Supplementary Information

Continuous Transfer of Liquid Metal Droplets Across a Fluid-Fluid Interface Within an Integrated Microfluidic Chip

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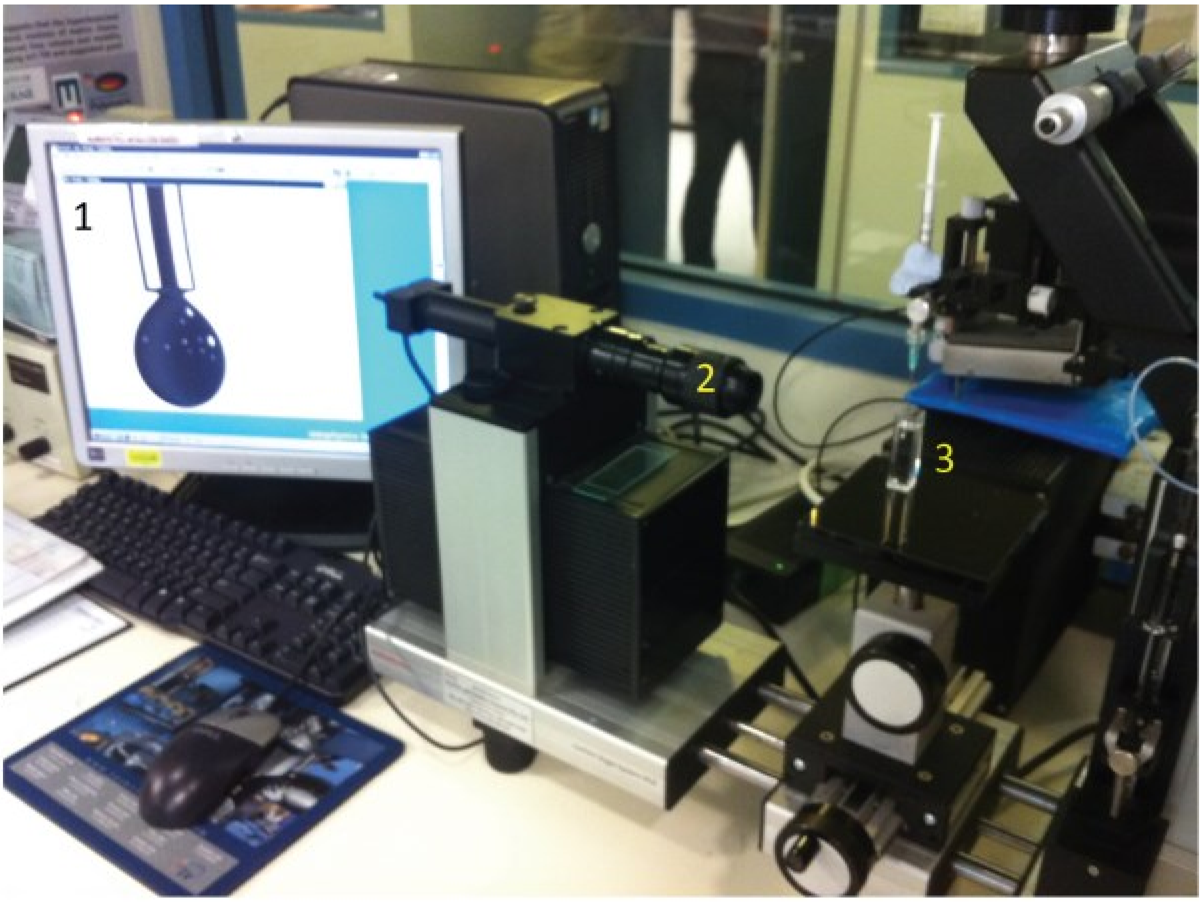
Supplementary Information 1: Interfacial tension measurements of galinstan-glycerol and galinstan-NaOH

Interfacial tension of galinstan in glycerol and NaOH is measured by using a goniometer (OCA20, DataPhysics Instruments GmbH, Germany) interfaced with CSA20 software to analyse the images (**Figure S1a.**).



1. Software used for interfacial tension measurements
2. Camera
3. Galinstan droplet

Figure S1a: Experimental setup for surface tension measurements with goniometer

Pendant drop shape analysis is used to determine the interfacial tensions with galinstan droplets ranging in volume from 15 to 20 µl (**Figure S1b**).

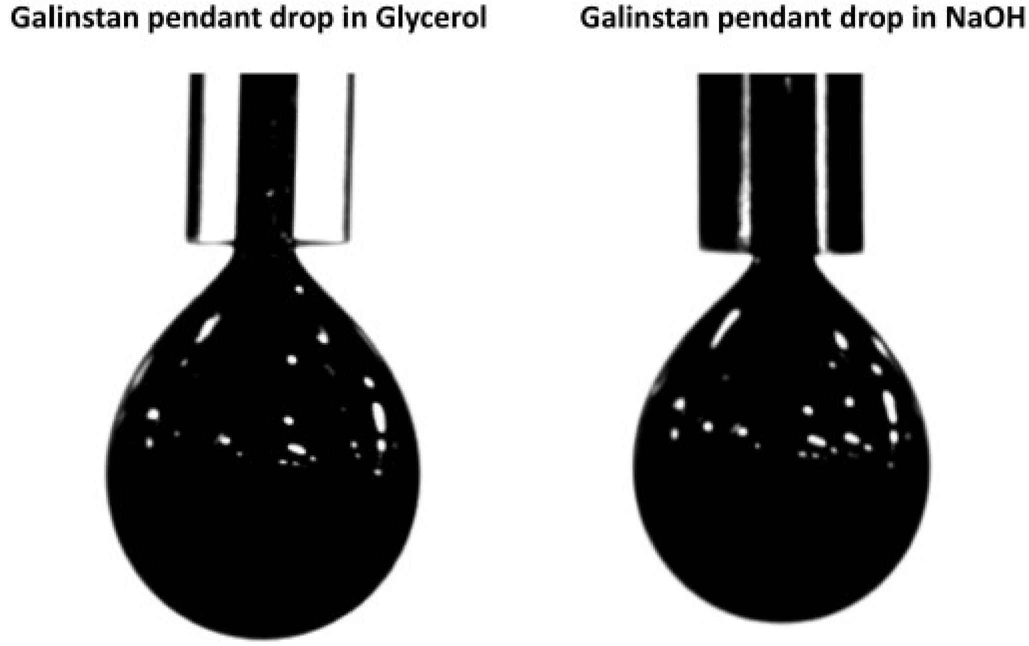


Figure S1b: Snapshots of galinstan pendant drop in glycerol (left) and NaOH (right).

Pendant drop shape analysis is a method for measuring interfacial tension between two fluids. This method requires a clean needle or tube for a drop of fluid, which hangs from this needle/tube in related fluid. The shape of the drop is a result of a balance between gravitational force and surface tension, from which the interfacial tension of drop-medium is calculated from the following equation [1]:

$$\gamma = (\Delta \rho g D^2)/H \qquad [S1]$$

where *D* is equatorial diameter, *d* is the distance *D* from the bottom of the drop (**Figure S1c**), $\Delta \rho$ is difference in fluid density, *g* is gravity, *H* is shape dependent parameter and depends of the value of "*S=d/D"* is the shape factor, and 1/H can be calculated from the following equation:

$$\frac{1}{H} = \frac{B^4}{S^a} + B_3 S^3 - B_2 S^2 + B_1 S - B_0 \qquad [S2]$$

where B_i (i=0, 1, 2, 3, 4) and a are empirical constants, as given in Table S1 [1].

Table S1. Values of B_i and a used in Pendant drop shape analysis [1]

Range of S	A	B_4	B_3	B_2	B_1	B_0
0.401-0.46	2.56651	0.32720	0	0.97553	0.84059	0.18069
0.46-0.59	2.59725	0.31968	0	0.46898	0.50059	0.13261
0.59-0.68	2.62435	0.31522	0	0.11714	0.15756	0.05285
0.68-0.90	2.64267	0.31345	0	0.09155	0.14701	0.05877
0.90-1.00	2.84636	0.30715	-0.69116	-1.08315	-0.18341	0.20970

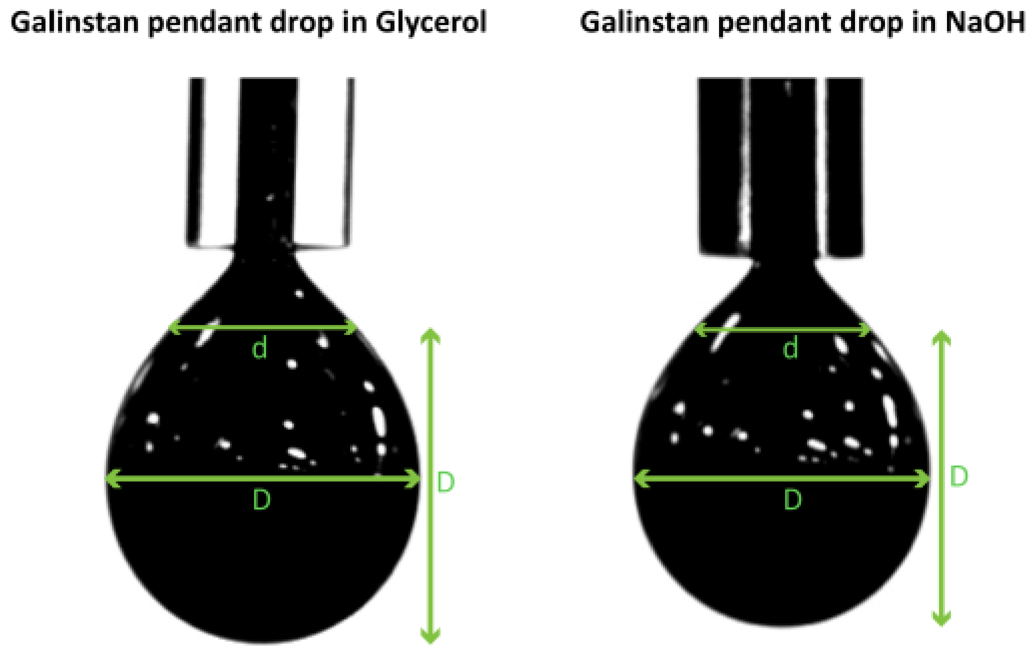
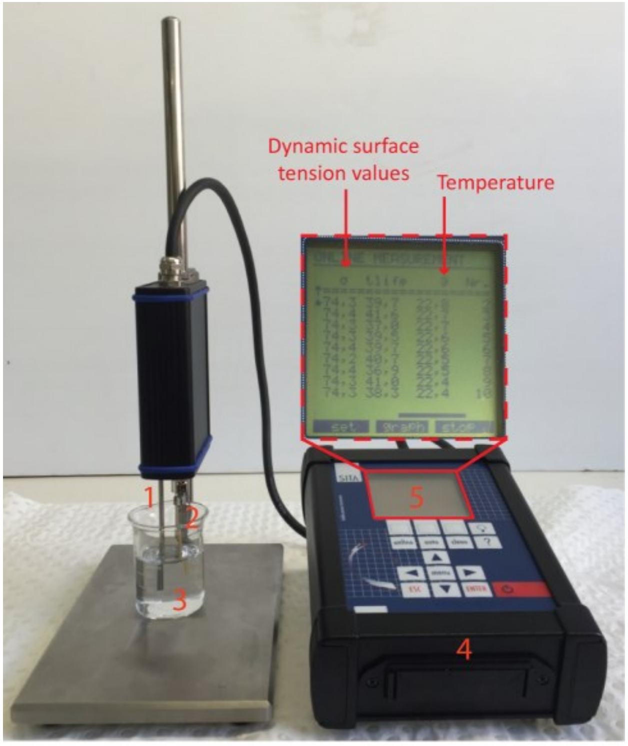


Figure S1c: Snapshots of galinstan pendant drop in glycerol (left) and NaOH (right) with equatorial diameter D and diameter d.

Using Pendant drop shape analysis, the interfacial tension of galinstan-glycerol and galinstan-NaOH are calculated as 537±13 mN/m and 480±12 mN/m, respectively.

Supplementary Information 2: Surface tension measurements of NaOH

Surface tension of NaOH is measured using bubble pressure method using SITA online t60 tensiometer. The pH value of NaOH is 12.8 and temperature is 22.5ºC. Under these conditions, the dynamic surface tension of NaOH is measured as 74 mN/m (**Figure S2**).



1. Temperature sensor
2. Capillary for bubble pressure
3. NaOH
4. Control unit
5. Monitor

Figure S2: Experimental setup for measuring the surface tension of NaOH with tensiometer

Supplementary Information 3: Calculation of Capillary number for flow focusing devices

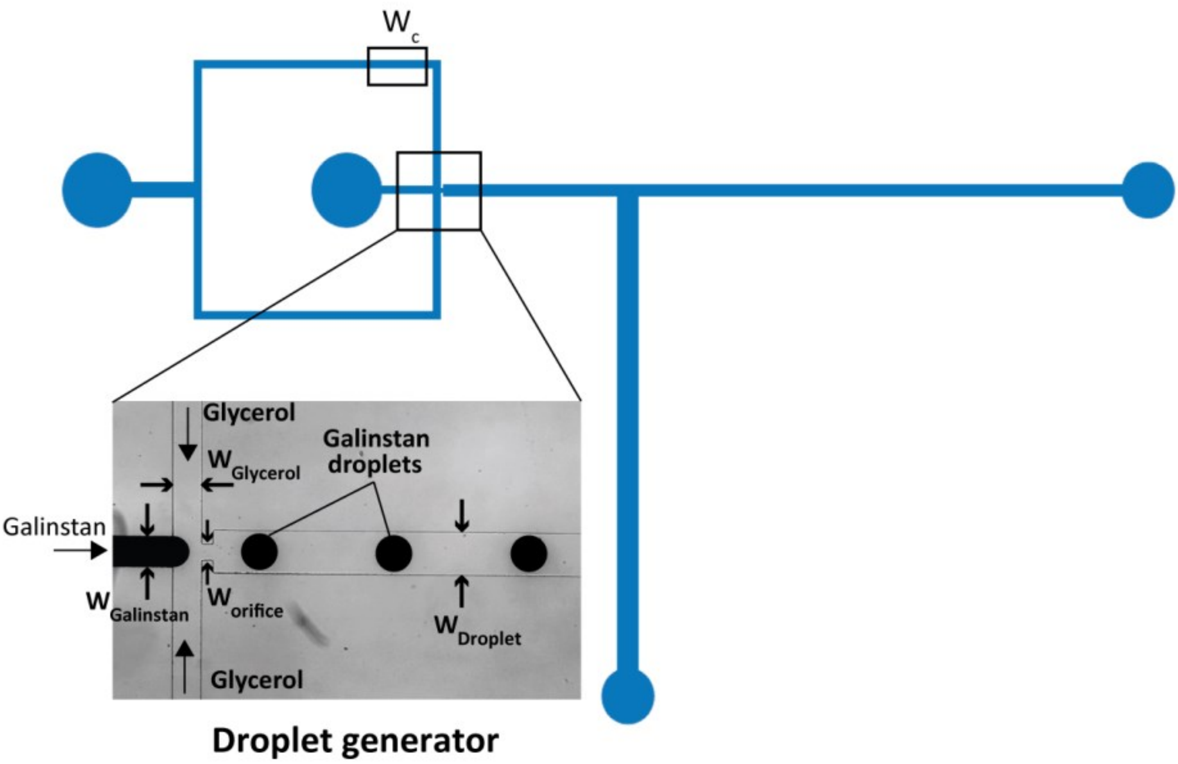


Figure S3: Microfluidic flow focusing chip design with important parameters in order to calculate Capillary number.

The more accurate value of Capillary number (Ca) in a flow focusing device can be calculated as follows [2]:

$$Ca = \frac{\mu_{glycerol}\ Q_{glycerol}\ a}{\gamma\ h\ W_{glycerol}} \left[\frac{1}{W_{orifice}} - \frac{1}{2W_{galinstan}}\right] \qquad \text{[S3]}$$

where $\mu_{glycerol}$ is the viscosity of glycerol, $Q_{glycerol}$ is the flow rate of glycerol, γ is the interfacial tension of glycerol and galinstan, h is the height of the microchannel, $2a$ is the width of galinstan inlet ($W_{Galinstan}$) channel, $W_{Glycerol}$ is the width of glycerol inlet channel, $W_{orifice}$ is the width of flow focusing orifice, and $W_{Galinstan}$ is the width of galinstan channel.

Supplementary Information 4: Result of CFD simulations for ethanol-NaOH flows

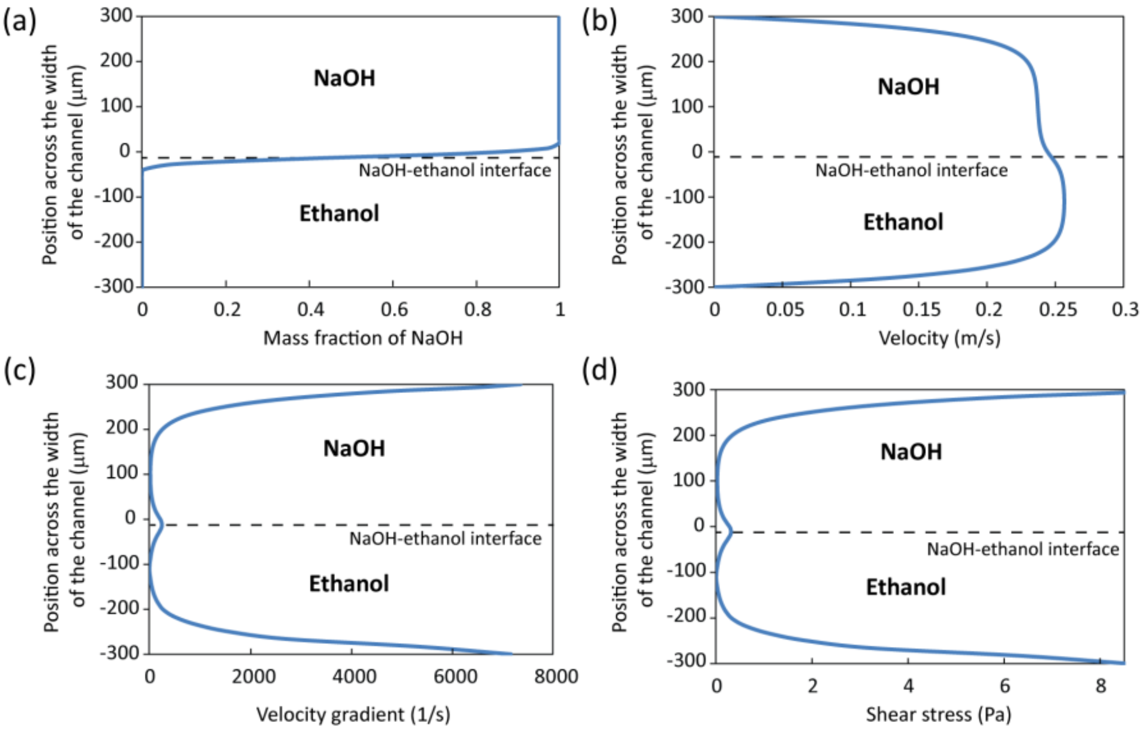


Figure S4: The results of CFD simulations predicting the hydrodynamic behaviour for the initial conditions of steady state confluent flow of ethanol and NaOH over the channel cross-section 5 mm downstream of the T-junction: (a) mass fraction; (b) velocity profile; (c) magnitude of lateral velocity gradient *(∂U/∂y)*; and (d) magnitude of shear stress. The flow rates of both ethanol and NaOH are set to 250 µl/min.

References:

1 J. Drelich, Ch Fang, C. L. White, Measurement of Interfacial Tension in Fluid-Fluid Systems, 2002, 3152-3166

2 W. Lee, L. M. Walker and S. L. Anna *Physics of Fluids*, 2009, **21**.