Supplemental Information

Droplet microfluidic method for measurement of ultralow interfacial tension in ternary fluid systems

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(a) Supporting data

The data supporting the article are listed in Table I for water-isopropanol mixtures and in Table II for ethanol-isopropanol mixtures. The uncertainty in interfacial tension γ at $\Phi_S \neq 0$ is estimated at about 20% of the measured value.

Table I: Data for water-IPA mixtures, including solvent concentration Φ_S, prefactor ratio k/k_0, and interfacial tension γ.

Φ_S	k/k_0	γ (mN/m)
0.00	1.000	42.70
0.10	0.790	21.05
0.20	0.716	15.67
0.30	0.672	12.96
0.40	0.652	11.84
0.50	0.637	11.04
0.60	0.635	10.93
0.70	0.601	9.27
0.80	0.569	7.87
0.90	0.514	5.80
0.95	0.396	2.65
0.99	0.223	0.47

Table II: Data for Ethanol-IPA mixtures, including solvent concentration Φ_S, prefactor ratio k/k_0, and interfacial tension γ

Φ_S	k/k_0	γ (mN/m)
0.00	1.000	0.750
0.10	0.864	0.483
0.20	0.688	0.244
0.30	0.641	0.197
0.40	0.571	0.140
0.50	0.522	0.107
0.60	0.503	0.095
0.70	0.419	0.055
0.80	0.197	0.006
0.90	0.139	0.002

(b) Uncertainty analysis

The measurement method of interfacial tension γ_i at solvent concentration $\Phi_S = i$ is based on the relationship $\gamma_i = \gamma_0(k_i/k_0)^3$, where γ_0 is the reference interfacial tension at $\Phi_S = 0$ and constants k_0 and k_i are prefactors in the relationship for the droplet spacing $L_0/h = k_0(\varphi Q_T^{1/3})^{-1}$ and $L_i/h =$

$k_i(\varphi Q_T^{1/3})^{-1}$ respectively. The Root-Sum-Square method is used to estimate the relative uncertainty $\Delta\gamma_i/\gamma_i \approx 20\%$ according to

$$\frac{\Delta\gamma_i}{\gamma_i} = \left[\left(\frac{\Delta\gamma_0}{\gamma_0} \right)^2 + \left(3\frac{\Delta k_i}{k_i} \right)^2 + \left(3\frac{\Delta k_0}{k_0} \right)^2 \right]^{1/2}.$$

The uncertainty of the Du Nouy ring method is estimated at $\Delta\gamma_0/\gamma_0 = 5\%$ of the measured value and the relative error in the determination of prefactor k_0 and k_i corresponds to $\Delta k_0/k_0 = \Delta k_i/k_i = 5\%$, which yields $\Delta\gamma_i/\gamma_i = 21.8\% \approx 20\%$

(c) Experimental validation

The experimental validation of the proposed microfluidic technique is conducted with immiscible fluid pairs made of a mixture of DI water and ethanol in a continuous phase of 100-cSt silicone oil. First, the surface tension of the mixture with air is measured using the Du Nouy ring method and compared with the literature [G. Vazquez, E. Alvarez, J. M. Navaza, *Journal of chemical and engineering data*, 1995, 40(3), 611-644] to validate the Du Nouy ring method in Fig. S1(a). Second, microfluidic experiments are conducted with the water-ethanol mixtures and 100-cSt silicone oil at low flow rate ratio $\varphi = 0.05$ and data are reported in Fig. S1(b).

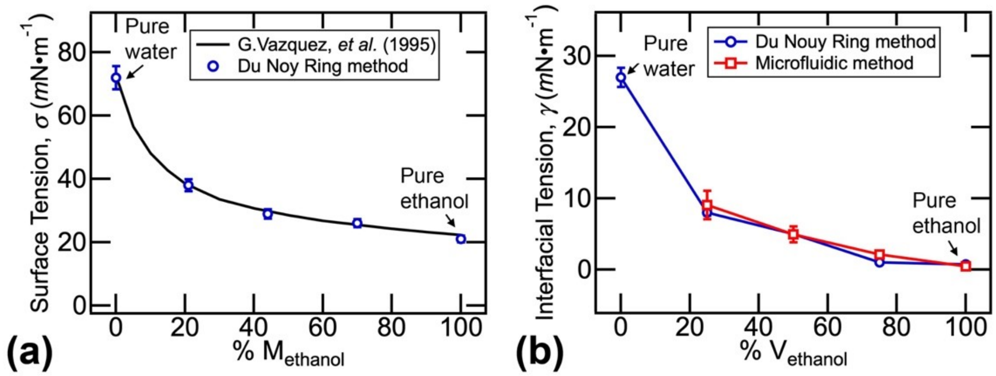

Fig. S1 Validation of interfacial tension measurements with immiscible fluids. (a) Measurements of surface tension σ between a mixture of DI water and ethanol with air at room temperature as a function of the percent mass of ethanol in the mixture. Symbols: Du Nouy ring method, relative error $\Delta\sigma/\sigma = 5\%$ (○). Solid line: published data. (b) Measurement of interfacial tension γ between a mixture of DI water and ethanol with 100-cSt silicone oil at room temperature as a function of the percent volume of ethanol in the mixture. Symbols: Du Nouy ring method, relative error $\Delta\gamma/\gamma = 5\%$ (○) and presented microfluidic method, relative error $\Delta\gamma/\gamma = 21.8\%$ (□).