



Supporting Information

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MICRO COUNTER-CURRENT LAMINAR FLOW FOR HIGHLY EFFICIENT SOLVENT EXTRACTION**

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Relations of the interfacial tension and viscous force in micro gas-liquid counter-current flow

The micro gas-liquid counter-current flows were formed in a microchip having a deep side channel and a shallow side channel as illustrated in Figure S1. The width and depth of the deep channel are 210 μm and 80 μm , respectively, while those of the shallow channel are 170 μm and 11 μm , respectively. The channel wall of the shallow side channel was modified with ODS.¹ Water flows in the deep side channel and air flows in the shallow side channel. The micro gas-liquid counter-current flow is expected to form when the hydrodynamic pressure is lower than critical Laplace pressure (Figure S1). The hydrodynamic pressure and Laplace pressure are induced by pressure-driven water flow and interfacial tension, respectively. In order to verify this hypothesis, the dependence of water flow rates on air flow rates V_{air} where water flow was leaked from the deep side channel to the shallow side channel was examined. The results are presented in Figure S2, where the water leakage flow rate (in $\mu\text{L min}$ units) was converted to a pressure difference ΔP by utilizing the Hagen-Poiseuille relationship. The solid line in Figure S2 indicates theoretical value calculated from the Young-Laplace equation as follows.

$$\Delta P_L = \frac{\gamma}{R} = \frac{2\gamma \sin(\theta - 90^\circ)}{d}$$

where γ is the interfacial tension, d is the depth of the shallow side channel of 11 μm , R is the radius of liquid curvature and θ is the advancing contact angle of water on the ODS-modified surface. The literature value of 72 mN m^{-1} was used for γ .² and θ was measured to be 115°. The critical pressure difference does not depend on the air flow rates. This result suggests that the dominant forces are interfacial tension and the viscous force of the aqueous phase while the viscous force of the gas phase is negligible.

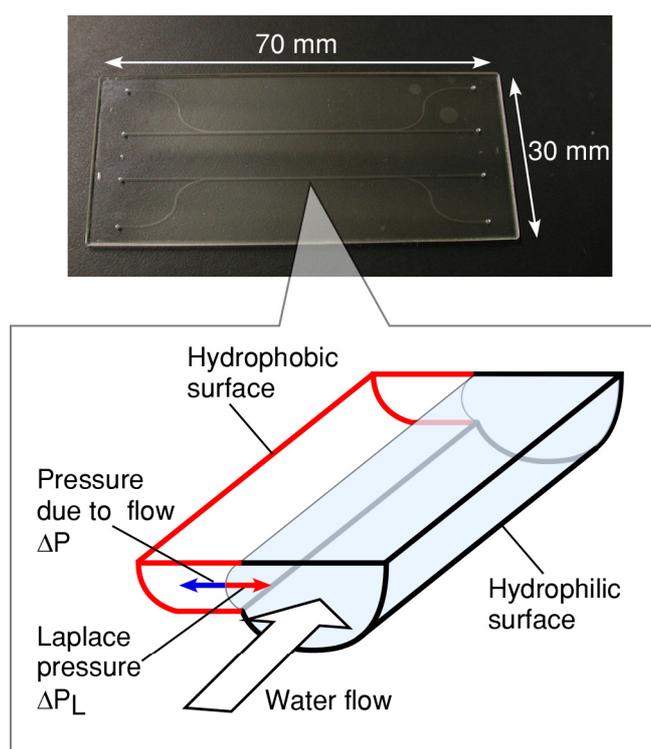


Figure S1. Picture of the microchip and illustration of cross sectional view. The microchip has a deep side channel and a shallow side channel. The shallow side channel wall has a hydrophobic surface made by modifying it with ODS. The forces at the gas-liquid interface that operate to form the micro gas-liquid counter-current flow are the pressure difference (ΔP) due to flow and the Laplace pressure (ΔP_L) due to gas-liquid interfacial tension.

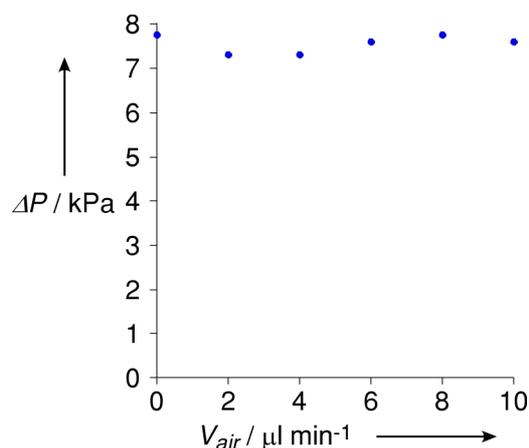


Figure S2. Pressure difference as a function of gas flow rates. Blue circles show experimental values when the water flow was leaked from the deep side channel to the shallow side channel.

Evaluation of color pictures of the micro counter-current flow

In order to verify the phase separation of the micro counter-current flow from the color pictures, the RGB color intensities in figure S3a and b were analyzed. The red color intensities of the aqueous phase and the green color intensities of the organic phase in left picture of figure 2b are described as $X_1 = \bar{X}_1 \pm \sigma_{X_1}$ and $Y_1 = \bar{Y}_1 \pm \sigma_{Y_1}$, respectively, where \bar{X}_1 and \bar{Y}_1 are the average of the color intensities at 20 points and σ is the standard deviation. Likewise, the color intensities in right picture of figure 2b are described as $X_2 = \bar{X}_2 \pm \sigma_{X_2}$ and $Y_2 = \bar{Y}_2 \pm \sigma_{Y_2}$, respectively. X_1 , X_2 , Y_1 and Y_2 are estimated as 166.5 ± 5.2 , 112.9 ± 6.0 , 68.3 ± 5.3 and 92.0 ± 3.6 , respectively. The ratios of red color intensity of the aqueous phase to that of the green color intensity of the organic phase are calculated as $X_1 / Y_1 = 1.66 \pm 0.17$ and $X_2 / Y_2 = 1.81 \pm 0.09$. These good agreements between two values verify the phase separation of the micro counter-current flow.

TLM signal intensity in the aqueous and toluene phases as a function of the flow rates

Figure S3 shows the TLM signal intensity in the aqueous and toluene phases as a function of the flow rates. Since the sensitivity of TLM depends on the optothermal properties of the solvent,^[3] the absolute size of the TLM signal intensity of the aqueous phase is different from that of the toluene phase. As the flow rate decreases, the TLM signal intensity of the toluene phase becomes lower and that of the aqueous phase becomes higher. This tendency indicates that the Co-complex is extracted from the toluene into the water.

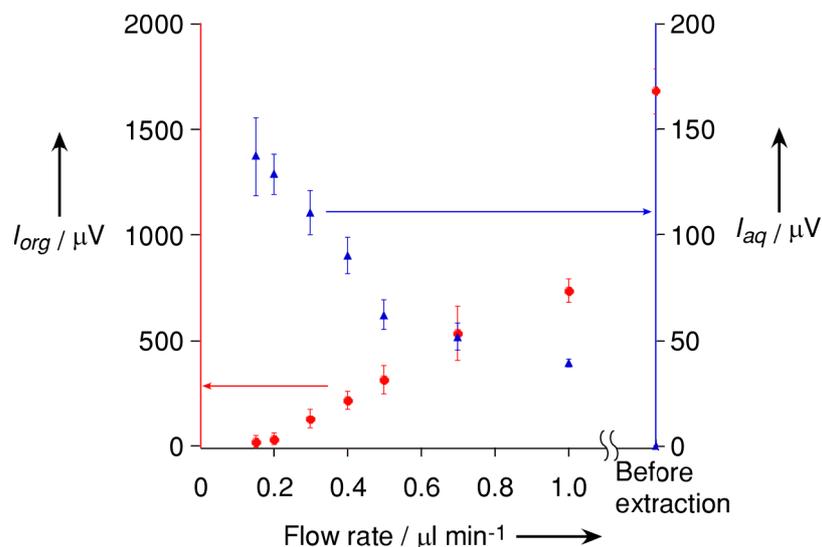


Figure S3. TLM signal intensity in the aqueous and organic phases for the extraction of Co-2-nitroso-5-dimethylaminophenol complex in toluene as a function of the flow rates. The red circles show the TLM signal intensity of the organic phase (I_{org}) and the blue triangles shows that of the aqueous phase (I_{aq}). b) The theoretical plate number (N) as a function of the flow rate.

Calculation of theoretical plate number

The following equation was used for calculating the theoretical plate number.

$$\left(\frac{V_{org}}{DV_{aq} + V_{org}} \right)^N = \frac{C_{org}}{C_{0org}} = \frac{S}{S_0} \quad (\text{S1})$$

where N , V_{aq} , V_{org} , C_{org} , C_{0org} , S and S_0 are the theoretical plate number, the volume of the aqueous phase, the volume of the toluene phase, the concentration of Co-complex in toluene after extraction, the concentration of Co-complex in toluene before extraction, the TLM signal intensity in toluene phase after extraction and the TLM signal intensity in toluene phase before extraction.

References

- [1] A. Hibara, S. Iwayama; S. Matsuoka, M. Ueno, Y. Kikutani, M. Tokeshi, T. Kitamori, *Anal. Chem.* **2005**, *77*, 943-947.
- [2] *CRC Handbook of Chemistry and Physics, 82nd ed.* (Eds.: D. R. Lide), CRC Press, Boca Raton, FL, **2002**.
- [3] S. E. Bialkowski, *Photothermal Spectroscopy Methods for Chemical Analysis*, Wiley & Sons, New York, **1996**.