Supporting Information

Formation, Dissolution, and Transfer Dynamics of a Millimeter-scale Thin Liquid Droplet in Glycine Solution by Laser Trapping

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S1: Estimation of Glycine Concentration in a Dense Droplet

As described in section 3-1, we successfully demonstrated the dense droplet formation through the characteristic surface deformation. Previous experiments by the authors showed that the intensity of the backscattered He-Ne laser at the glass/solution interface becomes lower during the droplet formation.[1] The result revealed that the droplet has a higher refractive index due to the increase of the solute concentration. Here, accurately estimating the absolute concentration of the formed droplet is essential. Fortunately, the high stability of the droplet enables us to estimate the concentration by picking up a potion from the surrounding solution. First, the droplet volume was estimated by using the 2D solution profile obtained at 300 sec. By assuming that the convex-shaped droplet has a parabolic shape, the profile can be fitted well with the parabolic function; $y = ax^2 + b$, where y and x are surface height and horizontal position on the cover glass (the droplet center is set to 0), respectively. The average volume measured in four samples was 2.2 μl.

Next, the concentration of the droplet was estimated in comparison with its surroundings. After the droplet was formed by 300 sec-laser irradiation, the trapping laser was turned off, and a 5 μl portion of the solution surrounding the droplet was carefully sampled with a micropipette as depicted in Figs. S1a (1)-(3). Since the droplet remains stably for about 1 min even without the laser beam, the surrounding solution can be safely and easily collected. After the extraction, the remaining solution on the glass substrate was allowed to sit for 5 min (Fig. S1a (4)), since it takes about 3 min to recover to the homogeneous solution as described in section 3-2. A 5 μl portion of the homogeneous solution was then sampled as shown in Fig. S1a (5), which is hereafter called “resultant solution”. Both the surrounding and the resultant solutions were diluted by adding a 1.5 ml of D_2O before pouring them into a quartz cuvette of 1 cm optical pass. The UV absorption measurement for each sample was performed with the reference sample of neat D_2O, and Fig. S1b plots their absorbance at 210 nm. The measurement results showed that the average solute concentration of the resultant solution is 6% higher than that of the surrounding solution.

Finally, the absolute solute concentration of the droplet was calculated. Figure S1a summarizes the corresponding molar concentration and volume of the initial solution, droplet, surrounding and resultant solutions. The solute concentration of the resultant solution (C_{res}) was calculated to be $1.06C_{sur}$ since its concentration was 6% higher than that of the surrounding solution. The estimated total volume of the sample after the 1^{st} extraction of 5 μl was 35 μl. Here, the loss of the solution by evaporation during the irradiation was not considered since earlier experiments confirmed that the solution thickness after the 300 sec-laser irradiation was almost identical to that before the irradiation. Therefore, the concentration and volume can be correlated with the following equation; $2.2 \times C_{dro} + 32.8 \times C_{sur} = 35 \times 1.06 \times C_{sur}$, which results in a relation equation of $C_{dro} = 2.0 \times C_{sur}$. At this stage, the concentration of the droplet was twice that of the surrounding solution. Next, we pay our attention to the process of Figs. S1a (1)-(2). The concentration can be correlated with each other according to the following equation; $40 \times C_{ini} = 2.2 \times C_{dro} + 37.8 \times C_{sur}$, giving a notable relation equation of $C_{dro} = 1.9 \times C_{ini}$. The equation revealed that the concentration of the droplet is 1.9 times that of the initial solution, *i.e.,* 7.0 M (supersaturation value; 2.6).

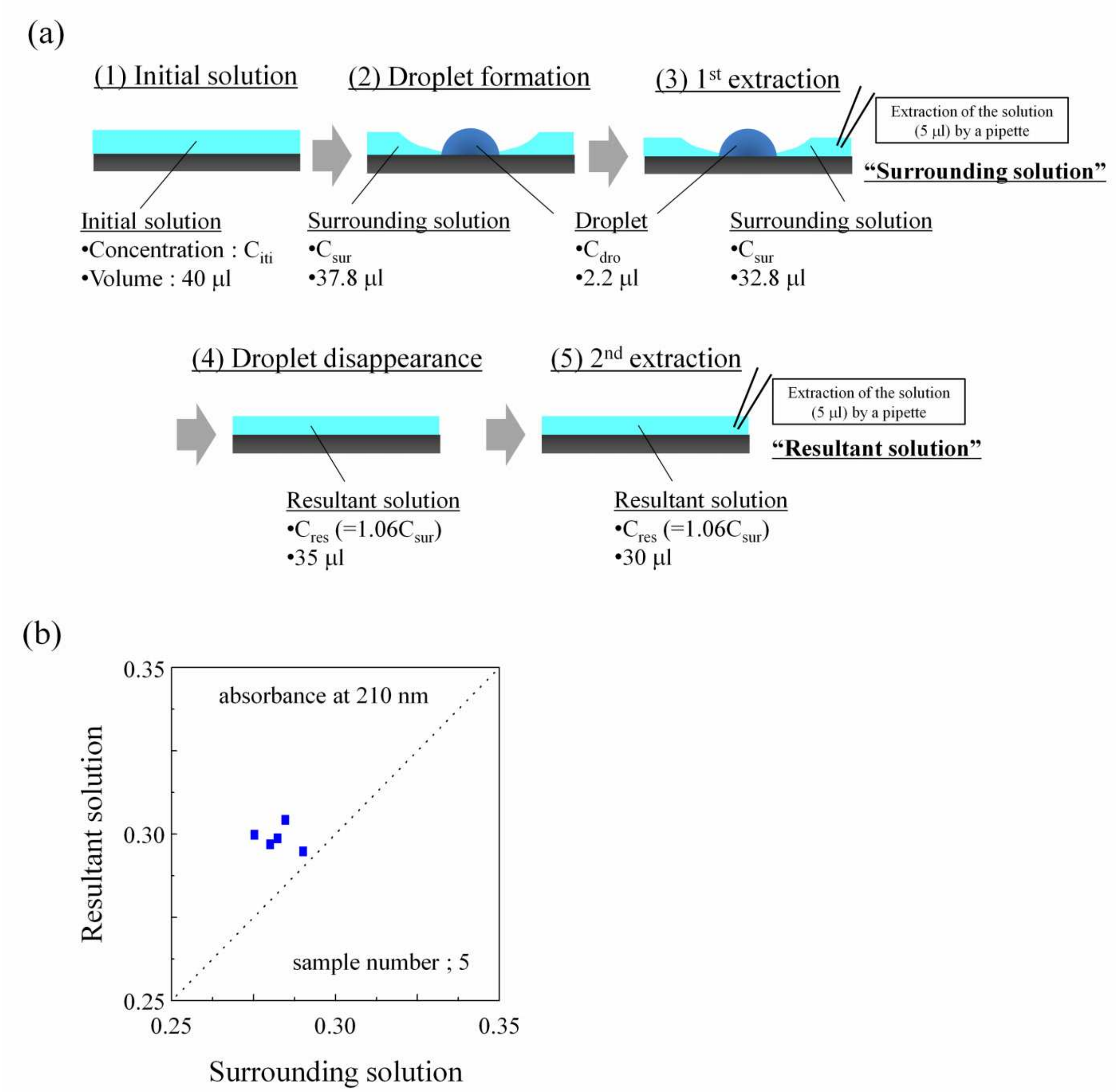

Figure S1. (a) Procedure for analyzing solute concentration in the droplet. Estimated molar concentration and volume before and after laser irradiation are included. (b) Relation of absorbance at 210 nm between the surrounding and the resultant solutions.

REFERENCES

(1) Yuyama, K.; Sugiyama, T.; Masuhara, H. *J. Phys. Chem. Lett.* **2010**, *1*, 1321–1325.