscosity ratio, $X = \mu_d/\mu_c$, Capillary number of the continuous phase, $Ca_c = U_c \mu_c / \sigma$, Reynolds number of the continuous phase, $Re_c = w_1 \rho_c U_c / \mu_c$, and geometrical ratios w_2/w_1 and L/w_1 . Thus, Equation 2.1 can be rewritten as:

$$\frac{L_{de}}{D} = f\left(\frac{\rho_d}{\rho_c}, \frac{\mu_d}{\mu_c}, Ca_c, Re_c, \frac{w_2}{w_1}, \frac{L}{w_1}\right) \quad (2.2)$$

The density ratio has been shown to have a negligible effect on the velocity of Taylor drops at the microscale [35], and its effect is not usually studied in microchannels. Based on the previous analysis, Table 2.4 summarizes the main dimensionless groups that characterize two-phase flows in small channels and their relevance.

Table 2.4. Main dimensionless numbers that govern two-phase flows in microscale.

Non-Dimensional Parameters		Function
Reynolds number of the continuous phase (Re_c)	$Re_c = \frac{w_1 \rho_c U_c}{\mu_c}$	Ratio between inertial and viscous forces; Definition of the flow regime of the continuous phase and transportation process optimization; It is independent of the properties of the dispersed phase.
Capillary number of the continuous phase (Ca_c)	$Ca_c = \frac{U_c \mu_c}{\sigma}$	Ratio between shear stress and surface tension; It is the most widely used non-dimensional parameter in a droplet squeezing process; Multiphase flow parameter deciding co-flow behavior [36, 37], flow regime [38], and passing/stuck [39].
Viscosity ratio between dispersed and continuous phases (X)	$X = \frac{\mu_d}{\mu_c}$	—

In spite of the aforementioned, several other dimensionless groups may arise from the recombination of the main dimensionless groups (Table 2.5). For example, the Weber (We) and Laplace numbers (La) result from the combination between Reynolds (Re) and Capillary numbers (Ca). We can also mention the Deborah number (De), which is a useful non-dimensional parameter when working with viscoelastic fluids.

Table 2.5. Dimensionless numbers sometimes used when working with two-phase flows.

Non-Dimensional Parameters		Function
Weber number (We)	$We = \frac{\rho U^2 w_1}{\sigma} = Re \times Ca$	Ratio between inertia and surface tension; Multiphase flow parameter useful in analyzing droplets formation and thin-film flows.
Laplace number (La)	$La = \frac{\rho \sigma w_1}{\mu^2} = \frac{Re}{Ca}$	It is independent of external dynamics parameters, such as velocity, and only includes the properties of the fluids and geometric characteristics of the channel.
Deborah number (De)	$De = \frac{\tau_c}{\tau_p}$	Rheology parameter quantifying viscoelastic effects [40]; τ_c is the relaxation time and τ_p the time of observation.

2.2.3.2. Pressure

Pressure is an operational parameter that is used to control the flow of the dispersed phase through the continuous phase [41, 42]. The critical pressure is a particularly relevant value, since it is defined as the maximum extra pressure that is required to squeeze a bubble/droplet through a microfluidic element (usually a constriction). This critical pressure can be described by the Young-Laplace equation, which relates the capillary pressure difference (ΔP), i.e., the pressure difference over an interface between two fluids, the surface tension, σ , and the principal radii of surface curvature, R_1 and R_2 :

$$\Delta P = \sigma \left(\frac{1}{R_1} + \frac{1}{R_2} \right) \quad (2.3)$$

For a spherical bubble flowing in a circular narrow channel, Equation 2.3 can be rewritten as:

$$\Delta P = \frac{2\sigma \cos \theta}{R} \quad (2.4)$$

where R is the channel radius, and θ the contact angle.

The Young-Laplace equation is the most widely used design criterion in two-phase flow microfluidics, being utilized to define the interfacial equilibrium between two static fluids (e.g., capillary surface). However, its application has some limitations: (i) it is only valid in quasi-static cases (non-static situations may result in sharper curvature or backflow) [43]; (ii) in systems involving non-Newtonian or viscoelastic fluids, since the effect of viscosity variation is not taken in consideration [44]; (iii) it is not valid in the presence of non-uniform stresses; and, (iv) wetting behavior is not accounted for when dealing with channels with sharp corners [45].

The pressure drop along the constricted channel is no longer linear due to the existence of the interface (Figure 2.2). The pressure drop along the constriction results from the contribution of three different factors: (i) the major pressure loss determined by Hagen-Poiseuille law, which linearly increases with the velocity of the phases; (ii) the minor pressure loss due to the contraction and to the expansion, which exhibits quadratic growth as a function of the velocity of the phases; and, (iii) the surface tension term, which dynamically changes with the droplet movement along the channel [42].

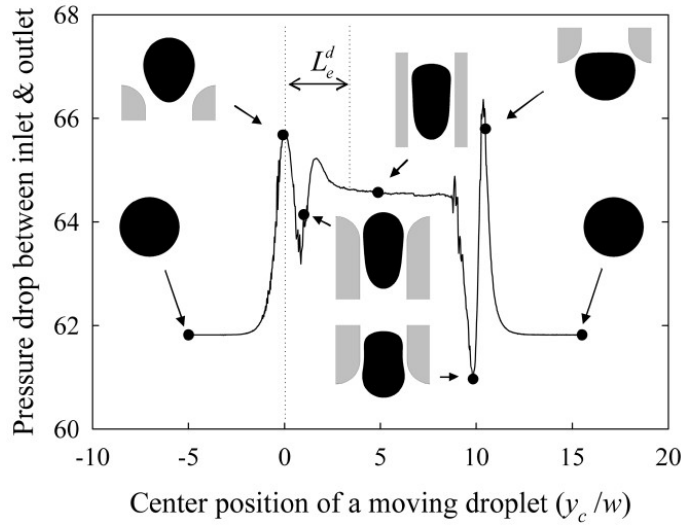


Figure 2.2. Pressure drop in a constriction between inlet and outlet with $X = 1$ and $Ca = 0.1$. L_e^d is the entry length (i.e., the moving distance of the droplet since it enters the constriction until the pressure becomes constant), w the constriction microchannel width, and y_c the center position of the moving droplet along the microchannel. Reproduced with permission from [46].

2.2.3.3. Transit time

The transit time (also called passage time or residence time) is the time that the bubble/droplet takes to move from the inlet to the outlet of a channel, and it is inversely proportional to the average velocity in the constricted channel [47]. This parameter can be calculated by the analytical expressions that are presented in Table 2.6. The transit time of a bubble/droplet passing through a constriction is influenced by the physical properties of continuous and dispersed phases, and by the bubble/droplet interactions with the confinement walls, by means of collision and friction [42, 48]. Accordingly, it is dependent on three types of parameters: operational conditions (pressure, velocity, or Capillary number), physical properties (contact angle, surface tension, dynamic viscosity, or shear modulus), and structures (channel length, confinement radius, or height of the confinement) [42, 49, 50]. The dependence on the physical properties of the fluids can be used to evaluate the stiffness and viscosity of the dispersed phase [42, 51].

Table 2.6. Analytical expressions available in the literature for the transit time of a bubble/droplet passing through a microfluidic element.

Transit Time (t_p)	Comments	References
$t_p = \frac{V_D + V_C - 2V_0}{\bar{U}A_C}$	V_D : volume of the droplet; V_C : volume of the constricted channel; V_0 : volume of the droplet inside the constricted channel at initial contact; $\bar{U}$ average flow velocity in the constricted channel: $\bar{U} = \frac{1}{A_C} \int_0^R U 2\pi r dr$ (R is the radius of the constricted channel); A_C : area of the constricted channel.	[47]
$t_p \propto (Ca - Ca_{crit})^{-1/3}$	Flow-induced droplet squeezing through lattice of spheres; Ca : Capillary number; Ca_{crit} : Capillary number below which trapping occurs.	[52]

2.3. Experimental studies

In the last two decades, many microdevices, comprising several types of the aforementioned geometrical elements, have been designed and fabricated, due to a large development of microfabrication technologies. Some have been specifically designed to promote two-phase flows with different flow patterns and characteristics. For example, as the fluids flow through a microfluidic element, it might develop vortexes at sharp corners, leading to flow pattern and topological changes, and irreversible pressure losses. Hence, two-phase flows in the basic microfluidic elements need to be deeply understood to prompt new innovations in microfluidic devices for bubble/droplet transport, control, breakup, and coalescence. Several experimental studies involving four types of microfluidic elements – namely, expansions, constrictions, T-junctions, and obstacles – have been reported in the literature and will be discussed in this section.

2.3.1. Expansions

Expansions are often used to promote coalescence in microchannels. The coalescence process is usually divided into three stages: the acceleration and elongation of the trailing bubble/droplet; the trailing bubble/droplet catching up with the leading one; and, drainage, thinning, and rupture of the thin film between the two bubbles/droplets [53].

Mazutis and Griffiths [54] presented a microfluidic approach to selectively control droplet coalescence in an expansion section. Several types of oils (with and without surfactant) were used as continuous phase, and pure water or solutions of water-sodium chloride, water-hydrochloric acid, and water-sodium phosphate as dispersed phase. The authors found that the coalescence efficiency depends on the contact time between droplets and the interfacial surfactant coverage of the droplets. This process was independent of the Capillary number, coalescence angle, nature of dispersed and continuous phases, and type of surfactants [54].

Fu *et al.* [55] studied bubble coalescence in non-Newtonian fluids in a microfluidic expansion. Two different cases of bubble coalescence were observed: (i) the coalescence of two different bubbles along the expansion section; and, (ii) at the entrance of the expansion section, one bubble breaks into two different ones, and then the two daughter bubbles recombine. The transition between both cases can be controlled by changing the gas and liquid flow rates. In both cases, three different mechanisms for bubble coalescence were observed: in-line equal-sized bubbles coalescence, in-line unequal-sized bubbles coalescence, and parallel unequal-sized bubbles coalescence.

2.3.2. Constrictions

Unlike numerical works (Subsection 2.4.1), experimental studies involving constrictions are scarce. Only a few experimental studies have been made to analyze the process of a bubble/droplet squeezing through this kind of microfluidic element.

Harvie *et al.* [56] combined experimental and numerical methods to study the deformation of a viscoelastic droplet passing through a microfluidic cylindrical 4:1:4

constriction. This work is a very good example of a study that uses experimental data to assess the numerical results. Two-dimensional (2D) simulations were compared with the experimental results for a three-dimensional (3D) planar constriction. The authors observed a good agreement between experimental and numerical results. Particularly, a forked tail inside the constriction was observed in both of the methods. However, the simulations were unable to predict the encapsulation, observed in the experiments, of the continuous phase after passing the constriction. The authors attributed this problem to an inadequate mesh resolution.

Chai *et al.* [57] experimentally studied the effect of geometry on pressure drop and on two-phase (nitrogen gas and deionized water) flow patterns. They studied one straight microchannel and two microchannel comprising alternated expansions and constrictions. They observed slug flow, annular, or single-phase liquid flow patterns, depending on the flow rates of both phases. The authors applied a homogeneous flow model (i.e., single-phase flow equation, with suitably averaged properties [58]) and a separated flow model (i.e., the two-phase are artificially separated into two streams – Lockhart–Martinelli correlation [59]) to predict pressure drop. The separated flow model predicted the experimental pressure drop better.

2.3.3. T-junctions

Among experimental works involving microfluidic elements, the great majority were carried out while using T-junctions. Droplet/bubble formation in this type of configurations has been widely studied by theoretical analyzes, experimental observations, and numerical simulations [60-67]. Furthermore, T-junctions are also useful to study the coalescence and breakup mechanisms. In this subsection, focus will only be placed on experimental works that involve the study of coalescence and breakup. Droplet formation will not be analyzed, since it is a subject outside the scope of the present review.

2.3.3.1. Droplet coalescence

Microdroplets are frequently used in microfluidics as reactors to assure the confinement of reactions and respective products, and the digitalization of chemical

processes. In digitalized microdroplet processes, the reactants are mixed through microdroplets coalescence. For this reason, geometrical elements that promote coalescence have been developed, and they are frequently reported in literature describing practical applications.

Christopher *et al.* [68] experimentally studied the effect of collision speed and Capillary number on the coalescence of droplets in a microfluidic T-junction. The working fluids were water-glycerol solutions (droplets) and silicone oil (continuous phase). The authors observed that, for two droplets to coalesce, the collision speed must be low enough, since droplets colliding at high speeds interact but do not merge, or one droplet can split the other into two smaller droplets. At low Capillary numbers, the droplets coalesced over the entire range of droplets sizes considered, and a critical Capillary number (Ca_{crit}) was found at 0.02 (i.e., for $Ca_{crit} > 0.02$ droplets do not coalesce).

Wang *et al.* [69] studied microdroplet coalescence while using water and alcohol as working fluids and two types of junctions: T-shape and Y-shape junctions with different collision angles (60° to 180°). As visible in Figure 2.3, after droplets contact (Figure 2.3b), two different possibilities may occur: the two droplets may coalesce (Figure 2.3d) or separate (Figure 2.3f). On one hand, the time interval from the contact of two droplets to their fusion is mainly affected by droplet size and the physical properties of the working fluids. On the other hand, the time interval from the contact of two droplets to their complete separation is strongly dependent of the droplet collision angle. It was also shown that another way to enhance the microdroplet coalescence was by using lower droplet collision angles.

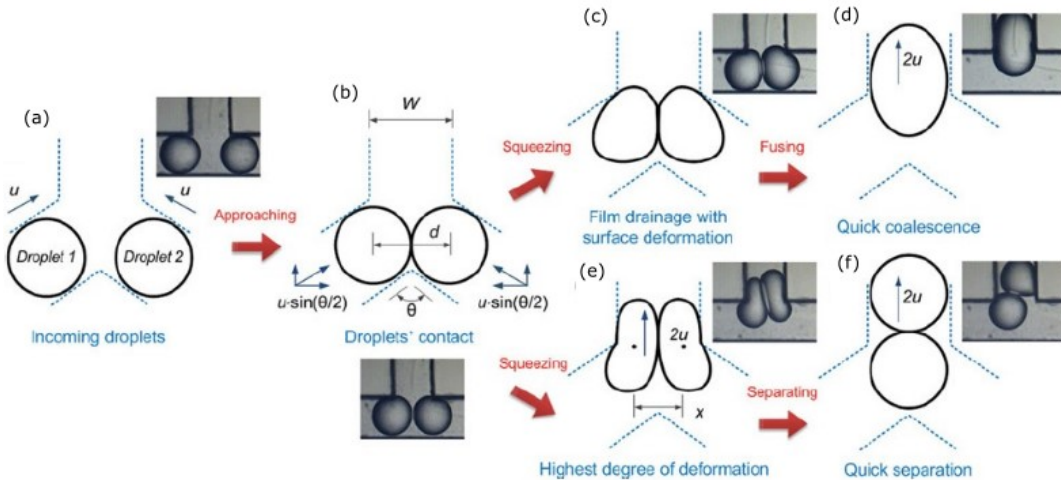


Figure 2.3. (a) droplet flow; (b) droplets contact; (c) squeezing; (d) fused droplet; (e) strongest deformation; and (f) separated droplets. Reproduced with permission from [69].

2.3.3.2. *Droplet and bubble breakup*

At a T-junction bifurcation, a single droplet or bubble may not break, leaving through one branch of the bifurcation (Figure 2.4a), or break into two smaller ones (Figure 2.4b). The outcome depends on the droplet/bubble initial aspect ratio and the strength of the deformation that was induced by the collision between the droplet/bubble and the channel walls. The breakup in symmetrical T-junctions is normally used to improve the production rate of bubbles or droplets. When the two bifurcation branches do not have the same width, the T-junction is called asymmetrical. Asymmetrical T-junctions are used to further manipulate the size of bubbles or droplets.

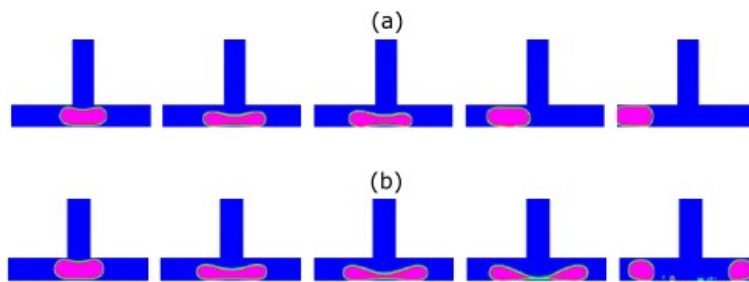


Figure 2.4. Droplet shapes from two-dimensional (2D) numerical simulations with time progressing from left to right in a symmetric T-junction: (a) non-breaking droplet; and (b) breaking droplet. Reproduced with permission from [15].

Link *et al.* [70] employed a T-junction to passively break a droplet into two daughter droplets. The flow could be tuned to induce symmetric or asymmetric breakups, resulting in daughter droplets of equal or unequal sizes, respectively. The device can be used to obtain droplets with a specific average size and size distribution. Link *et al.* [70] also presented an analytical model to explain the transition between breaking and non-breaking in terms of the stretching of droplet in an extensional flow, based on the classical Rayleigh–Plateau instability (i.e., a liquid cylinder under the influence of surface tension is unstable if its length is as long as its base perimeter).

Leshansky and Pismen [15] developed a 2D theory for droplet breakup in a T-junction that was driven by pressure decrement in a narrow gap. The authors found that the normalized critical droplet extension, l/w (l is the deformed droplet length and w the width of the microchannel), depends on the Capillary number. The authors argued that the squeezed neck of a droplet shows an arc shape, and the dependence of l/w on the Capillary number was $l/w \sim Ca^{-1/5}$, with these findings being in good agreement with a previous study conducted by Link *et al.* [70].

Garstecki *et al.* [71] and Jullien *et al.* [16] experimentally studied the mechanism of droplets breakup in a microfluidic T-junction at small Capillary numbers: $0.00008 < Ca < 0.008$, and $0.0003 < Ca < 0.27$, respectively. Garstecki *et al.* [71] found that the dynamics of breakup are dominated by the pressure drop across the droplet as it passes through the T-junction, and not by shear stresses. When the droplet fills almost the entire cross-section of the channel, this pressure drop results from the high resistance to the flow of the continuous phase in the thin film separating the droplet from the microchannel walls. Jullien *et al.* [16] studied two liquid-liquid systems (fluorinated oil droplets in deionized water and water droplets in hexadecane) over a wide range of flow rates and fluid viscosities, and they observed three different behaviors: non-breakup, breakup with tunnelling (there is a visible gap between the droplet and the walls before breakup), and breakup with permanent obstruction (droplets keep obstructing the T-junction before breakup).

Fu *et al.* [72] and Lu *et al.* [73] conducted too studies similar to [16] for gas-liquid systems. Fu *et al.* [72] studied the breakup regime map of nitrogen bubbles in glycerol-water solutions, in the presence of a surfactant, for Capillary numbers ranging between 0.001 and 0.1. By changing the gas and the liquid flow rates, the authors observed four different flow patterns: breaking with permanent obstruction, breaking with partial

obstruction, breaking with permanent gaps, and non-breaking. The experimental results suggest that the bubble breakup in a T-junction is similar to the droplet breakup [15, 16, 70, 71]. Lu *et al.* [73] analyzed the evolution of the gas-liquid interface (nitrogen-turpentine oil) during the breakup of bubbles, and observed the same four patterns. The breakup was irreversible and fast when the normalized minimum width of the bubble neck was less than the critical value (0.5-0.6); otherwise, the breakup was reversible and slow.

Wang *et al.* [74] and Ziyi *et al.* [75] studied the partial obstructed breakup of bubbles in microfluidic junctions. Wang *et al.* [74] used an asymmetric T-junction, and they concluded that this process can be divided into three stages: squeezing, transition, and pinch-off, according to the evolution of the minimum width of the bubble neck, w_m . This evolution can be described by a power-law relationship between w_m and time. It was also observed that the critical neck width of the breaking bubble is related to the Capillary number ($0.0004 \leq Ca \leq 0.006$) and the length of the original bubble ($2.74 \leq l/w \leq 4$). Very recently, Ziyi *et al.* [75] reported a study about the partially obstructed breakup of bubbles in microfluidic Y-junctions. In Figure 2.5, it is possible to see the effects of the channel angle (30° to 150°), Capillary number, and bubble length on the bubble rupture process. The authors found that the curve of the bubble neck during the breakup is nearly a parabola, rather than the aforementioned circular arc that was proposed by Leshansky and Pismen [15].

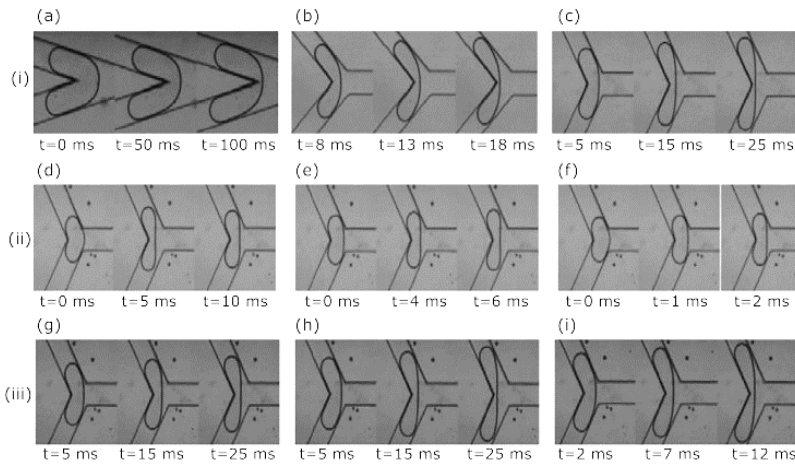


Figure 2.5. (i) Effect of the channel angle on bubble breakup: (a) 30° ; (b) 90° ; (c) 150° ; (ii) Effect of the Capillary number, Ca , on bubble breakup: (d) $Ca = 0.021$; (e) $Ca = 0.025$; (f) $Ca = 0.029$; (iii) Effect of the bubble length, L , on bubble breakup: (g) $L = 1.03$ mm; (h) $L = 1.24$ mm; and (i) $L = 1.42$ mm. Adapted with permission from [75].

In a study similar to the one conducted by Ziyi *et al.* [75], Chen *et al.* [76] analyzed nitrogen-water two-phase flow splitting at microchannel junctions with different branch angles (30° to 150°). The test range covered slug, slug-annular, and annular flow patterns. The authors found that the phase split at the junction depends on the flow pattern at the inlet. For slug flow at the inlet, the dominant phase in both outlet branches is the gas phase. On the other hand, for annular flow at the inlet, the dominant phase at the outlet branches is the liquid phase. Furthermore, for all types of inlet flow patterns, the fraction of liquid taken off through the branches (i.e., the ratio between the liquid mass flow rate of the branches and the liquid mass flow rate at the inlet) did not decrease with an increasing branch angle.

2.3.4. Obstacles

As aforementioned, the presence of obstacles in microchannels constitutes another relevant geometrical element, and some important experimental studies that are related with their interaction with flowing droplets can be found in literature.

Protière *et al.* [77] studied the breakup of long viscous droplets passing through a circular obstacle partially blocking the microchannel. The authors observed that, when a droplet moving above a critical velocity collides with an obstacle, it breaks into two uneven droplets. Below the critical velocity, the droplet only bypasses the obstacle through one of the gaps. Therefore, for a viscosity ratio equal to 8 ($X = 8$), droplet will break for Capillary numbers above 0.02.

Zaremba *et al.* [78] studied, experimentally and numerically, the passage of a droplet through a groove. Hexadecane and surfactant were used as continuous phase, and distilled water colored with ink as the dispersed phase. The numerical simulations were carried out while using the Volume of Fluid (VOF) methodology implemented in ANSYS Fluent[®] (ANSYS, Inc., Canonsburg, PA, USA). The contact angle was set to 180° (i.e., the droplet did not touch the walls of the microchannel). The coupling of experimental and numerical techniques provided a detailed description of the interactions between the two immiscible liquid phases that were altered by the geometry of the groove. The droplet divides into two parts in both experimental and numerical studies, with one of them being immobilized before the groove.

2.4. Numerical studies

Numerical modelling can play a crucial role on increasing researchers' ability to understand complex two-phase flows in microfluidic elements. Modelling must deal with interactions between bubbles/droplets of the dispersed phase, between the dispersed phase and the continuous phase, and between the dispersed phase and the walls of the microfluidic elements. Numerical methods must be able to properly track the interface to fulfil these objectives, including the evolution of the contact line at the wall surface. The main outputs of the numerical modelling are the evolution of the shape of the dispersed and continuous phases, and the velocity and pressure fields. These data may deliver important insights into optimum design of microfluidic devices.

In numerical simulations of two-phase flows, the interface tracking methodologies applied must describe interface shape evolution with high level of detail. Additionally, the appearance of small gaps between the dispersed phase and the microchannel wall requires high local mesh densities, which is also numerically challenging. Wörner has also discussed these issues [79]. This author compared different numerical methods that were implemented in commercial and open-source software that have already been used to study this kind of processes, and identified the advantages and benefits of the numerical approaches that are available to the research community. This section is focused on the main findings of numerical works involving two-phase flow in the main types of microfluidic elements (i.e., constrictions, contractions, T-junctions, and obstacles).

2.4.1. Constrictions

As aforementioned, constrictions are one of the most common examples of microfluidic elements, and there is a large number of numerical studies that involve this geometrical element. Furthermore, these works regard systems with Newtonian fluids and non-Newtonian fluids, so, they are addressed below separately since the subjacent principles are different.

2.4.1.1. Newtonian fluids

Harvie *et al.* [30, 56, 80] studied droplet deformation in a 4:1:4 axisymmetric microfluidic constriction applying the volume-of-fluid method. The authors analyzed the effect of Reynolds number (Re) and surface tension strength – $S = 1/(We + Ca)$ – on droplet deformation while using low viscosity Newtonian droplets and a high viscosity Newtonian continuous phase (viscosity ratio, X , equals 0.001) [80]. For high surface tension strength, droplets break into slugs within the constriction (Figure 2.6a). For low surface tension strength and moderate Reynolds number, the droplets form thin filaments, and remain with this shape after passing the constriction (Figure 2.6b). When surface tension strength is small and the Reynolds number is low, the droplet assumes the form of a “string of sausages”, due to the influence of large amplitude instabilities on the droplet surface (Figure 2.6c).

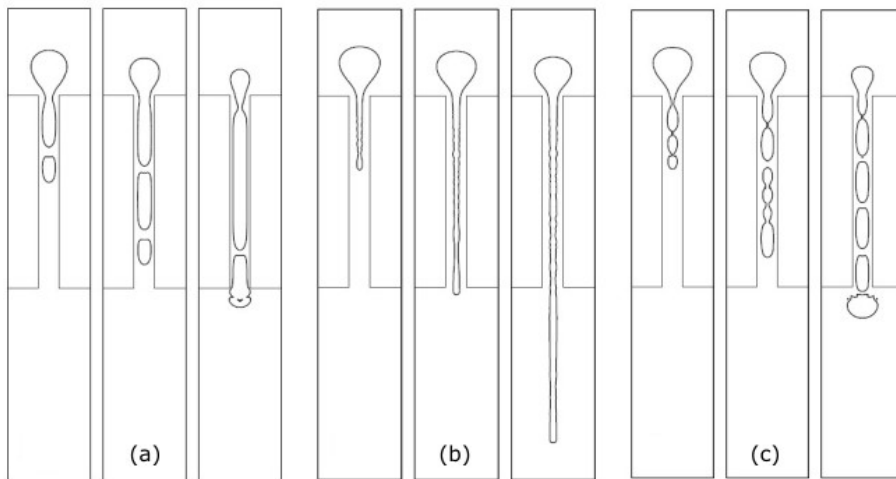


Figure 2.6. Low-viscosity droplet deformation behavior in constrictions for Newtonian fluids with $X = 0.001$: (a) $Re = 2.13 \times 10^{-3}$ and $S = 2.13 \times 10^1$; (b) $Re = 6.59 \times 10^{-1}$ and $S = 3.97$; and (c) $Re = 6.76 \times 10^{-3}$ and $S = 6.71$. Adapted with permission from [80].

Park and Dimitrakopoulos [81] studied the passage of elastic capsules and isolated droplets through constrictions. The elastic capsules had a spherical undisturbed shape and an elastic membrane enclosing a Newtonian fluid with a viscosity $\lambda\mu$, with μ being the viscosity of the continuous phase and λ the viscosity ratio between phases. The authors studied a range of viscosity ratios between 0.01 and 5 ($0.01 < \lambda < 5$) and a

range of Capillary numbers between 0.01 and 0.1, for both capsules and droplets. In this numerical study, an interfacial spectral boundary element (SBE) method for membranes, as developed by the same authors [82], was used to describe the movement of the elastic capsule. The authors utilized their fully-implicit time-integration spectral boundary element algorithm [83], as well as their membrane spectral boundary element algorithm by imposing surface-tension interfacial conditions, to determine the droplet dynamics [84]. The numerical results show that both droplets and capsules assume an elongated shape with a maximum length and a minimum height in the contraction region, changing to a flattened parachute in the expansion region. Therefore, the deformation of the droplet/capsule is only related to the hydrodynamic effects that were introduced in the flow by the constriction [81]. However, while the droplet velocity consistently decreases with decreasing viscosity ratios, the capsule velocity is insensitive to the viscosity ratio. Nevertheless, the behavior of capsules (regardless of the viscosity ratio) corresponded better to that of very viscous droplets.

Zhang *et al.* [43, 45] developed two relevant numerical work, where the deformation of a droplet passing through a microfluidic constriction was computed while using the VOF methodology, implemented in the commercial software ANSYS Fluent[®]. The authors studied the influence of channel geometry (e.g., circular, square, rectangular, and triangular) on the pressure field and droplet deformation. On one hand, for circular, square, and triangular channels, the pressure starts to increase before the droplet enters the constriction, and then decreases after optimum inlet velocity. On the other hand, for rectangular channels, the authors did not verify the existence of an optimum velocity, and the pressure continuously decreases with the increase of the inlet velocity [43]. Zhang *et al.* [45] presented analytical expressions to calculate the critical pressure on the four mentioned cross-sectional shapes. The circular cross-section provided the highest critical pressure and, in decreasing order, being followed by the square, triangular, and rectangular channels. Additionally, it was shown that the use of a rectangular cross-section with a high aspect ratio might result in droplet breakup at high-speed values.

Zhang *et al.* [47] also developed an analytical model to predict the pressure that is required to squeeze a viscous droplet ($1 < X < 50$) through a circular constriction. The model is based on five assumptions: (i) low Reynolds number; (ii) axisymmetric geometry; (iii) elasticity, wall-adhesion, and bending elasticity of the droplet

negligible; (iv) one-phase in the radial direction; and, (v) spherical droplet in the curvature change part. For low Reynolds ($Re = 1.8$) and Capillary ($Ca = 0.0072$) numbers, an analytical model was able to accurately predict the squeezing pressure obtained numerically by solving the Navier-Stokes equations.

2.4.1.2. Non-Newtonian fluids

Khayat [85] presented a 2D numerical study of two-phase flow of viscoelastic fluids in a hyperbolic convergent-divergent constriction, while using the Boundary Element Method (BEM) and the linear Oldroyd-B constitutive model. The droplet and the continuous phase were assumed to be incompressible, with a viscosity ratio equal to 3. The author concluded that, under small deformation rates, the fluid elasticity tends to enhance the droplet deformation. In a subsequent paper, Khayat *et al.* [86] examined the effects of shear and elongation on a droplet deformation in the same geometry. As in the previous study, the flow was simulated while using the BEM method. Both, the dispersed phase (droplet) and the continuous phase, could be either Newtonian or Maxwell fluids. The droplet deformation is different in these two cases: a high-viscous Newtonian droplet slowly deforms at the beginning, but then the process accelerates; a viscoelastic droplet initially deforms quickly, but then the deformation rate tends to slow down.

More recently, the contributions to the understanding of the dynamics of non-Newtonian droplets in microfluidic elements have increased, with several numerical studies being performed on axisymmetric channels [30, 87] and on 2D planar constrictions [24, 56].

Harvie *et al.* [30] focused on two types of liquid-liquid systems flowing through constrictions: droplets of a shear thinning fluid dispersed on a Newtonian fluid and droplets of a Newtonian fluid being dispersed on a shear thinning fluid. In both cases, the Carreau model described the viscosity of the shear thinning fluid. In the first case, the authors studied low-viscosity Newtonian continuous phases ($10^{-3} < Ca < 10^3$) and observed that the droplets deformation was similar to that of Newtonian droplets with a viscosity that was equal to the average viscosity of the shear thinning liquid. In the second case, the authors observed that droplet deformation was smaller than in the first case. Harvie *et al.* [56] also experimentally studied the deformation of a

viscoelastic droplet surrounded by a low-viscosity Newtonian liquid flowing through a thin 3D planar constriction. The viscoelastic effects were described by the Oldroyd-B model. The results were compared with the numerical results obtained for a 2D constriction (already mentioned in Subsection 2.3.2). The numerical model employed captures the deformation process well, with special emphasis on the forked tail that is characteristic of a viscoelastic droplet inside a constriction. The authors also observed the presence of an elastic stress inside the droplet, which might cause the entrainment and subsequent encapsulation of continuous phase fluid within the droplet as it leaves the constriction.

Chung *et al.* [24, 46, 88] studied the effect of viscoelasticity (Deborah number between 0.4 and 2.2) on droplet dynamics in a 5:1:5 planar constriction while using the finite element-front tracking method (FE-FTM). Again, the Oldroyd-B model described the viscoelastic fluid. According to the fluids' characteristics, two different types of deformation were observed [24]. On one hand, a Newtonian droplet in a viscoelastic continuous phase results in an "ellipsoid-like drop", and, on the other hand, a Newtonian droplet and a Newtonian continuous phase results in a "bullet-like drop". In a subsequent paper, the same authors investigated the excess of pressure (Δp^+) that is required to move the droplet through the constriction [46]. The excess of pressure was roughly proportional to the viscosity ratio (X), and inversely proportional to the Capillary number (Ca). The excess of pressure was strongly linked to the film thickness (δ_f) between the interface of the droplet and the walls in the thinner region of the microchannel; this film thickness increases with the Capillary number. Thus, a decrease in the Capillary number results in a decrease in the film thickness and an increase of the excess of pressure. Chung *et al.* [88] found that droplet deformation depends on the Capillary number, with the effect being more pronounced for smaller viscosity ratios. The authors also studied the formation of bilateral symmetric vortices in the droplet and found that circulation intensity inside the droplet increases with decreasing viscosity ratios.

2.4.2. Contractions

A microfluidic contraction has a simple geometry, but the flow has strong and distinct regions of extensional and shear strains, which are able to considerably deform

the shape of a droplet. Several numerical studies have been done regarding droplet dynamics in contraction microchannels, assuring a good level of knowledge on this subject. Nevertheless, the great majority of these numerical works involve 2D models, which may present some limitations.

Khayat *et al.* [89] used a boundary element method to investigate the effect of contraction geometry, droplet initial shape, and fluids rheology on droplet deformation. The geometry of the contraction was characterized by the convergence ratio. Two convergence ratios were studied, three and ten. The authors investigated the influence of the viscosity ratio ($X = 0.5, 4, 10$) on the deformation of circular and elliptic droplets positioned upstream to the contraction. For circular droplets and $X = 0.5$, the droplet deforms at the contraction inlet, while, for $X = 10$, the droplet behaves like a rigid solid particle. Similarly, the deformation of elliptic droplets decreased as X increases, being larger due to the elongated initial shape of the droplet. As expected, for the same flow rate, the deformation was larger for the channel, with the convergence ratio equal to 10.

Christafakis and Tsangaris [25] analyzed the influence of the Capillary, Reynolds, and Weber numbers and viscosity ratio on the droplet dynamics in a 2D contraction microchannel while using the level-set (LS) method. These authors simulated systems with three or four consecutive droplets, and the droplet shapes, their deformation and migration compare well with literature data. The values of the non-dimensional numbers were chosen to avoid the breakup or coalescence of the droplets (Ca between 0.1 and 1, $Re = 20$, $X = 4$, and We between 2 and 20). The Taylor deformation factor (Subsection 2.2.1) was used to describe the geometrical state of the droplets and to evaluate their deformation.

As aforementioned, 3D models must be used when dealing with the deformation of droplets in non-circular microchannels, since the droplet curvature and the hydrodynamics is not accurately described by 2D models. Accordingly, Hoang *et al.* [38] performed 3D simulations, together with a theoretical analysis of the droplet dynamics in a planar microchannel contraction. Both phases were assumed to be incompressible Newtonian fluids, and inertial effects were neglected. A volume of fluid method implemented in the commercial software ANSYS Fluent[®] was used, and the surface tension was treated as a body force by using the continuum surface force model, to track the interface [90]. The authors identified three different regimes: trap,

squeeze, and breakup (Figure 2.7). They proposed simple theoretical models, which were found to agree well with the numerical data, in order to elucidate the transitions between each regime. Droplet deformation and retraction along of the microchannel were analyzed.

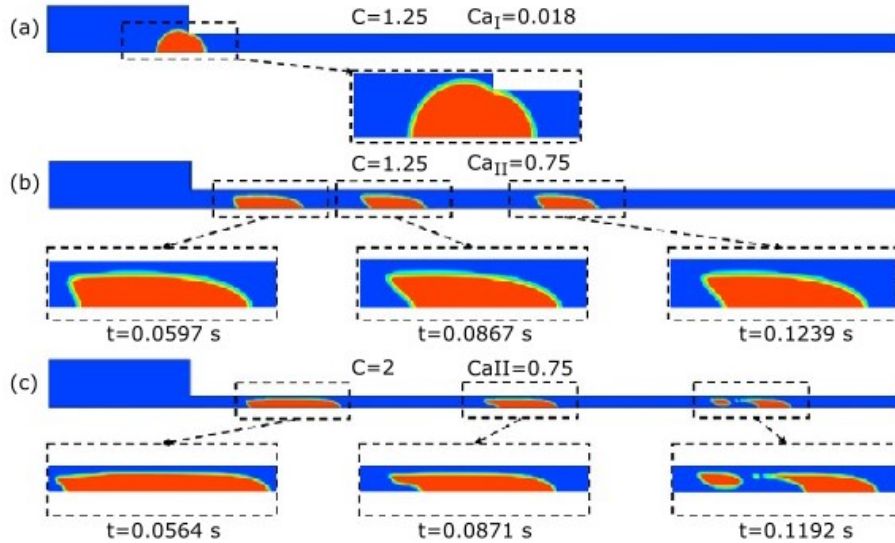


Figure 2.7. Droplet cross-section at the symmetric plane viewed from top: (a) trap regime; (b) squeeze regime; and (c) breakup regime. C is a non-dimensional number representing the contraction level ($C = D/W$, where D is the droplet diameter, and W the contraction microchannel width). In the thicker part of the microchannel (i.e., before the contraction), the inlet velocity is employed as the characteristic velocity, and the Capillary number is denoted as Ca_I , while the average velocity in the thinner region of the microchannel (i.e., after the contraction) is used to define the Capillary number Ca_{II} . Adapted with permission from [38].

2.4.3. T-junctions

The breakup mechanism of droplets on T-shaped junctions is also a topic that has been addressed by computational approaches and, hence, some numerical studies are available in the literature.

Bedram and Moosavi [91] studied the breakup of non-uniform droplets through an asymmetric T-junction while using the Volume of Fluid (VOF) methodology. The T-junction comprised one inlet and two outlets with different dimensions. The authors also developed an analytical theory in the limit of the thin-film approximation. In this

theory, for low Capillary numbers, they assumed that the pressure drop along the continuous phase mainly occurs in the thin film between the top wall of the microchannel and the droplet. While using the analytical solution, the authors calculated the smallest film thickness as a function of the Capillary number and of the ratio between the dimensions of the two outlet microchannels. A good agreement between analytical and numerical data was obtained, namely for the critical droplet length as a function of the Capillary number and for the velocity of the droplet as a function of the average velocity of the continuous phase. Smaller droplets can be obtained by increasing the Capillary number and, in this way, it is possible to decrease the breakup time and increase the pressure drop.

Mora *et al.* [23] developed a 3D numerical work about breakup dynamics of an isolated Taylor droplet in a rectangular T-junction microchannel while using a volume-of-fluid based solver from an open-source Computational Fluid Dynamics (CFD) code (OpenFOAM®). S-CLSVOF was the algorithm used in this study, which is a method that combines the VOF (Volume of Fluid) and LS (Level-set) methods. On one hand, VOF provides accurate mass conservation and, on the other hand, LS an exact representation of the interface. A new mathematical correlation was proposed for the dimensionless neck thickness (i.e., minimum width of the droplet at junction) of the droplet versus Capillary number, dimensionless time, and dimensionless droplet length. In the breakup regime, the authors identified two different behaviors: non-obstructed breakup and obstructed breakup. The first one can also be divided into two subcategories, named breakup with permanent tunnel (i.e., a gap is clearly visible between the droplet and the channel), and breakup partially obstructed.

2.4.4. Obstacles

In the literature, there are also some numerical approaches available regarding the dynamics involved in the passage of a droplet through obstacles in microchannels. Chung *et al.* [92] presented an interesting study, both numerical and experimental. The FE-FTM was employed to study the deformation of a 2D droplet for interface tracking and a calculation of interfacial tension. Oil was considered as continuous phase, and water as dispersed phase. Four different cases were studied: a droplet flowing through a confined microchannel with a single cylinder obstacle (Figure 2.8a), a single square

obstacle (Figure 2.8b), successive cylinder obstacles (not shown), and an array structure of cylinders (Figure 2.8c). The authors studied the effects of the obstacle shape, droplet size, and Capillary number (Ca) on droplet dynamics. For droplet size control, the cylinder obstacle showed better performance than the square obstacle. Low Ca droplets and enough distance between cylinders were necessary to have an independent/efficient wetting of the cylinder's surfaces. Another relevant finding was that droplet size did not play a significant role on the droplet dynamics.

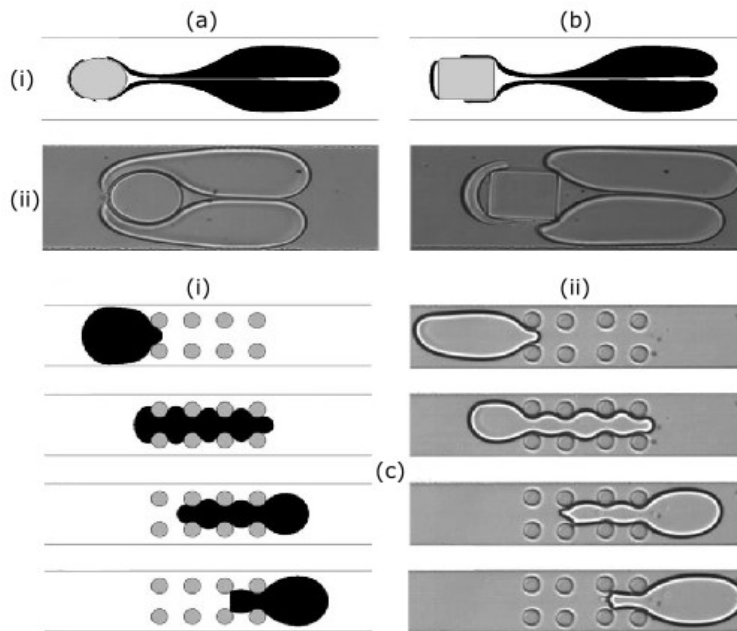


Figure 2.8. Droplet passing through the obstacles: (a) single cylinder; (b) single square; and, (c) array of cylinders; (i) numerical results; (ii) experimental results. Adapted with permission from [92].

2.5. Perspectives and challenges for the future

Several researchers have studied, from a fundamental perspective, microfluidic elements to manipulate bubbles and droplets, covering a wide range of geometries and fluid properties. However, on one hand, the experimental studies do not cover the full range of cases of interest and the numerical studies are frequently standalone studies without validation with experimental results. On the other hand, much remains to be

done to apply the results from fundamental studies to the design of new devices for practical applications.

Two-phase flows in microchannels are affected by the surface properties of the walls of the channel. The surface properties influence the contact angle when the bubble/droplet gets in contact with the wall. This effect has been neglected in the literature on two-phase flows through microfluidic elements. From an experimental perspective, the control of the surface properties at the microlevel during microfabrication procedures is required to develop devices that can manipulate droplets through the local contact angle. On the numerical perspective, the numerical methods must deal with contact angle, which is a difficult subject that requires numerical innovations and experimental validation.

Microfluidic elements can be used in practical applications, such as microparticle fabrication, chemical reactors, mixers, and particle processing and separation. Most of the main functions that are required for these applications, such as droplet breakup and coalescence, have already been studied at the element level. However, practical applications require the scale-up of the device through parallelization. New studies are needed to understand how microfluidic elements can be used to conduct large scale production. Additionally, new applications need to be identified, exploring the surface properties effects and the combined effects of different elements. For example, droplets can be split or redirected by surface patches with different hydrophobicities and two-phase flows in complex networks can be used to improve the mixing. These geometries can be optimized by computational methods, i.e., mixing, droplet size, size distribution, and pressure drop can be improved applying topological optimization.

2.6. Conclusions

In the last years, microstructured devices played an important role in the technological development of several areas, including medicine, biotechnology, cosmetics, food, materials, pharmaceuticals, and energy. Two-phase flows and, consequently, the presence of bubbles and droplets is recurring in such devices. Recently, with the rapid development of microfabrication technologies, many new elements and topologies of microstructured devices (constrictions, contractions, expansions, obstacles, or T-junctions) have been designed and manufactured. Since

these elements affect the flow, shape, breakup, and coalescence of bubbles and droplets, playing with their geometrical features can be crucial in attaining different types of goals in practical scenarios. The systematization of the data published about the flow of droplets/bubbles in such elements is the main motivation of this review. Theoretical, numerical, and experimental analysis are presented, covering gas-liquid and liquid-liquid flows, with non-Newtonian and Newtonian fluids.

The deformation of bubbles/droplets passing through a microfluidic element depends on the physical properties and flow rates of dispersed and continuous phases, and on the type of confinement involved. For a specific element, the Reynolds and Capillary numbers of both phases and the viscosity ratio are the main dimensionless numbers governing the deformation. Several analytical expressions available in the literature are presented to calculate the Deformation Factor of bubbles/droplets passing through microfluidic elements.

The most relevant experimental studies involving bubbles/droplets coalescence and/or breakup in expansions, constrictions, T-junctions, and obstacles are reviewed. Expansions are often used to promote bubbles/droplets deformation and coalescence. Symmetrical T-junctions are useful for promoting coalescence, but also breakup, in order to improve the production rate of bubbles or droplets. There are four possible flow patterns in the droplet/bubble breakup in T-junctions: breaking with permanent obstruction, breaking with partial obstruction, breaking with permanent gaps (i.e., a gap is clearly visible between the droplet and the channel), and non-breaking. Asymmetrical T-junctions are applied to further manipulate the size of bubbles or droplets. Constrictions can be used to fragment bubbles and droplets, and they have the potential to be applied in microparticle production. Obstacles can be used to produce complex two-phase flows, promoting the bubble/droplet breakup and chaotic flow in both phases. Combined in complex arrangements, obstacles could be used to promote the mixing of reactants in the continuous phase.

In numerical simulations of a droplet/bubble moving through microfluidic elements, the reliability of the interface tracking methodology applied is a key component due to the high level of detail that is required to describe the interface shape evolution. Different numerical methods are presented, such as the Volume of Fluid (VOF), the Spectral Boundary Element (SBE), the Boundary Element Method (BEM),

the Finite Element-Front Tracking Method (FE-FTM), the Level-set (LS) method, and the S-CLSVOF method.

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Nomenclature

Acronyms	Definition
2D	Two-dimensional
3D	Three-dimensional
BEM	Boundary Element Method
CFD	Computational Fluid Dynamics
FE-FTM	Finite Element-Front Tracking Method
LS	Level-set
SBE	Spectral Boundary Element
S-CLSVOF	Coupled Level-set and Volume of Fluid method
VOF	Volume of Fluid

Greek	Definition	Units
δ_f	Film thickness	[m]
ΔP	Capillary pressure difference	[Pa]
Δp^+	Excess of pressure	[Pa]
θ	Contact angle	[°]
λ	Viscosity ratio	—
μ	Dynamic viscosity	[Pa s]
μ_c	Dynamic viscosity of the continuous phase	[Pa s]
μ_d	Dynamic viscosity of the dispersed phase	[Pa s]
ρ	Fluid density	[kg m ⁻³]
ρ_c	Density of the continuous phase	[kg m ⁻³]
ρ_d	Density of the dispersed phase	[kg m ⁻³]
σ	Interfacial tension	[N m ⁻¹]
τ_c	Relaxation time	[s]
τ_p	Observation time	[s]

Latin	Definition	Units
A	Area of the droplet/bubble	[m ²]
A_C ; A_H	Area of the constricted channel	[m ²]
A_D	Area of the un-deformed droplet/bubble	[m ²]
B	Minimum length of the droplet/bubble during the deformation process	[m]

C	Contraction level	—
Ca	Capillary number	—
Ca_c	Capillary number of the continuous phase	—
Ca_{crit}	Critical Capillary number	—
Ca_I	Capillary number before the constricted channel	—
Ca_{II}	Capillary number in the constricted channel	—
d	Diameter of the constricted channel	[m]
D	Un-deformed droplet/bubble diameter	[m]
De	Deborah number	—
H	Height of the constricted channel	[m]
L	Un-deformed droplet/bubble length; maximum length of the droplet/bubble during the deformation process	[m]
$L_{de} ; l$	Deformed droplet/bubble length	[m]
L_e^d	Entry length	[m]
La	Laplace number	—
P	Perimeter of the droplet/bubble	[m]
r	Un-deformed droplet/bubble radius	[m]
r_H	Hydraulic radius of the constricted channel	[m]
R	Channel radius	[m]
$R_1 ; R_2$	Principal radii of surface curvature	[m]
Re	Reynolds number	—
Re_c	Reynolds number of the continuous phase	—
S	Surface tension strength	—
t_p	Transit time	[s]
U	Flow velocity	[m s ⁻¹]
$\bar{U}$	Average flow velocity in the constricted channel	[m s ⁻¹]
U_c	Velocity of the continuous phase	[m s ⁻¹]
V_0	Volume of the droplet inside the constricted channel at initial contact	[m ³]
V_C	Volume of the constricted channel	[m ³]
V_D	Volume of the droplet	[m ³]
w_1	Channel width before the constricted channel	[m]
$W_c ; w_2 ; w ; W$	Width of the constricted channel	[m]
w_m	Minimum width of the bubble neck	[m]
We	Weber number	—
X	Viscosity ratio	—
y_c	Center position of the moving droplet along the microchannel	[m]

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Numerical and experimental analysis of droplet dynamics in constricted milli and microchannels

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*Numerical and experimental analysis of droplet dynamics in constricted milli and microchannels***Abstract**

This study investigates the dynamics of droplets flowing through constricted channels at milli and microscales using a combination of experimental and numerical approaches. Experiments were conducted in a 3D-printed millichannel with three immiscible fluid pairs, spanning a wide range of viscosity ratios between dispersed and continuous phases ($10^{-2} < X < 10^2$) and Capillary numbers of the continuous phase ($10^{-4} < Ca_c < 10^{-1}$). Numerical simulations, employing the Volume of Fluid (VOF) methodology and performed with Ansys FLUENT® 2021 R2, complemented the experiments by exploring two geometrical scales: milli (aspect ratio: 5:3 at the inlet channel; 1:3 in the constricted region) and micro (aspect ratio: 5:1.5 before the contraction; 1:1.5 after contraction). For milliscale simulations, X ranged from 10^{-1} to 10^2 and Ca_c from 10^{-3} to 10^{-1} , while in microscale simulations, $10^{-2} < X < 10^2$ and $10^{-4} < Ca_c < 10^0$. A comprehensive analysis of velocity profiles, film thickness, and droplet deformation was conducted numerically. Strong agreement between experimental and numerical results for average droplet velocity and length within the constricted region validated the computational model's robustness. This work elucidates the influence of Ca_c and X on droplet deformation, velocity, and film thickness, providing valuable insights into the optimization of multiphase microfluidic systems.

3.1. Introduction

Simultaneous flow of immiscible liquids is a phenomenon that is widely present in many natural and engineering scenarios of different scale ranges. The great impact of this phenomenon in several fields, together with its hydrodynamic complexity, has been acting as an important driving force for a considerable amount of research. The identification and characterization of flow patterns is one of the most relevant topics in this research. For instance, in a simple geometry like a conventional pipe, the phase distribution of a liquid-liquid flow can be classified according to four main groups [1]: 1) stratified flow – both liquids are in separated layers with a well-defined interface; 2) dispersed flow – small elementary volume units (i.e., droplets) of one phase flow through a continuous phase; 3) intermittent flow; and 4) annular flow. Depending on the flow pattern, the characteristic features and hydrodynamic behavior will be quite different, which can be crucial for the performance of the processes involved.

In recent years, the use of miniaturized technologies and devices has received increased attention since it can offer several advantages compared to conventionally sized systems – such as less materials consumption (samples or reagents), quicker processes and cheaper manufacturing [2, 3]. On the other hand, on smaller scales, it should be noted that the predominance of the governing forces changes when compared to systems with conventional scales. Nowadays, this fact still implies a need for deeper knowledge to effectively control and optimize several practical applications. Particularly regarding dispersed flow pattern, manipulating droplets is the core issue of many systems and processes in multiphase flows, such as emulsions, drug encapsulation, crystallization, materials synthesis, chemical mixing and reaction [4, 5].

In fluidic channels, droplets are generated by injecting a liquid phase into another immiscible liquid, being the T-junction or a flow-focusing device the most common configurations to generate droplets [6-9]. Different flow behaviors can be observed at the cross-junction of devices for droplets generation [10, 11]: 1) viscous displacement – the dispersed phase invades the continuous phase side channels; 2) tubing – the continuous phase occupies most of the cross-section of the outlet channel; 3) dripping – the thread of the continuous phase breaks and the cap formed stays within the

focusing section; 4) jetting – the thread breaks and the cap stays in the outlet channel; and 5) threading – the thread is stable.

The shape and size distribution of droplets are within the main features to be addressed in fluidic dispersed flow. So, a good level of control of droplet deformation and breakup mechanisms can be of utmost importance in the intensification of associated phenomena like mixing, heat and/or mass transfer and reaction [12, 13]. The analysis of information about these mechanisms is usually based on a set of characteristic dimensionless numbers [14]: Reynolds (Re) and Capillary (Ca) numbers of continuous (denoted by the index c) and dispersed (index d) phases, and viscosity ratio between phases (X) – Equation 3.1:

$$\text{Re} = \frac{\rho U l}{\mu} \quad ; \quad \text{Ca} = \frac{U \mu}{\sigma} \quad ; \quad X = \frac{\mu_d}{\mu_c} \quad (3.1)$$

where ρ is the density, U the velocity, l the characteristic length, μ the dynamic viscosity, and σ is the surface tension between both phases.

Despite those groups, several others may arise from the recombination of them. For example, the Weber number (We) results from the recombination between Reynolds and Capillary numbers – $\text{We} = \text{Re} \times \text{Ca}$. Additionally, it can also be mentioned the Deborah number (De), which is a useful non-dimensional group when working with viscoelastic fluids [15].

The presence of sudden changes (i.e., accidents) in the path of dispersed flow has been proven to effectively promote droplet deformation or breakup [3, 16, 17]. Accidents inside channels, like expansions and contractions, where flow velocity varies due to the abrupt changes on the channel dimensions, affect droplet behavior significantly [16]. The increase in flow velocity due to channel contraction increases the shear force which may lead to an early breakup of the droplet into the continuous phase. So, an inappropriate design of the channel geometry can cause undesired droplet behavior. As an example, if droplets are used as drug carriers in small vessels, they should only split when the targeted site has been reached [18]. Therefore, it is highly desirable to systematically investigate and quantify the conditions for droplet breakup in complex channels, especially where expansions and contractions are involved. In the literature about constricted channels, there are three different regimes with

particular relevance in the deformation, retraction, and breakup of a droplet [19, 20]: 1) trap regime – the droplet does not deform conformably to the constricted channel, becoming immobilized as it gets trapped at the contraction; 2) squeeze regime – the droplet deforms and passes through the contraction; and 3) breakup regime – the droplet breaks up into smaller droplets inside the contraction.

Numerical approaches to droplet dynamics in constricted channels have been addressed in the literature with a good level of detail. Based on the Boundary Element Method (BEM), Khayat *et al.* [21] studied the effect of contraction geometry, viscosity ratio between dispersed and continuous phases, and droplet initial shape (circular or elliptical) on droplet deformation when passing through a contraction. The droplet and the continuous phase were assumed to be viscous and incompressible. The authors concluded that the deformation of circular and elliptical droplets decreases as the viscosity ratio increases, being more significant for elliptic droplets. Then, still focusing on a contraction geometry and using the same approach, Khayat *et al.* [15] numerically explored the effects of shear and elasticity on droplet deformation. They studied Newtonian and Maxwell fluids and verified that a Newtonian droplet slowly deforms at the beginning, but then the deformation process accelerates. The Maxwell droplet starts by deforming more quickly, but the deformation rate tends to slow down afterwards. Chung *et al.* [22, 23] applied the Finite Element-Front Tracking Method (FE-FTM) and the Oldroyd-B model to infer how viscoelasticity (Deborah number between 0.4 and 2.2) affected droplet dynamics in a two-dimensional 5:1:5 planar constriction. On one hand, a Newtonian droplet in a Maxwell continuous phase led to an “ellipsoid-shaped drop”, and on the other hand, a Newtonian droplet in a Newtonian continuous phase resulted in a “bullet-shaped drop” [22]. It was also investigated the excess of pressure that is required to move the droplet through the constriction [23]. The excess of pressure was roughly proportional to the viscosity ratio and inversely proportional to the Capillary number of the continuous phase. In an additional study, Christafakis and Tsangaris [24] analyzed the effects of some characteristic parameters in a two-dimensional contraction for the following ranges of values: 1) Capillary number of the continuous phase between 0.1 to 1; 2) Weber number of the continuous phase between 2 and 20; 3) Reynolds number of the continuous phase equal to 15 or 20; 4) ratio between droplet size and inlet size 0.24 to 0.8; and 5) viscosity ratio equal to 4. These non-dimensional numbers were chosen to avoid the breakup or coalescence of the droplets. The Level-set (LS) method was the interface tracking methodology

applied, and the simulations included the flow of three or four consecutive droplets. Droplet shapes, their deformation and migration were well compared qualitatively and visually with results available in the literature. Harvie *et al.* [2, 25] investigated droplet deformation in a 4:1:4 constricted channel with axisymmetric geometry by applying the Volume of Fluid (VOF) methodology. The effect of Reynolds number and surface tension strength, both related to the continuous phase, were considered for exploring droplet deformation. For high surface tension strength, droplets break into slugs within the constriction. The droplets form thin filaments at low surface tension strength and moderate Reynolds number, remaining with this shape after passing the constriction. When surface tension strength is small, and Reynolds number is low, the droplet assumes the form of a “string of sausages” due to large amplitude instabilities on the droplet surface [25].

Most of the numerical investigations in the publications referred previously are restricted to two dimensions or use axisymmetric geometries. However, these simulations might differ significantly from what should be observed experimentally, especially for non-circular channels. Two-dimensional models, for instance, could not adequately forecast droplet generation or deformation in a non-circular channel due to the curvature of the droplet and the hydrodynamics near the corners of the walls [26, 27]. To conduct a thorough investigation, the dynamics of droplets in such geometry should be addressed by a three-dimensional approach. A limited number of authors discussed droplet dynamics in constricted channels based on simulations in 3D domains. Zhang *et al.* [28] belong to that small group since they conducted representative three-dimensional investigations using the VOF methodology implemented in the commercial software ANSYS Fluent®. The authors studied the influence of the constriction’s cross-section shape (e.g., circular, square, rectangular, and triangular) on the pressure field and droplet deformation [28]. The circular cross-section provided the highest critical pressure and was followed by the square, triangular, and rectangular channels in decreasing order. Droplet dynamics in the transitions from trap to squeeze and from squeeze to break up in a three-dimensional contraction channel with a rectangular cross-section were investigated by Hoang *et al.* [20]. This numerical work addressed a wide range of Capillary numbers and contraction ratios and also used the VOF methodology implemented in ANSYS Fluent®. Based on a system composed by an oil droplet flowing in water, Imani *et al.* [29] explored the effects of capillarity-wettability on droplet deformation through a

constricted 3D channel with two different contact angles on opposing faces of the constriction. It was verified that the wettability of the channel greatly influences the droplet dynamics, including pressure drop, liquid-liquid interface and liquid-wall interface areas.

To the best of our knowledge, unlike numerical works, experimental studies involving constrictions are scarce. Only a few experimental studies have been made to analyze the process of a droplet squeezing through this kind of fluidic element. Harvie *et al.* [26] combined experimental and numerical methods to study the deformation of a Maxwell droplet passing through a cylindrical 4:1:4 constriction. The authors observed a good agreement between experimental and numerical results. Particularly, a droplet with a forked tail inside the constriction, which is characteristic of a Maxwell droplet in a Newtonian continuous phase, was observed in both approaches. Patlazhan *et al.* [30] also presented a study that combines numerical and experimental approaches to investigate the elongation of droplets as a function of the ratio between the initial diameter of droplets and the cross-sectional area of an abrupt contraction. These two works are good examples of studies using experimental data to assess the numerical results.

In this study, numerical simulations that replicate the flow of a single droplet through a continuous liquid phase in a three-dimensional constricted channel were performed with relevance to milli and microscales. This is a topic of high relevance for multiphase systems in chemical and biomedical processes. Despite the increasing application of droplet-based microfluidics in drug delivery, emulsification, and multiphase reaction engineering, the impact of geometric constrictions on droplet behavior, particularly in three-dimensional systems, remains insufficiently understood. Experiments were also performed in a 3D-printed constricted millichannel with different fluid pairs to investigate droplet dynamics. The use of 3D printing techniques to study the flow of a droplet through a constricted channel is new and has not yet been reported in the literature. Droplet velocity and length inside the constricted part of the channel are analyzed by comparing experimental and numerical data. Additionally, numerical simulations were used to examine the impact of Ca_c and X on droplet deformation, velocity, and film thickness.

3.2. Experimental techniques and procedures

Experiments with a 3D-printed millichannel were conducted using three different two-phase systems to study the droplet flow through a constricted channel. The physical properties of the fluids are presented in Subsection 3.2.2. The millichannel comprises a flow-focusing region for droplet generation followed by a constricted zone. The flow-focusing part is composed of two inlets, one for the continuous phase and another one for the dispersed phase, as illustrate in Figure 3.1.

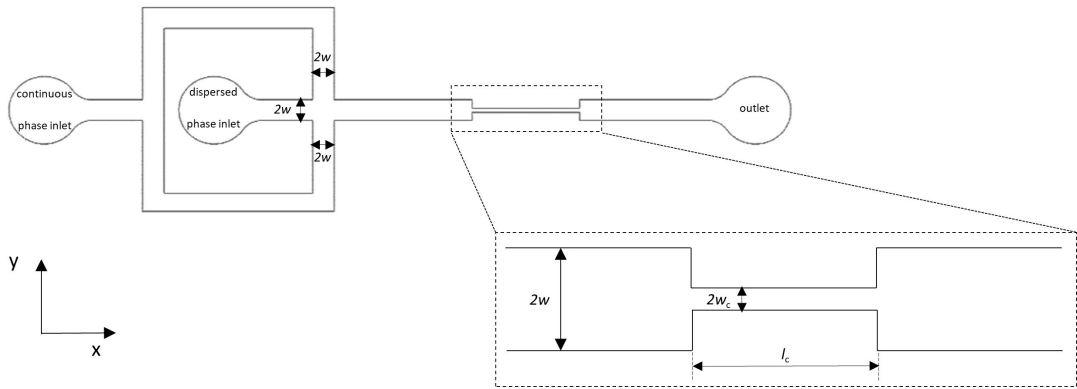


Figure 3.1. Top view of the millifluidic device with a flow-focusing region and a constricted zone. The millichannel height ($2h$) is 1.5 mm, and the other main channel dimensions are listed in Table 3.1.

3.2.1. Fabrication method of the millifluidic device

The geometry design of the millifluidic device used in the experiments was made with Autodesk AutoCAD 2022[®]. The channel was manufactured by the stereolithography 3D printing technique [31]. In this additive manufacturing process, an ultraviolet (UV) laser beam repeatedly strikes the printing material to produce the object layer by layer. The printing materials are photosensitive thermoset polymers in the liquid form. The 3D printer used to produce the millichannel was a Form 3 from Formlabs[®] and the material chosen was the Clear Resin V4[®], which is transparent and hydrophilic. The main advantages of the stereolithography are its high surface resolution and precision [31, 32]. In Figure 3.1, it is schematically represented the

millifluidic device that was used to perform the experiments, and the main dimensions are listed in Table 3.1.

Table 3.1. Millichannel dimensions (Figure 3.1).

Geometric parameters	Dimensions [mm]
w	1.25
w_c	0.25
l_c	12.5
h	0.75

Once the geometry design was ready (Figure 3.1), the resulting stereolithography file (STL) was converted to G-code using PreForm 3.40.0[®], and in the end imported to the 3D printer (Figure 3.2a) for manufacture. The resulting millifluidic device ($94.4 \times 28.9 \times 4.00 \text{ mm}^3$) can be seen in Figure 3.2b. Furthermore, three connections were implemented for both inlets (i.e., continuous and dispersed phase) and for the outlet, which are also visible in Figure 3.2b.

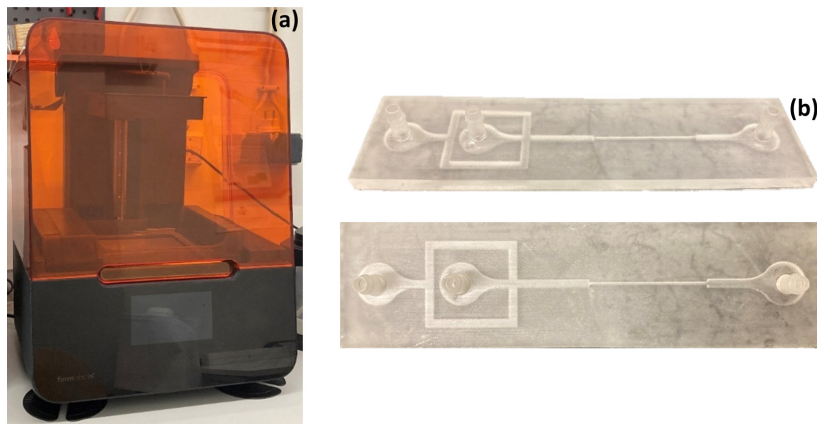


Figure 3.2. 3D printer Form 3 from Formlabs[®] (a), and the 3D printed millichannel using the Clear Resin V4[®] (b).

3.2.2. Working fluids

Three fluid pairs were used in the experiments: 1) water droplet in mineral oil (Sigma-Aldrich® 330779 light mineral oil) with a surfactant, SPAN® 80 (Sigma-Aldrich® 85548) 3% w/w; 2) mineral oil droplet in an 85% w/w aqueous glycerol solution with a surfactant, Sodium Dodecyl Sulfate (SDS from Sigma-Aldrich® L3771 $\geq 98.5\%$ GC) 0.4% w/w; and 3) mineral oil droplet in water with SDS 2% w/w. The surfactants were used to obtain monodisperse droplets of small size, and to prevent coalescence. SDS is the surfactant normally preferred with aqueous continuous phases [33] while SPAN® 80 is the most frequent choice with a mineral oil continuous phase [34]. Additionally, orange food coloring was added to the continuous phase to enhance visualization and improve the differentiation between the two phases (Figure 3.3b).

The test fluids were characterized regarding some essential physical properties: 1) viscosity (μ) of both Newtonian phases was measured by a rotational rheometer (Physica MCR301, Anton Paar) with a Peltier temperature control system set at 20 °C; 2) density (ρ) of both phases was determined with a density probe at ~ 23 °C; and 3) interfacial tension (σ) was also obtained at ~ 23 °C in a Du Noüy ring tensiometer (Force Tensiometer Sigma 701).

The experimental data gathered for the referred physical properties are listed in Table 3.2. The presented results for all properties are the average values from several measurements performed with the mentioned experimental equipment.

Table 3.2. Physical properties of the fluids.

Phases	Fluids	μ [Pa s]	ρ [kg/m ³]	σ [mN/m]	X
Continuous	Mineral oil + 3% SPAN [®] 80	0.038	854	6	0.03
Dispersed	Water	0.001	998		
Continuous	85% aqueous glycerol solution + 0.4% SDS	0.109	1222	24	0.3
Dispersed	Mineral oil	0.036	838		
Continuous	Water + 2% SDS	0.0012	1001	9	30
Dispersed	Mineral oil	0.036	838		

3.2.3. Flow visualization and characterization

The dispersed and continuous phases were injected into the fluidic device with the help of two syringe pumps (Harvard Apparatus PHD ULTRA[™]), and the droplet movement was recorded through images taken with a smartphone (iPhone SE 2nd Generation, 4K resolution at 60 fps), as visible in Figure 3.3. The size and velocity of the droplets were measured using ImageJ[®].

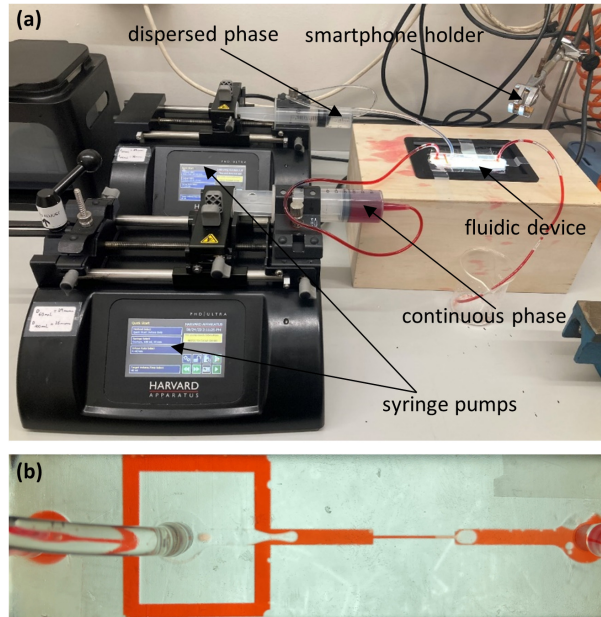


Figure 3.3. Experimental setup (a) and top view image example of the millifluidic device taken with the smartphone (b).

To illustrate a comparison between experiments (orange and transparent-white colors of Figure 3.4) and the corresponding numerical simulations (blue and red colors of Figure 3.4), we selected an intermediate case with $Ca_c = 0.09$ and $X = 0.3$. This is depicted in Figure 3.4, showing: (a) the initial droplet; (b) the droplet entering the contraction; (c) the droplet within the constricted channel; and (d) the droplet exiting the contraction. As shown in the figure, the similarities between the experiment and simulation are clear, although the droplet length inside the constricted channel is slightly larger in the simulation compared to the experiments. This small discrepancy will be further analyzed in Section 3.4 (Results and discussion).

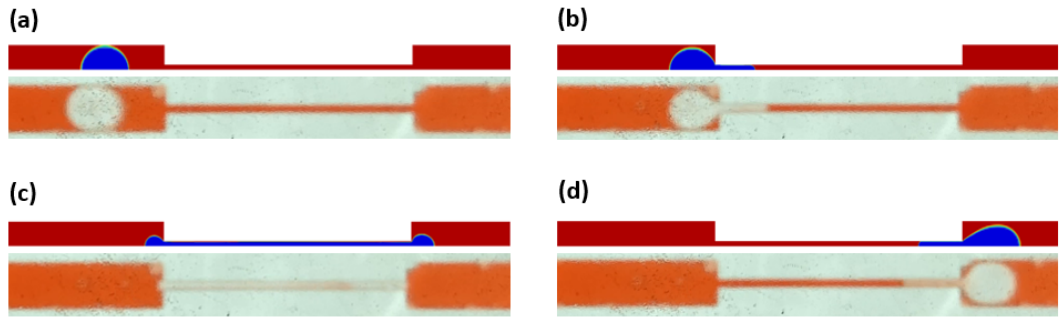


Figure 3.4. Comparison between experiments (orange and transparent-white colors) and the corresponding numerical simulations (blue and red colors) for $Ca_c = 0.09$ and $X = 0.3$: (a) initial droplet; (b) droplet entering the constriction; (c) droplet inside the constricted channel; and (d) droplet exiting the constriction.

3.3. Numerical methods

3.3.1. Governing equations

The continuity and momentum equations were solved under laminar flow conditions, and they are already pre-implemented in the ANSYS Fluent[®] software:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{U}) = 0 \quad (3.2)$$

$$\frac{\partial (\rho \vec{U})}{\partial t} + \nabla \cdot (\rho \vec{U} \vec{U}) = -\nabla p + \nabla \cdot (\bar{\bar{\tau}}) + \vec{F}_\sigma \quad (3.3)$$

where p stands for pressure, $\vec{U}$ for the velocity vector, $\bar{\bar{\tau}}$ for the stress tensor, and $\vec{F}_\sigma$ represents the surface tension force term. The term associated with gravitational forces is neglected, since the cases in study are in milli and microscale where these forces are negligible.

To solve the liquid-liquid interface, the Volume of Fluid (VOF) method was chosen:

$$\frac{\partial \alpha}{\partial t} + \vec{U} \cdot \nabla \alpha = 0 \quad (3.4)$$

where α is the volume fraction ($\alpha = 0$ for the continuous phase, $0 < \alpha < 1$ at the interface, and $\alpha = 1$ for the dispersed phase).

The continuum surface model was applied to account for the surface tension forces at the interface:

$$\vec{F}_\sigma = \sigma k \frac{\rho \nabla \alpha}{0.5(\rho_c + \rho_d)} \quad (3.5)$$

where the local mean interface curvature is expressed as $k = -\nabla \cdot \frac{\nabla \alpha}{|\nabla \alpha|}$

3.3.2. ANSYS Fluent® simulations

As already mentioned, numerical simulations of individual droplets flowing through a constricted channel were carried out using the commercial CFD software ANSYS Fluent® 2021 R2. The two different scales addressed in the numerical simulations were: 1) milliscale with dimensions similar to the millichannel used in the experiments – Figure 3.1; and 2) microscale channel represented in Figure 3.5 with the main dimensions of Table 3.3.

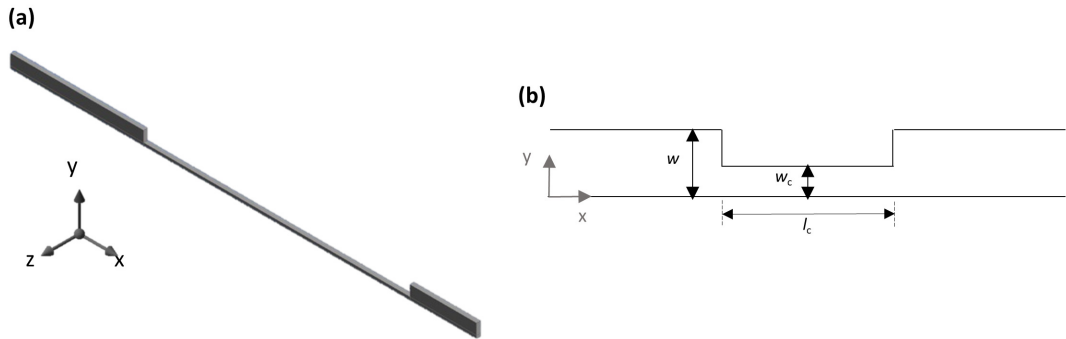


Figure 3.5. Microfluidic device utilized in the ANSYS Fluent® simulations (a) along with its schematic front view (b). The microchannel height ($2h$) is $30 \mu\text{m}$, and the other main channel dimensions are listed in Table 3.3.

Table 3.3. Microchannel dimensions (Figure 3.5).

Geometric parameters	Dimensions [μm]
w	50
w_c	10
l_c	1000
h	15

Both scale simulations were based on 3D domains with the corresponding dimensions shown in Figure 3.1 and Figure 3.5. Figure B.1 in Appendix B provides a color map of the dimensionless magnitude velocity (U/U_i) throughout the constricted microchannel for $X = 0.3$ and $\text{Ca}_c = 0.06$: full-domain (a), and quarter-domain of the entire channel using two symmetry planes (b). Since the results obtained with the full domain were symmetric, axial symmetry around the channel's central planes (i.e., the xy-plane and xz-plane) was assumed in all simulations to reduce computational time and effort. The simulations were initialized with a droplet shape, consisting of a quarter of a sphere placed before the contraction, as shown in Figure 3.6.

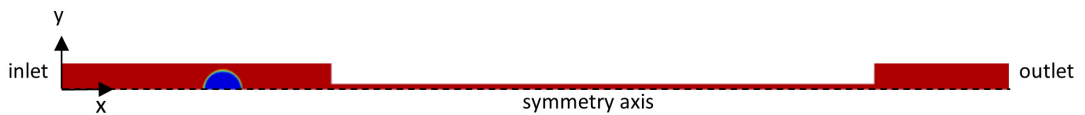


Figure 3.6. Front view of the microfluidic domain showing the droplet in its initial position for all performed numerical simulations.

For $X = 3$ and $\text{Ca}_c = 0.06$, three different simulations were carried out by varying the number of hexahedral cells in the domain (i.e., 7.05×10^5 , 1.41×10^6 , and 2.82×10^6 elements). The chosen variable to analyze the mesh independence was the dimensionless droplet velocity (U_d/U_i), as visible in Figure B.2 in Appendix B. The intermediate mesh, consisting of 1.41×10^6 elements, was used for all microscale numerical simulations, as the results demonstrated to be mesh independent. The mesh was generated using the ANSYS Fluent® Meshing, with finer refinement in the constriction zone. A higher mesh density close to the two symmetry planes of the

channel was also considered, due to the presence of the droplet in those regions. The mesh in the microfluidic contraction region is represented in Figure 3.7a, along with two zoom details for a better visualization – Figure 3.7b and Figure 3.7c.

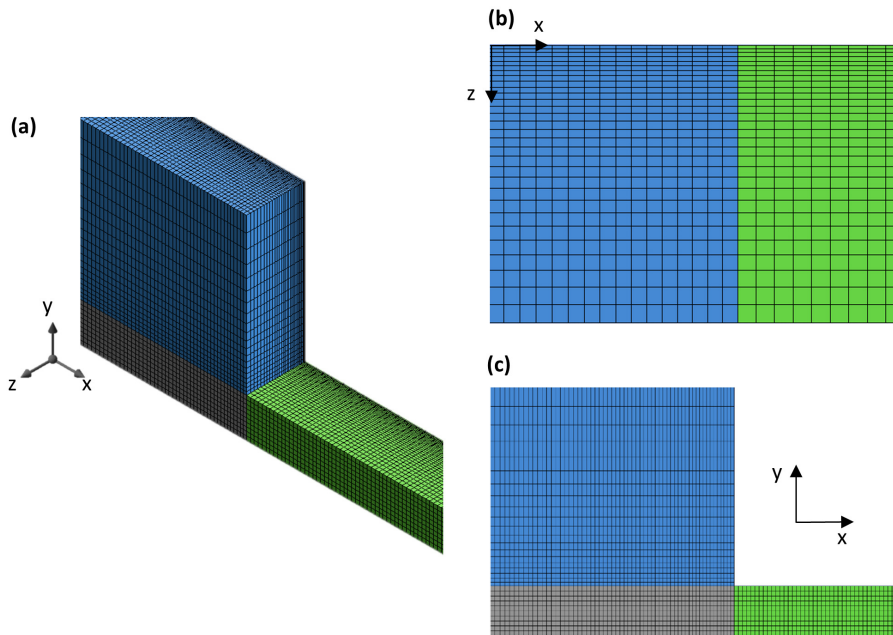


Figure 3.7. Mesh created in ANSYS Fluent® Meshing in the microfluidic contraction region (a), mesh top view zoom detail (b), and mesh front view zoom detail (c).

Regarding the boundaries of the domain, a uniform velocity profile was set at the inlet, while a pressure-outlet condition was applied at the outlet. The walls were modelled with a no-slip boundary condition, and a full non-wetting boundary condition with a static contact angle of 180° was considered.

The simulations were conducted in the transient state to capture the temporal evolution of the droplet shape, and the conditions applied always guaranteed that the flow was under laminar regime.

The liquid-liquid interface was tracked using the VOF methodology, applied with a geometric reconstruction scheme that assumes a Piecewise Linear Interpolation Calculation (PLIC) approach to describe the interface between phases. The continuum surface model was selected to account for the effects of surface tension at the liquid-liquid interface.

The numerical schemes used to solve the governing equations included: 1) PISO (Pressure-Implicit with Splitting of Operators) algorithm for pressure-velocity coupling; 2) PRESTO! (PREssure STaggering Option) method for pressure interpolations; 3) QUICK (Quadratic Upstream Interpolation for Convective Kinematics) scheme for solving the momentum equation; and 4) Green-Gauss Node Based method for computing scalar gradients.

Additionally, an explicit time scheme was used to solve the conservative mass equations with a maximum Courant number of 0.25. The time step was variable and determined by the Courant number. At each time step, the convergence criterion was based on the residuals of the momentum and continuity equations, which were monitored and kept below 10^{-6} .

3.4. Results and discussion

In order to visualize the extent of the conditions addressed in this work, Figure 3.8 shows a summary map, in logarithmic scales, of the simulations performed at the micro and milliscales (sixteen and six respectively), and the experiments (nine) made at the milliscale. The Capillary number of the continuous phase (Ca_c) ranges from 10^{-4} to 10^0 , and the viscosity ratio (X) between the dispersed and continuous phases varied from 10^{-2} to 10^2 . With the results gathered from this set of simulations and experiments, it is intended to provide a relevant contribution for a better understanding of the features involved in the behavior of droplets flow through constricted channels.

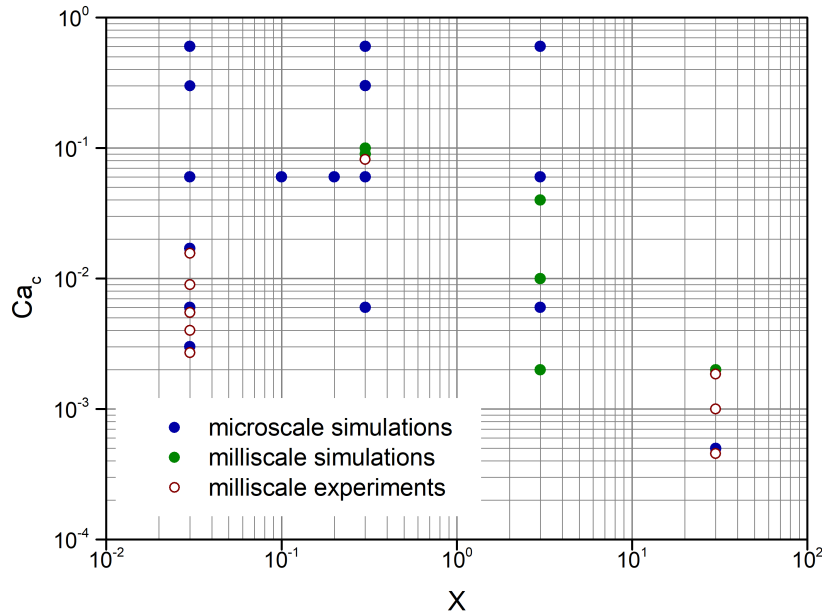


Figure 3.8. Map of the numerical simulations (blue circles – microscale; green circles – milliscale), and of the milliscale experiments (open red circles): Capillary number of the continuous phase (Ca_c) versus the viscosity ratio between the dispersed and continuous phases (X).

3.4.1. Numerical method validation

The first step of the numerical work was to validate the applicability of the CFD model described in the previous section to the systems addressed in this study. For that purpose, a comparison between six experimental tests and the corresponding numerical simulations were performed. Two different parameters, both measured in the constricted part of the channel, were evaluated: droplet velocity (U_d) and droplet length (x_d).

Figure 3.9 presents on the left side the dimensionless average droplet velocity (U_d/U_i) – blue symbols – and on the right side the dimensionless average droplet length (x_d/w_c) – green symbols. Both parameters are plotted as a function of the Capillary number of the continuous phase for three viscosity ratios: $X = 0.03$, $X = 0.3$ and $X = 30$; solid symbols correspond to numerical results and open symbols to experimental data.

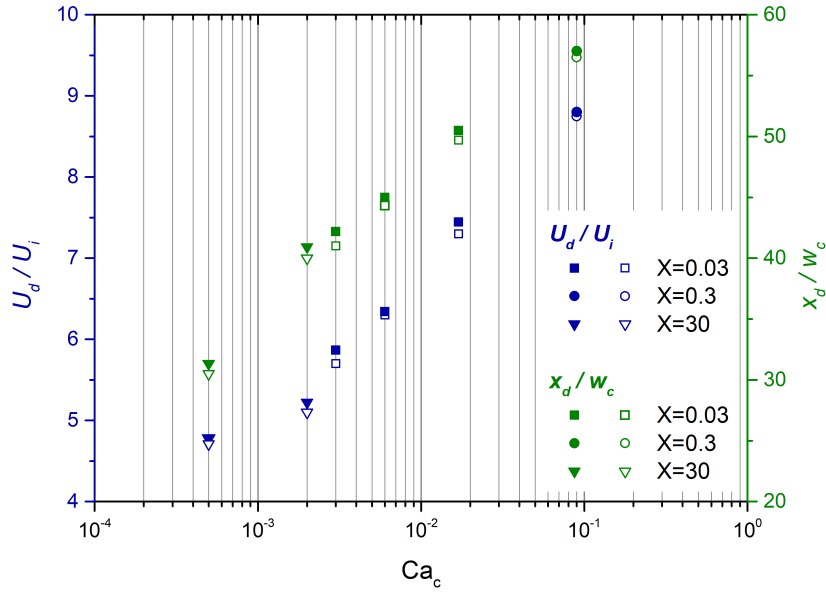


Figure 3.9. Dimensionless average droplet velocity (U_d/U_i) and dimensionless average droplet length (x_d/w_c) when the droplet is flowing inside the constricted part of the channel. On the left side, the graph represents U_d/U_i and on the right side x_d/w_c . Both parameters are plotted as a function of the Ca_c , in logarithmic scale, for a set of three X : solid symbols – numerical simulations; open symbols – experiments.

The observation of Figure 3.9 clearly points out that the simulation results are in good agreement with the experimental data. The deviation between numerical and experimental results is low across all cases, ranging from 0.3% to 1.4%.

This good agreement allows us to validate the numerical procedure and use with confidence the simulation results obtained for all the conditions addressed in this work. Furthermore, the observed agreement shows that the surfactant added to the continuous phase in the experiments does not alter the droplet flow pattern.

3.4.2. Microscale numerical simulation of the single-phase flow of the continuous phase

Initially, numerical simulations of the single-phase flow of the continuous phase were conducted at the microscale using a 3D domain with the corresponding

dimensions shown in Figure 3.5. These simulations were conducted to validate the boundary conditions and achieve a fully developed velocity profile before introducing the dispersed phase. This ensures that the single-phase flow solution serves as the initial state for the multiphase flow simulation.

Figure 3.10 presents a set of dimensionless axial velocity profiles (U_x/U_i) as a function of the dimensionless transversal position (y/w) regarding the single-phase flow of the continuous phase in the microscale channel for $X = 0.03$ and $Ca_c = 0.06$. These profiles correspond to five distinct axial locations along the domain: before the contraction (A and B), before the expansion (C), and after the expansion (D and E).

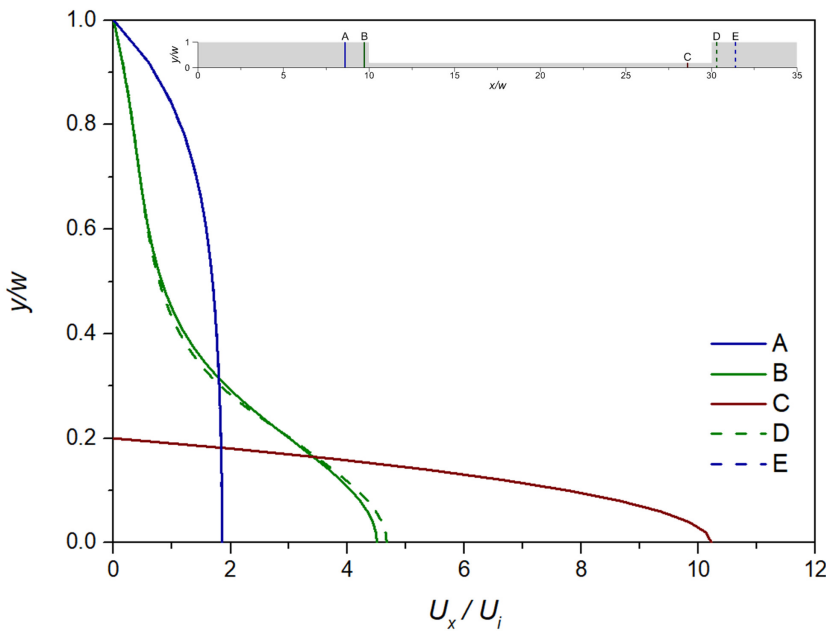


Figure 3.10. Dimensionless axial velocity profiles (U_x/U_i) without droplet at five different positions for $X = 0.03$ and $Ca_c = 0.06$: before the contraction (A and B), before the expansion (C), and after the expansion (D and E).

The dimensionless velocity profile in location A is fully developed reaching $U_x/U_i \approx 1.9$ at the center of the channel ($y/w = 0$). In profile B, near the inlet of the contraction, the perturbation in the velocity field is clearly visible as it differs from the fully developed profile seen in A. This profile has a convex curvature, reaching $U_x/U_i \approx 4.5$ at near $y/w = 0$. As it moves into the wall ($y/w = 1$), it becomes concave, and the velocity decreases bending down toward zero due to the

boundary layer effect. The velocity profile in the narrowest channel before the expansion (C) is again fully developed, and a peak value of approximately $U_x/U_i \approx 10.2$, which is 2 times the average flow velocity in the constricted channel. This is significantly higher than the maximum velocity before the contraction, indicating a large acceleration of the flow through the constricted section of the channel, which is a direct consequence of a lower cross-section area available in this zone. After the expansion, the velocity profiles (D and E) are almost coincident with the ones obtained before the contraction, i.e., B and A respectively. In particular, the evolution from profile D to E reflects the flow recovery towards a fully developed state in the laminar regime after the perturbation introduced by the expansion.

Figure 3.11 provides a color map of the dimensionless magnitude velocity (U/U_i) throughout the constricted channel, covering the contraction and expansion regions for $X = 0.03$ and $Ca_c = 0.06$. After presenting velocity profiles at specific locations, this illustration intends to provide a wider perspective of the continuous phase flow. Before the contraction, it is confirmed that the flow is fully developed. In the constricted section of the channel, the flow develops over a short length. After the expansion, it is also confirmed the re-establishment of the initially fully developed flow.

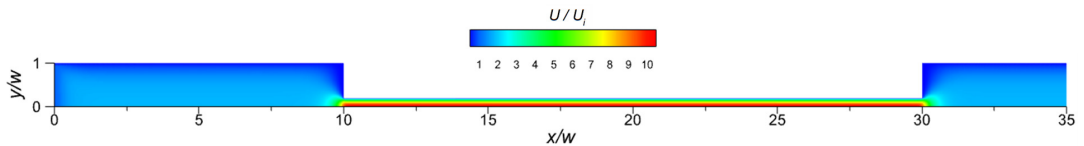


Figure 3.11. Dimensionless magnitude velocity field (U/U_i) for the single-phase flow of the continuous phase for $X = 0.03$ and $Ca_c = 0.06$.

3.4.3. Microscale numerical simulations: droplet moving before the contraction

The behavior of the velocity field when a droplet flows through the continuous phase is another aspect that provides interesting information and tendencies. Based on the numerical data obtained in the present work, this kind of information can be extracted when the droplet is flowing in the larger region before the contraction. For that purpose, dimensionless axial velocities (U_x/U_i) were plotted against the dimensionless transversal position (y/w) in several axial locations. This was done by

cutting the droplet roughly in the middle, i.e., by a horizontal plane containing the axial axis. The droplet velocity was always measured at the droplet's nose. Figure 3.12 shows the compilation of these plots for nine microscale simulations mentioned in Figure 3.8. The aspect ratio (width to height ratio) of the inlet channel is 5:1.5, while for the narrowest channel is 1:1.5, as shown in Figure 3.10.

The analysis covers the effect of the Capillary number of the continuous phase for fixed X values – (a), (b), and (c). As already mentioned, the data used to build these plots concerns instants when the droplet is approaching the constricted zone of the channel. The corresponding droplet shapes are also provided together with horizontal lines delimiting the separation between phases (i.e., continuous phase above the line and dispersed below), to quickly visualize which part of the velocity profiles are inside of the droplet. It is important to remember that each simulation was initialized with a quarter-spherical droplet with a pre-defined size close to the inlet boundary of the domain.

According to the data (a), (b), and (c), the effect of Ca_c in the velocity profiles is attenuated as the viscosity ratio X increases. For values of $X \geq 0.3$, the velocity profiles are practically independent of Ca_c . There is also a significant decrease in the dispersed phase velocities with increasing X and, by mass conservation, the velocity in the continuous phase increases. This fact is particularly evident in the values at the center of the channel ($y/w = 0$), which decrease by more than a half when X increases from 0.03 to 3. It is also possible to conclude that only for $X = 3$, the profiles became concave inside the droplet, which means that the continuous phase velocities close to the interface are greater than the ones on the droplet side. For the simulations with $Ca_c = 0.006$, the droplet shapes appear to be very similar and barely deformed, and so, almost independent of the viscosity ratio. With the increase of Ca_c , the droplet seems to undergo a stronger deformation, increasing the film thickness of the continuous phase; this deformation is more noticeable for the higher viscosity ratio.

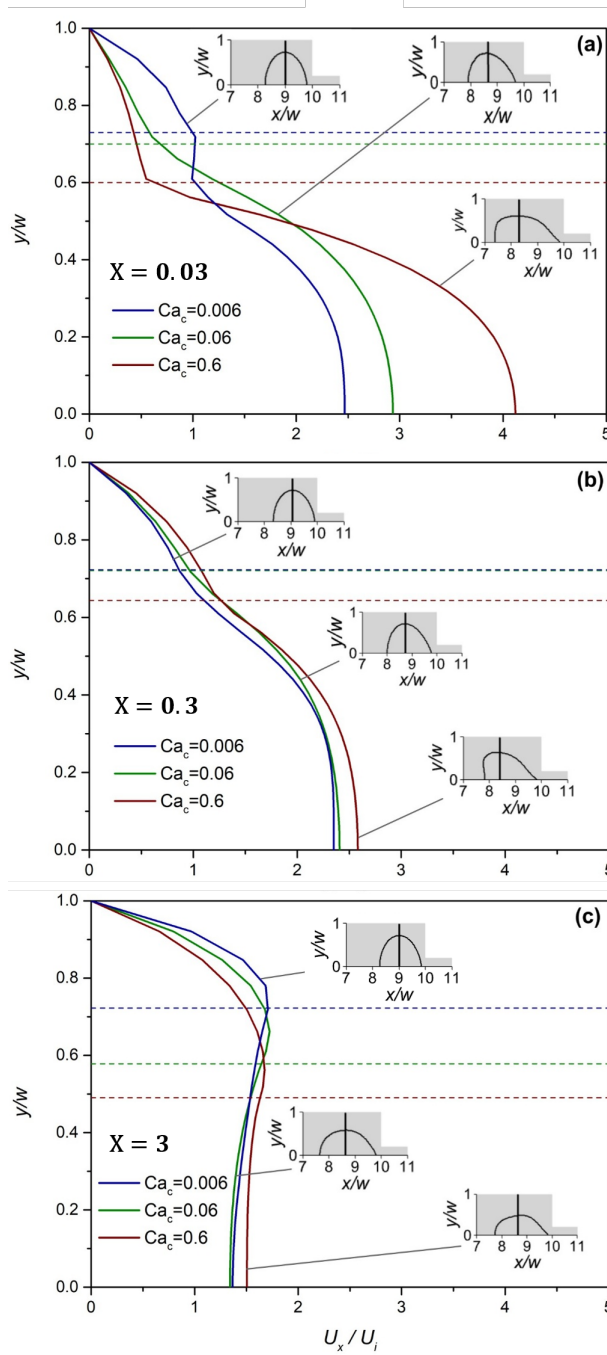


Figure 3.12. Dimensionless axial velocity profiles (U_x/U_i) obtained when the droplet is moving before the contraction. Each graph was produced for a fixed value of X and three values of Ca_c : (a) $X = 0.03$; (b) $X = 0.3$; (c) $X = 3$. The horizontal lines indicate the region of each velocity profile that belongs to each phase: continuous phase above the line and dispersed phase below the line.

3.4.4. Microscale numerical simulations: droplet moving inside the constricted part of the channel

The approach to build Figure 3.12 was also applied when the droplets were completely inside the constricted part of the domain. Figure 3.13 shows a compilation of dimensionless axial velocities (U_x/U_i) as a function of the dimensionless transversal coordinate inside the contraction (y/w_c). The profiles were extracted at a specific axial location, i.e., roughly in the middle of the droplet. The data is presented for fixed X values – (a), (b), and (c), and for each X , it was addressed three values of Ca_c . The aspect ratio (width to height ratio) of the inlet channel is 5:1.5, while for the narrowest channel is 1:1.5, as in the previous figures.

Based on Figure 3.13, the overall effects of Ca_c and X on the behavior of the velocity profiles are somehow similar to the trends described in the discussion of Figure 3.12. Despite this similarity, the role of Ca_c is much more pronounced for lower values of X . Nonetheless, regardless of the viscosity ratio, the velocities within the droplet increase as Ca_c increases. The higher velocity values are identified at the center of the channel for the lowest viscosity ratio ($X = 0.03$) and the highest Ca_c number ($Ca_c = 0.6$). It is also observed that there is a stronger deformation of the droplet with the increasing of Ca_c , which is consistent to what was already mentioned above.

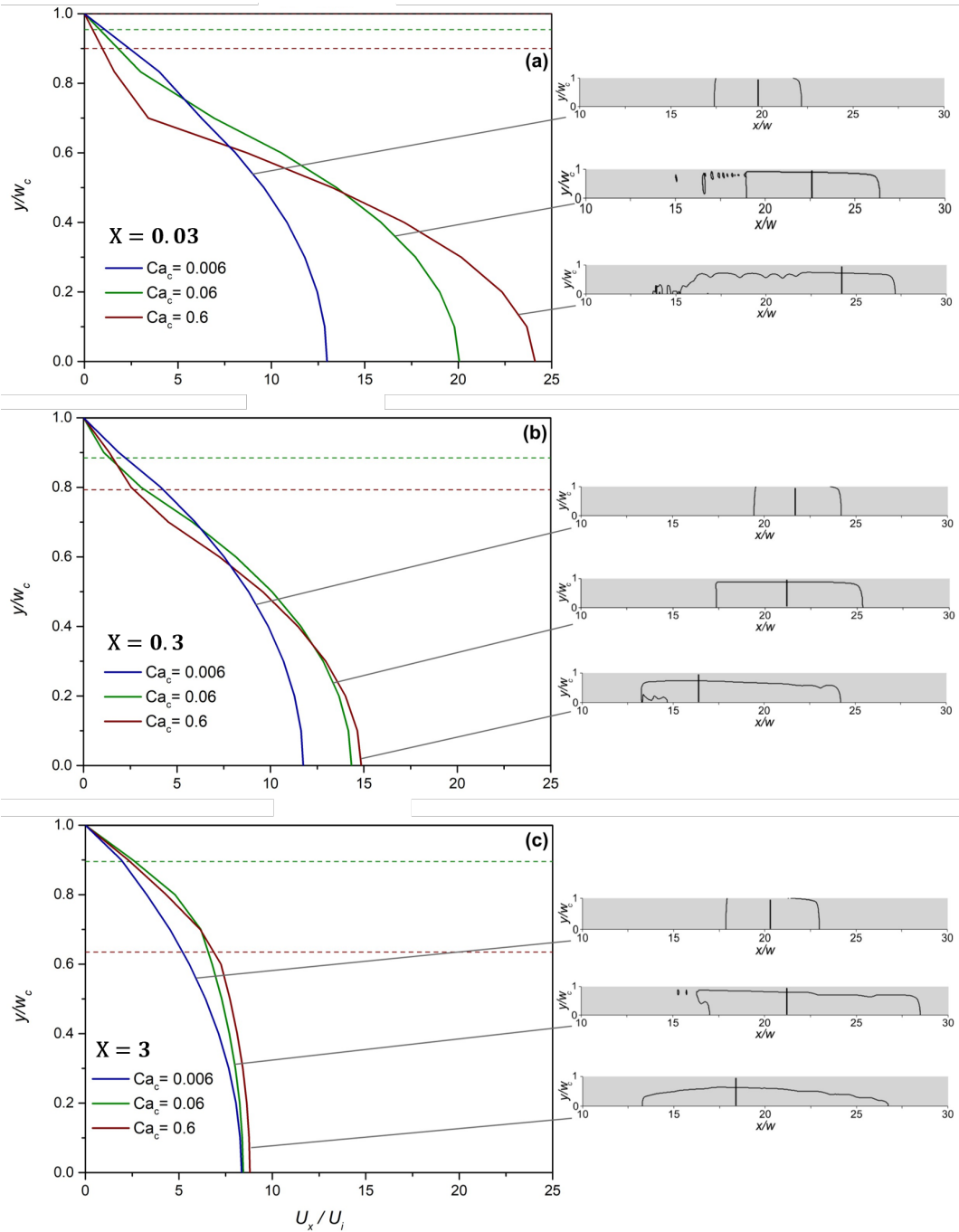


Figure 3.13. Dimensionless axial velocity profiles (U_x/U_i) when the droplet is inside the constricted part of the microchannel. Each graph was obtained for a fixed value of X and three values of Ca_c : (a) $X = 0.03$; (b) $X = 0.3$; (c) $X = 3$. The horizontal lines represent the separation between phases: continuous phase above the line and dispersed phase below the line. For $Ca_c = 0.006$, since the film thickness is almost zero, the separation line is not present.

Figure B.3 in Appendix B shows the evolution of the dimensionless droplet velocity (U_d/U_i) along the constriction as a function of the dimensionless axial coordinate (x/w) for $X = 3$ and three increasing Ca_c values. The reference position used to determine the droplet velocity was always placed at the nose of the droplet. In all cases, the droplet reaches a constant velocity upon entering the constriction.

In order to provide more complete and deeper information on the velocity field of the continuous phase, the streamlines in this phase were plotted considering the droplet stationary (i.e., translating the data to a reference frame attached to the droplet), $U_x - U_d$, in Figure 3.14, Figure 3.15 and Figure 3.16.

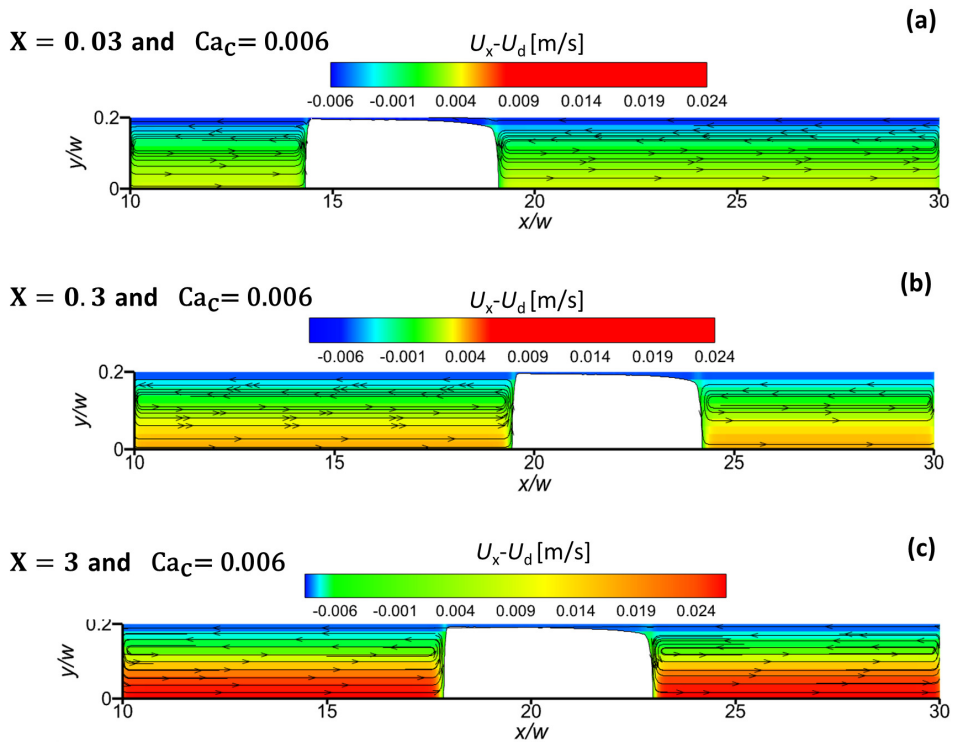


Figure 3.14. Streamlines $U_x - U_d$ when the droplet is inside the constricted part of the microchannel for a fixed value of $Ca_c = 0.006$ and three values of X : (a) $X = 0.03$; (b) $X = 0.3$; (c) $X = 3$.

According to Figure 3.14, for a reduced value of Ca_c , i.e., a strong effect of surface tension, the flow pattern in the continuous phase is only slightly influenced by viscosity ratio of the phases. In general terms, there is a thin film of continuous phase that moves in the opposite direction to the droplet, while the bulk of the continuous

phase moves almost as a piston block with the droplet. In this block, recirculation is noticeable, with the continuous phase close to the axis moving at a speed slightly higher than that of the droplet, and the liquid placed further from the axis moving at a lower speed. The liquid recirculation rate increases for increasing values of X .

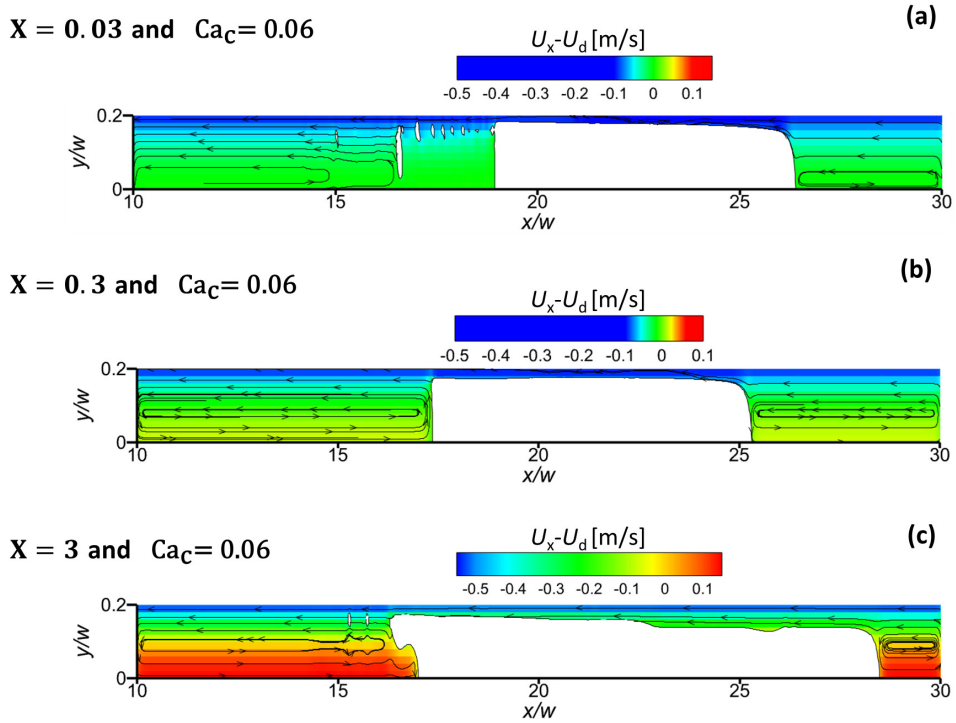


Figure 3.15. Streamlines $U_x - U_d$ when the droplet is inside the constricted part of the microchannel for a fixed value of $Ca_c = 0.06$ and three values of X : (a) $X = 0.03$; (b) $X = 0.3$; (c) $X = 3$.

In Figure 3.15, as the effect of surface tension decreases (by increasing Ca_c), the thickness of the liquid film between the droplet and the wall tends to increase, and this effect is accompanied, as already mentioned, by a more pronounced deformation of the droplet. Compared with the flow patterns for smaller values of Ca_c , the recirculation velocity in the bulk of the continuous phase, behind and in front of the droplet increases. This increase in recirculation is a consequence of the increase in the thickness of the liquid film and of the respective quantity of continuous phase that moves slowly than the droplet.

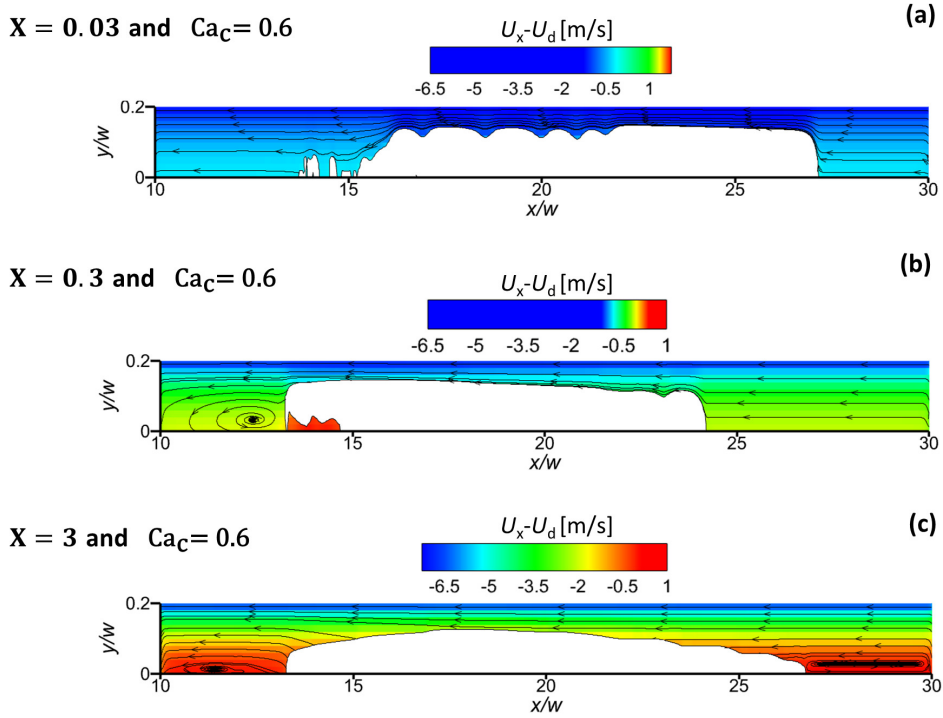


Figure 3.16. Streamlines $U_x - U_d$ when the droplet is inside the constricted part of the microchannel for a fixed value of $Ca_c = 0.6$ and three values of X : (a) $X = 0.03$; (b) $X = 0.3$; (c) $X = 3$.

According to Figure 3.16, for higher values of Ca_c , with a weak effect of surface tension, the importance of the viscosity ratio is noticeable. When this ratio is low, the droplet flows faster than the continuous phase and in the opposite direction, since the whole map has blue colors. There is a clear decrease (i.e., less presence of blue colors) in this velocity relative to the droplet for increasing values of X , i.e., an observer following the droplet sees the continuous phase moving in the opposite direction to the droplet at a lower velocity. For the intermediate value of X , a toroidal vortex is noticeable in the region behind the droplet which accompanies the droplet in its movement, usually called the droplet's wake. For $X = 3$, as mentioned in Figure 3.14 and Figure 3.15, it is once again visible very near the central plane ($y = 0$) a high recirculation velocity of the continuous phase, behind and in front of the droplet.

Using the same approach as in the three previous figures, Figure B.4 and Figure B.5 in Appendix B present additional flow details of droplet behavior during entry into and exit from the contraction for $Ca_c = 0.06$.

3.4.5. Film thickness

The film thickness quantifies the thin layer of the continuous phase flowing between the droplet and the wall of the constriction. Figure 3.17 presents the numerical results obtained for this hydrodynamic feature in dimensionless form (δ/w_c) as a function of the Ca_c for six different values of X . While it is true that the film thickness is not uniform along the droplet length in some cases, the data presented in Figure 3.17 always follow one criterion: the film thickness was measured at its thinnest point. Two different colors were used to distinguish the two sets of scales addressed in the numerical simulations: blue symbols for the microscale (aspect ratio in the inlet channel 5:1.5 and 1:1.5 in the narrowest channel), and green symbols for the milliscale (aspect ratio in the inlet channel 5:3 and 1:3 in the narrowest channel). For the experimental results, the average film thickness is not represented in Figure 3.17, due to the difficulty of accurately measuring it from the collected images.

When analyzing the figure, the conclusions to be drawn are aligned with those mentioned in previous figures:

- For each viscosity ratio, as Ca_c increases the average film thickness also increases, indicating that higher deformation leads to a thicker layer of continuous phase between the droplet and the channel wall;
- At low Ca_c values ($Ca_c < 10^{-2}$), the film is very thin for all viscosity ratios. This indicates that at low Ca_c , where surface tension dominates, the droplet remains very close to the channel wall, flowing at almost the same velocity as the continuous phase – plug flow.

In addition to these conclusions, another important one can be drawn from the figure:

- Normalized thickness values are independent of the channel aspect ratio, as thicknesses are similar across different scales (i.e., micro and milli) for comparable X and Ca_c values.

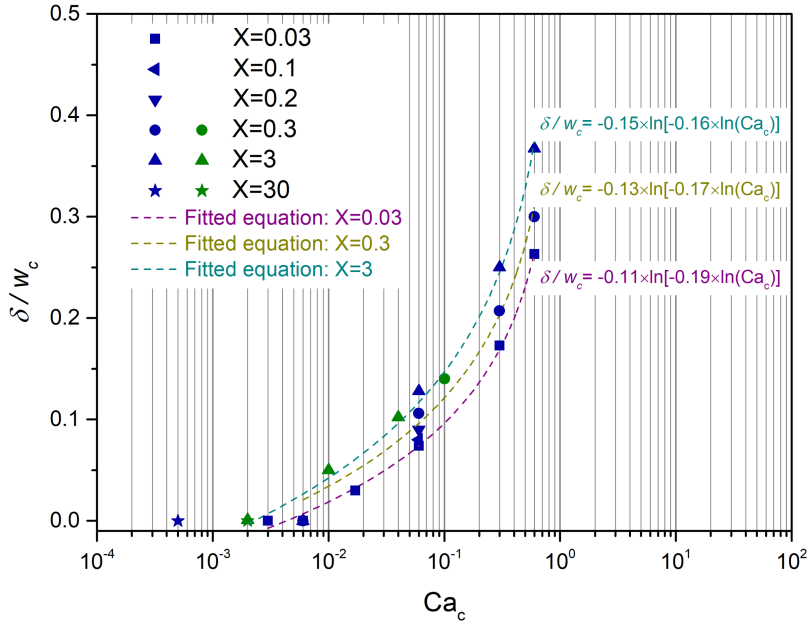


Figure 3.17. Dimensionless average film thickness (δ/w_c) developed when the whole droplet is flowing inside the constricted part of the channel. The graph represents this feature as a function of the Ca_c , in logarithmic scale, for a set of six X values (blue – microscale; green – millyscale).

3.4.6. Average droplet velocity

Figure 3.18 presents the dimensionless average droplet velocity (U_d/U_i) as a function of the Ca_c on a logarithmic scale. The plot compiles data from both microscale simulations (blue symbols), millyscale simulations (green symbols), and millyscale experiments (open red symbols) that collectively address six different values of viscosity ratio.

As mentioned previously, the agreement between simulation and experimental data suggests robustness in predicting numerically the droplet behavior under similar conditions.

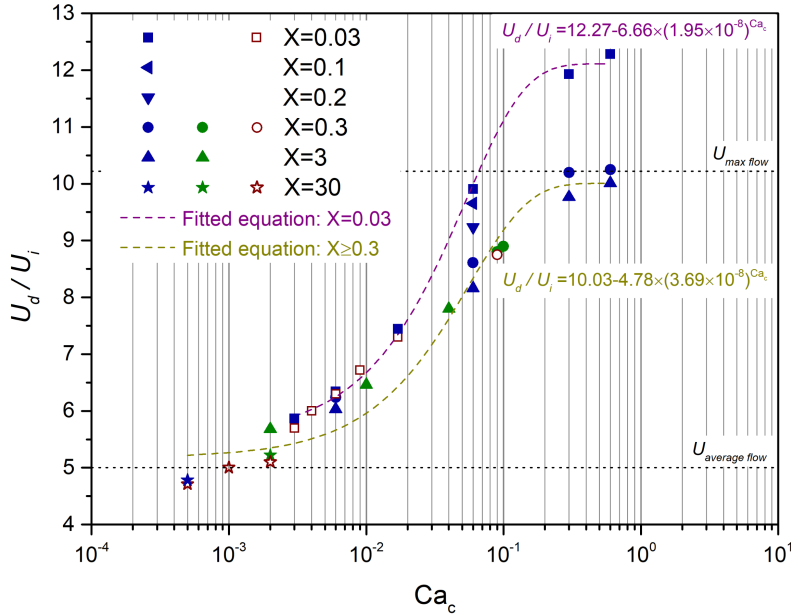


Figure 3.18. Dimensionless average droplet velocity (U_d/U_i) when the droplet is flowing completely inside the constricted part of the channel. The graph represents U_d/U_i as a function of the Ca_c , in logarithmic scale, for a set of six X (blue symbols – microscale simulations; green symbols – milliscale simulations; open red symbols – milliscale experiments).

As expected, as Ca_c increases the average droplet velocity generally increases for all viscosity ratios, indicating that a higher Capillary number of the continuous phase led to faster droplets through the constriction. For a specific Ca_c , the droplet velocity tends to decrease as X increases.

An important conclusion drawn from the figure is that for values of $X \geq 0.3$, the droplet velocity seems to be independent of this ratio.

There are two specific cases where the droplet velocity is greater than the continuous phase velocity (i.e., they are located above the line that corresponds to the maximum flow velocity): 1) $X = 0.03$ and $Ca_c = 0.3$; and 2) $X = 0.03$ and $Ca_c = 0.6$. The second case aligns with the observations made from the streamlines shown in Figure 3.16a.

For $X = 30$ and $Ca_c = 5 \times 10^{-4}$, there seems to be a slight error in the droplet velocity, as these two cases are situated below the average flow velocity line. This may be attributed to limitations when working with very small Ca_c .

As previously observed in the analysis of film thickness data, the normalized droplet velocity is not a function of the channel aspect ratio.

In none of the experiments or simulations was droplet partition observed. It is worth noting that our study employed long constricted domains, whereas Li *et al.* [35] investigated very short and abrupt constrictions, observing droplet breakup experimentally when $Ca_c < 0.036$ and for $L < 3$, where L is the dimensionless initial droplet size.

3.5. Conclusions

In the present study, the flow of a droplet through a constricted channel was investigated experimentally and numerically, leading to the following key conclusions:

- The numerical predictions demonstrated strong agreement with experimental data, showcasing their accuracy in capturing droplet behavior in constricted channels. No droplet breakup was observed in any of the experiments or simulations. Additionally, regarding the experiments, the presence of surfactants in the continuous phase did not significantly affect the flow patterns;
- The influence of Ca_c on dimensionless axial velocity profiles (U_x/U_i) is attenuated as X increased, with velocity profiles becoming nearly independent of X for $X \geq 0.3$;
- For $Ca_c < 10^{-2}$ (i.e., where surface tension dominates), droplets interface remained close to the channel wall, moving at a velocity nearly equal to that of the continuous phase (plug flow). Droplet deformation and the continuous phase film thickness increased with Ca_c , with this effect being most pronounced for higher values of X ;
- As expected, as Ca_c increases, the droplet velocity (U_d) also increased across all X . For a fixed Ca_c , U_d decreased with increasing X , but for $X \geq 0.3$, U_d became almost independent of X ;

Despite differences in channel geometries, behaviors observed at both milli and microscales, such as film thickness and droplet velocity, were overall consistent and

coincident, indicating that the primary flow characteristics are governed by the dimensionless parameters (Ca_c and X).

It is very important to notice that the findings presented on this study may be relevant for the design and optimization of microfluidic systems in applications requiring precise droplet control, like controlled drug delivery and emulsification processes.

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Nomenclature

Acronyms	Definition
3D	Three-dimensional
BEM	Boundary Element Method
CFD	Computational Fluid Dynamics
FE-FTM	Finite Element-Front Tracking Method
LS	Level-set
PISO	Pressure-Implicit with Splitting of Operators
PLIC	Piecewise Linear Interpolation Calculation
PRESTO!	PREssure STaggering Option
QUICK	Quadratic Upstream Interpolation for Convective Kinematics
SDS	Sodium Dodecyl Sulfate
STL	Stereolithography
UV	Ultraviolet
VOF	Volume of Fluid

Greek	Definition	Units
α	Volume fraction	—
δ	Film thickness	[m]
μ	Dynamic viscosity	[Pa s]
μ_c	Dynamic viscosity of the continuous phase	[Pa s]
μ_d	Dynamic viscosity of the dispersed phase	[Pa s]
ρ	Fluid density	[kg m ⁻³]
ρ_c	Density of the continuous phase	[kg m ⁻³]
ρ_d	Density of the dispersed phase	[kg m ⁻³]
σ	Interfacial tension	[N m ⁻¹]
$\bar{\tau}$	Stress tensor	[Pa]

Latin	Definition	Units
Ca	Capillary number	—
Ca_c	Capillary number of the continuous phase	—
De	Deborah number	—
$\vec{F}_\sigma$	Surface tension force term	[N m ⁻³]

h	Height of the channel	[m]
k	Interface curvature	[m ⁻¹]
l	Characteristic length	[m]
L	Dimensionless initial droplet size	—
l_c	Length of the constricted channel	[m]
p	Pressure	[Pa]
Re	Reynolds number	—
t	Time	[s]
U	Flow velocity	[m s ⁻¹]
$\vec{U}$	Velocity vector	[m s ⁻¹]
$U_{average\ flow}$	Average flow velocity	[m s ⁻¹]
U_d	Droplet velocity	[m s ⁻¹]
U_i	Velocity at the inlet	[m s ⁻¹]
$U_{max\ flow}$	Maximum flow velocity	[m s ⁻¹]
U_x	Axial velocity	[m s ⁻¹]
w	Channel width before the constricted channel	[m]
w_c	Width of the constricted channel	[m]
We	Weber number	—
x	Axial coordinate	[m]
x_d	Droplet length	[m]
X	Viscosity ratio	—
y	Transversal coordinate	[m]

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Droplet deformation and breakup in 3D-printed millichannels featuring arrays of rectangular obstacles with different configurations

A. T. S. Cerdeira, J. B. L. M. Campos, J. M. Miranda, and J. D. P. Araújo, "Droplet deformation and breakup in 3D-printed millichannels featuring arrays of rectangular obstacles with different configurations," *Chemical Engineering Science*, vol. 326, p. 123493, 2026, doi: [10.1016/j.ces.2026.123493](https://doi.org/10.1016/j.ces.2026.123493)

Droplet deformation and breakup in 3D-printed millichannels featuring arrays of rectangular obstacles with different configurations

Abstract

The present study investigates the deformation and breakup of droplets in 3D-printed millichannels containing an array of rectangular obstacles with different configurations: transverse, longitudinal, and herringbone. Experiments were performed with two different viscosity ratios between phases ($X = 0.026$ and $X = 30$) under a range of flow conditions, with systematic variations of the flow rate ratio ($\emptyset$) and the Capillary number of the dispersed phase (Ca_d). Flow visualization combined with image processing techniques enables a quantitative characterization of droplet area and velocity distributions at the inlet, across the array of obstacles, and at the outlet region. The results show that, while the droplet area at the inlet region is determined by the flow rate, the outlet area distributions are predominantly governed by the configuration of the array of rectangular obstacles and porosity, with negligible influence from $\emptyset$. For a constant $\emptyset$, as Ca_d increases, viscous stresses become stronger relative to interfacial tension, promoting droplet deformation and breakup. Among the tested designs, the herringbone configuration produced the smallest droplets, followed by the transverse and longitudinal arrays. Additionally, porosity was found to play a critical role: lower porosity enhanced droplet confinement and promoted faster breakup, while higher porosity weakened droplet-obstacle interactions and reduced breakup efficiency. These findings highlight the effectiveness of array of obstacles as passive strategies to promote droplet breakup and size uniformity, offering new design guidelines for droplet manipulation in millifluidic devices.

4.1. Introduction

Droplets are liquid entities surrounded by an immiscible continuous phase with a shape highly dependent on the interfacial tension. The flow of droplets through channels filled with solid obstacles are frequently present in a wide range of applications, such as immiscible displacement during oil recovery, ink-jet printing, fuel cells, and droplet-based fluidics [1, 2]. When passing through a gap between obstacles, a droplet deforms due to the multiple interactions with the solid walls. Beyond deformation, solid obstacles in channels can work as passive methods to promote the breakup of the droplets not requiring the use of an external field [2, 3]. Array structures composed of dozens to hundreds of obstacles are also employed in small scale fluidic devices due to their ability to manipulate fluid behavior [4]. These arrays of obstacles serve critical roles in processes such as phase separation [5], mixing [6], and fluidic control [7], which are fundamental to a variety of industrial, biomedical, and environmental applications. In phase separation processes, they enhance the selective partitioning of immiscible fluids by increasing the interfacial area and promoting efficient mass and heat transfer. This is particularly valuable in chemical reactors and extraction units where precise control over liquid-liquid interfaces leads to improved separation efficiency, reactants conversion, and energy savings [5]. Mixing is significantly improved by introducing an array of obstacles that induce chaotic advection and break flow symmetry. Passive mixers leveraging these obstacles enable rapid homogenization of reactants without the need for external power sources or moving parts, making them highly suitable for biochemical assays, point-of-care diagnostics, and drug delivery systems where fast and uniform mixing is essential [6]. Such mixing enhancements reduce reaction times and increase reaction yields in reactors used for synthesis and analysis. Furthermore, optimized array of obstacles facilitates the rapid removal or concentration of components in fluidic heat exchangers, making them ideal for compact cooling systems in electronics and microprocessors.

Fluidic control, encompassing the precise manipulation of droplet and multiphase flows, benefits from an array of obstacles by regulating droplet deformation, breakup, coalescence, and trajectory. This control is vital for droplet-based fluidics, which supports digital fluidic platforms used in single-cell analysis, droplet-based

polymerase chain reaction (PCR), and high-throughput screening [7]. Obstacle-induced droplet dynamics also enable sorting, gating, and filtering functions critical to lab-on-a-chip technologies. The performance of these processes is closely linked to parameters such as the wetted surface area, droplet penetration rate, and penetration time through the array of obstacles. Optimizing these factors improves heat and mass transfer efficiencies, which directly impact device throughput, energy consumption, and overall process reliability [4]. Thus, the design and arrangement of the array of obstacles are central to enhance microfluidic technologies across diverse fields, from chemical engineering and biotechnology to environmental monitoring.

The main dimensionless numbers to describe the deformation and breakup of droplets flowing through channels with solid obstacles are the Reynolds (Re) and Capillary (Ca) numbers of continuous (denoted by the index *c*) and dispersed (index *d*) phases, and the viscosity ratio (*X*) between phases [8] – Equation 4.1:

$$\text{Re}_c = \frac{\rho_c U_c L}{\mu_c} \quad ; \quad \text{Re}_d = \frac{\rho_d U_d L}{\mu_d} \quad ; \quad \text{Ca}_c = \frac{U_c \mu_c}{\sigma} \quad ; \quad \text{Ca}_d = \frac{U_d \mu_d}{\sigma} \quad ; \quad X = \frac{\mu_d}{\mu_c} \quad (4.1)$$

where ρ is the volumetric mass, U the velocity, L the characteristic length, μ the dynamic viscosity, and σ is the surface tension between both phases.

Besides those dimensionless groups, deformation and breakup phenomena are significantly influenced by the geometry of the obstacles, and the surface properties of the wall material (reflected in the contact angle of the interface between phases). In micro and millifluidic devices, the inertial forces are usually negligible and the droplet deformation is independent of the Reynolds number. Additionally, one of the Capillary numbers can be substituted by the flow rate ratio, $\emptyset = q_d / q_c$, where q_d is the flow rate of the dispersed phase and q_c is the flow rate of the continuous phase. So, the deformation of a droplet, or their normalized size, after passing through a fluidic device is then dependent of the following factors: geometry, $\emptyset$, Ca_d and X .

In recent years, droplet manipulations (i.e., formation, coalescence and breakup) have attracted significant attention with the purpose of obtaining droplets with uniform sizes [9]. Experiments, theoretical analyzes, and numerical simulations [2, 7, 10-15] have been made to investigate the breakup of a droplet in channels with cylindrical, square or rectangular obstacles (Figure 4.1).



Figure 4.1. Schematic top view examples of channels with a single obstacle: a) cylindrical; b) square; and c) rectangular.

These works show that breakup only occurs when the Capillary number of the continuous phase exceeds a minimum value [3]. A pioneering study by Link *et al.* [10], using droplets of water in a continuous phase of hexadecane with SPAN[®] 80 3% w/w, demonstrated that channel geometry alone could reliably mediate droplet breakup. By forcing droplets through a narrow T-junction, they showed that extensional flow fields, combined with confinement, cause deterministic breakup into smaller daughter droplets. This geometrically mediated mechanism introduced a design framework where droplet breakup is governed by the balance between flow-induced elongation and interfacial tension, setting the stage for subsequent studies on obstacle-driven breakup. In this study, the authors also experimentally analyzed the breakup of a droplet using a single square obstacle. They observed that the sizes of the broken droplets are related to the obstacle position. Chung *et al.* [11] numerically simulated the dynamics of both Newtonian and viscoelastic droplets passing through a single cylindrical obstacle. The results showed that the breakup of the Newtonian droplet depends on the droplet size, while for the viscoelastic droplet, the normal stress difference ($\tau_{xx} - \tau_{yy}$) becomes the key factor for the breakup of the droplets. In another work, Chung *et al.* [7] presented an interesting numerical and experimental investigation to study the deformation of a droplet passing through different types of obstacles (i.e., a single cylindrical obstacle and a square obstacle). Oil was used as continuous phase and water as dispersed phase, and the authors studied the effects of the obstacle shape, the initial droplet size, and the Capillary number of the continuous phase in a two-dimensional channel. The cylindrical obstacle performed better than the square obstacle for droplet size control, since more non-trivial satellite droplets were produced, and an additional dead zone was observed near the front stagnation point of the square obstacle. Using the hexadecane properties for the droplet and the water properties for the continuous phase, Lee and Son [14] numerically studied the breakup dynamics and the volume ratio of daughter droplets. They disclosed that the

droplet breakup pattern with or without re-merging depends on the width, length, location, and inclination of the obstacle. Li *et al.* [2] numerically explored the phenomena of deformation and breakup of a droplet passing through a two-dimensional channel with a cylindrical obstacle. Four cases with viscosity ratio (X) ranging from 0.02 to 50 were considered, and they confirmed that the viscosity ratio is an important parameter which strongly affects the droplet dynamics. For $X > 1$, droplets undergo larger elongation and deformation that may ultimately result in breakup, whereas for $X < 1$, droplets exhibit comparatively small deformations and tend to preserve their shape when passing through the obstacle. Salkin *et al.* [13] theoretically and experimentally studied the breakup phenomena of large droplets (i.e., slugs) passing through a rectangular obstacle with different lengths (200-800 μm). Two liquid-liquid systems were used: 1) different water-glucose mixtures as dispersed phases and hexadecane as continuous phase: $0.3 < X < 2.3$; and 2) deionized water dispersed in a viscous silicone oil: $X = 0.02$. The authors focused on the effects of both phases viscosities, μ_c and μ_d , to identify the critical droplet size and critical Capillary number of the continuous phase for breakup. They found that the evolution of this critical Capillary number with the droplet size is nonmonotonic if $\mu_d > \mu_c$ (i.e., $X > 1$) and monotonic when $\mu_d < \mu_c$ (i.e., $X < 1$).

Despite several studies involving a single obstacle, there is a lack of information about systems with droplets crossing a channel with an array of obstacles (Figure 4.2).

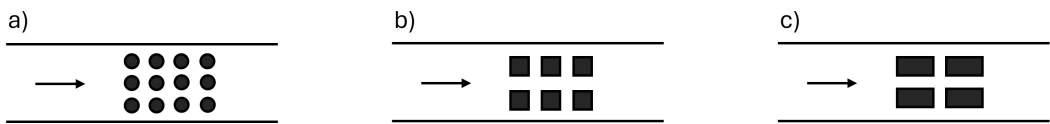


Figure 4.2. Schematic top view examples of channels with an array of obstacles: a) cylindrical; b) square; and c) rectangular.

An array of obstacles may be used to convert a polydisperse population of droplets into a relatively monodispersed population [16]. Another application for this array of obstacles is the promotion of an efficient mixing between two miscible liquids in a fluidic system [17].

The passage of a single droplet through an array of obstacles in a channel was experimentally and numerically investigated by Chung *et al.* [7] and Wang *et al.* [4].

The promotion of efficient mixing in a fluidic device was studied by Wang *et al.* [18] and Bhagat *et al.* [6].

Chung *et al.* [7] presented a numerical and experimental investigation to study the deformation of a water droplet surrounded by oil passing through different array of obstacles (i.e., an array of cylindrical obstacles aligned in a single row and a 2×4 array of cylindrical obstacles). Two requirements for selective wetting of each obstacle (i.e., the droplet individually wets or adheres to each obstacle in sequence, rather than bypassing some, or merging across them) were established: 1) a low Capillary droplet number, Ca_d , where surface tension dominates, causing droplets to deform slowly and interact more predictably with each obstacle. This is in agreement with the work of Link *et al.* [10], who showed that controlled breakup in geometric constrictions happens only when Ca_d is below a critical value, where the droplet is confined enough to stretch but not so much that it skips over the obstacle; and 2) a distance between obstacles larger than the channel width (i.e., if obstacles are too close together, the droplet does not have time to fully relax or deform before reaching the next obstacle). Wang *et al.* [4] conducted a three-dimensional numerical parametric study to examine the effects of droplet initial velocity, obstacle dimensions (i.e., width and length), and obstacle surface wettability. The authors found that when the water droplet passes through the array of obstacles, the droplet dynamics in the longitudinal direction is governed by the competition between the dynamic pressure and the capillary force. In contrast, the capillary effect dominated the permeation process in the lateral and vertical directions.

Wang *et al.* [18] theoretically and numerically studied the effect of using from zero to eighteen cylindrical obstacles disposed of in linear, square or triangular configurations to improve mixing in a “Y-shape” channel using two fluids flowing parallel to each other (water and ethanol). They found that with an increase in the number of obstacles, the mixing performance can be improved by affecting the flow pattern, and that the triangular arrangement of obstacles changes the flow directions, also enhancing the mixture process. Bhagat *et al.* [6] conducted experimental and numerical investigations using arrays of cylindrical, triangular, and diamond obstacles, demonstrating their potential for integration into lab-on-a-chip devices to achieve mixing at low Reynolds numbers. The mixing process was evaluated via a fluorescence technique with two fluids (fluorescein solution in deionized water and deionized

water). Their results showed that asymmetric obstacle placement induces chaotic advection and enhances mixing in the Stokes flow regime ($Re < 1$), with mixing efficiency improving as the number of obstacles increases due to the generation of velocity gradients and streamline stretching. Although the study primarily addressed mixing rather than droplet breakup, it supports the concept that strategic obstacle placement can be employed to manipulate flow characteristics and interfacial deformation.

Recent investigations have provided valuable insights into droplet deformation and breakup in channels with single obstacles of different geometries, highlighting the role of parameters such as obstacle shape, viscosity ratio, and Capillary number of the dispersed phase [2, 7, 10-15]. However, most of these studies have been limited to single obstacles or simple configurations, while systematic studies involving array of obstacles remain scarce [4, 7, 18]. In particular, there is a lack of experimental data on the effects of obstacle arrangement, porosity, and flow conditions on droplet breakup and size distribution in fluidic devices. Moreover, previous works have primarily focused on numerical analyzes or specific mixing applications [6, 18], leaving an evident gap in understanding how different geometrical configurations of array of obstacles can be exploited to achieve controlled droplet breakup and size regulation in practical fluidic systems.

In this study, experiments were performed with two fluid pairs ($X = 0.026$ and $X = 30$) to investigate the deformation and breakup of droplets in 3D-printed millichannels containing an array of rectangular obstacles with different geometrical configurations (transverse, longitudinal, and herringbone arrays – Figure 4.3). By systematically varying the flow conditions (ϕ and Ca_d), as well as the geometrical configuration of the array of obstacles and porosity, this study establishes how an array of obstacles can be employed as efficient passive strategy to promote droplet breakup and produce droplets with uniform sizes. The novelty of this work lies in the direct comparison of distinct arrays of obstacles, coupled with the use of high-precision 3D printing technique and robust quantitative analysis of droplet area and velocity distributions. Furthermore, this study presents the first comprehensive experimental comparison between different configurations of complex arrays of obstacles in millichannels. Porosity is identified as a critical design parameter, revealing how lower-porosity array of obstacles enhance confinement, droplet-obstacle interactions,

and breakup efficiency. This work also demonstrates that outlet droplet size distributions converge to geometry-controlled steady states, independent of inlet conditions. In conclusion, these findings establish arrays of obstacles as effective passive strategies for achieving uniform droplet populations. By leveraging geometric features rather than external fields, array of obstacles enables scalable, energy-efficient droplet manipulation, offering new pathways for compact and adaptive millifluidic devices.

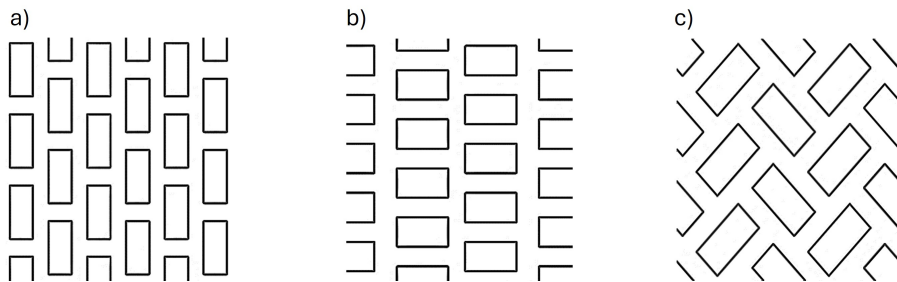


Figure 4.3. Schematic top view representation of the array of rectangular obstacles: a) transverse; b) longitudinal; and c) herringbone.

4.2. Experimental techniques and procedures

Experiments were conducted in 3D-printed millichannels featuring different array of rectangular obstacles. The millichannels comprise a flow-focusing region for droplet generation followed by a region with obstacles, as visible in Figure 4.4. The flow-focusing part is composed by two inlets, one for the continuous phase and the other for the dispersed phase.

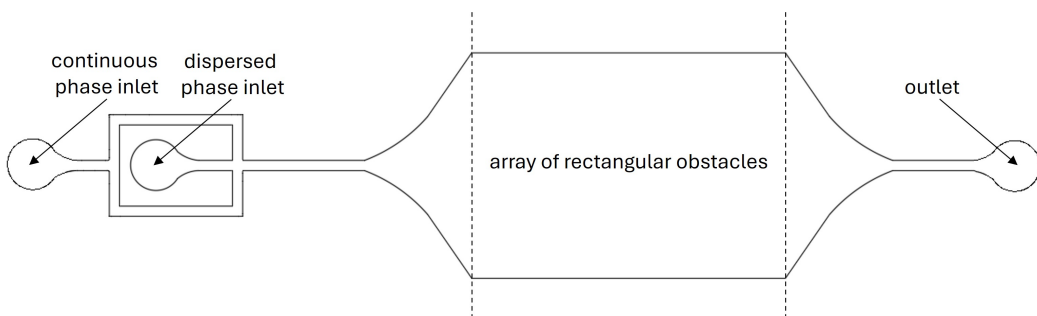


Figure 4.4. Schematic top view representation of the millifluidic device.

Three different configurations with an array of rectangular obstacles were experimentally tested and presented in more detail in Subsection 4.2.1: 1) transverse – Figure 4.3a; 2) longitudinal – Figure 4.3b; and 3) herringbone – Figure 4.3c. In the transverse array, the obstacles are oriented with their larger surface perpendicular to the flow, and arranged in staggered columns such that alternate rows are laterally offset. The longitudinal array consists of staggered rows with the larger surface parallel to the flow direction. In the herringbone array configuration, the obstacles are oriented at 45° relative to the flow, with successive columns alternately rotated 45° clockwise and 45° anticlockwise.

All configurations exhibit similar porosities, ε , (i.e., the ratio between the void volume and the total volume of the millichannel region that comprises the array of obstacles): $\varepsilon = 55\%$ for the transverse array and $\varepsilon = 56\%$ for the longitudinal and herringbone arrays.

Two different fluid pairs (described in Subsection 4.2.2) were used to study the deformation of droplets in these channels. Additionally, the efficacy of these fluidic devices to promote the breakup of droplets, obtaining droplets with uniform sizes, will also be analyzed.

4.2.1. Fabrication method of the millifluidic devices

The geometry design of all millifluidic devices used in the experiments was made with Autodesk AutoCAD 2022[®]. These devices were manufactured by the stereolithography 3D printing technique [19]. In this additive manufacturing process, an ultraviolet (UV) laser beam repeatedly strikes the printing material to produce the object layer by layer. The printing materials are photosensitive thermoset polymers in the liquid form. The 3D printer used to produce the millichannel was a Form 3 from Formlabs[®] and the material chosen was the Clear Resin V4[®], which is transparent and hydrophilic. The main advantages of the stereolithography are its high surface resolution and precision [19, 20]. The three different configurations that were experimentally studied are presented in Figure 4.5. The main dimensions of the corresponding millifluidic devices are listed in Table 4.1.

To demonstrate the integrity and geometric accuracy of the printed structures, the Appendix C includes an optical microscope image of part of the array of rectangular obstacles (Figure C.1). The measured dimensions are consistent with those reported in Table 4.1.

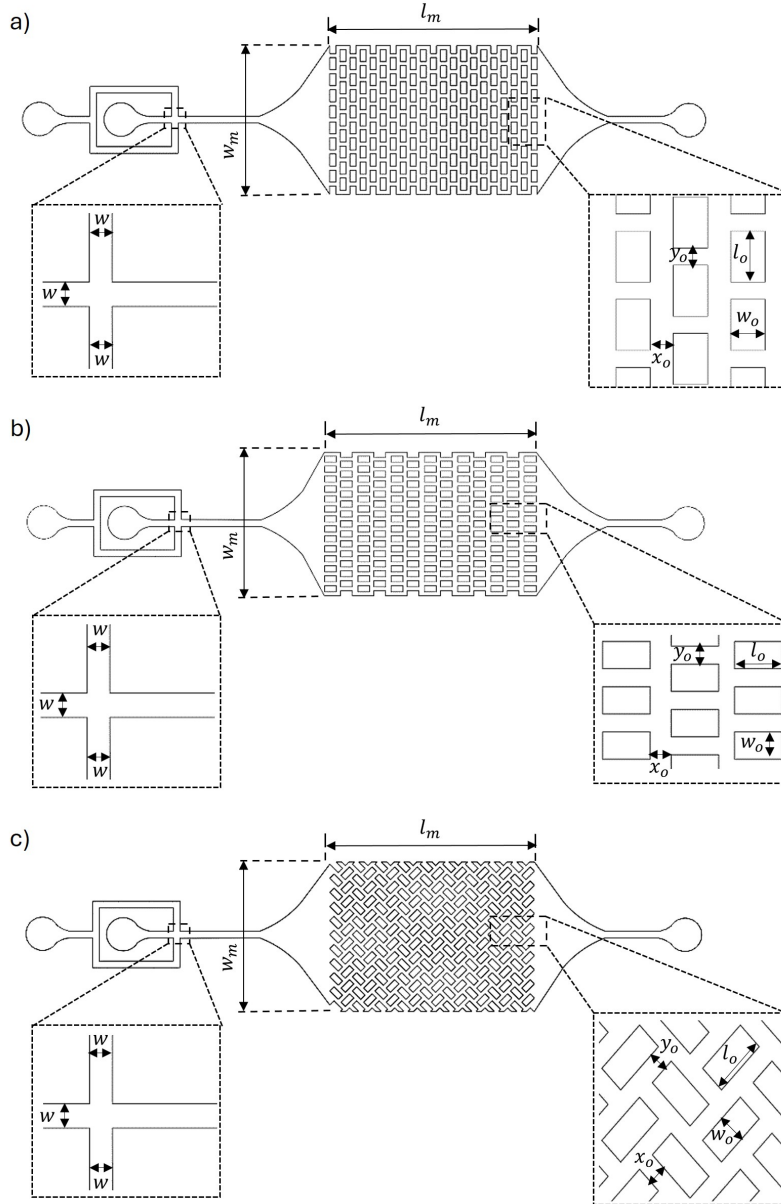


Figure 4.5. Top view of millifluidic devices with an array of rectangular obstacles: a) transverse; b) longitudinal; and c) herringbone. The millichannels height (h) is 1.50 mm, and the other main channels dimensions are listed in Table 4.1.

Table 4.1. Millichannels dimensions (Figure 4.5).

Configuration	w [mm]	h [mm]	l_m [mm]	w_m [mm]	x_o [mm]	y_o [mm]	l_o [mm]	w_o [mm]
Transverse 	1.00	1.50	30.9	22.2	0.600	0.600	1.80	0.900
Longitudinal 	1.00	1.50	31.8	21.6	0.600	0.600	1.80	0.900
Herringbone 	1.00	1.50	30.8	22.5	0.600	0.600	1.80	0.900

Once the geometry design was ready (Figure 4.5), the resulting stereolithography files (STLs) were imported to the 3D printer for manufacture and the resulting millifluidic devices ($94.4 \times 28.9 \times 4.00 \text{ mm}^3$) can be seen in Figure 4.6. Furthermore, three connections were implemented for both inlets (i.e., continuous and dispersed phase) and the outlet which are also visible in Figure 4.6.

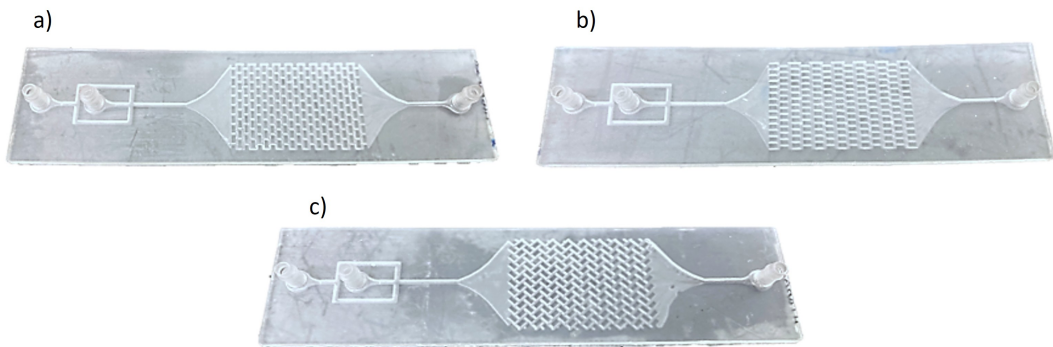


Figure 4.6. 3D-printed millichannels using the Clear Resin V4[®] with an array of rectangular obstacles: a) transverse; b) longitudinal; and c) herringbone.

4.2.2. Working fluids

Two fluid pairs were used in the experiments: 1) water droplet in mineral oil (Sigma-Aldrich® 330779 light mineral oil) with a surfactant, SPAN® 80 (Sigma-Aldrich® 85548) 3% w/w; and 2) mineral oil droplet in water with a surfactant, Sodium Dodecyl Sulfate (SDS from Sigma-Aldrich® L3771 $\geq 98.5\%$ GC) 2% w/w.

The surfactants were used to obtain monodisperse droplets of small size, and to prevent coalescence. SDS is the surfactant normally preferred with aqueous continuous phases [21] while SPAN® 80 is the most frequent choice with a mineral oil continuous phase [22]. Additionally, orange food dye was always added to the continuous phase to enhance visualization and improve the differentiation between the two phases (Figure 4.7b).

Since the Clear Resin V4® is hydrophilic, a surface treatment is required to enable the formation of water droplets in mineral oil with SPAN® 80. To make the millichannel hydrophobic, a car glass water repellent was injected into the millifluidic device. After a few minutes of contact and a subsequent rinse with water, the device was ready for the experiments.

The test fluids were characterized regarding some essential physical properties: 1) viscosity (μ) of both Newtonian phases was measured by a rotational rheometer (Physica MCR301, Anton Paar) with a Peltier temperature control system set at 20 °C; 2) volumetric mass (ρ) of both phases was determined with a density probe at ~ 23 °C; 3) interfacial tension (σ) was also obtained at ~ 23 °C in a Du Noüy ring tensiometer (Force Tensiometer Sigma 701).

The experimental data gathered for the referred physical properties are listed in Table 4.2.

Table 4.2. Physical properties of the fluids.

Phases	Fluids	μ [Pa s]	ρ [g/cm ³]	σ [mN/m]	X
Continuous	Mineral oil + SPAN [®] 80 3% w/w	0.038	0.85		
Dispersed	Water	0.0010	1.0	5.8	0.026
Continuous	Water + SDS 2% w/w	0.0012	1.0		
Dispersed	Mineral oil	0.036	0.84	8.9	30

The contact angles of both fluid pairs on the untreated and surface-treated millichannels were measured (Figure C.2 in Appendix C). As expected, after surface treatment with the car glass water repellent, a low contact angle was obtained for the continuous phase (23°), while a higher contact angle was measured for the dispersed phase (79°). In contrast, for the untreated millichannel, similar contact angles were measured for both phases, with a low contact angle of 14° for the dispersed phase and 22° for the continuous phase. Despite this, the untreated millichannel still enabled stable droplet generation with this fluid pair, as illustrated in Figure 4.10 for the lowest value of Ca_d .

4.2.3. Flow visualization and characterization

The dispersed and continuous phases were injected into the millifluidic device with the help of two syringe pumps (Harvard Apparatus PHD ULTRA[™]), and the droplet movement was registered through videos recorded with a smartphone (iPhone SE 2nd Generation, 4K resolution at 60 fps), as visible in Figure 4.7. The spatial calibration was such that 1 pixel corresponds to 0.03 mm, and was obtained using a reference length of known dimensions on the millichannel.

The slight defocusing observed on the right side in Figure 4.7b is due to the optical limitations of the smartphone camera. However, droplet detection and tracking were performed using two plugins – described in more detail below – based on intensity contrast and edge detection. This approach proved to be sufficiently robust to

accurately identify and measure droplets even in regions with low to moderate loss of focus.

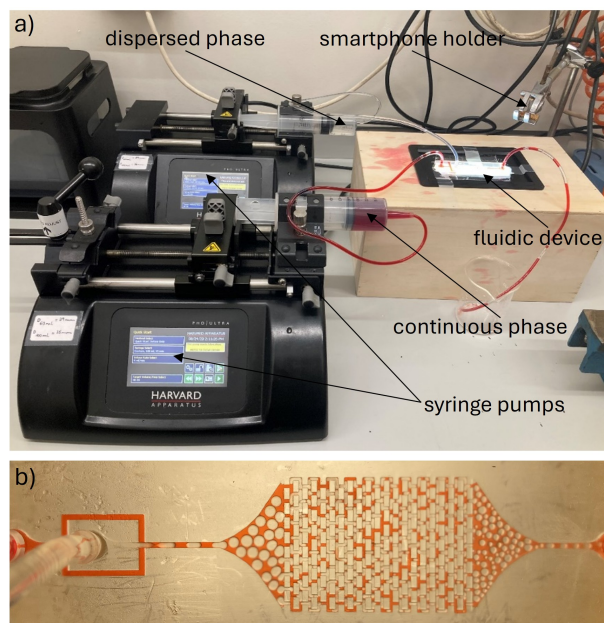


Figure 4.7. a) Experimental setup [23]; and b) top view example image of the millifluidic device with transverse array of rectangular obstacles taken with the smartphone. In this experiment, mineral oil was used as dispersed phase (transparent-white color) and water with SDS 2% w/w as continuous phase (orange color due to the use of an orange food dye to enhance visualization between phases).

For each of the three devices under study, three different regions of interest were considered: 1) inlet; 2) array of rectangular obstacles; and 3) outlet – Figure 4.8.

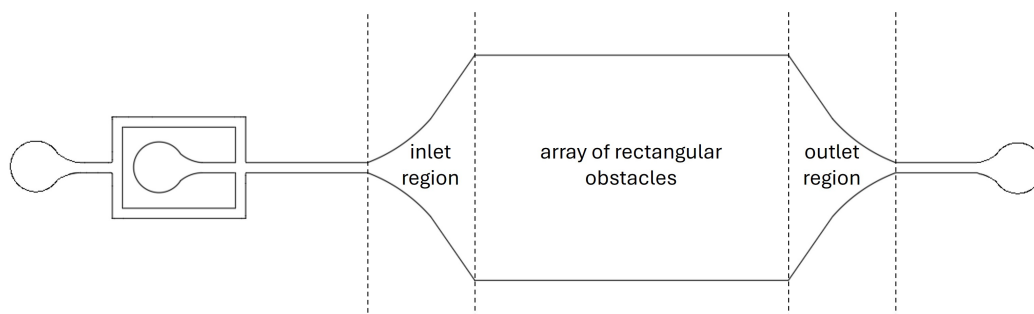


Figure 4.8. Regions of interest for the three different configurations: inlet (before the obstacles); array of rectangular obstacles; and outlet (after the obstacles).

For each region represented in Figure 4.8, the position, area and velocity of the droplets were determined using the Droplet Tracker (2013) [24], which is an ImageJ® (version 1.52a) plugin – Subsubsection 4.2.3.2. Prior to applying the Droplet Tracker, the original video frames were processed with an orange mask over the obstacles and outside the millichannel to facilitate more accurate droplet identification – Figure 4.9a. Then, a segmentation of all acquired images (i.e., black and white images in which droplets are represented in black and the obstacles/empty spaces of the channel are in white) was performed through the use of Trainable Weka Segmentation (version 3.9.4) [25], which is a Fiji® (version 1.53q) plugin – Subsubsection 4.2.3.1. The post-processing of the obtained results was made through a Python script.

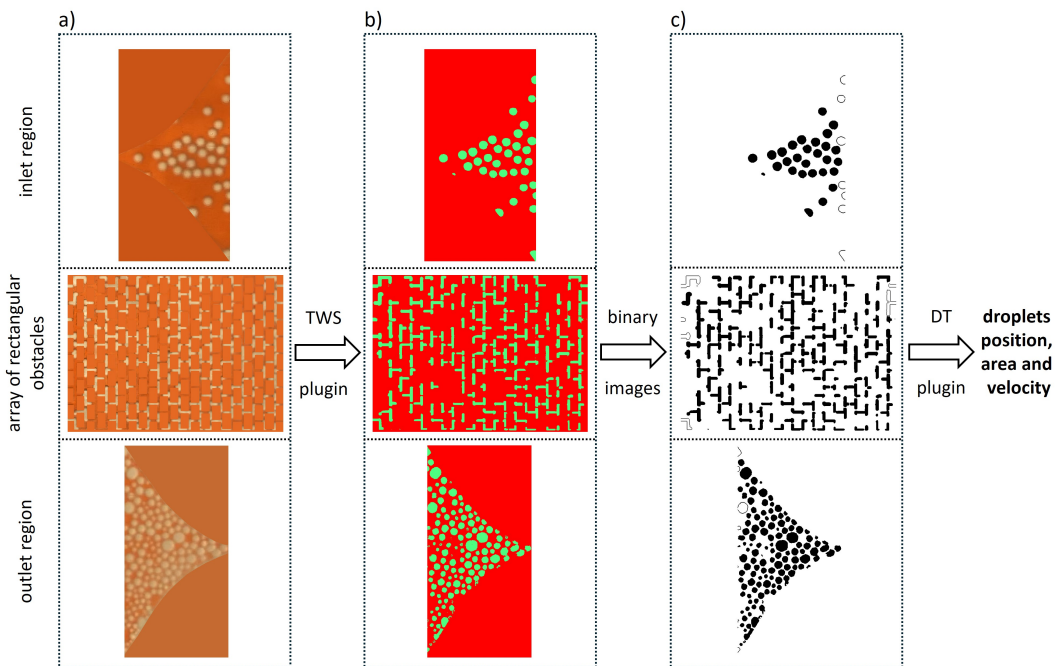


Figure 4.9. Images processing example for the three regions under analysis according to Figure 4.8 – inlet, array of rectangular obstacles and outlet: a) original frames with an orange mask on the obstacles and outside the millichannel; b) resulting frames after applying the TWS plugin; and c) conversion of the previous images to binary images for application of the DT plugin.

4.2.3.1. Trainable Weka Segmentation (TWS)

Trainable Weka Segmentation integrates a collection of machine learning algorithms with selected image features to generate pixel-based segmentations. The method reformulates the segmentation task as a pixel classification problem, where each pixel is assigned to a specific segment or class. In this study, two classes were defined: droplets and obstacles/empty channel regions – Figure 4.9b. The process begins with a labeled set of input pixels represented in the feature space, which serves as training data for the chosen classifier. After training, the classifier can reliably label the remaining pixels in the dataset as well as those in previously unseen images.

4.2.3.2. Droplet Tracker (DT)

Droplet Tracker is a particle tracker with a specific focus on tracking droplets in fluidic devices. DT is used for tracking binary, black-on-white droplet blobs – Figure 4.9c. In this way, after having all images classified, DT was used to obtain several droplet features: 1) centroid through time; 2) area; and 3) velocity. Droplets spanning several pixels (typically more than 10 pixels in diameter) could be reliably detected and analyzed using the DT plugin. However, it is important to notice that this tool has some limitations since DT tracks droplets across stack frames using a nearest-neighbor approach. If a droplet in two successive frames is within the user-specified "velocity" number of pixels, it is considered the same droplet as in the previous frame. At the end of the process, a CSV file can be generated with all the output fields, and a Python script can be used to post-process all obtained data.

4.3. Results and discussion

In subsections 4.3.1 - 4.3.5, the dispersed phase used in the experiments was mineral oil, and the continuous phase was water containing SDS 2% w/w and colored orange with food dye to enhance phase contrast (viscosity ratio $X = 30$). The last subsection (i.e., Subsection 4.3.6) presents and discusses the results for both investigated viscosity ratios ($X = 0.026$ and $X = 30$).

4.3.1. Flow visualization

A qualitative assessment of the droplet flow behavior in millifluidic devices with a rectangular array of obstacles was performed through a parametric study, varying the dispersed phase flow rate (q_d) and the continuous phase flow rate (q_c) within the range 0.5 to 4 mL/min for $X = 30$ (Figure 4.10). Figure 4.10 shows representative images of the herringbone configuration, illustrating the droplet distribution at steady flow as a function of $\emptyset = q_d / q_c$ and Ca_d ; identical flow conditions were also experimentally reproduced for transverse and longitudinal arrays, with the corresponding results discussed in the following subsections. For all cases, Ca_d was evaluated using the dispersed phase velocity measured at the flow-focusing region of the millichannel. This velocity was calculated as the ratio of the imposed q_d to the cross-sectional area of the channel at that location.

For $X = 0.026$, the Re_c and Re_d are within the following ranges: $5.33 \times 10^{-2} \leq Re_c \leq 4.26 \times 10^{-1}$ and $2.38 \times 10^0 \leq Re_d \leq 1.90 \times 10^1$. For $X = 30$, the corresponding ranges are: $3.97 \times 10^0 \leq Re_c \leq 3.17 \times 10^1$ and $1.11 \times 10^{-1} \leq Re_d \leq 8.89 \times 10^{-1}$. For all cases, the Reynolds number was calculated using the average velocity measured at the array of obstacles, with the characteristic length defined as the spacing between obstacles (x_o in Figure 4.5). Inertial effects within this Reynolds number range are typically negligible or absent; therefore, the influence of Re will not be considered in this work.

The observed flow patterns behavior reveals two aspects: 1) lower Ca_d value (i.e., first column of Figure 4.10) – droplet breakup occurs in a highly regular manner, characteristic of the dripping regime. The uniformity in droplet area and spacing suggests a shear-dominated regime, where the continuous phase exerts sufficient viscous stress to periodically pinch off droplets; and 2) higher Ca_d values (i.e., second to fourth columns of Figure 4.10) – the dispersed phase forms large and continuous jets that fail to breakup into discrete droplets at the flow-focusing region. The continuous phase flow was insufficient to induce breakup, resulting in unstable interfaces and loss of droplet control; however, small droplets were still observed at the outlet region, confirming the high efficiency of the array of obstacles in promoting

droplet breakup. For $Ca_d = 4.75 \times 10^{-2}$, a notable feature was the presence of large portions of the dispersed phase remaining attached to the obstacles.

For the lowest value of Ca_d (i.e., $Ca_d = 2.38 \times 10^{-2}$), when holding the dispersed phase flow rate constant and increasing the continuous phase flow rate (i.e., decreasing $\emptyset$), a clear decrease in droplet area at the inlet region was observed, which indicates that increasing shear from the continuous phase promotes earlier detachment and smaller droplet formation.

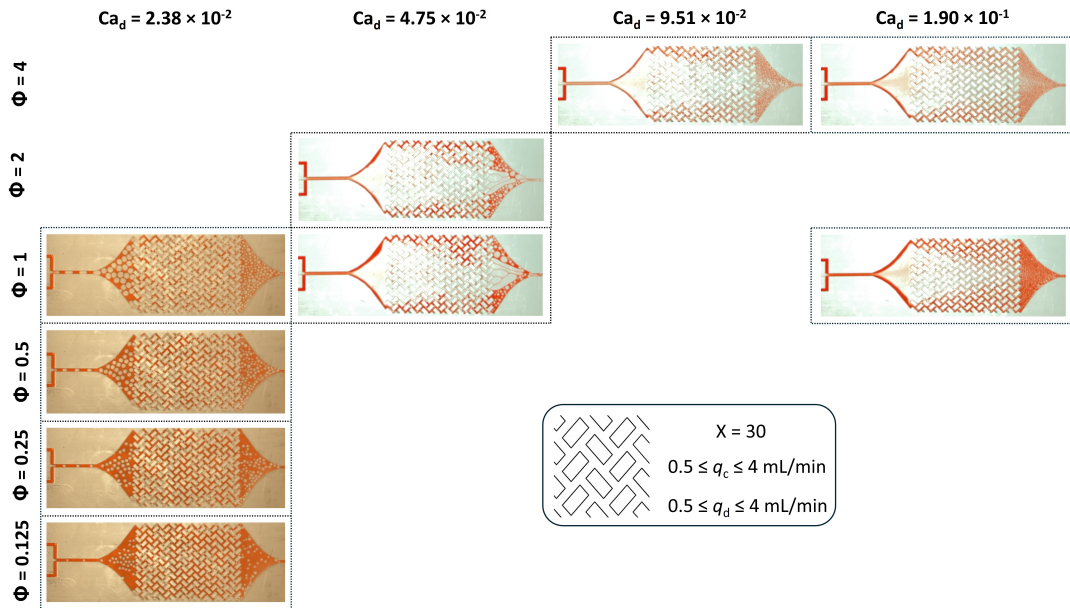


Figure 4.10. Flow patterns behavior in the millifluidic device with herringbone configuration for $X = 30$, as a function of $\emptyset$ and Ca_d . In these experiments, mineral oil was used as dispersed phase (transparent-white color) and water with SDS 2% w/w as continuous phase (orange color due to the use of an orange food dye to enhance visualization between phases).

4.3.2. Effect of $\emptyset$ on droplet formation

As mentioned in Section 4.2, three different array of rectangular obstacles were experimentally studied (Figure 4.5). For each configuration, the dispersed phase flow rate was always kept equal to 0.5 mL/min, and for the continuous phase, four different flow rates were addressed: 0.5 mL/min, 1 mL/min, 2 mL/min, and 4 mL/min.

Accordingly, four $\emptyset$ values (i.e., $0.125 \leq \emptyset \leq 1$) are considered for discussion. The value of Ca_d was kept constant at 2.38×10^{-2} , and $X = 30$.

The droplet area distributions at the inlet (Figure 4.11) is exclusively controlled by the flow regime established within the droplet formation channel, which only depends on the flow rates and is independent of the array of obstacles configuration (i.e., channel geometry). For $0.125 \leq \emptyset \leq 0.25$, the droplets are small and exhibit a narrow area distribution at the inlet region. For $0.5 \leq \emptyset \leq 1$, an increase in droplet area is observed at the inlet region, accompanied by a slightly wider spread in the area distribution. The histograms of the droplet area at the inlet region seems to follow a normal distribution, as visible in Figure 4.11.

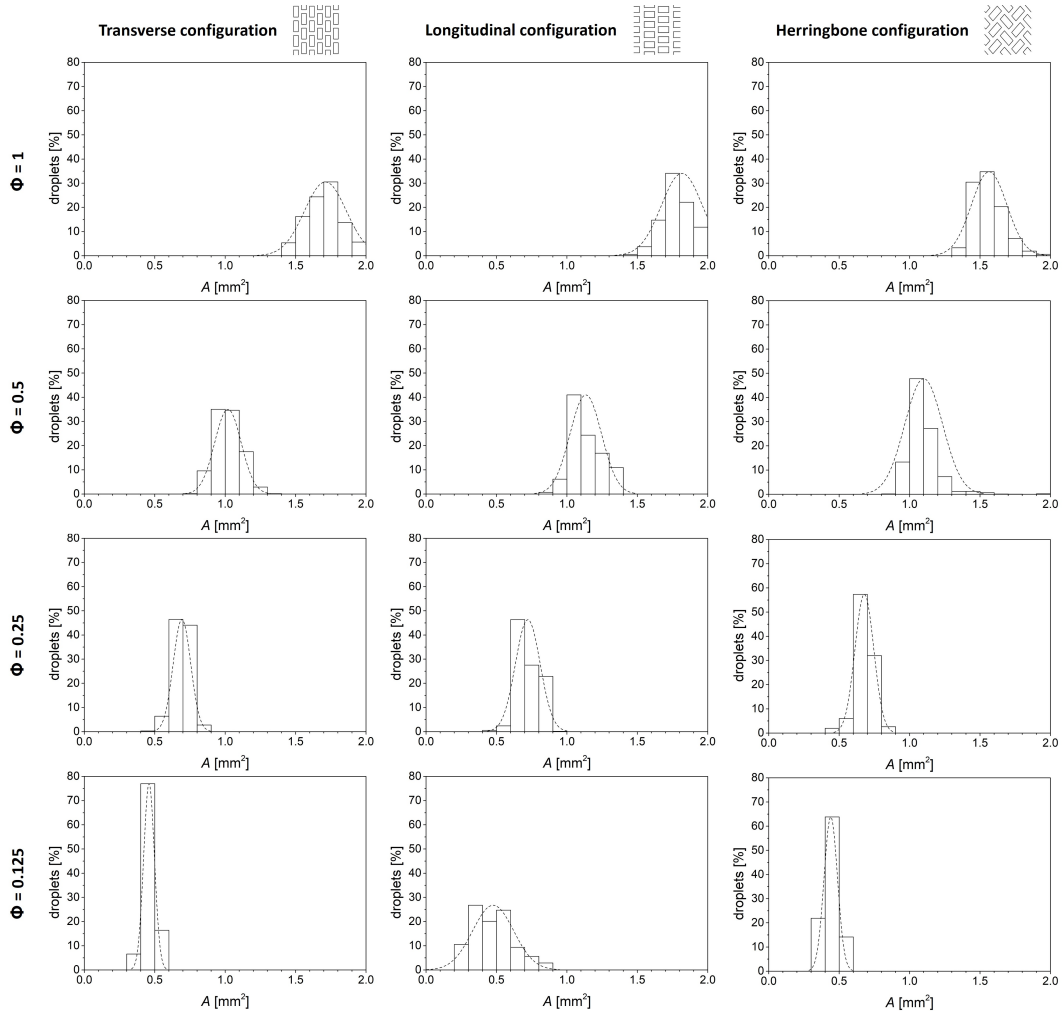


Figure 4.11. Droplet area (A) distribution at the inlet region for the three millichannel configurations considered in this study. The conditions addressed included four distinct $\emptyset$ values, $X = 30$, and $Ca_d = 2.38 \times 10^{-2}$. The superimposed dashed line represents a fitted normal distribution curve.

4.3.3. Effect of the array configuration

Following a similar procedure, Figure 4.12 presents histograms for the droplet area at the outlet for all three configurations. The same four $\emptyset$ values were addressed, Ca_d was kept constant at 2.38×10^{-2} , and $X = 30$. In contrast to the inlet region (Figure 4.11), where droplet area and distribution are strongly dependent on the flow

conditions, the outlet distributions display a different behavior. For each configuration, the mean droplet area remains nearly constant across the four $\emptyset$ values tested, indicating that the flow rate ratio has a negligible influence on the final droplet area after interaction with the array of obstacles. This trend is particularly evident in the herringbone configuration, where the central tendencies of the distributions show minimal variation, with overlapping peaks across the four $\emptyset$. This behavior suggests that the array of rectangular obstacles exerts a dominant influence over droplet breakup and deformation processes. As droplets traverse the array of obstacles region, their area and shape become increasingly regulated by the geometric confinement and local shear stresses induced by the obstacles, leading to a convergence toward a steady area distribution that is independent of upstream conditions. This implies that the role of the inlet flow regime is substantially diminished downstream, as the droplets are forced to adapt to the spatial constraints and flow bifurcations imposed by the configuration. Minor differences in outlet droplet area are observed between configurations, with the herringbone tending to produce smaller droplets than transverse and longitudinal configurations. Under the operating conditions investigated, no droplet merging or coalescence was observed along the millichannel. Furthermore, as it can be seen in Figure 4.12, the great majority of histograms of the droplet area at the outlet region also follows a normal distribution.

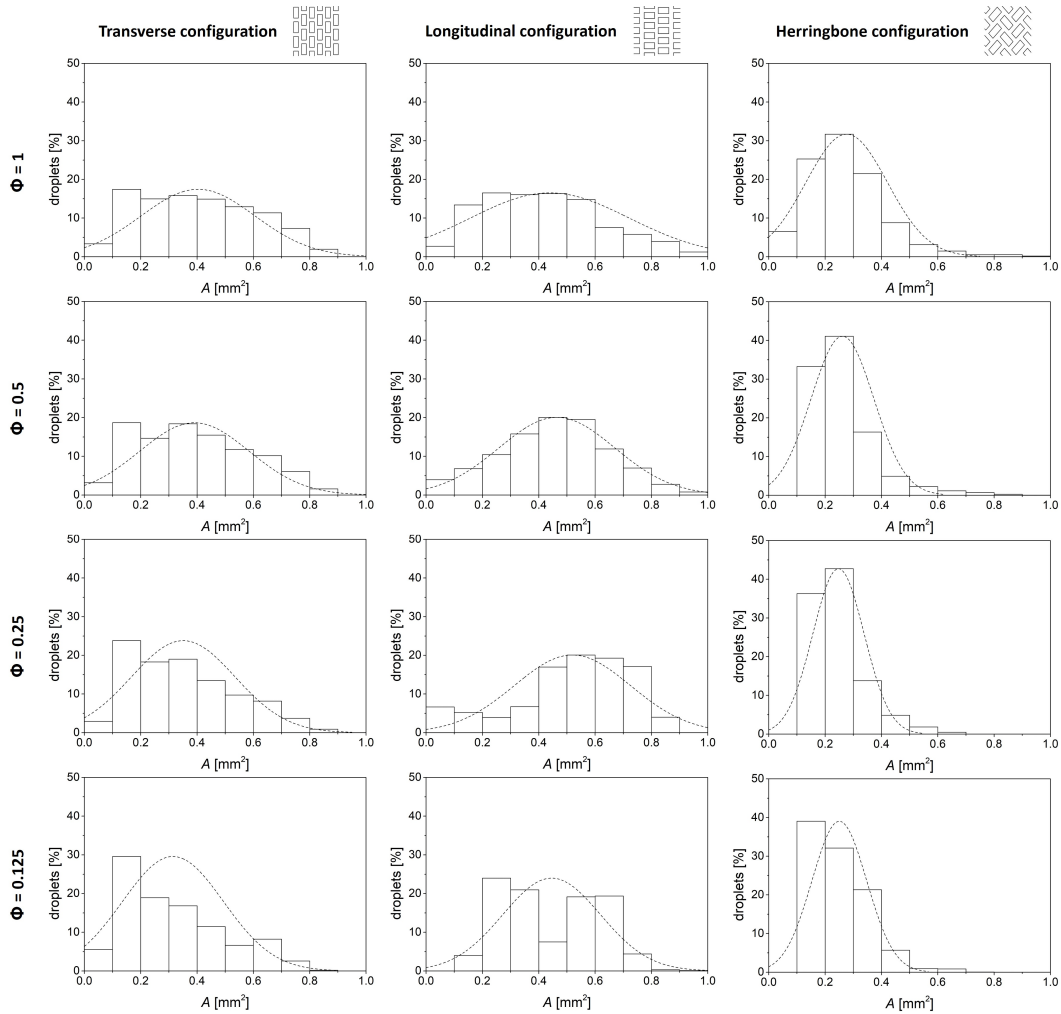


Figure 4.12. Droplet area (A) distribution at the outlet region for the three millichannel configurations considered in this study. The conditions addressed included four distinct $\emptyset$ values, $X = 30$, and $Ca_d = 2.38 \times 10^{-2}$. The superimposed dashed line represents a fitted normal distribution curve.

To provide a clear comparison of droplet areas at the inlet and outlet regions, Figure 4.13 presents the mean and standard deviation of all droplet area distributions measured at the inlet (Figure 4.11) and at the outlet (Figure 4.12). As already mentioned, one important conclusion can be drawn from Figure 4.13a: for each $\emptyset$, the droplet area at the inlet region is almost independent of the array of obstacles configuration, being this effect more noticeable for low $\emptyset$ values (i.e., $0.125 \leq \emptyset \leq$

0.25). In Figure 4.13b, it is clearly visible that, for each array of obstacles configuration, the droplet area at the outlet region is almost independent of $\emptyset$. Herringbone configuration generates the smallest droplets among the three, followed by the transverse configuration, and with the longitudinal configuration producing the largest droplets.

From Figure 4.13, it is also possible to conclude that for $\emptyset = 0.125$, the initial droplet area before the array of obstacles (i.e., at the inlet region) – Figure 4.13a – is practically the same as the one obtained at the outlet region – Figure 4.13b. This indicates that at low flow rate ratios, the array of obstacles does not significantly reduce droplet area, as the droplets, generated in the droplet formation channel, are already small and have areas comparable to those measured at the outlet region. This effect is particularly evident for the longitudinal configuration.

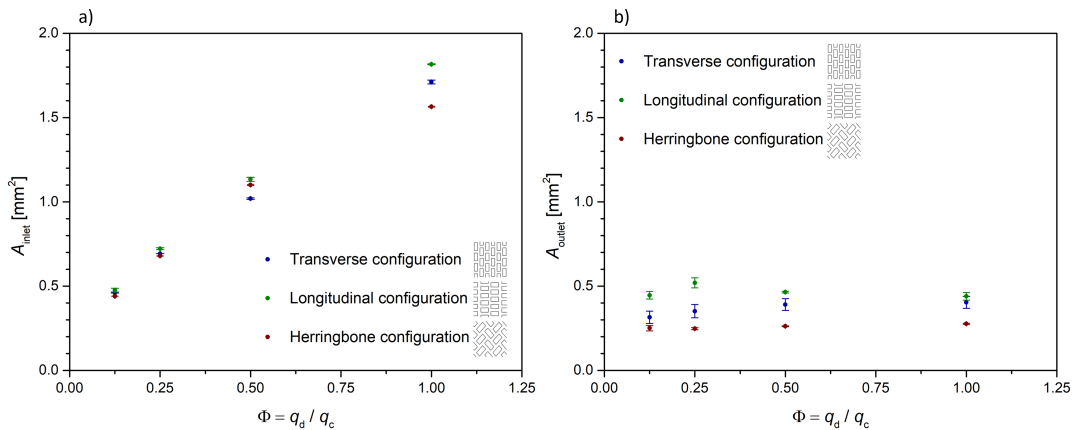


Figure 4.13. Mean droplet area and standard deviation for each configuration and $\emptyset$, based on normal distributions with $X = 30$ and $\text{Ca}_d = 2.38 \times 10^{-2}$: a) inlet region (A_{inlet}); and b) outlet region (A_{outlet}).

To evaluate the effectiveness of the obstacles in fragmenting droplets, the ratio between droplet diameters at the outlet (D) and the gap distance between two obstacles (y_0 from Figure 4.5) was calculated for all configurations and $0.125 \leq \emptyset \leq 1$. The value of Ca_d was kept constant at 2.38×10^{-2} and $X = 30$. A ratio of 1 indicates that the droplet diameter equals the gap distance. For the three configurations studied, most values were below 1, demonstrating that the outlet droplet diameters are smaller than the obstacle spacing and confirming the effectiveness of the configurations in

promoting breakup. Figure 4.14 shows the D/y_o distributions, revealing two main observations: 1) $\emptyset$ has minimal effect on D/y_o ; and 2) D/y_o is dependent on the obstacle configuration. These trends are consistent with the findings discussed in Figure 4.13b, where outlet droplet area was shown to depend on the channel geometry rather than the flow rate ratio. Additionally, the majority of the distributions follow a normal distribution at the outlet, as shown in Figure 4.14.

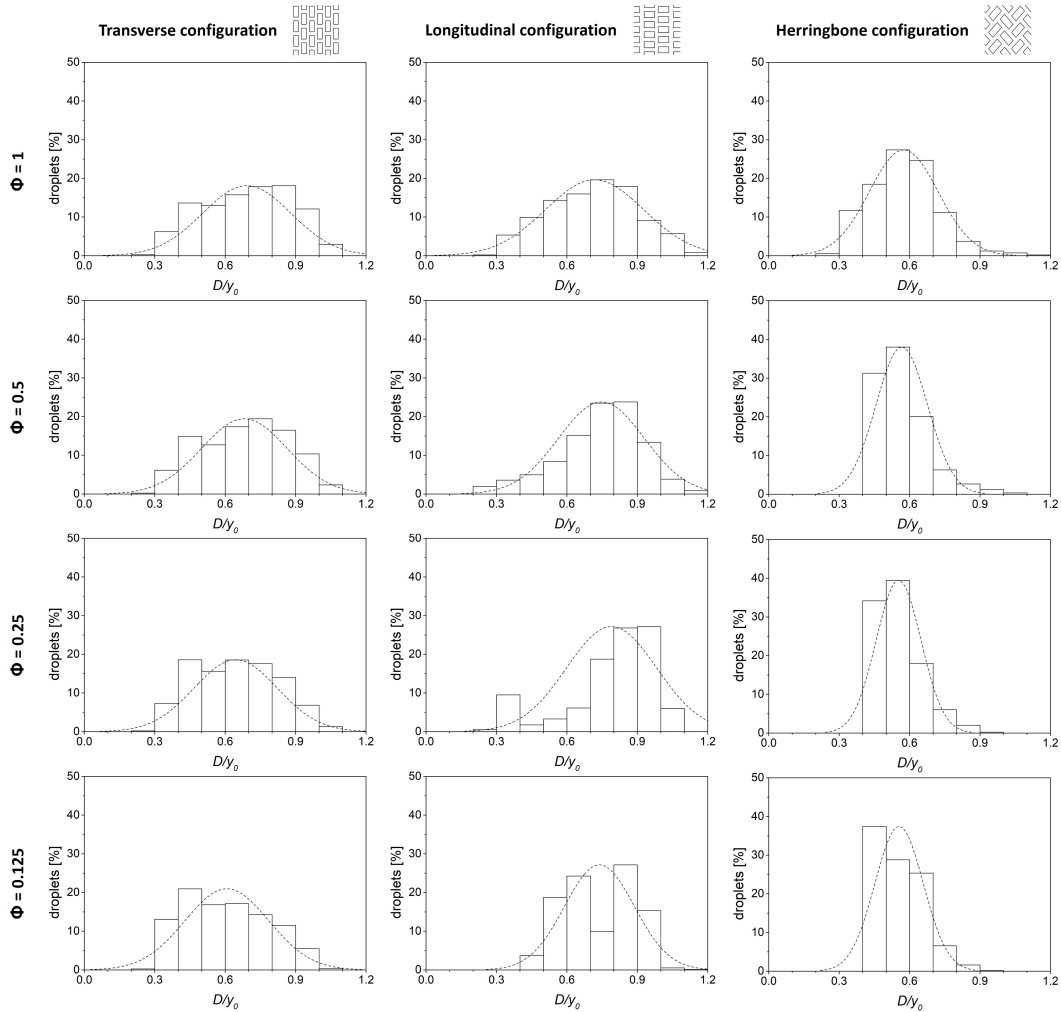


Figure 4.14. Dimensionless droplet diameter (D/y_o) at the outlet region for the three millichannel configurations considered in this study. The conditions addressed included four distinct $\emptyset$ values, $X = 30$, and $Ca_d = 2.38 \times 10^{-2}$. The superimposed dashed line represents a fitted normal distribution curve.

In Figure 4.15, it is possible to confirm that the mean D/y_0 is almost the same regardless of the flow rate ratio used for each configuration. As already mentioned, herringbone configuration can further reduce the size of the droplets, being followed by transverse configuration and longitudinal configuration.

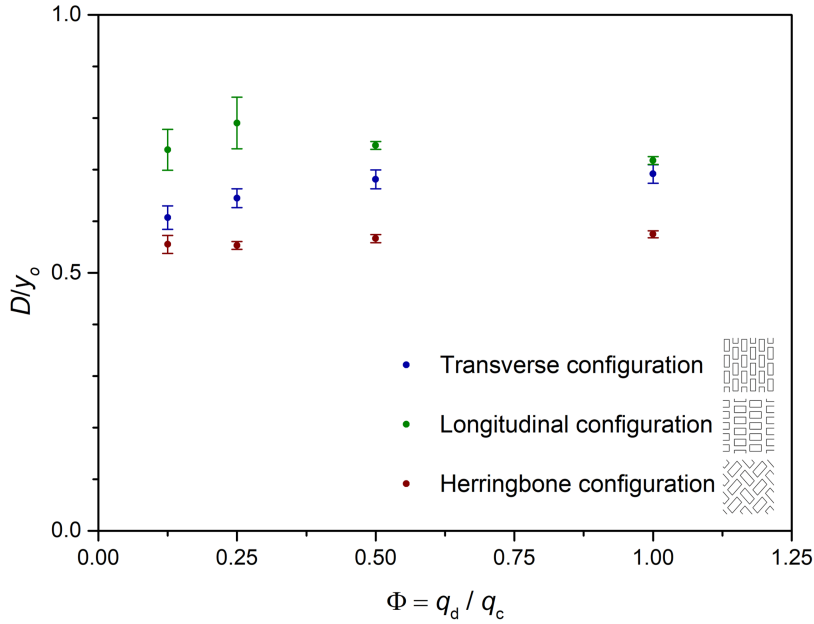


Figure 4.15. Dimensionless mean droplet diameter (D/y_0) and standard deviation for each configuration and Φ , based on normal distributions (Figure 4.14) with $X = 30$ and $Ca_d = 2.38 \times 10^{-2}$ at the outlet region.

4.3.4. Effect of porosity (ϵ)

Figure 4.16 presents the evolution of the mean droplet area along the streamwise direction x of the millichannel. These results concern the three configurations under study and the four flow rate ratios already considered with $X = 30$ and $Ca_d = 2.38 \times 10^{-2}$. In all cases, a consistent trend is observed: droplet area decreases progressively along the array of obstacles region, reaching a plateau approximately 15 mm downstream, which then persists until the end of the channel. This behavior indicates that the length of the array of obstacles is sufficient to achieve the minimal equilibrium droplet area observed at the outlet. The trend reflects the cumulative effect

of hydrodynamic interactions within the obstacles region, which act to restructure the dispersed phase toward a characteristic geometry-specific droplet area.

At the highest flow rate ratio (i.e., $\emptyset = 1$), droplets are initially larger and undergo more pronounced reductions in area along the channel, consistent with stronger deformation and breakup mechanisms. Conversely, at the lowest flow rate ratio (i.e., $\emptyset = 0.125$), droplets are small and show minimal area variation along x , suggesting that equilibrium is reached almost immediately, and that inlet and outlet areas are very similar. These observations align well with the conclusions drawn from Figure 4.13.

As previously noted, the systems addressed exhibit similar porosities (ε): $\varepsilon = 55\%$ for the transverse array and $\varepsilon = 56\%$ for the longitudinal and herringbone arrays. This may explain why droplet area stabilizes at approximately the same value ($x \approx 15$ mm) in all three configurations.

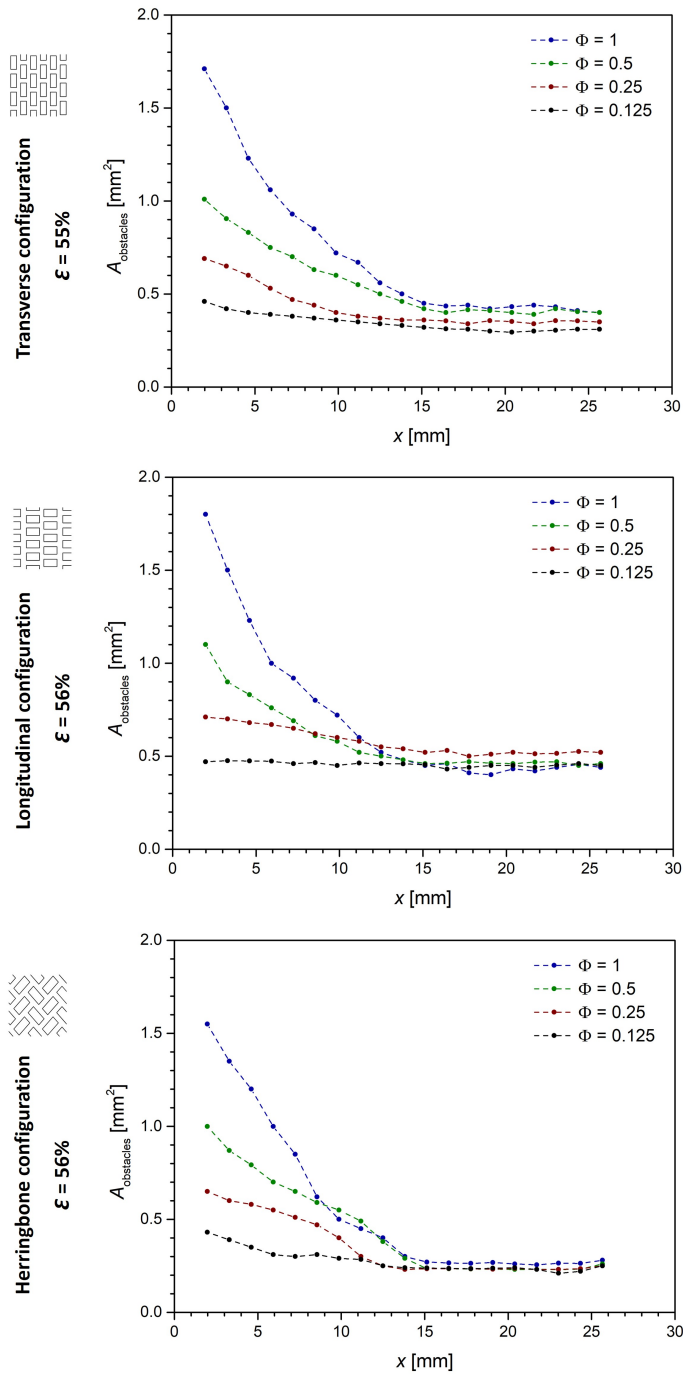


Figure 4.16. Mean droplet area distribution at the array of rectangular obstacles region ($A_{obstacles}$) along the position x of the millichannel (i.e., in the flow direction) for the three configurations under study with $X = 30$ and $Ca_d = 2.38 \times 10^{-2}$.

To investigate the effect of porosity on droplet area, two new geometrical configurations were designed (Figure 4.17) and experimentally tested for the same flow conditions (i.e., $X = 30$, $Ca_d = 2.38 \times 10^{-2}$, and $0.125 \leq \emptyset \leq 1$). Both feature a transverse array similar to that shown in Figure 4.5, differing only in the size of the rectangular obstacles (l_o e w_o), while all other geometric parameters listed in Table 4.1 remain unchanged. The obstacle dimensions for the three transverse arrays, along with their corresponding ε values, are reported in Table 4.3.

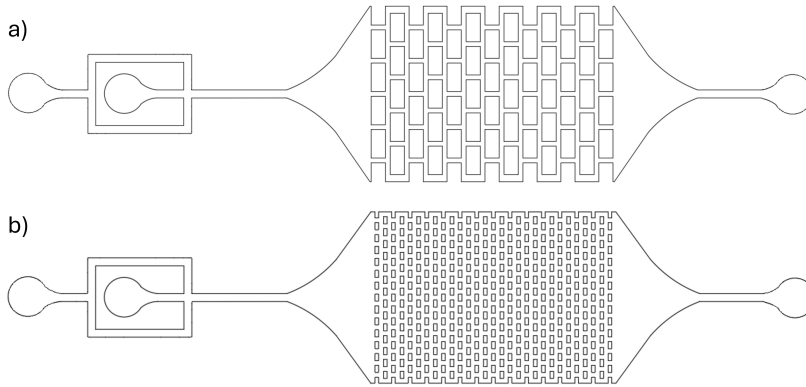


Figure 4.17. Top view of millifluidic devices with a transverse array of rectangular obstacles: a) $\varepsilon = 36\%$; and b) $\varepsilon = 75\%$.

Table 4.3. Dimensions of rectangular obstacles: transverse configuration from Figure 4.5 ($\varepsilon = 55\%$) and configurations from Figure 4.17 ($\varepsilon = 36\%$ and $\varepsilon = 75\%$).

Figure	ε	l_o [mm]	w_o [mm]
Figure 4.17a	36%	3.60	1.80
Figure 4.5a	55%	1.80	0.900
Figure 4.17b	75%	0.900	0.450

Following the procedure described at the beginning of Subsection 4.3.4, Figure 4.18 shows the evolution of the mean droplet area along the streamwise direction x of the millichannel for the configurations in Figure 4.17 and the four considered $\emptyset$ values. A consistent trend is observed: the droplet area decreases progressively along the

rectangular obstacle array before reaching a plateau at $x \approx 10$ mm for $\varepsilon = 36\%$, and $x \approx 20$ mm for $\varepsilon = 75\%$. As previously noted for the transverse array in Figure 4.5 ($\varepsilon = 55\%$), the plateau was reached at $x = 15$ mm (Figure 4.16).

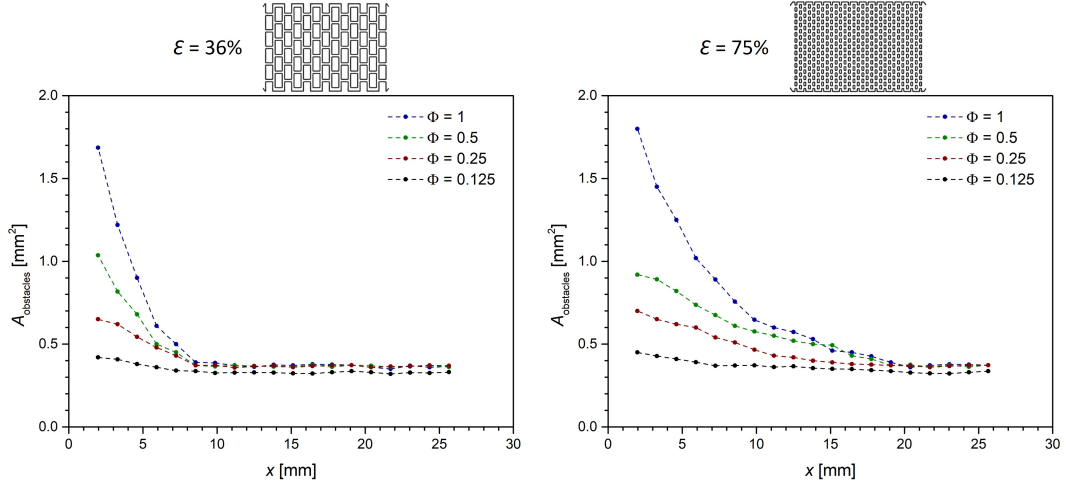


Figure 4.18. Mean droplet area distribution at the array of rectangular obstacles region ($A_{obstacles}$) along the position x of the millichannel (i.e., in the flow direction) for the transverse configurations in Figure 4.17 with $X = 30$ and $Ca_d = 2.38 \times 10^{-2}$.

Lower porosity implies stronger confinement, forcing droplets to undergo greater deformation and increasing their interaction frequency with the obstacles. This confinement amplifies local shear and extensional stresses, effectively lowering the critical Capillary number required for breakup and promoting faster interface destabilization relative to interfacial tension. Additionally, the higher flow resistance associated with low porosity raises the local pressure drop, further intensifying droplet deformation and breakup efficiency. In contrast, higher porosity reduces confinement and droplet-obstacle interactions, resulting in weaker hydrodynamic stresses and a higher effective threshold for breakup, which diminishes breakup efficiency. These conclusions align well with the observations made from [12, 15].

4.3.5. Droplet velocities

Following a similar procedure to the one applied in Subsection 4.3.2, the distributions of the droplet velocities at the outlet region were produced for all configurations and $0.125 \leq \emptyset \leq 1$. The value of Ca_d was kept constant at 2.38×10^{-2} and $X = 30$. The resulting histograms are presented in Figure 4.19, and it is visible that most droplet velocities distributions follow a log-normal behavior.

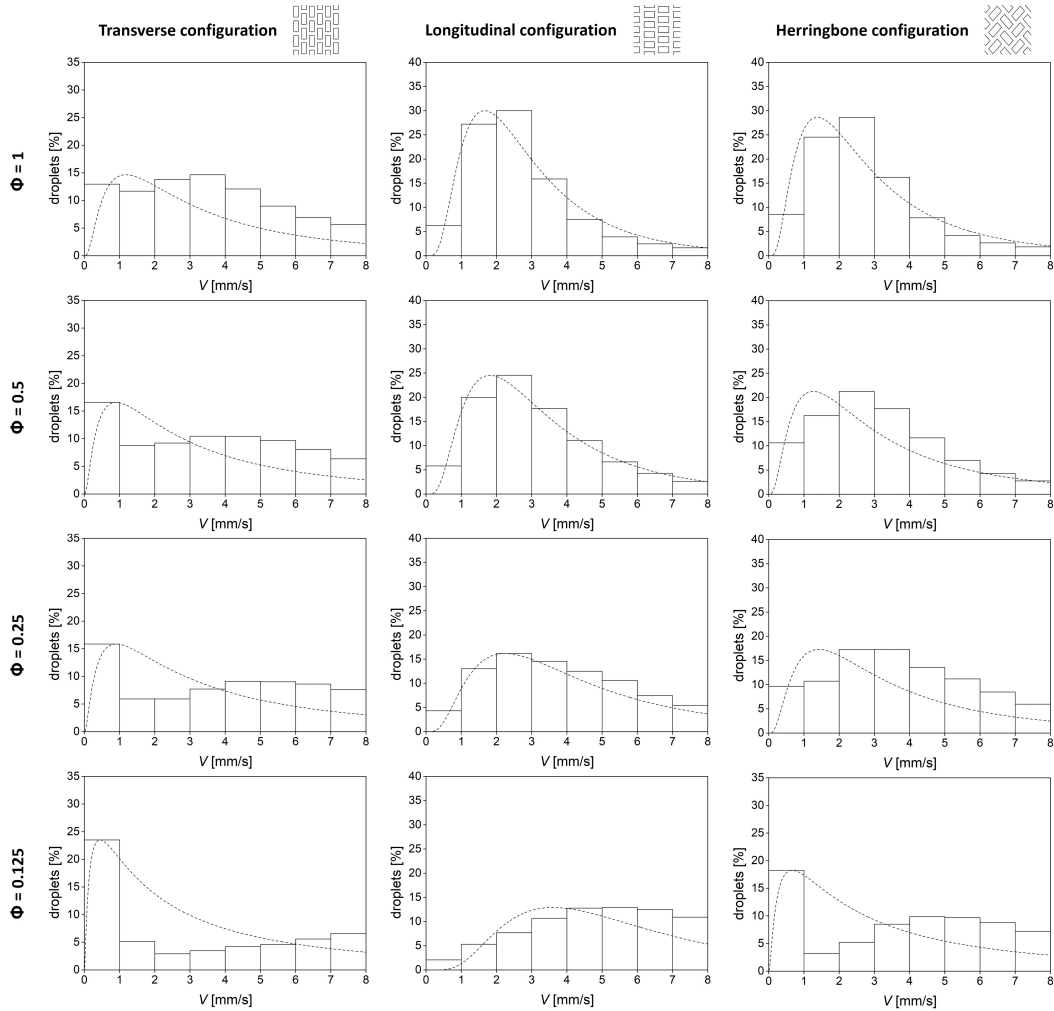


Figure 4.19. Droplet velocity (V) distribution at the outlet region for the three millichannel configurations considered in this study. The conditions addressed included four distinct $\emptyset$ values, $X = 30$, and $Ca_d = 2.38 \times 10^{-2}$. The superimposed dashed line represents a fitted log-normal distribution curve.

From the observation of the histograms in Figure 4.19, two main ideas seem to arise: 1) the droplet velocity at the outlet region (V) decreases as the flow rate ratio increases, noting that this trend applies specifically to the case studied here (i.e., constant q_d with four different q_c values); 2) keeping $\emptyset$ constant, the droplet velocities at the outlet are quite similar in the three configurations. The mean values of all distributions shown in Figure 4.19 were summarized in the plots of Figure 4.20, along with the mean droplet velocity values at the inlet region. For both regions, and for each $\emptyset$ value, the ratios V_{inlet} / V_{mean} and V_{outlet} / V_{mean} are nearly independent of the array configuration (V_{mean} is the average flow velocity at the flow-focusing region). In Figure 4.20a, a slight increase in the dimensionless mean droplet velocity (V_{inlet} / V_{mean}) with $\emptyset$ is observed across all three configurations. In Figure 4.20b, the dimensionless mean droplet velocity (V_{outlet} / V_{mean}) shows a more pronounced increase with $\emptyset$ for all configurations.

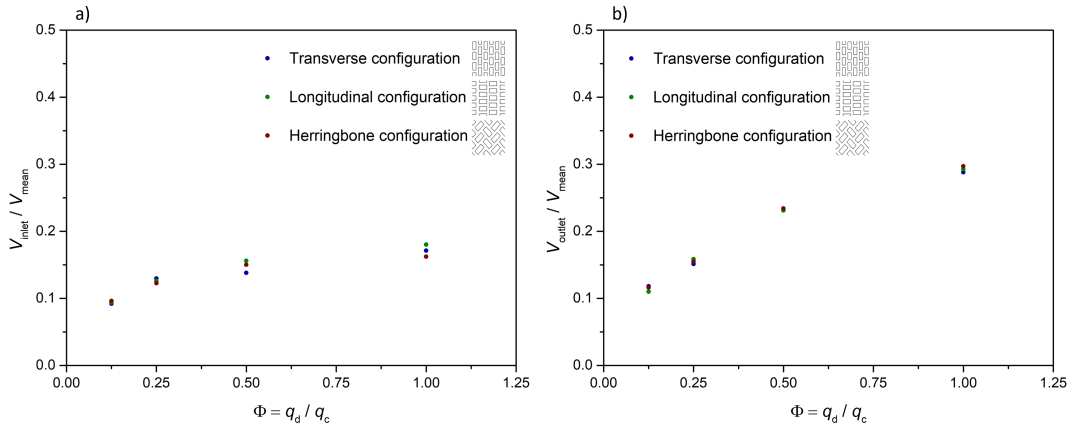


Figure 4.20. Dimensionless mean droplet velocity for each configuration and $\emptyset$, based on log-normal distributions with $X = 30$ and $Ca_d = 2.38 \times 10^{-2}$: a) inlet region (V_{inlet} / V_{mean}); and b) outlet region (V_{outlet} / V_{mean}).

4.3.6. Effect of Ca_d and X

To assess the effect of the Capillary number of the dispersed phase (Ca_d), the nine cases shown in Figure 4.10 for a viscosity ratio of $X = 30$ (green symbols) were compiled in Figure 4.21, where the mean droplet area at the outlet region is plotted for

the herringbone configuration. In addition, nine further experiments were performed using water (colored orange with food dye to enhance phase contrast) as dispersed phase, and mineral oil with SPAN[®] 80 3% w/w as continuous phase, corresponding to a viscosity ratio of $X = 0.026$ (blue symbols). These results address six Φ values (i.e., $0.125 \leq \Phi \leq 4$).

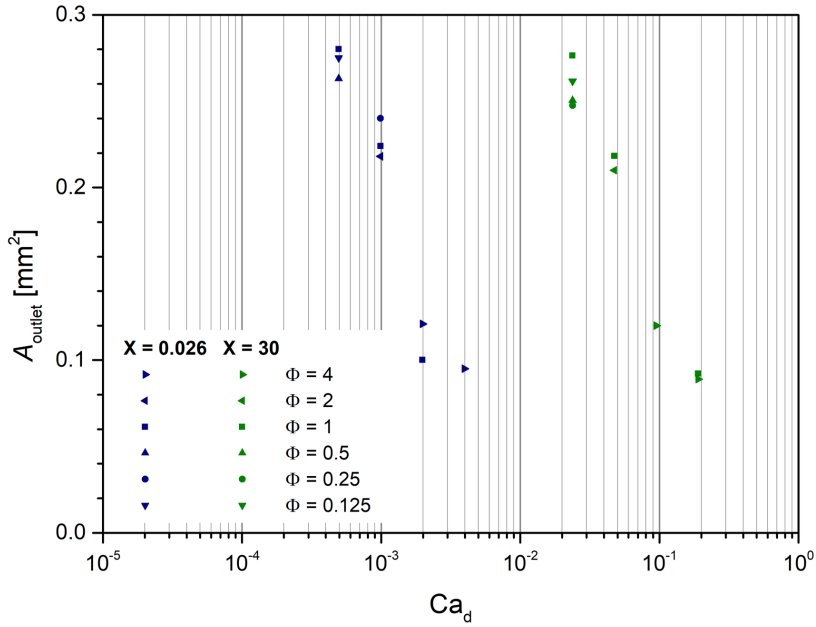


Figure 4.21. Mean droplet area at the outlet region (A_{outlet}) in the herringbone configuration as a function of Ca_d for viscosity ratios $X = 0.026$ (blue symbols) and $X = 30$ (green symbols), within the range $0.125 \leq \Phi \leq 4$.

It should be noted that for $Ca_d = 4.75 \times 10^{-2}$ (second column of Figure 4.10), where large portions of the dispersed phase remained attached to the obstacles, these larger droplets were excluded from the calculation of the mean droplet area at the outlet.

Furthermore, due to the fluid properties employed in the experiments, the same Ca_d values obtained for $X = 30$ could not be replicated for $X = 0.026$.

A clear decreasing trend in droplet size with increasing Ca_d has also been reported in previous studies, where higher values of Ca_d enhance viscous stresses relative to

interfacial tension, thereby promoting droplet deformation and eventual breakup [2, 7, 11, 13].

As previously observed, for different flow rate ratios and identical Ca_d , the droplet areas at the outlet converge to nearly identical values, indicating that the area of the droplets exiting the device becomes almost independent of $\emptyset$. This fact could be easily observed in Figure 4.21 also for $X = 0.026$ and $X = 30$.

Figure 4.21 highlights another remarkable finding: despite the large difference in X values, a specific droplet area at the outlet can be achieved using the same $\emptyset$ value, regardless of the Ca_d . This may suggest that the viscosity ratio between phases does not affect the droplet areas at the outlet region.

4.4. Conclusions

In the present study, the deformation and breakup of droplets flowing through 3D-printed millichannels containing an array of rectangular obstacles with different configurations (i.e., transverse, longitudinal and herringbone) were experimentally investigated. Experiments were performed with two different viscosity ratios between phases: $X = 0.026$ and $X = 30$. This work led to the following key conclusions:

- Array of rectangular obstacles significantly influences droplet deformation and breakup in millichannels;
- Stereolithography-based fabrication, quantitative image analysis, and controlled flow conditions enabled robust comparison of the performance between different millichannels configurations;
- Droplet area at the inlet region depends on flow conditions, while distributions at the outlet region converge to geometry-controlled state that is independent of the flow rates;
- Herringbone array most effectively reduces droplet area, followed by transverse and longitudinal arrays;
- Lower-porosity array enhance confinement, droplet-obstacle interactions, and breakup, whereas higher porosity diminishes these effects;

- Droplet formation transitions from uniform dripping at low Ca_d (i.e., $Ca_d = 2.38 \times 10^{-2}$) to unstable jetting at high Ca_d (i.e., $4.75 \times 10^{-2} \leq Ca_d \leq 1.90 \times 10^{-1}$) for $X = 30$. A clear decreasing trend in droplet area at the outlet region with increasing Ca_d was observed for both regimes, confirming the high efficiency of the array of obstacles in promoting droplet breakup.

In summary, the findings established that an array of obstacles is an effective passive strategy for regulating droplet breakup and size uniformity, providing valuable design principles for advanced millifluidic devices in chemical processing, biomedical diagnostics, and environmental monitoring.

Acknowledgements

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Nomenclature

Acronyms	Definition
3D	Three-dimensional
CSV	Comma-Separated Values
DT	Droplet Tracker
PCR	Polymerase chain reaction
SDS	Sodium Dodecyl Sulfate
STL	Stereolithography
TWS	Trainable Weka Segmentation
UV	Ultraviolet

Greek	Definition	Units
ε	Porosity	[%]
μ	Dynamic viscosity	[Pa s]
μ_c	Dynamic viscosity of the continuous phase	[Pa s]
μ_d	Dynamic viscosity of the dispersed phase	[Pa s]
ρ	Fluid density	[kg m ⁻³]
ρ_c	Density of the continuous phase	[kg m ⁻³]
ρ_d	Density of the dispersed phase	[kg m ⁻³]
σ	Interfacial tension	[N m ⁻¹]
τ_{xx}	Normal stress in x-direction	[Pa]
τ_{yy}	Normal stress in y-direction	[Pa]
$\emptyset$	Flow rate ratio	—

Latin	Definition	Units
A_{inlet}	Droplet area at the inlet region	[m ²]
$A_{obstacles}$	Droplet area at the array of obstacles region	[m ²]
A_{outlet}	Droplet area at the outlet region	[m ²]
Ca	Capillary number	—
Ca_c	Capillary number of the continuous phase	—
Ca_d	Capillary number of the dispersed phase	—
D	Droplet diameter	[m]

h	Height of the channel	[m]
L	Characteristic length	[m]
l_m	Length of the array of obstacles	[m]
l_o	Length of the obstacle	[m]
q_c	Continuous phase flow rate	[m ³ s ⁻¹]
q_d	Dispersed phase flow rate	[m ³ s ⁻¹]
Re	Reynolds number	—
Re _c	Reynolds number of the continuous phase	—
Re _d	Reynolds number of the dispersed phase	—
U	Flow velocity	[m s ⁻¹]
U_c	Continuous phase velocity	[m s ⁻¹]
U_d	Droplet velocity	[m s ⁻¹]
$V ; V_{outlet}$	Droplet velocity at the outlet region	[m s ⁻¹]
V_{inlet}	Droplet velocity at the inlet region	[m s ⁻¹]
V_{mean}	Average flow velocity at the flow-focusing region	[m s ⁻¹]
w	Width of the channel at the flow-focusing region	[m]
w_m	Width of the array of obstacles	[m]
w_o	Width of the obstacle	[m]
x	Axial coordinate	[m]
$x_o ; y_o$	Gap distance between two obstacles	[m]
X	Viscosity ratio	—

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Production of microparticles with biomedical interest using array of obstacles devices

*Production of microparticles with biomedical interest using array of obstacles devices***Abstract**

The controlled production of biocompatible microparticles with tailored size and transport properties is essential for advanced biomedical applications, including drug delivery, tissue engineering, and blood-analogue systems. The present work, investigates the fabrication of microparticles using a combined milli and microfluidic strategy incorporating arrays of passive obstacles to promote droplet breakup and size refinement. Two fluid systems of biomedical relevance were studied: perfluorodecalin (PFD) droplets dispersed in sodium alginate solution and sodium alginate droplets dispersed in mineral oil. Monodisperse droplets were first generated in a 3D-printed millifluidic flow-focusing device featuring a herringbone array of obstacles and subsequently injected into a polydimethylsiloxane (PDMS) microchannel containing an array of transverse rectangular obstacles.

Droplet breakup was analyzed at three locations: millifluidic outlet, microchannel inlet, and microchannel outlet. At the milliscale, experiments were conducted over a viscosity ratio (X) ranging from 10^{-2} to 10^2 , and Capillary numbers of the dispersed phase (Ca_d) between 10^{-4} and 10^0 . At the millifluidic outlet, droplet area distributions exhibited weak dependence on flow rate and viscosity ratios between the two phases, indicating that geometric confinement dominates breakup behavior. A clear inverse correlation between droplet area and Capillary number of the dispersed phase was observed for each value of X under study. After entering the array of obstacles within the microfluidic channel, droplets underwent successive breakup events, resulting in a diameter reduction from approximately $60\text{ }\mu\text{m}$ at the microchannel inlet to $\approx 10\text{ }\mu\text{m}$ at the microchannel outlet.

These results demonstrate that passive arrays of obstacles provide an efficient and scalable hydrodynamic mechanism for producing uniform micrometric droplets, and independent of viscosity ratio within the explored range. The proposed two-stage strategy can enable the generation of oxygen-carrying PFD microdroplets and alginate-based microparticles with dimensions suitable for biomedical applications, including oxygen carriers and cell encapsulation.

5.1. Introduction

The controlled production of functional and biocompatible microparticles represents a central goal in modern biomedical engineering, as microscale materials with tailored morphology, composition and transport properties are instrumental for progress in drug delivery, tissue regeneration and artificial physiological systems [1-3]. Over the past two decades, the integration of polymer chemistry with microfluidic technology has revolutionized the way that such particles are synthesized, enabling the formation of monodisperse droplets under precisely defined hydrodynamic conditions [4-9]. Among the materials explored for biomedical microparticles [2, 3, 10], alginate has gained prominence due to its excellent biocompatibility, biodegradability, low toxicity, low cost of extraction and processing, ionotropic gelation behavior, and non-immune response in the human body [11-15]. Alginate mechanical properties can be easily tuned [13] through the adjustment of alginate concentration, divalent cation concentration, and crosslinking density (influenced by the type of cations used and their interaction with the alginate chain) [11-14]. When coupled with perfluorocarbon compounds such as PFD, alginate systems can be engineered to mimic both the rheological behavior and the oxygen-transporting functions of blood, creating new opportunities for advanced oxygen-therapeutic systems and physiological model fluids [16-19].

As already mentioned, microfluidic droplet generation provides a powerful way for fabricating microparticles under controlled hydrodynamic conditions. Classical device architectures (i.e., T-junctions, co-flow, and flow-focusing [4-6, 20]) allow for continuous production of monodisperse droplets through a balance of viscous and interfacial stresses [4-7, 9]. More recently, passive microchannel modifications, such as the inclusion of arrays of obstacles, have been used to enhance droplet breakup and

mixing without external actuation, improving production throughput and uniformity [21-25].

In the present work, two biomedical-relevant two-phase fluid systems were investigated: PFD droplets dispersed in a sodium alginate solution, and sodium alginate droplets dispersed in mineral oil. Monodisperse droplets were initially generated using a 3D-printed millifluidic flow-focusing device equipped with a herringbone obstacle array. These droplets were then injected into a PDMS microchannel containing a transverse array of rectangular obstacles. Droplet breakup was systematically analyzed at three key locations along the flow path: the millifluidic device outlet, the microchannel inlet, and the microchannel outlet (as illustrated later in Figure 5.4). At the microchannel outlet, droplets of approximately 10 μm were obtained; from a biomedical standpoint, the production of small, monodisperse droplets is particularly relevant for applications such as oxygen delivery systems and cell encapsulation.

5.2. State of the art

The following subsections provide a state-of-the-art overview of the biomedical relevance of alginate and PFD, briefly review encapsulation strategies that combine these materials, and examine how arrays of obstacles in microfluidic devices facilitate the production of microparticles with biomedical applications.

5.2.1. Alginate: structure and biomedical applications

Alginate is a naturally derived, linear polysaccharide composed of β -D-mannuronic (M) and α -L-guluronic (G) acid residues arranged in varying block sequences. Its ability to form hydrogels through ionic crosslinking with divalent cations such as Ca^{2+} or Ba^{2+} has made it one of the most widely used biomaterials for encapsulation technologies [11, 26-31]. The gelation mechanism, often described by the “egg-box” model, enables mild processing under physiological conditions, preserving the functionality of encapsulated drugs, proteins and cells [11, 26]. The resulting hydrogels are biocompatible, biodegradable and mechanically tunable, with

permeability that can be modulated by polymer concentration, M/G ratio and crosslinking ion type [11, 26]. Due to these properties, alginate microparticles have been extensively studied as carriers for drugs, proteins, and growth factors, enabling diffusion-controlled or stimuli-responsive release [11, 26, 32]. In drug-delivery applications, alginate microparticles have been used to encapsulate antibiotics, anticancer drugs and peptides, and controlling delivery of bioactive molecules [11, 26-29].

Beyond controlled release, alginate hydrogels have been used to encapsulate living cells for transplantation and bioprocessing [11, 26]. Their mild crosslinking chemistry maintains cell viability, while the hydrated polymer matrix permits diffusion of oxygen and nutrients yet preventing immune rejection [11, 26]. Alginate's transparency, porosity, and mechanical softness make it an excellent three-dimensional microenvironment for cell culture and encapsulation [11, 26-31].

5.2.2. Perfluorodecalin: structure and biomedical applications

Perfluorodecalin ($C_{10}F_{18}$) is a fully fluorinated bicyclic compound distinguished by extraordinary chemical and thermal stability, high density, and exceptional solubility for respiratory gases. The physicochemical properties of PFD derive from its dense fluorine shell, which creates weak intermolecular van der Waals interactions, low surface tension and a high capacity for dissolving oxygen ($\approx 403 \text{ mL O}_2 \text{ L}^{-1}$ at 25°C) and carbon dioxide [16, 33-35].

Early research by Clark and Gollan [33] demonstrated that mammals could survive while breathing oxygen-saturated perfluorocarbon liquids, establishing the basis for liquid breathing. Riess [16] developed the fundamental principles of perfluorocarbon emulsions for biomedical use, emphasizing their chemical inertness and rapid gas-exchange kinetics. Subsequent generations of PFD-based emulsions, often stabilized with phospholipid or block-copolymer surfactants, demonstrated improved biocompatibility and were exploited as oxygen-therapeutic emulsions, topical wound dressings and experimental blood substitutes [36-41]. The use of neat PFD liquids in ophthalmic surgery, particularly as temporary vitreous substitutes to stabilize retinal detachments, remains an established medical practice, benefiting from PFD's high

density and optical transparency [38]. In tissue engineering and regenerative medicine, perfluorocarbon emulsions have been incorporated within hydrogel matrices to overcome oxygen-transport limitations in three-dimensional cultures [32, 42, 43]. Embedding PFD microdroplets within polymer networks increases the effective oxygen-diffusion coefficient and reduces hypoxic gradients, leading to improved viability of encapsulated cells such as hepatocytes or pancreatic islets [32, 42].

Nonetheless, there are several challenges in stabilizing PFD emulsions, particularly due to their hydrophobic and oleophobic nature. Therefore, alternative approaches for encapsulation, such as the use of alginate offer significant advantages, since encapsulating substances in alginate microparticles improves their stability in aqueous environments [32].

5.2.3. Microfluidic production of microparticles

The fabrication of complex microparticles relies critically on advances in microfluidic technology. The field of droplet-based microfluidics involves the controlled generation of monodisperse droplets through the manipulation of immiscible fluid streams within microchannels operating under laminar flow [4-7]. Droplets can serve as microreactors or templates for solid and gel particles, with their size dictated primarily by the competition between interfacial and viscous forces [4-8, 44, 45]. Common channel configurations used for droplets formation include T-junctions, co-flow and flow-focusing devices, where phase interaction and shear rate govern the transition between dripping and jetting regimes [4-6, 46, 47]. Carneiro *et al.* [46, 47] systematic investigation of PDMS droplet generation under varied viscosity ratios and Capillary numbers established ranges for dripping and jetting transitions. Such microfluidic control enables reproducible synthesis of hydrogel beads with narrow size distributions (coefficient of variation < 2%), a feat unattainable by conventional emulsification methods [4-7, 48].

Moreover, microfluidic encapsulation permits multicore and core-shell configurations through sequential emulsification or coaxial flows, offering new means of combining immiscible materials like alginate and PFD within a single microparticle [32, 36, 37]. These architectures provide new paths for the co-encapsulation of oxygen

carriers such as PFD within alginate matrices, yielding hybrid microparticles that integrate biocompatibility with gas-transport functionality.

Microfluidic encapsulation techniques also facilitate real-time control of physicochemical conditions during particle formation [32, 36, 37]. The small dimensions and high surface-to-volume ratios intrinsic to microchannels lead to rapid mass transfer and heat dissipation, promoting gentle processing conditions for sensitive biomolecules and living cells. Consequently, microfluidics is increasingly recognized as the most powerful technology for producing microparticles with reproducible physicochemical characteristics tailored to specific biomedical applications [2, 3, 10].

5.2.4. Array of obstacles in microdevices and droplet dynamics

Recent studies have provided valuable insights into droplet deformation and breakup in channels with single obstacles of different geometries, highlighting the role of various parameters such as obstacle shape, viscosity ratio, and Capillary number of the dispersed phase [4, 21, 49-54]. However, most of these studies have been limited to single obstacles or simple configurations, while systematic studies involving array of obstacles remain scarce [21, 23, 24].

Array of obstacles induce complex shear and extensional flow fields that deform, fragment or reorganize droplets without external actuation [21-24]. Deterministic lateral displacement theory, originally formulated for particle separation, describes how obstacles can impart lateral trajectories depending on size and flow streamline topology [22, 43, 55]. Applied to two-phase flows, such arrays enable droplet breakup, coalescence suppression and mixing enhancement [21-24]. These findings align with prior studies showing that obstacle-induced elongational stresses can overcome interfacial tension even at low Capillary numbers, offering an efficient passive mechanism for emulsification [21-24]. Moreover, by systematically varying the flow conditions, as well as the geometrical configuration of the array of obstacles and its porosity, it was shown how an array of obstacles can be employed as an efficient passive strategy to promote droplet breakup and produce droplets with uniform sizes.

In addition, a robust methodology for quantitative analysis of droplet area and velocity distributions was already developed during the work of this PhD thesis [25].

Collectively, the synergy between alginate's biocompatible hydrogel chemistry, PFD's exceptional oxygen solubility, and the precision of array of obstacles microdevices has the potential to provide a powerful framework for engineering microparticles with biomedical relevance. The ongoing refinement of passive microfluidic geometries, combined with advances in materials formulation and in-channel gelation strategies, holds promise for the scalable manufacturing of oxygen-carrying, drug delivery systems, and physiologically relevant model fluids.

5.3. Experimental techniques and procedures

Experiments at the microscale were conducted using an array of transverse rectangular obstacles (Subsubsection 5.3.1.2) with two fluid pairs. The physical properties of both phases are reported in Subsection 5.3.2.

A two-stage milli-to-microfluidic approach was designed to achieve controlled production of microparticles. In the first stage, monodisperse droplets were produced using a 3D-printed millifluidic flow-focusing device equipped with a herringbone obstacle array. The droplets were subsequently injected into a microfluidic channel with transverse obstacles, where their transport and breakup were further analyzed (Subsubsection 5.3.1.1).

5.3.1. Fabrication methods of the fluidic devices

The geometry design of all fluidic devices used in the experiments was made with Autodesk AutoCAD 2022®. Two different manufacturing methods were employed: 1) stereolithography (SLA) 3D printing technique for the millichannel (Subsubsection 5.3.1.1); and 2) soft lithography for the microchannel (Subsubsection 5.3.1.2).

5.3.1.1. Milliscale fluidic device for droplet generation

The millifluidic device was manufactured by the stereolithography 3D printing technique [56]. The 3D printer used was a Form 3 from Formlabs® and the material chosen was the transparent and hydrophilic Clear Resin V4®. A detailed explanation of this channel fabrication method and the main dimensions of the herringbone array were presented in the previous work of Cerdeira *et al.* [25].

Two fluid pairs were used in the experiments, as summarized later in Table 5.2. Since Clear Resin V4® is hydrophilic, no surface treatment was required when working with PFD droplets. However, surface treatment was necessary to enable the formation of sodium alginate aqueous droplets. For that purpose, a car glass water repellent was injected into the millifluidic device. After a few minutes of contact and a subsequent rinse with water, the millichannel walls were hydrophobic and ready for the experiments.

The millichannel comprise a flow-focusing region for droplet generation followed by a region with obstacles – herringbone configuration (Figure 5.1). The flow-focusing part is composed by two inlets, one for the continuous phase and the other for the dispersed phase. The dispersed and continuous phases were injected into the millifluidic device with the help of two syringe pumps (Harvard Apparatus PHD ULTRA™).

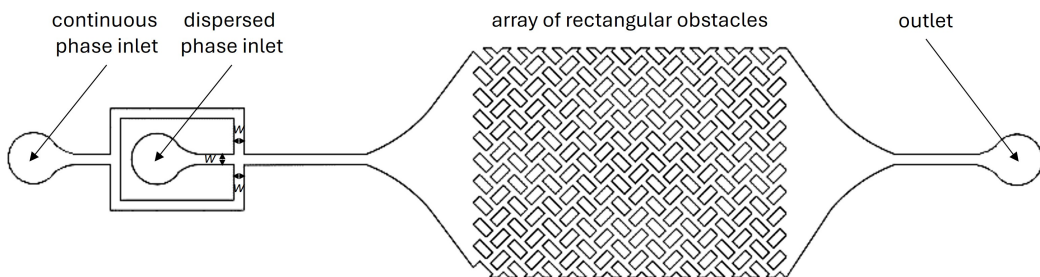


Figure 5.1. Schematic top view representation of the millifluidic device used for droplet generation: $w = 1.00$ mm. The microchannel height (h) is 1.50 mm.

5.3.1.2. *Microfluidic device with an array of rectangular obstacles*

The design of the microfluidic device (Figure 5.2) was made in Autodesk AutoCAD 2022®, and the resulting computer-aided design (CAD) file was sent to a specialized laboratory (International Iberian Nanotechnology Laboratory – INL, Braga, Portugal) to produce the corresponding mold. This mold was fabricated by the combination of direct write laser lithography and reactive ion etching [57]. Additionally, the mold was ordered with 30 μm height.

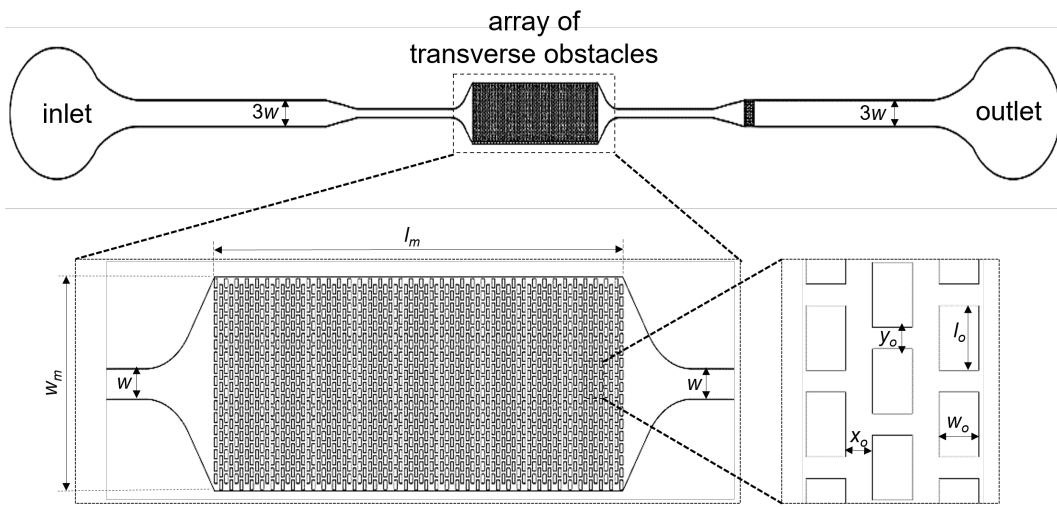


Figure 5.2. Schematic top view representation of the microfluidic device with an array of transverse obstacles. The microchannels height (h) is 30 μm , and the other main channels dimensions are listed in Table 5.1.

After the design and the production of the mold, the following step was the fabrication of the microchannel. Microfluidic research devices are generally fabricated in PDMS, which is a silicone elastomer, chemically inert, thermally stable, hydrophobic and transparent [58-60].

Microchannels were fabricated by soft lithography using a two-part kit composed by a PDMS pre-polymer and a cross-linker/curing agent (Dow Corning Sylgard® 184 kit) [47, 58, 61]. A well-mixed solution of PDMS and curing agent in a ratio of 5:1 was poured into the mold, and a thin layer of PDMS and curing agent in a ratio of 20:1 was spread by spin-coating in a glass slide (for sealing channels). This ensures that the entire microchannel walls material have the same wetting properties. Afterwards, the

PDMS mold and the coated glass slide were cured in an oven at 80 °C for 20 minutes. Then, the cured PDMS was peeled off from the mold and sealed to the coated glass slide. To ensure a good sealing, the microchannel was left in the oven to cure for approximately 12 hours at 80 °C.

When working with PFD droplets, air plasma surface treatment was applied to change the PDMS surface using a low-pressure Plasma Reactor (Diener[®] electronic GmbH, model ZEPTO). The intention was to change the surface behavior from hydrophobic to hydrophilic in order to have more affinity with the continuous sodium alginate aqueous phase. This technique is one of the most efficient ways to modify the PDMS surface because of the short treatment time and the easiness of operation. However, it is important to notice that approximately two hours after air plasma surface treatment, the microchannel reverts to hydrophobic behavior; therefore, experiments should be conducted promptly after the treatment. This phenomenon is due to the migration of uncured PDMS oligomers from the bulk to the surface and the rearrangement of highly mobile polymer chains at room temperature to minimize the surface energy [62].

The main dimensions of the microfluidic device represented in Figure 5.2 are listed in Table 5.1 (CAD file and INL mold). To confirm if the microchannel height corresponded to what was expected, a cross-section of the microchannel was cut, placed in a glass slide, and observed using a microscope (DMI 5000M, Leica Microsystems GmbH). The dimensions were measured using the Photron FASTCAM Viewer Software[®] (PFV4), and these data are also presented in Table 5.1.

Table 5.1. Microfluidic device with an array of transverse obstacles (Figure 5.2): CAD file and INL mold dimensions.

Microchannel dimensions	w [μm]	h [μm]	l_m [μm]	w_m [μm]	x_o [μm]	y_o [μm]	l_o [μm]	w_o [μm]
CAD file	100.0	30.00	1988	700.0	10.00	10.00	30.00	15.00
INL mold	98.8	26.4	1967	692	9.0	9.3	29.3	14.4

5.3.2. Working fluids

Two fluid pairs were used in the experiments: 1) sodium alginate (Sigma-Aldrich® W20152) 2% w/w aqueous solution droplet in mineral oil (Sigma-Aldrich® 330779 light mineral oil) with 5% w/w SPAN® 80 (Sigma-Aldrich® 85548); and 2) perfluorodecalin (Sigma-Aldrich® P9900) droplet in a sodium alginate 3% w/w aqueous solution with TWEEN® 80 (Sigma-Aldrich® P1754) 1% w/w. The surfactants (i.e., SPAN® 80 and TWEEN® 80) were used to prevent coalescence.

The test fluids were characterized regarding the relevant physical properties: 1) viscosity (μ) of both Newtonian phases was measured by a rotational rheometer (Physica MCR301, Anton Paar) with a Peltier temperature control system set at 20 °C; 2) volumetric mass (ρ) of both phases was determined with a density probe at ~23 °C; 3) interfacial tension (σ) was also obtained at ~23 °C in a Du Noüy ring tensiometer (Force Tensiometer Sigma 701).

The experimental data gathered for the referred physical properties are listed in Table 5.2.

Table 5.2. Physical properties of the fluids.

Phases	Fluids	μ [Pa s]	ρ [g/cm ³]	σ [mN/m]	$X=\mu_d/\mu_c$
Continuous	Mineral oil + SPAN® 80 5% w/w	0.0349	0.8568		
Dispersed	Sodium alginate 2% w/w aqueous solution	0.0322	1.008	3.247	0.92
Continuous	Sodium alginate 3% w/w aqueous solution + TWEEN® 80 1% w/w	0.0821	1.022	11.41	0.062
Dispersed	Perfluorodecalin	0.00510	1.923		

5.3.3. Flow visualization and characterization

After droplet generation at the millifluidic device of Figure 5.1, the droplets were collected and then injected into the microchannel with the array of obstacles (Figure 5.2) using a syringe pump operating in withdrawal mode to minimize sedimentation effects during the injection (neMESYS CETONI GmbH).

Droplet images were captured via a high-speed camera (FASTCAM Mini UX100, Photron) mounted on an inverted epifluorescence microscope (DMI 5000M, Leica Microsystems GmbH). A schematic representation of the experimental setup is shown in Figure 5.3.

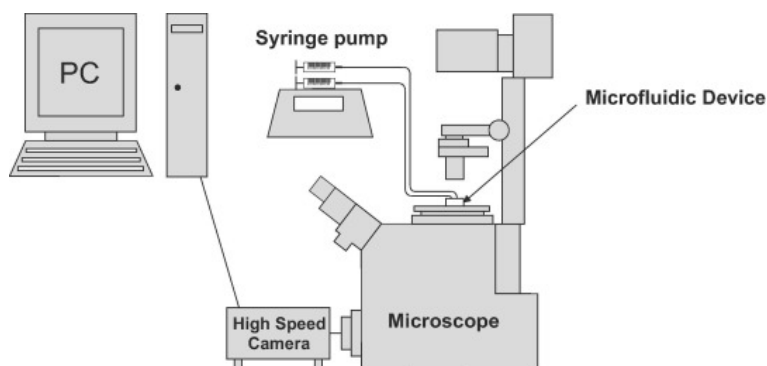


Figure 5.3. Schematic representation of the experimental setup. Adapted from Carneiro *et al.* [46].

Droplets position and area were determined using the Droplet Tracker (2013) [63], which is an ImageJ[®] (version 1.52a) plugin. Prior to applying the Droplet Tracker, a segmentation of all acquired images (i.e., black and white images in which droplets are represented in black and the obstacles/empty spaces of the channel are in white) was performed through the use of Trainable Weka Segmentation (version 3.9.4) [64], which is a Fiji[®] (version 1.53q) plugin. The post-processing of the obtained results was made through a Python script. A detailed description of the images processing method is presented in the previous work of Cerdeira *et al.* [25].

5.4. Results and discussion

This section includes a systematic analysis of droplet size distributions obtained at different regions of the experimental apparatus: 1) at the outlet of the millifluidic device used to generate the droplets – Figure 5.4a; 2) at the inlet of the microchannel with an array of transverse obstacles – Figure 5.4b; and 3) at the outlet of that microchannel – Figure 5.4b. Two fluid systems were investigated: PFD droplets in a sodium alginate solution ($X = 0.062$) and sodium alginate droplets in mineral oil ($X = 0.92$).

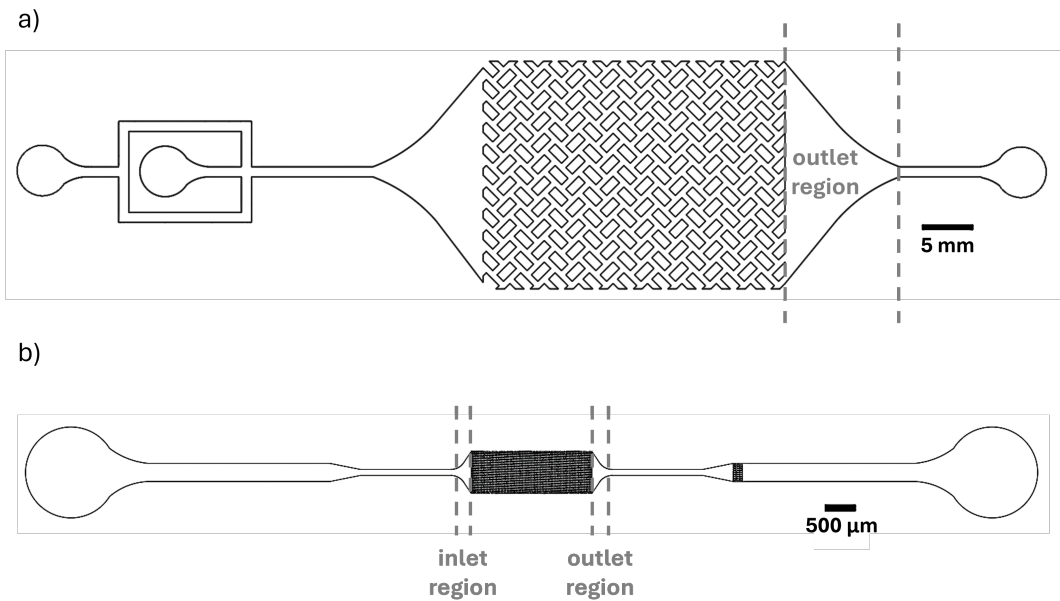


Figure 5.4. Interest regions of the two channels: a) millichannel with the outlet region (after the obstacles); and b) microchannel with the inlet region (before the obstacles) and the outlet region.

At the milliscale, the dispersed phase flow rate (q_d) was fixed at 0.5 mL/min, while four different flow rates were addressed for the continuous phase (q_c): 0.5 mL/min, 1 mL/min, 2 mL/min, and 4 mL/min. Accordingly, four flow rate ratios ($\emptyset = q_d/q_c$), ranging from 0.125 to 1 were considered. The value of Ca_d was maintained constant at 5.31×10^{-4} for $X = 0.062$, and at 1.18×10^{-2} for $X = 0.92$.

To evaluate the influence of Ca_d , q_d was subsequently varied, resulting in $\emptyset$ values ranging from 0.125 to 4, and Ca_d spanning from 10^{-4} and 10^0 .

At the microscale, experiments were conducted at a single, constant flow rate ($q = 1 \mu\text{L}/\text{min}$).

Figure 5.5 shows representative PFD and alginate droplet images captured at the microchannel inlet and outlet regions, illustrating their distributions under steady flow conditions.

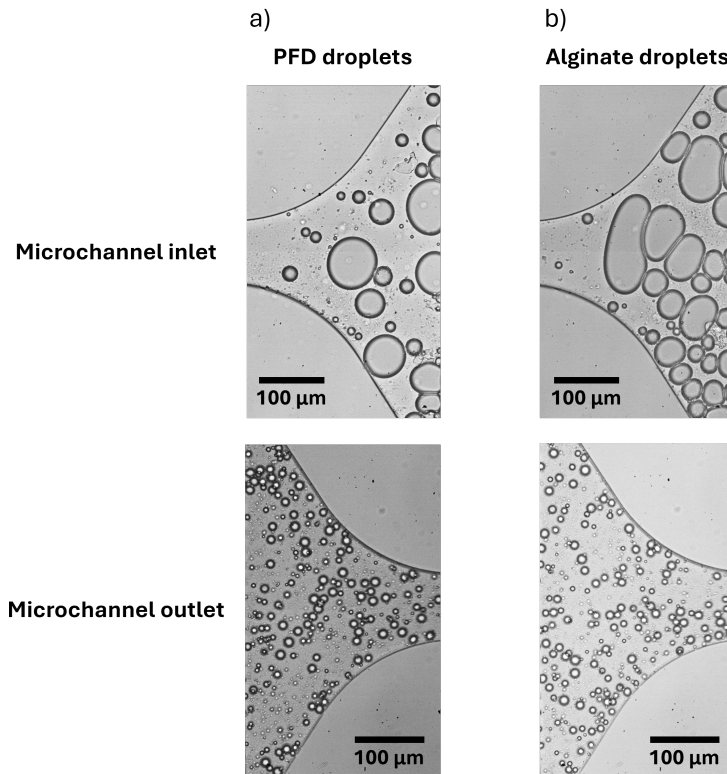


Figure 5.5. Representative droplet images at the inlet, and outlet regions of the microchannel (Figure 5.4b): a) PFD droplets; and b) alginate droplets.

5.4.1. Droplet area at the outlet region of the millichannel

Figure 5.6 presents the droplet area distributions measured at the outlet of the millifluidic device after passing through the herringbone array of obstacles. For comparison, and to consider a viscosity ratio beyond the range addressed in this work, data for mineral oil droplets ($X = 30$ and Capillary number of the dispersed phase $Ca_d = 2.38 \times 10^{-2}$) from the previous study of Cerdeira *et al.* [25] were also included.

The operating conditions for the PFD droplets were $X = 0.062$ and $Ca_d = 5.31 \times 10^{-4}$, and $X = 0.92$ and $Ca_d = 1.18 \times 10^{-2}$ for the sodium alginate droplets.

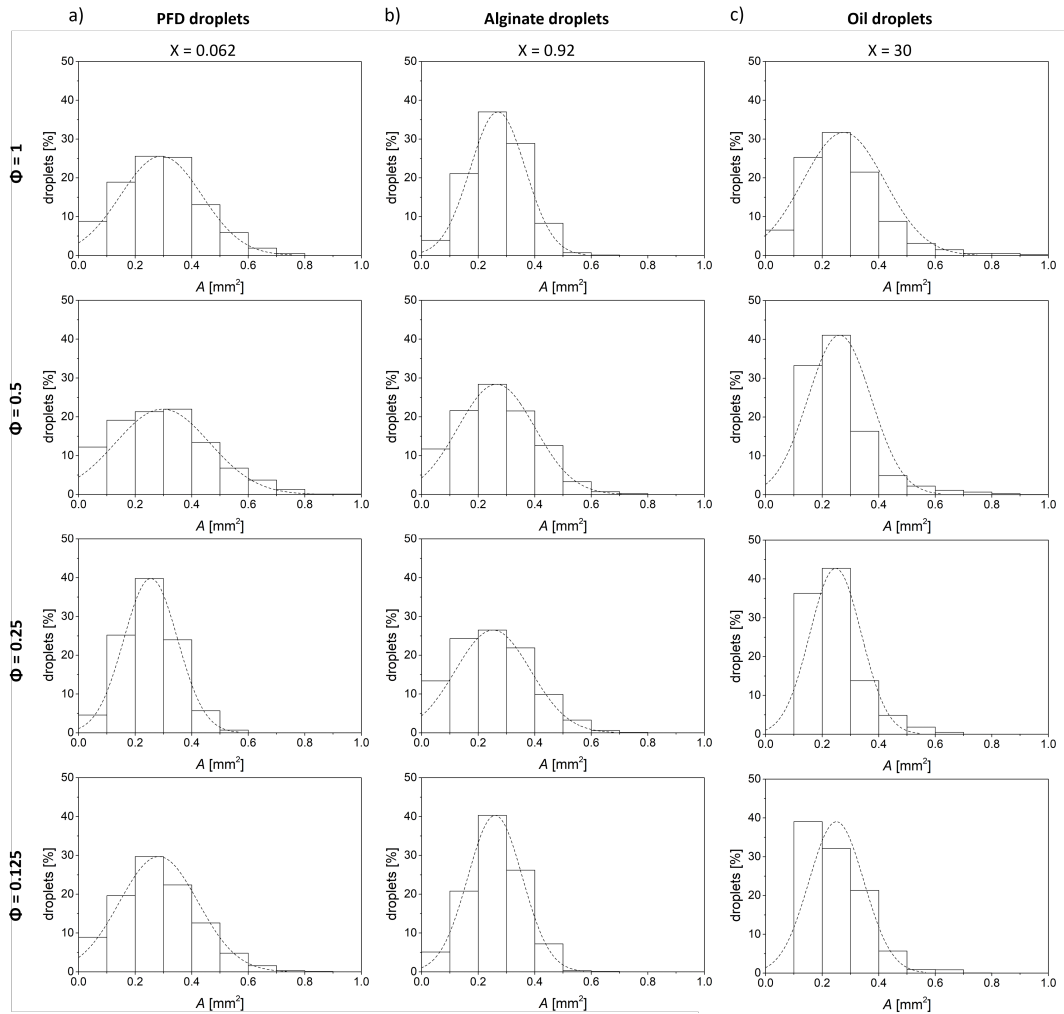


Figure 5.6. Droplet area (A) distribution at the outlet region of the millichannel considering four distinct $\emptyset$ values: a) perfluorodecalin droplets ($X = 0.062$ and $Ca_d = 5.31 \times 10^{-4}$); b) sodium alginate droplets ($X = 0.92$ and $Ca_d = 1.18 \times 10^{-2}$); and c) mineral oil droplets (results from Cerdeira *et al.* [25] with $X = 30$ and $Ca_d = 2.38 \times 10^{-2}$). The superimposed dashed line represents a fitted normal distribution curve.

For all tested flow rate ratios ($\emptyset$) values, the distributions exhibit a nearly Gaussian profile. The low dispersion around the mean confirms the effectiveness of the herringbone configuration in producing monodisperse droplets prior to injection into

the microchannel. As already verified in previous work [25], this suggests that $\emptyset$ exerts minimal influence on the final droplet size after passage through the array of obstacles, with similar mean droplet areas obtained for all $\emptyset$ values.

Furthermore, the experimental data indicate that the viscosity ratio between phases does not significantly affect the droplet area measured at the outlet region of the millichannel. This finding suggests that the array of herringbone obstacles imposes a dominant geometric constraint that overrides rheological differences between fluid pairs. As droplets traverse successive constrictions, they experience repeated elongation and squeezing events governed primarily by channel confinement, obstacle spacing, and local pressure gradients. Under the operating conditions addressed, no droplet merging or coalescence was observed along the millichannel. These geometric effects appear to define a characteristic breakup scale that is relatively insensitive to viscosity contrast within the explored range. Consequently, droplets with markedly different viscosity ratios (i.e., PFD/sodium alginate system and mineral oil/water system) converge toward similar mean outlet areas, as also visible in Figure 5.7.

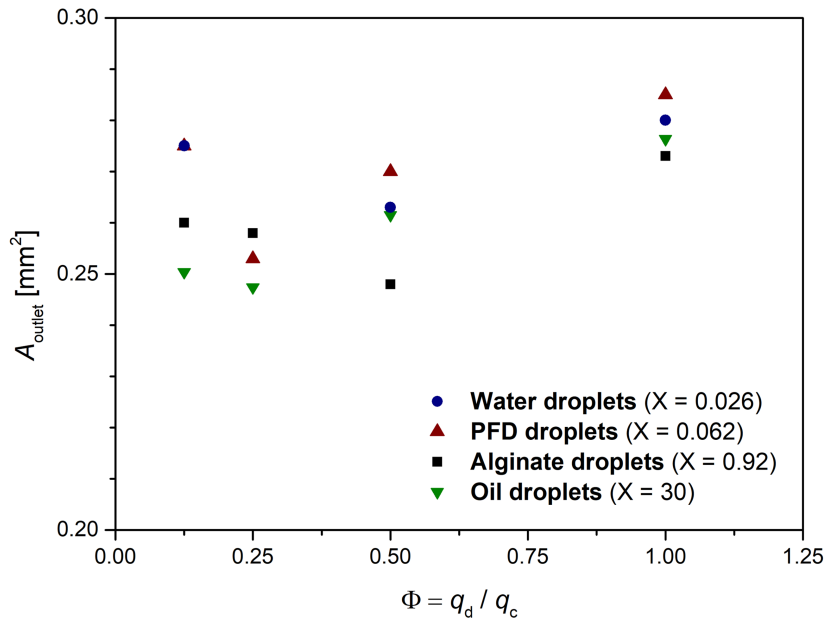


Figure 5.7. Mean droplet area at the outlet region (A_{outlet}) of the millichannel with different fluid droplets and $\emptyset$, based on the normal distributions of Figure 5.6. Water droplet results from Cerdeira *et al.* [25] with $X = 0.026$ and $Ca_d = 4.95 \times 10^{-4}$.

To provide a clear comparison between different fluid systems, Figure 5.7 presents the mean values of all droplet area distributions measured at the outlet of the millichannel (Figure 5.6). Again, for comparison purposes, the results obtained with water droplets ($X = 0.026$ and $Ca_d = 4.95 \times 10^{-4}$) from the previous study [25] were also included. In Figure 5.7, it is visible that the droplet area at the outlet region is almost independent of $\emptyset$.

Figure 5.8 correlates A_{outlet} with Ca_d for all investigated droplet fluids (i.e., water, PFD, sodium alginate, and mineral oil). A clear decrease in droplet area with the increase in Ca_d is observed for each value of X under study. This is in agreement with previous studies reporting the same trend between droplet size and Ca_d , as higher Ca_d values amplify viscous stresses relative to interfacial tension, facilitating droplet deformation and eventual breakup [21, 25, 49, 50, 52].

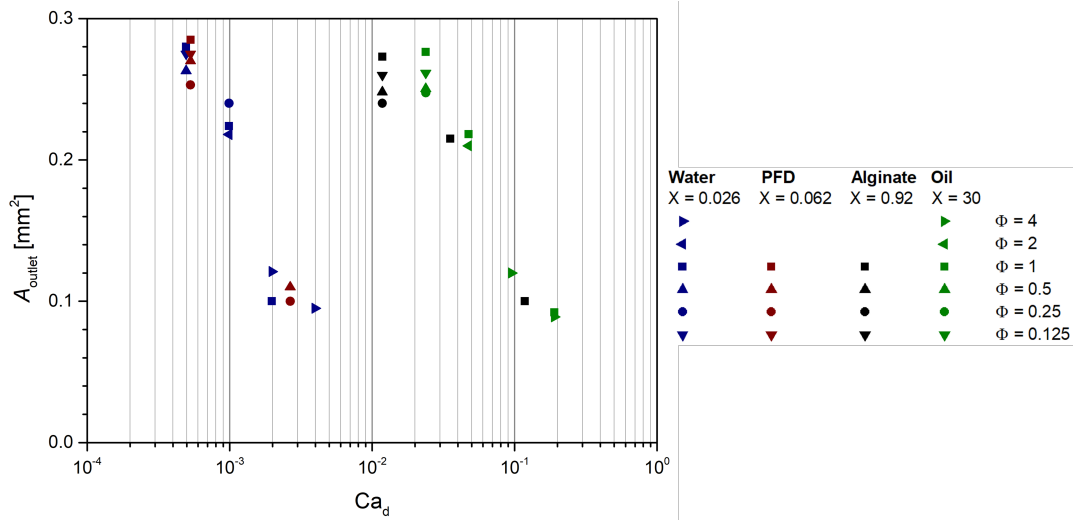


Figure 5.8. Mean droplet area at the outlet region (A_{outlet}) of the millichannel as a function of Ca_d with different fluid droplets: water (blue symbols), PFD (red symbols), sodium alginate (black symbols), and mineral oil (green symbols), within the range $0.125 \leq \emptyset \leq 4$.

Overall, the millifluidic channel provides controlled, reproducible droplet populations with tunable size, suitable for subsequent deformation and breakup studies in the microchannel with an array of obstacles.

5.4.2. Droplet area distributions in the microchannel

The droplets produced at the milliscale were injected into the microfluidic device containing an array of transverse rectangular obstacles, as explained in the experimental techniques and procedures section. It is important to note that in this subsection and the next, $\emptyset$ sample values refer to droplets collected at the exit of the millichannel for a specific $\emptyset$, since in the microscale experiments, a single and identical flow rate ($q = 1 \mu\text{L}/\text{min}$) was used for all experiments.

Figure 5.9 and Figure 5.10 present the droplet area distributions measured at the inlet and outlet regions of the microchannel, respectively.

At the inlet region (Figure 5.9), the droplet area distributions differ from those measured at the outlet of the millifluidic device (Figure 5.6). Specifically, droplet areas at the microchannel inlet are roughly 100 times smaller than the corresponding areas at the millichannel outlet, indicating substantial breakup during transfer between devices and also during the injection process. For the two fluid pairs studied, droplet areas were very similar across the four $\emptyset$ sample values, with PFD droplets (A around $5000 \mu\text{m}^2$) consistently larger than those of sodium alginate (A around $3000 \mu\text{m}^2$). Another notable observation is that droplet area distributions become narrower as the $\emptyset$ sample value decreases. This is likely because lower $\emptyset$ values correspond to a lower droplet flow rate (q_d), resulting in fewer droplets, reduced droplet interactions, less breakup, and consequently narrower area distributions.

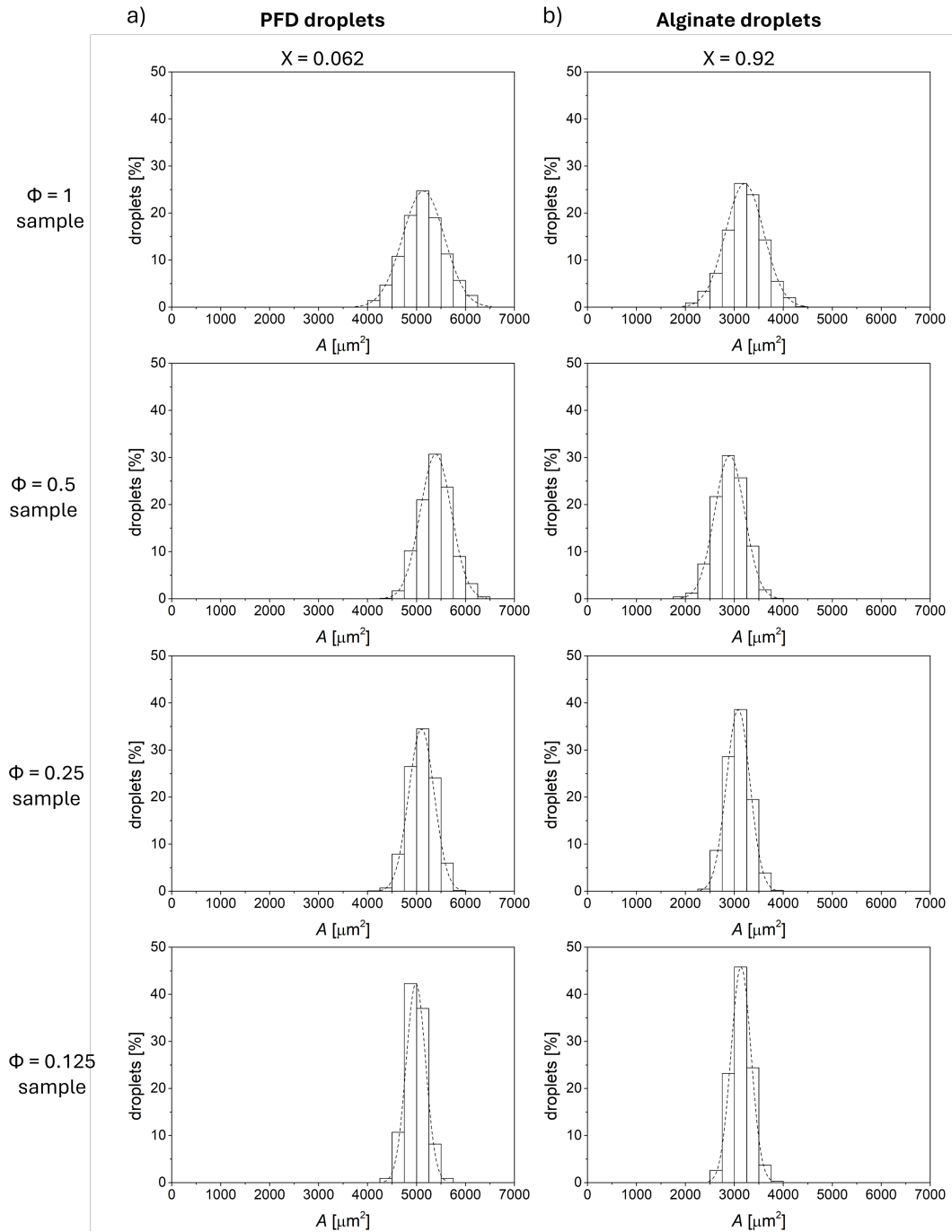


Figure 5.9. Droplet area (A) distribution at the inlet region of the microchannel for four sample Φ values and $q = 1 \mu\text{L/min}$: a) perfluorodecalin droplets ($X = 0.062$ and $\text{Ca}_d = 5.31 \times 10^{-4}$); and b) sodium alginate droplets ($X = 0.92$ and $\text{Ca}_d = 1.18 \times 10^{-2}$). The superimposed dashed line represents a fitted normal distribution curve.

At the outlet region (Figure 5.10), the mean droplet area for each fluid system remains nearly constant across the four $\emptyset$ sample values, indicating that even at the microscale, the flow rate ratio has little influence on the final droplet size after passing through the obstacle array. Furthermore, only minor differences in outlet droplet area are observed for both fluid systems ($A \approx 75 \mu\text{m}^2$), with droplets nearly 55 times smaller than at the microchannel inlet. This demonstrates the high efficiency of the array of obstacles in promoting breakup and generating uniform droplet sizes, despite the different inlet distributions for each fluid pair (i.e., PFD droplets being larger than alginate droplets at the inlet). In addition, as shown in Figure 5.10, the outlet droplet area histograms exhibit a normal distribution.

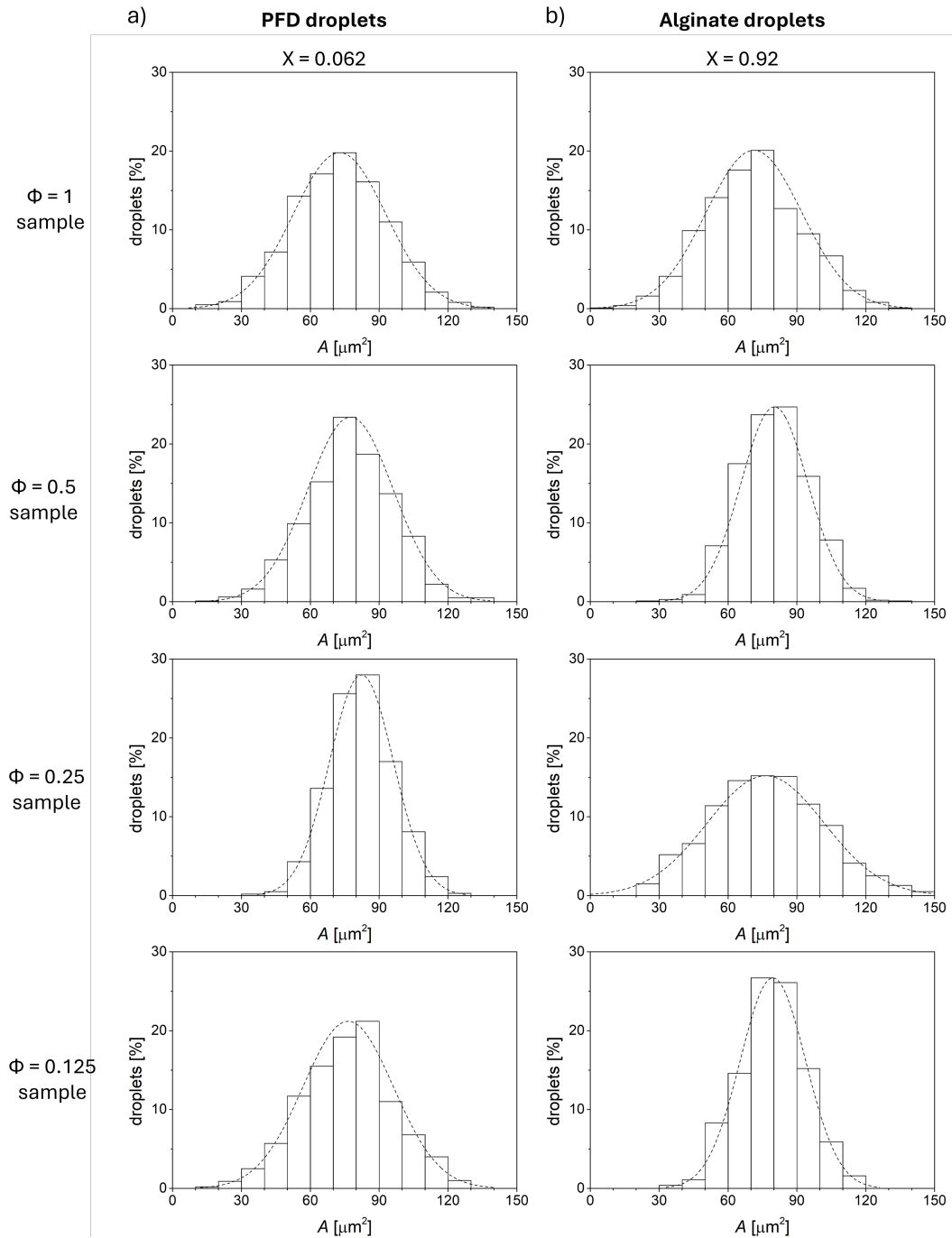


Figure 5.10. Droplet area (A) distribution at the outlet region of the microchannel for four sample Φ values and $q = 1 \mu\text{L/min}$: a) perfluorodecalin droplets ($X = 0.062$ and $\text{Ca}_d = 5.31 \times 10^{-4}$); and b) sodium alginate droplets ($X = 0.92$ and $\text{Ca}_d = 1.18 \times 10^{-2}$). The superimposed dashed line represents a fitted normal distribution curve.

5.4.3. Droplet size comparison in all considered interest regions

Figure 5.11 summarizes the mean droplet diameter (D) measured at three locations for both fluid pairs: 1) outlet of the millifluidic device – red symbols; 2) inlet of the microchannel – blue symbols; and 3) outlet of the microchannel – green symbols. Sodium alginate droplets were denoted by closed circle symbols, whereas PFD droplets were represented by open triangle symbols. It is important to note that two different approaches were used to calculate droplet diameter: 1) for the millichannel and microchannel outlets, droplets were assumed to be spherical, as they do not fill the entire channel; and 2) for the microchannel inlet, where droplets occupy the full channel height, the equivalent spherical diameter was calculated using $D = [(6 \times A \times h)/\pi]^{1/3}$, where A is the mean droplet area from Figure 5.9, and h is the microchannel height. Consequently, the calculation of droplet diameters involves an associated error; however, its influence is expected to be small [65].

At the millichannel outlet, droplets from both fluid pairs exhibited a diameter of approximately 600 μm , consistent with the gap between two obstacles, which is also 600 μm (millichannel dimensions reported in previous work of Cerdeira *et al.* [25]). During transfer and injection into the microchannel, the droplet size decreased, with a mean diameter of about 60 μm – ten times smaller than at the millichannel outlet. This size reduction may be attributed to the transition from the millichannel to the narrower microchannel. Finally, at the microchannel outlet, droplets measured around 10 μm in diameter, corresponding to the gap between two obstacles (i.e., x_o or y_o in Table 5.1).

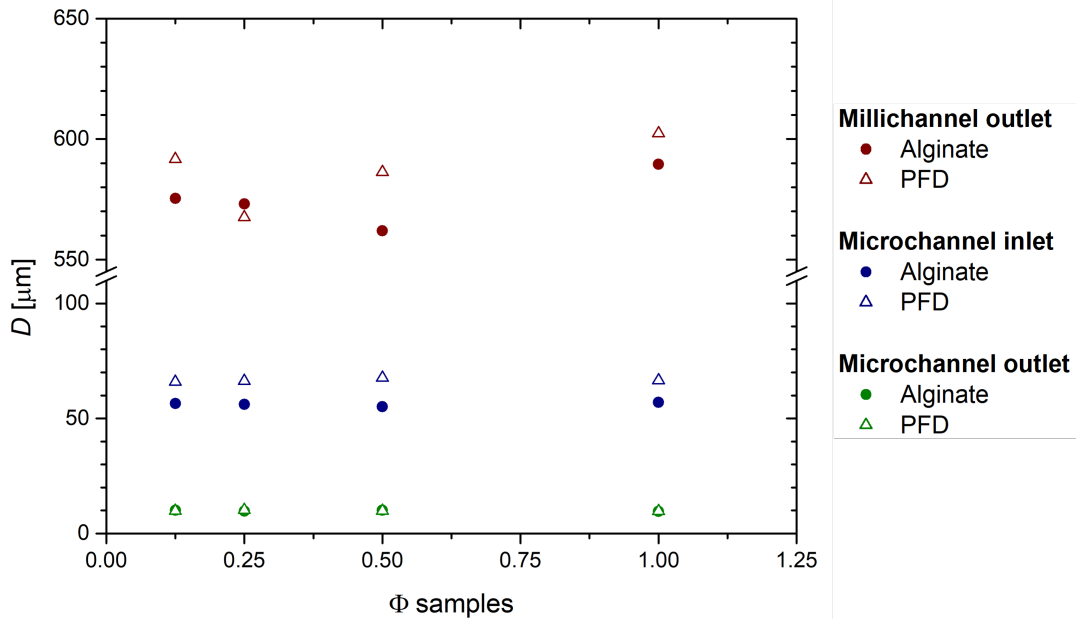


Figure 5.11. Mean droplet diameter (D), determined from normal distributions, at the outlet of the millichannel Figure 5.6, red symbols), the inlet of the microchannel (Figure 5.9, blue symbols), and the outlet of the microchannel (Figure 5.10, green symbols) for different fluid droplets and Φ samples: PFD (open triangle symbols), and sodium alginate (closed circle symbols).

From a biomedical perspective, these findings are highly relevant. The ability to reduce droplet size downstream of initial generation could be advantageous for producing:

- Small PFD droplets encapsulated in alginate matrices for oxygen-therapeutic systems, where enhanced surface-area-to-volume ratio improves gas transport;
- Alginate microparticles with diameters in the micrometric range suitable for blood-analogue fluids or cell-encapsulation applications.

Importantly, the process relies exclusively on passive hydrodynamic mechanisms, avoiding external fields or complex actuation. This enhances robustness, scalability, and compatibility with sensitive biomaterials.

The combination of controlled droplet generation at the milliscale and secondary hydrodynamic refinement at the microscale represents a promising strategy for manufacturing monodisperse, oxygen-carrying, biopolymer-based microparticles. It is

important to reinforce that 10 μm microparticles may be directly relevant to applications in artificial oxygen carriers, drug delivery, tissue engineering, and physiologically relevant blood-analogue systems.

5.5. Conclusions

This study demonstrates that arrays of passive obstacles integrated within millifluidic and microfluidic devices provide a robust and scalable platform for the controlled production of microparticles with biomedical relevance. By combining initial droplet generation at the milliscale with secondary hydrodynamic refinement at the microscale, it was possible to achieve progressive and reproducible droplet size reduction across three distinct regions of interest.

At the outlet of the millifluidic device, droplet area distributions were nearly Gaussian and independent of flow rate ratio and viscosity ratio, indicating that geometric confinement imposed by the herringbone obstacle array dominates breakup dynamics. Although droplet size decreased with increasing Capillary number of the dispersed phase, viscosity contrast between phases did not significantly affect the final outlet size within the investigated range.

Upon injection into the microchannel containing an array of transverse rectangular obstacles, droplets experienced further breakup driven by obstacle-induced elongational stresses. A reduction in mean droplet diameter from approximately 600 μm (millifluidic outlet) to 60 μm (microchannel inlet) – likely due to the transition from the millichannel to the narrower microchannel –, and ultimately to $\approx 10 \mu\text{m}$ (microchannel outlet) was achieved. The final droplet size corresponded closely to the obstacle spacing, confirming that passive geometric constraints define a characteristic breakup scale. Importantly, similar final sizes were obtained for both fluid systems (PFD/alginate and alginate/mineral oil), reinforcing the dominant role of microchannel geometry over rheological differences.

From a biomedical perspective, the production of $\approx 10 \mu\text{m}$ droplets is particularly relevant for applications such as oxygen-therapeutic systems, and cell encapsulation. The entirely passive nature of the breakup mechanism eliminates the need for external

fields or complex actuation, enhancing device simplicity, robustness, and compatibility with sensitive biomaterials.

Overall, the synergy between alginate hydrogel chemistry, oxygen-carrying perfluorocarbon phases, and obstacle-based microfluidic design establishes a versatile framework for engineering functional microparticles. Future work may explore in-channel gelation, encapsulation of living cells, and scaling strategies toward continuous manufacturing of oxygen-carrying biopolymer microparticles for translational biomedical applications.

Nomenclature

Acronyms	Definition
3D	Three-dimensional
CAD	Computer-aided design
G	α -L-guluronic
INL	International Iberian Nanotechnology Laboratory
M	β -D-mannuronic
PFD	Polydimethylsiloxane
PDMS	Perfluorodecalin
SLA	Stereolithography

Greek	Definition	Units
μ	Dynamic viscosity	[Pa s]
μ_c	Dynamic viscosity of the continuous phase	[Pa s]
μ_d	Dynamic viscosity of the dispersed phase	[Pa s]
ρ	Fluid density	[kg m ⁻³]
σ	Interfacial tension	[N m ⁻¹]
$\emptyset$	Flow rate ratio	—

Latin	Definition	Units
A	Droplet area	[m ²]
A_{outlet}	Droplet area at the outlet region	[m ²]
Ca_d	Capillary number of the dispersed phase	—
D	Droplet diameter	[m]
h	Height of the channel	[m]
l_m	Length of the array of obstacles	[m]
l_o	Length of the obstacle	[m]
q	Flow rate	[m ³ s ⁻¹]
q_c	Continuous phase flow rate	[m ³ s ⁻¹]
q_d	Dispersed phase flow rate	[m ³ s ⁻¹]
w	Width of the channel at the flow-focusing region	[m]
w_m	Width of the array of obstacles	[m]
w_o	Width of the obstacle	[m]

$x_o ; y_o$	Gap distance between two obstacles	[m]
X	Viscosity ratio	–

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Conclusions and future work

*Conclusions and future work***6.1. Thesis conclusions**

The current PhD thesis addresses the dynamics of droplets flowing through confined geometries in milli and microfluidic systems, with particular emphasis on deformation and breakup mechanisms promoted by fluidic geometrical elements, such as constrictions and arrays of obstacles. Through an approach that combined experimental investigations and numerical simulations, this work contributes to a deeper understanding of multiphase flows in structured channels and provides design guidelines for milli and microfluidic devices aimed at droplet manipulation and microparticle production.

Microstructured devices have gained increasing importance in fields such as biotechnology, medicine, energy, materials processing, and chemical engineering. In these systems, two-phase flows are commonly present, and the interaction between droplets and microfluidic geometric elements – such as contractions, expansions, constrictions, T-junctions, and obstacles – play a decisive role in controlling interfacial phenomena. Droplet deformation and breakup are governed primarily by the Capillary number of the continuous phase (Ca_c) and viscosity ratio (X) between the dispersed and continuous phases, together with channel geometry. These parameters determine the balance between viscous, inertial, and interfacial forces and ultimately dictate the flow regimes and droplet evolution within microfluidic geometrical elements.

The experimental and numerical investigation of droplet flow through constricted channels demonstrated that the Volume of Fluid (VOF) methodology is capable of accurately predicting droplet behavior under confined geometries. A strong agreement between simulations and experiments was obtained for droplet velocity and deformation characteristics. The results showed that increasing the Ca_c enhances droplet deformation and increases the film thickness between the droplet and the channel wall. At low Ca_c , where surface tension dominates, droplets move in a plug-like regime with velocities close to the continuous phase velocity. Although droplet velocity decreases with increasing viscosity ratio, this influence becomes negligible for viscosity ratios above approximately 0.3. Furthermore, despite variations in channel geometry, the behaviors observed at both the milli and microscales – such as film thickness and droplet velocity – remained consistent, indicating that the primary flow characteristics are governed by dimensionless parameters. (i.e., Ca_c and X).

The role of passive geometrical fluidic elements in promoting droplet breakup was further investigated experimentally in 3D-printed millifluidic channels, comprising a flow-focusing region followed by an array of rectangular obstacles disposed in transverse, longitudinal, or herringbone configurations. The results clearly demonstrated that the obstacle configuration and channel porosity (ranged from 36% to 75%) significantly influence droplet deformation and breakup. While droplet size at the channel inlet is primarily governed by the continuous and dispersed flow rates imposed at the flow-focusing region, the outlet droplet size distributions converge toward a geometry-controlled dimension that is independent of the flow rates. Among the configurations studied, the herringbone arrangement was the most effective in

reducing droplet size, followed by the transverse and longitudinal arrays. Lower-porosity structures increased droplet confinement and intensified droplet-obstacle interactions, resulting in more efficient breakup. Additionally, increasing the Capillary number of the dispersed phase raises viscous stresses relative to surface tension, contributing to larger droplet deformation and a progressive reduction in droplet size. These findings demonstrate that arrays of obstacles constitute an efficient passive strategy for controlling droplet breakup and achieving improved droplet size uniformity.

Based on the aforementioned conclusions, a two-stage milli/microfluidic strategy was developed for the controlled production of microparticles with potential biomedical applications. Monodisperse droplets were first generated in a 3D-printed millifluidic flow-focusing device incorporating a herringbone obstacle array, where breakup was primarily governed by geometric confinement. The droplets were subsequently introduced into a microfluidic channel containing transverse obstacles, where additional hydrodynamic stresses induced successive breakup events. The microscale array of obstacles reduced the droplet diameter from $\approx 60 \mu\text{m}$ at the inlet to $\approx 10 \mu\text{m}$ at the microchannel outlet. Importantly, the final droplet size was more dependent on the geometric features of the obstacle array rather than the viscosity ratio between the phases, indicating the dominant role of passive geometric constraints in defining the breakup scale. Furthermore, a clear decrease in droplet size with the Capillary number of dispersed phase was observed for each value of X under study.

The proposed strategy was validated using fluid systems of biomedical relevance, namely perfluorodecalin dispersed in sodium alginate solution and sodium alginate droplets dispersed in mineral oil. The production of droplets with diameters around $10 \mu\text{m}$ is particularly relevant for biomedical applications such as oxygen carriers and cell encapsulation. Since the breakup mechanism mainly relies on passive hydrodynamic interactions with channel geometries, the approach avoids the need for external fields or active actuation, improving device simplicity, robustness, and compatibility with sensitive biological materials.

Overall, the results of this PhD thesis demonstrate that carefully designed microfluidic geometries – particularly constrictions and arrays of obstacles – provide powerful and scalable tools for controlling droplet dynamics in multiphase flows. The findings contribute to the fundamental knowledge about droplet deformation and

breakup in confined environments and offer practical design principles for the development of advanced milli and microfluidic systems for chemical processing, materials synthesis, and biomedical engineering.

6.2. Future work

This PhD thesis provides important contributions to the understanding and control of droplet dynamics in structured milli and microfluidic systems, but several research directions remain open and could further expand the scope and applicability of the present work. Future developments may focus on further improving the fundamental physical insights about multiphase flow phenomena, exploring novel device architectures, and improving the biomedical applications based on the produced microparticles.

The first direction for future work involves broadening the experimental and numerical approaches to address a wider range of fluid properties. In the present study, the influence of the viscosity ratio and Capillary number of the continuous phase was explored within a broad but still limited range ($10^{-2} < X < 10^2$ and $10^{-4} < Ca_c < 10^{-1}$). Additionally, future studies could investigate more complex rheological behaviors, including shear-thinning or viscoelastic fluids, which are commonly found in biological and polymeric systems. Understanding how non-Newtonian effects influence droplet deformation, breakup, and transport in structured channels would significantly enhance the applicability of these devices in biomedical and industrial contexts.

Another important extension concerns the exploration of additional microfluidic geometries and obstacle configurations. Although transverse, longitudinal, and herringbone obstacle arrays were investigated, other designs – such as asymmetric obstacles, or obstacles of varying sizes within the same array – may further intensify droplet breakup efficiency and size control. Systematic studies combining experimental measurements and Computational Fluid Dynamics (CFD) simulations could help identify optimized geometries capable of producing narrower droplet size distributions or achieving specific breakup thresholds under controlled hydrodynamic conditions. Furthermore, to improve mixing efficiency under two-phase flow

conditions, appropriate configurations of arrays of obstacles should be carefully considered.

Exclusively from a numerical perspective, there are two relevant paths worth mentioning: 1) performing CFD simulations of droplets flowing through domains filled with arrays of obstacles to replicate and extend the work presented in this document; 2) incorporate more complex/complete models able to capture additional physical effects would also be highly relevant. For example, simulations including surfactant transport, interfacial rheology, or mass transfer between phases would provide a more comprehensive description of droplet behavior in realistic systems. Coupling CFD with particle tracking or population balance models also have a high potential to enable powerful predictive tools for droplet size evolution in complex microfluidic networks.

Further investigation into scale-up strategies represents another promising research direction. While the present work demonstrates the effectiveness of a combined milli/microfluidic approach, the development of parallelized or modular microfluidic architectures would enable higher throughput production of droplets and microparticles. Such developments are particularly important for translating microfluidic technologies from laboratory-scale demonstrations to practical industrial or biomedical manufacturing processes.

Future studies could also explore integrating additional functionalities within microfluidic devices. For example, in-channel gelation or crosslinking of alginate droplets should allow the direct on-chip production of solid hydrogel microparticles. The encapsulation of living cells, therapeutic agents, or nanoparticles inside these droplets would open new possibilities in drug delivery, cell therapy, and tissue engineering.

Finally, further work is needed to fully explore the biomedical potential of the microparticles produced. In particular, the perfluorodecalin droplets generated in this work may serve as promising candidates for artificial oxygen carriers or blood-analogue fluids. Deeper studies could therefore address oxygen transport properties, biocompatibility, long-term stability, and flow behavior in microvascular-inspired environments. Such studies would be essential steps toward the translation of these systems into clinically relevant technologies.

Overall, the continued integration of microfluidic design, multiphase flow physics, and biomedical engineering offers significant opportunities for the development of advanced droplet-based technologies. The insights gathered in this PhD thesis provide a strong foundation for future research on controlling interfacial phenomena and enabling scalable production of functional microparticles for a wide range of applications.

Supplementary material (Chapter 2)

Supplementary material (Chapter 2)

The present appendix provides a state-of-the-art review covering the last six years. Chapter 2 also reviews the main topics addressed in this work; however, it is a journal article published during this PhD thesis that focuses exclusively on studies reported before 2020. Accordingly, this previous review was updated with recent studies on droplet flow through contractions, expansions, constrictions, and obstacle-based microfluidic geometrical elements, which are addressed below.

A.1. Contractions, expansions and constrictions

The dynamics of microdroplets flowing through contractions, expansions and constrictions remain a central topic in multiphase microfluidics. These configurations are common in lab-on-a-chip devices, emulsification platforms, biomedical microreactors, and porous-media analogues. Since 2020, substantial advances have been made in understanding the influence of geometry (e.g., abrupt constrictions with a 90° entry angle or gradual constrictions obtained by varying the contraction/expansion entry angle, as well as the length and depth of the constricted channel), flow conditions through changes in non-dimensional parameters, surfactants, contact angle, and rheological effects on droplet deformation, breakup, and coalescence.

A.1.1. Effect of geometry

The influence of the entry geometry of the contraction has been numerically investigated by Hoang *et al.* (2023) [1]. It involved distinct regimes – trap, squeeze, and breakup – and extended their earlier work published in 2018 [2]. A simple theoretical model was proposed to characterize the transition between the trap and squeeze regimes, which depends on the entry angle. In contrast, the entry angle was found to have no effect on the transition from the squeeze to the breakup regime. Furthermore, the study showed that droplet breakup cannot be predicted solely from the Capillary number of the continuous phase (Ca_c); instead, local extensional stresses induced by entry angle and contraction channel ratio play a determining role. These findings provide practical design criteria for microfluidic devices where controlled droplet breakup is required.

Singla *et al.* (2024) [3] examined numerically the dynamics of droplets in three-dimensional constricted microchannels, focusing on the influence of constriction length and depth. They performed comparative qualitative and quantitative analyzes of the numerical simulations with previous experimental and numerical studies [4-9]. Constriction geometry alters the balance between viscous and interfacial forces, with droplet breakup occurring in longer and narrower constrictions. Regime maps were developed to identify the critical Ca_c for breakup and to relate droplet aspect ratio to constriction geometry, showing a clear link between aspect ratio and breakup onset.

Additionally, they investigated multiple constrictions with phase shifts (P) between the top and bottom walls (Figure A.1), which led to zig-zag droplet shapes, causing lateral deformations and velocity variations. Overall, the study confirms that constriction geometry is a key factor controlling droplet evolution, deformation, and breakup.



Figure A.1. Schematic representation of the multiple constrictions (P is the phase shift). Adapted from Singla *et al.* [3].

A.1.2. Effect of flow conditions

He *et al.* (2020) investigated experimentally the breakup of both pancake droplets (i.e., droplets predominantly compressed in the vertical direction, resulting in a flattened, pancake-like shape) and spherical droplets flowing through a constriction, using silicon oil as the continuous phase and water as the dispersed phase. Pancake droplets display a novel breakup mode, with rupture occurring at the rear meniscus with daughter droplet sizes dependent on the mother droplet size. Furthermore, the critical Ca_c shows opposite trends: pancake droplets break at high Ca_c , whereas spherical droplets break at low Ca_c . A theoretical model accounting for the velocity difference between the droplet and the surrounding fluid was presented, accurately predicting breakup and showing good agreement with experimental observations.

Li *et al.* (2021) [10] investigated the behavior of water droplets in mineral oil and aqueous glycerol droplets in hexadecane (with several SPAN[®] 80 concentrations used in both cases) passing through a short, abrupt constriction. The behavior of water droplets in mineral oil and glycerol aqueous droplets in hexadecane (several SPAN[®] 80 concentrations were used in both cases) passing through a short, abrupt. Three breakup behaviors – total, partial, and none – occur depending on droplet size, velocity, and continuous phase viscosity. Larger or faster droplets tend to resist breakup, while breakup occurs via dynamic snap-off at the droplet neck – the narrowest region of the droplet formed as it stretches through the contraction. Using Ca_c and dimensionless droplet size (i.e., ratio of the droplet area to the constriction area in the

top view), a phase diagram was constructed to predict breakup states, with critical curves delimiting regions of total, partial, and non-breakup regimes.

Wang *et al.* (2022) [11] developed a work that examines n-heptane droplet breakup in a square microchannel with a short square constriction, producing slug flow instead of the usual dripping flow. The breakup occurs in three stages – shear, squeezing, and pinch-off – with the dominance of the squeezing stage. Droplet size is influenced by mother droplet length and the continuous phase polyethylene oxide concentration. The constriction geometry used in this work also improved the uniformity of daughter droplet size compared with previous studies [10, 12, 13].

Zou *et al.* (2024) [14] further improved the understanding of breakup onset by experimentally identifying the transition between pure deformation and breakup conditions in contraction flows. The analysis of droplet interface evolution shows that breakup is triggered when the minimum neck shrinks below a critical width, resulting in an irreversible pinch-off. Otherwise, the droplet undergoes deformation without rupture. This behavior can be described using two characteristic times: the breakup time (t_b) and the transit time through the contraction (t_d). When $t_b < t_d$, the droplet ruptures; when $t_b > t_d$, only deformation occurs. Both times depend on droplet length, flow rate, and continuous phase viscosity, which can be captured using the dimensionless droplet length and Ca_c . This approach represents a shift toward mechanistic models capable of forecasting droplet behavior in confined geometries.

While contractions and constrictions have received considerable attention, expansions also play a key role in droplet breakup dynamics. Salari *et al.* (2020) [15] demonstrated that when bubbles and droplets enter an expansion, they can undergo periodic breakup distinct from classical dripping or squeezing regimes. In addition to Ca_c , previously shown to control droplet breakup in constrictions [16, 17], the results indicate that at high-inertia conditions, the Weber number of the continuous phase (We_c) significantly influences the transition from non-breakup to breakup regimes. Moreover, they identify distinct periodic breakup modes, including symmetric and asymmetric patterns, which are governed by the Ohnesorge number (i.e., the ratio of viscous forces to the combined effects of inertial and surface tension forces) of the continuous phase (Oh_c). Expansion-induced breakup arises from sudden reductions in confinement, leading to interface destabilization and extensional stress redistribution.

A.1.3. Effect of surfactants

Zhang *et al.* (2023) [18] used a three-dimensional (3D) Lattice Boltzmann (LB) method to simulate surfactant-covered droplets moving through an abrupt constricted microchannel. Increasing bulk surfactant concentration (Ψ_b) reduces interfacial tension, thereby lowering flow resistance and the pressure needed to pass the constriction. Consequently, this increases droplet velocity (t_d decreases) and stabilizes the neck, reducing the number of breakups. Comparing the behavior of surfactant-laden and clean droplets showed that the surfactant effect diminishes at higher Ca_c due to interface expansion and surfactant depletion. As Ψ_b increases, the critical Ca_c for breakup decreases, highlighting the combined influence of surfactant concentration and viscous forces on droplet breakup behavior.

A.1.4. Effect of contact angle

Imani *et al.* (2022a) [19] performed two-dimensional microscale numerical simulations in COMSOL[®], employing the phase-field method to track the interface between two immiscible fluids. In this approach, the interface is not explicitly defined; instead, a continuous scalar field (the phase-field variable) varies smoothly across a thin transition region to distinguish the phases [20]. They investigated the passage of an oil droplet in water through abrupt constrictions with different widths, as well as constrictions with different shapes (Figure A.2). The effects of wall wettability, constriction width, droplet size, and their coupled influence were analyzed. The main findings were: 1) droplet displacement was highly sensitive to wettability, with a contact angle of 45° promoting easier oil droplet passage through the constriction. Increasing the contact angle (60° and 75°) caused the droplets to adhere to the surface, while larger contact angles (120° and 135°) increased resistance to motion; 2) the threshold pressure (i.e., the minimum pressure required for the droplet to pass through the constriction) increased as the constriction width decreased and the droplet size increased; 3) the combination of higher contact angles (i.e., 120° and 135°), larger droplets, and narrower constrictions produced the greatest resistance and can trigger droplet breakup; and 4) viscosity ratio had little impact, longer constrictions lowered the threshold pressure, and circular constrictions reduced resistance compared to

rectangular or trapezoidal geometries (see Figure A.2). These results offer valuable guidance for enhanced oil recovery, microfluidic device optimization, and contaminant transport in porous media.

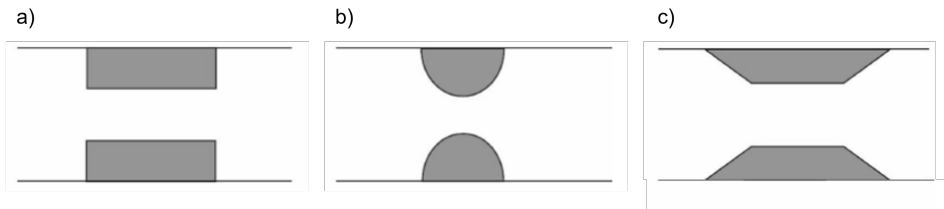


Figure A.2. Schematic representation of the different constriction shapes: a) rectangular; b) circular; and c) trapezoidal. Adapted from Imani *et al.* [19].

In another study, Imani *et al.* (2022b) [21] used three-dimensional direct numerical simulations to further investigate oil droplet dynamics in water by varying the wall contact angles. A key finding unique to 3D simulations was that the fluid-fluid menisci often adopt saddle-shaped interfaces, exhibiting positive curvature in one direction and negative curvature in the orthogonal direction (see Figure A.3) – an effect typically neglected in two-dimensional simulations, highlighting the importance of fully three-dimensional studies.

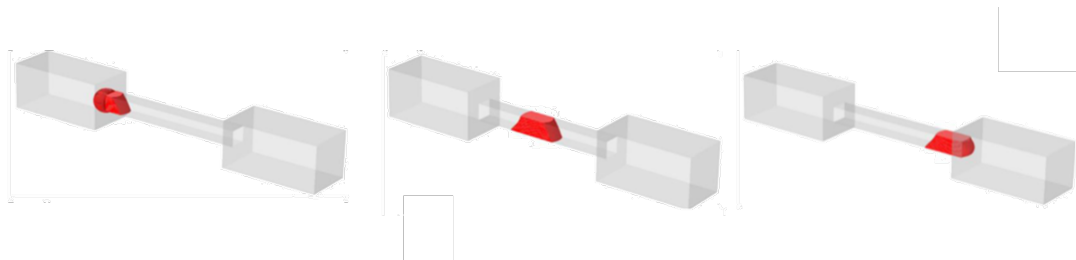


Figure A.3. Droplet dynamics within a constricted channel, where the upper wall exhibits a contact angle of 60° and the lower wall 120° . Adapted from Imani *et al.* [36].

A.1.5. Effect of non-Newtonian fluids

The role of rheology in droplet dynamics has also gained higher attention in recent years. Zhao *et al.* (2020) [22] investigated the breakup of oil droplets in 3D pore throat structures within viscoelastic non-Newtonian polyacrylamide (HPAM) continuous phase, focusing on factors affecting droplet breakup. Experiments showed that

increasing HPAM concentration reduces the critical breakup length (i.e., the minimum droplet length required for breakup under a given flow conditions) and the size of the first daughter droplets. Isolating polymer elasticity effects revealed that droplets in elastic polymer solutions break more readily than in Newtonian fluids. An exponential model predicted droplet size, providing guidance for polymer use in oilfield applications like fracturing and polymer flooding.

Patlazhan *et al.* (2026) [23] investigated shear-thinning xanthan gum droplets co-flowing with a Newtonian continuous phase (silicone oil) through a planar abrupt contraction. Combining experiments and numerical simulations, this work demonstrated strong agreement between the two approaches, enabling the analysis of droplet deformation and interfacial instabilities over varying Ca_c and a confinement parameter, defined as the ratio of the droplet's initial diameter (i.e., before entering the contraction) to the microchannel height. The results showed that breakup was governed by the non-uniform viscosity within the droplet, with secondary and satellite droplets forming at higher Ca_c as the volume-averaged viscosity ratio decreases sharply within the contraction due to intensified shear and extensional stresses. These results underscore the critical influence of viscosity heterogeneity in shear-thinning droplets on deformation and breakup, providing predictive criteria and a framework for viscosity-guided control in microfluidic devices and emulsification processes involving shear-thinning droplets.

Finally, Kootenaei *et al.* (2026) [24] work investigated how orifice geometry and non-Newtonian (aqueous solutions of sodium carboxymethyl cellulose – SCMC) rheology jointly influence a Newtonian droplet transport in constricted microchannels. The expansion region of the constriction promotes sustained droplet deformation and strong internal recirculation patterns, whereas the contraction region generates early but transient shear zones. The interaction between geometry and shear-thinning rheology affected pressure evolution and lubrication film formation, with constriction position controlling where viscosity reduction occurs. The study highlights geometry as an independent design parameter, enabling tailored droplet dynamics for applications like enhanced mixing or reduced pressure drops.

A.2. Obstacles

In recent years, studies addressing droplet/bubble dynamics in the presence of obstacles remain relatively limited compared to the extensive literature on microdroplets flowing through constrictions. This section is organized into three parts, addressing droplet flow through: 1) a single obstacle (Figure A.4a); 2) an array of obstacles (Figure A.4b); and 3) obstacles integrated within T-junctions (Figure A.4c).

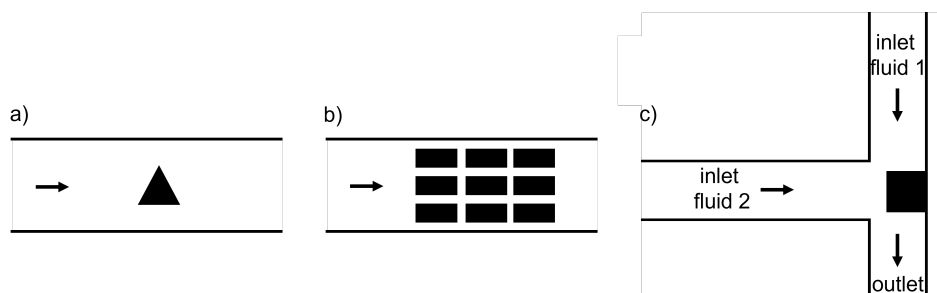


Figure A.4. Schematic representative illustrations of several obstacle-based fluidic geometrical elements: a) single obstacle; b) array of obstacles; and c) obstacle integrated within T-junction.

A.2.1. Single obstacle

Han *et al.* (2024) [25] investigated the breakup dynamics in a microchannel with a V-shaped obstacle structure to promote the production of monodisperse, high-throughput microbubbles for applications in analytical chemistry and fine chemical engineering. Extensive two-dimensional finite element simulations in COMSOL[®], employing the phase-field method to track the interface, were performed to analyze bubble breakup mechanisms inside microchannels containing these obstacles. The study identified four distinct breakup behaviors: non-breakup (Figure A.5a), breakup with permanent gaps (Figure A.5b), breakup with partial obstruction (Figure A.5c), and breakup with permanent obstruction (Figure A.5d). In all cases, the breakup process occurred in three successive stages – squeezing, transition, and pinch-off – governed by the combined effects of squeezing pressure, viscous shear forces, and interfacial tension. Finally, increasing interfacial tension led to larger bubble diameters and lower generation frequencies, as stronger molecular cohesion within the bubble promoted more stable neck contraction prior to breakup.

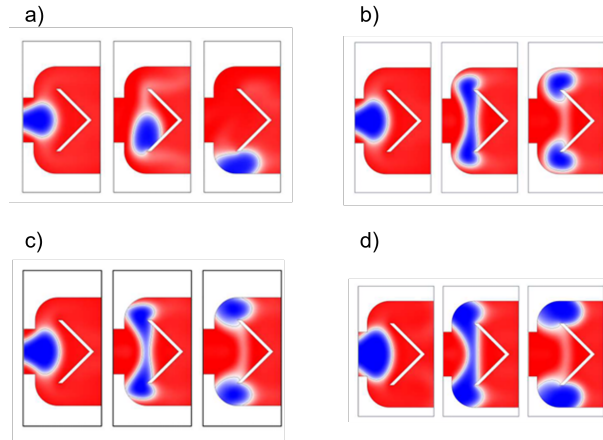


Figure A.5. Bubble breakup behaviors: a) no breakup; b) breakup with permanent gaps; c) breakup with partly obstruction; and d) breakup with permanent obstruction. Adapted from Han *et al.* [25].

Ho *et al.* (2025) [26] employed two-dimensional simulations using the front-tracking method to numerically investigate the effect of a semicircular-head obstacle on the breakup behavior of a compound droplet (i.e., a droplet containing one or more smaller fluid droplets inside it, also called a core-shell droplet or double emulsion) flowing through a microchannel. In the front-tracking method, a fixed Eulerian grid is used for the entire domain, while the interface is represented by connected marker points (or a mesh) that move with the local fluid velocity. This method is particularly suitable for compound droplets, as it can accurately capture both the inner and outer interfaces. The influence of key parameters – including the We_c , inner droplet size, and obstacle size – was systematically analyzed to determine conditions leading to breakup or non-breakup. Results show that at low We_c ($We_c \leq 0.008$), the compound droplet deforms around the obstacle and enters one of the two downstream channels without breaking. As the We_c number increases (i.e., surface tension decreases), breakup becomes more likely, particularly for smaller inner droplets and smaller obstacles. However, breakup can still be suppressed even at relatively high We_c ($We_c = 0.1$) when both the inner droplet and the obstacle are large compared to the outer droplet. In such cases, the inner droplet shifts toward one side, dragging the outer interface with it and preventing breakup. The study further demonstrated that, in breakup scenarios, smaller obstacle sizes or smaller inner droplet radii promoted faster separation. Despite the geometric symmetry of the channel, breakup could occur

asymmetrically due to imbalances introduced by the internal droplet motion and the high surface tension of the inner phase, which resists deformation and affects the flow distribution between the two branches.

A.2.1. Array of obstacles

Wang *et al.* (2021) [27] employed a nonorthogonal multiple-relaxation-time Lattice Boltzmann (LB) model to simulate the passage of a water droplet through an array of rectangular obstacles. A comprehensive parametric analysis was performed to investigate the effects of impact velocity, obstacle array opening fraction, and surface wettability. The results indicated that longitudinal droplet motion was governed by the balance between dynamic and capillary pressures, with the droplet travel length increasing approximately linearly over time. Larger opening fractions enhanced droplet velocity, while lateral spreading remained limited. Surface wettability of the obstacle array had a strong influence on droplet behavior: hydrophilic surfaces facilitated faster and deeper droplet passage, whereas hydrophobic surfaces significantly hindered it.

A.2.3. Obstacles incorporated into T-junctions

Jamalabadi *et al.* (2020) [28] presented a three-dimensional numerical investigation of four T-junction microchannels featuring an obstacle located at three different positions – before the dispersed phase inlet (Figure A.6a), directly above Figure A.6b), and after the dispersed phase inlet (Figure A.6c and Figure A.6d) – to evaluate its influence on flow and thermal characteristics. The system includes two inlets: water enters through one inlet, while oil is introduced through the other inlet. The results indicated that when the obstacle is split equally around the dispersed phase inlet (Figure A.6c), a greater number of smaller oil droplets were produced. A constant heat flux was applied at the obstacle region. When the dispersed phase entered the continuous phase (water), the temperature difference increased the heat transfer rate, resulting in a sharp rise in the Nusselt number (Nu). The highest Nu occurred when the obstacle was positioned directly above the dispersed phase inlet (Figure A.6b).

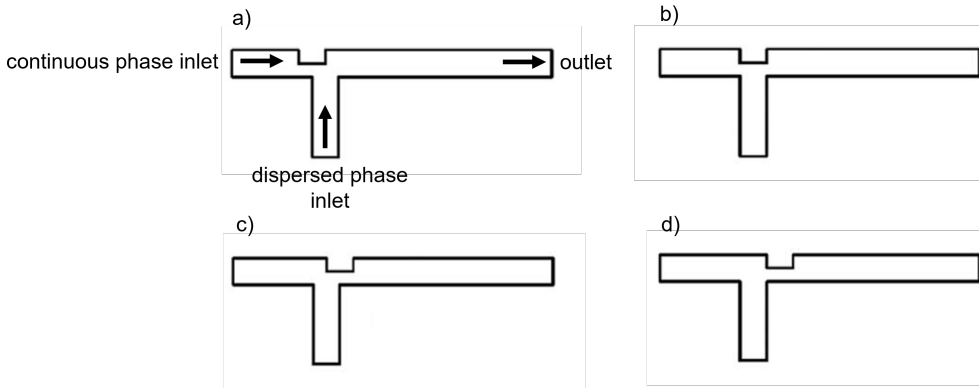


Figure A.6. Schematic of microchannels with an obstacle located at four different positions relative to the dispersed phase inlet: a) before; b) directly above; and c)-d) after. Adapted from Jamalabadi *et al.* [28].

A numerical investigation of droplet formation in a microfluidic T-junction containing a semicylindrical obstacle has been made by Nath *et al.* (2023) [29]. A three-dimensional finite element method using the level-set approach was employed to simulate the interaction between two fluid phases. The effects of obstacle radius, obstacle position, and Ca_c on droplet generation were analyzed, considering the combined impact of pressure forces exerted by the obstacle and fluid momentum on the dispersed phase. Three obstacle positions were studied: position A (Figure A.7a), position B (Figure A.7b), and position C (Figure A.7c). Results show that larger dimensionless obstacle radii (i.e., the ratio between droplet radius, R , and the microchannel width, w) generally promoted droplet formation. However, at the largest radius ($R/w = 0.9$), less continuous fluid impacts the dispersed phase. For $R/w = 0.3$, position C resulted in the most efficient droplet generation; for $R/w = 0.7$, droplet formation was most favorable at position B; while for $R/w = 0.9$, position A was optimal. Regarding droplet characteristics, at position A, droplet size and breakup time decreased steadily with increasing obstacle radius. For positions B and C, these parameters decreased up to a critical radius, then increased with further radius growth.

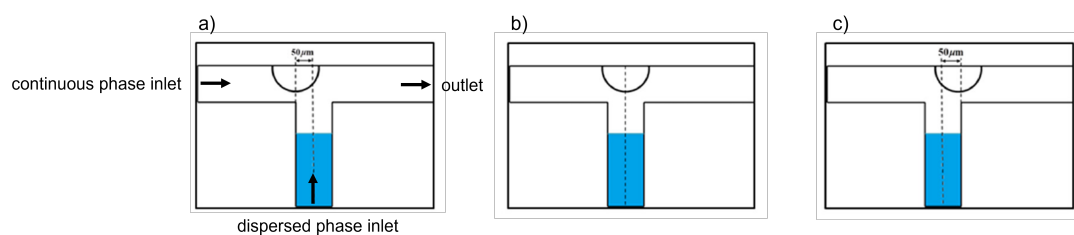


Figure A.7. Schematic of microchannels with an obstacle located at three different positions: a) position A; b) position B; and c) position C. Adapted from Nath *et al.* [29].

Nomenclature

Acronyms	Definition
3D	Three-dimensional
HPAM	Polyacrylamide
LB	Lattice Boltzmann
SCMC	Sodium carboxymethyl cellulose

Greek	Definition	Units
Ψ_b	Bulk surfactant concentration	[%]

Latin	Definition	Units
Ca_c	Capillary number of the continuous phase	—
Nu	Nusselt number	—
Oh_c	Ohnesorge number of the continuous phase	—
P	Phase shift	[m]
R	Droplet radius	[m]
t_b	Breakup time	[s]
t_d	Transit time through the contraction	[s]
w	Microchannel width	[m]
We_c	Webber number of the continuous phase	—

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Supplementary material (Chapter 3)

A. T. S. Cerdeira, J. B. L. M. Campos, J. M. Miranda, and J. D. P. Araújo, "Numerical and experimental analysis of droplet dynamics in constricted milli and microchannels," *Chemical Engineering and Processing – Process Intensification*, vol. 216, p. 110448, 2025, doi: [10.1016/j.cep.2025.110448](https://doi.org/10.1016/j.cep.2025.110448)

B.1. 3D full-domain versus 3D quarter-domain simulations

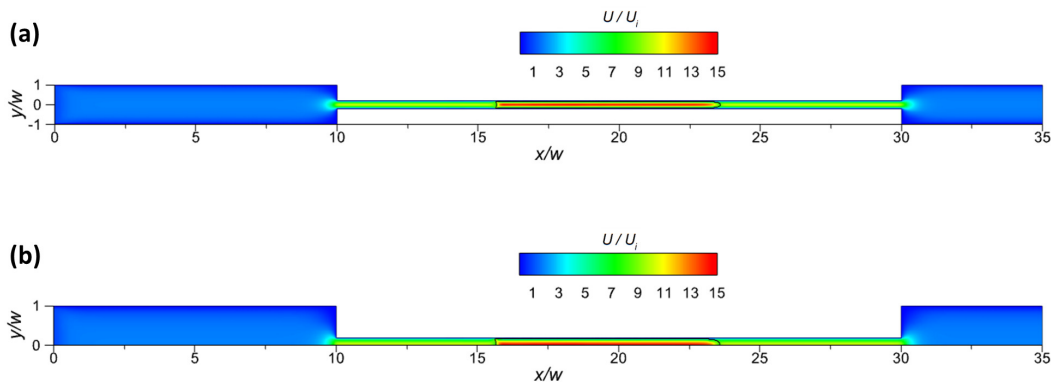


Figure B.1. Dimensionless magnitude velocity field (U/U_i) for the two-phase flow when $X = 0.3$ and $Ca_c = 0.06$: (a) full-domain; (b) quarter-domain.

B.2. Mesh tests

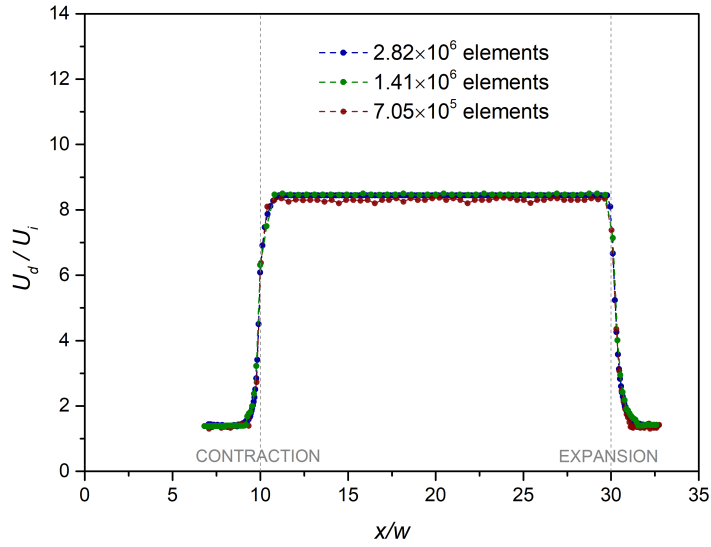


Figure B.2. Mesh independency tests: dimensionless droplet velocity (U_d/U_i) along the constriction for $X = 3$ and $Ca_c = 0.06$.

B.3. Droplet velocity along the constricted microchannel

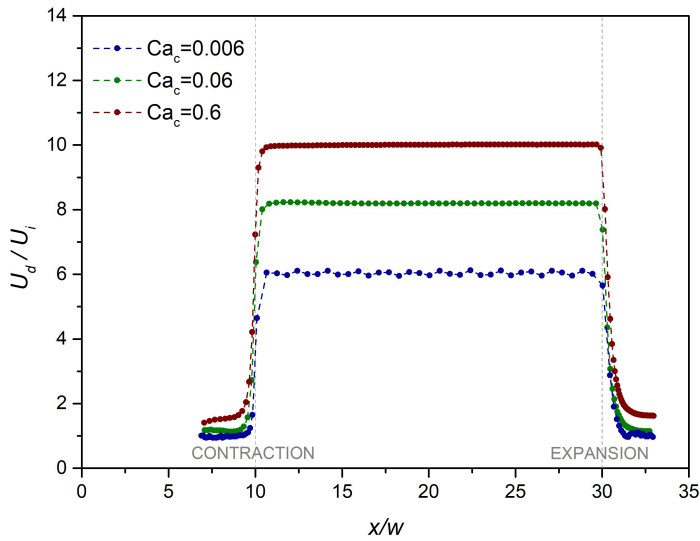


Figure B.3. Evolution of the dimensionless droplet velocity (U_d/U_i) along the constriction for $X = 3$.

B.4. Droplet entering the contraction

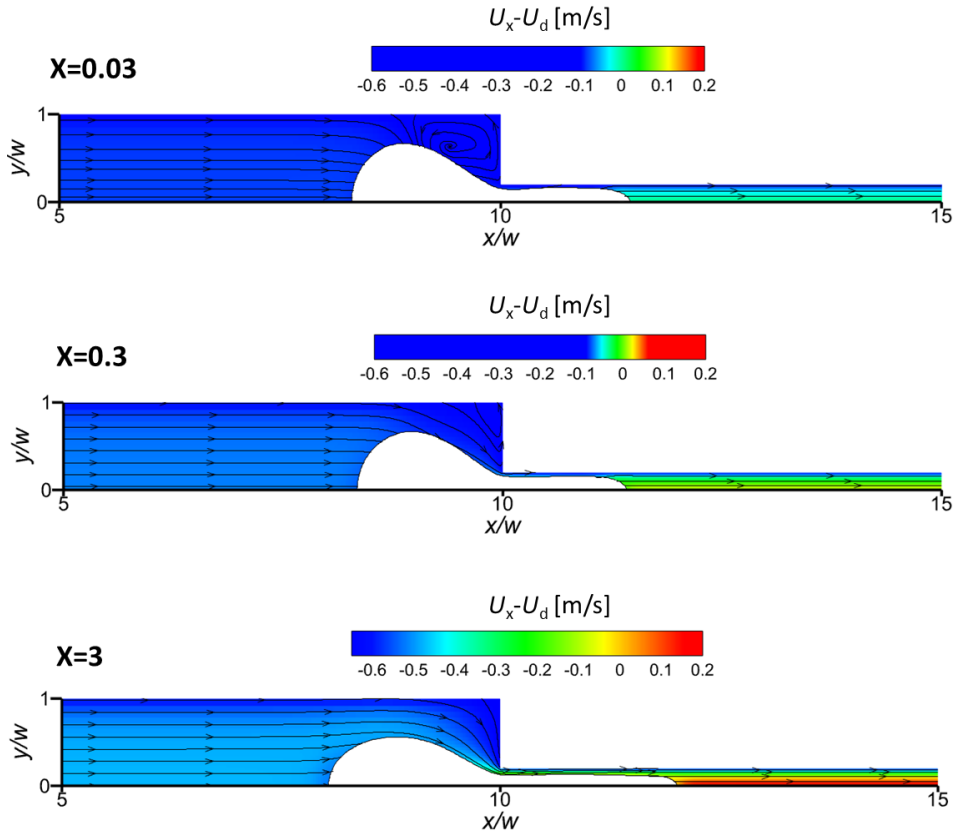


Figure B.4. Streamlines $U_x - U_d$ when the droplet is entering the constricted part of the microchannel for $Ca_c = 0.06$.

B.5. Droplet exiting the contraction

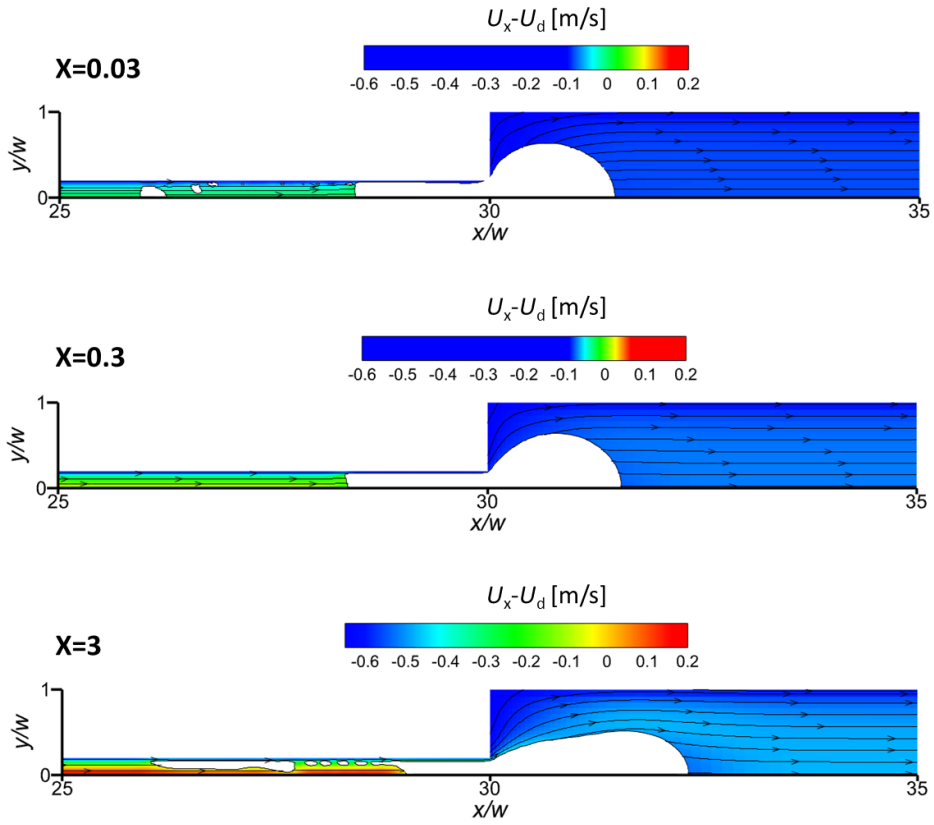


Figure B.5. Streamlines $U_x - U_d$ when the droplet is exiting the constricted part of the microchannel for $Ca_c = 0.06$.

Supplementary material (Chapter 4)

A. T. S. Cerdeira, J. B. L. M. Campos, J. M. Miranda, and J. D. P. Araújo, "Droplet deformation and breakup in 3D-printed millichannels featuring arrays of rectangular obstacles with different configurations," *Chemical Engineering Science*, vol. 326, p. 123493, 2026, doi: [10.1016/j.ces.2026.123493](https://doi.org/10.1016/j.ces.2026.123493)

Supplementary material (Chapter 4)

C.1. Obstacle dimensions after printing

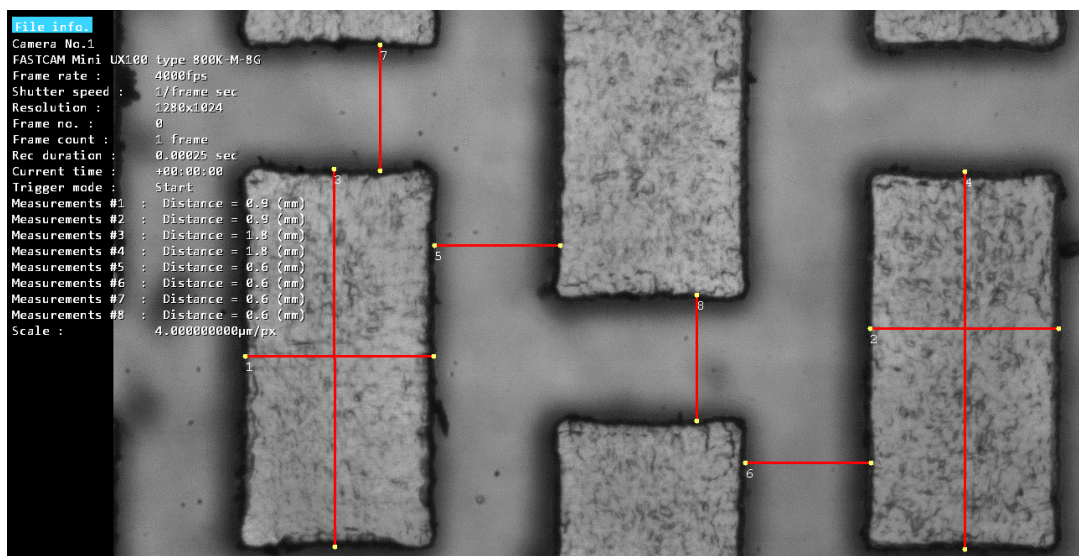


Figure C.1. Optical microscope image of the transverse array with the associated obstacle dimensions.

C.2. Contact angles

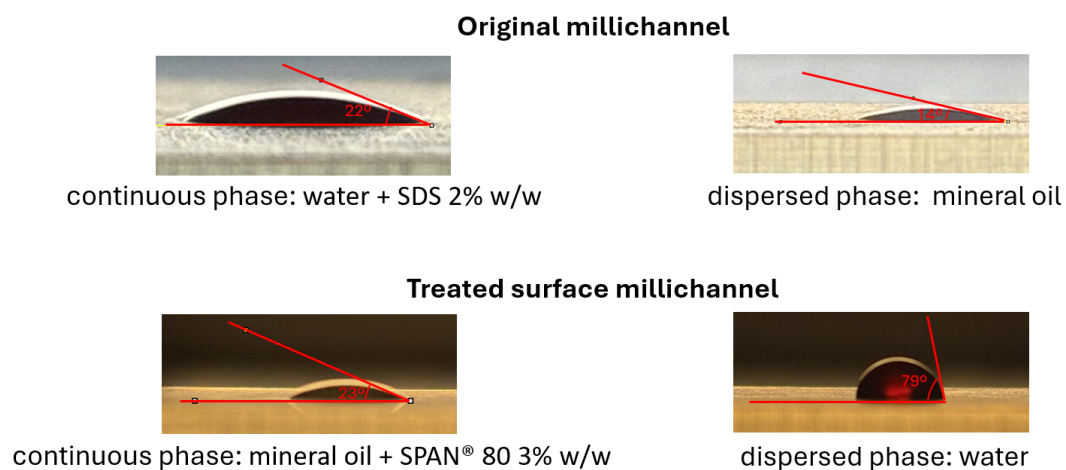


Figure C.2. Contact angles of both fluid pairs on the original and treated surface millichannels.

