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Regular Article

Prediction and control of drop formation modes in microfluidic generation of double emulsions by single-step emulsification

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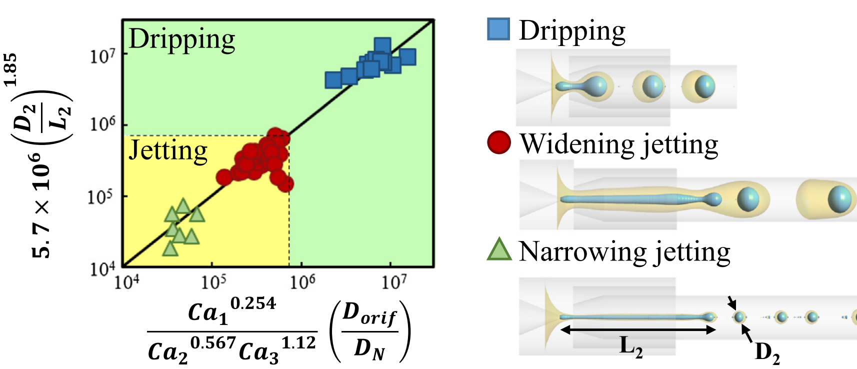
GRAPHICAL ABSTRACT



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ABSTRACT

Hypothesis: Predicting formation mode of double emulsion drops in microfluidic emulsification is crucial for controlling the drop size and morphology.
Experiments and modelling: A three-phase Volume of Fluid-Continuum Surface Force (VOF–CSF) model was developed, validated with analytical solutions, and used to investigate drop formation in different regimes. Experimental investigations were done using a glue-free demountable glass capillary device with a true axisymmetric geometry, capable of readjusting the distance between the two inner capillaries during operation.
Findings: A non-dimensional parameter (ζ) for prediction of double emulsion formation mode as a function of the capillary numbers of all fluids and device geometry was developed and its critical values were determined using simulation and experimental data. At $\log\zeta > 5.7$, drops were formed in dripping mode; the widening jetting occurred at $5 < \log\zeta < 5.7$; while the narrowing jetting was observed at $\log\zeta < 5$. The ζ criterion was correlated with the ratio of the break-up length to drop diameter. The transition from widening to narrowing jetting was achieved by increasing the outer fluid flow rate at the high capillary

Abbreviations: CNC, computer numerical control; CSF, continuum surface force; DC, Dow Corning; I.D., internal diameter; O.D., outer diameter; OTMS, octadecyltrimethoxysilane; PDMS, polydimethylsiloxane; PVA, polyvinyl alcohol; VOF, volume of fluid.

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Nomenclature

C'	constant, –
Ca	capillary number, –
D	diameter, m
D_1	inner droplet diameter, m
D_2	outer droplet diameter, m
f	drop generation frequency, s^{-1}
f'	volume fraction, –
F_b	body force, N m^{-3}
F_σ	interfacial force, N m^{-3}
L	break-up length, m
in	maximum instability, –
$\hat{n}$	unit normal, –
P	pressure, Pa
Q	volume flow rate, $m^3\ s^{-1}$
r	radial distance, m
r_c	radius of collection capillary, m
t	time, s
V	velocity, m s^{-1}
X	axial distance, m
x	direction (coordinates), m
α	diameter ratio, –
κ	curvature of interface, m^{-1}
μ	dynamic viscosity, kg·(m s)$^{-1}$
ρ	density, kg m^{-3}
σ	interfacial tension, N m^{-1}

Subscripts

1	inner phase
2	middle phase
3	outer phase
1,2	between inner and middle phase
2,3	between middle and outer phase
c	capillary
ci	injection capillary
co	outer capillary
continuous	continuous phase
dispersed	dispersed phase
ef	effective
j	jet
N	injection nozzle
orif	orifice

Volume of fluid–continuum surface force model
Glass capillary device

number of the inner fluid. The drop size was reduced by reducing the distance between the two inner capillaries and the minimum drop size was achieved when the distance between the capillaries was zero.

1. Introduction

Core-shell capsules attract great interest for potential applications ranging from controlled release of drugs and nutrients [1–4], encapsulation of fragrances [5], to CO_2 capture and energy storage [6–8]. The conventional fabrication strategies for core-shell capsules such as complex coacervation [9], internal phase separation [10], layer-by-layer electrostatic deposition [11], interfacial polymerisation [12], and spray drying [13] often require multi-stage processing and are associated with low encapsulation efficiency, low reproducibility and a lack of control over the size of the capsules and the shell thickness. In addition, the synthesised particles are usually highly polydispersed [14].

Microfluidic emulsification is a promising strategy for production of monodispersed double emulsions with controlled drop size and morphology. Double emulsion drops can be created through single-step or two-step emulsification methods, using flow-focusing [15,16], cross-flow [17,18] or co-flow [19,20] drop makers. Utada et al. [21] developed a three-phase 3D glass capillary device for single-step generation of double emulsions by combining co-flow and counter-current flow focusing flow patterns. The device consists of two tapered round capillaries, coaxially aligned within a square capillary and separated at some distance from each other, Fig. 1a and b. The device provides precise control over the droplet size and shell thickness. The wettability of each capillary can be independently modified, which is advantageous compared to polydimethylsiloxane (PDMS) devices that suffer from poor wettability control [22]. However, the distance between the round capillaries is fixed during operation and the device is difficult to clean after use.

The drop formation in capillary devices occurs in two distinct instability modes, absolute and convective [23]. Dripping is the result of an absolute instability; the perturbations that lead to drop pinch-off grow at a fixed spatial location and at a frequency that is intrinsic to the system, leading to monodispersed drops. Jetting occurs due to convective instability, which is associated with the advection of the perturbations along the interface of the jet, which causes random variation in the pinch-off location. Therefore, the drops generated in jetting mode are polydispersed [23,24]. The mechanism of drop formation in each mode is governed by the interaction between viscous, inertial, interfacial, and gravitational forces [25]. However, the effect of gravity is negligible when the drop diameter is in the micrometre range [26,27].

In dripping mode, the interfacial force is dominant and the inertial forces are negligible. The drops are formed in the vicinity of the injection nozzle (in co-flow geometry) or within one orifice diameter downstream of the orifice of the collection capillary (in flow-focusing geometry) once the viscous force, exerted by the continuous phase, exceeds the pinning force arising from the interfacial tension [27,29].

Two types of jetting mode have been observed in capillary devices, narrowing and widening [30]. Narrowing jetting is associated with formation of a relatively thin and long jet that eventually breaks into small drops; this mode occurs when the viscous force generated by the continuous phase is significantly greater than the interfacial force. On the other hand, widening jetting occurs when the inertial forces of the jet dominate the interfacial tension force. The widening shape of the jet has been attributed to the deceleration of the jet [31].

The formation of single emulsion drops in two-phase glass capillary devices is well characterised [27,30–33]. In three-phase glass capillary devices, inner and outer drops can be formed in the same or different modes and thus, versatile drop morphologies can be achieved. Due to complexity of the coaxial jet