

## Microfluidic Generation of Ultrafine Double Emulsions from Femto- to Atto-liter Scale Induced by Diffusive Phase Separation

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Microfluidic emulsification techniques have attracted significant research interest because of the precise and robust controllability of the size, morphological uniformity, and composition of the double emulsions. However, the minimum achievable droplets are in the range from pico- to nano-liter scale (typically  $>30\ \mu\text{m}$  in external droplet diameter) because double emulsions are created at the expense of more complex microfluidic systems. Here, we demonstrate a novel PDMS-glass hybrid microfluidic device which enables to generate highly uniform ultrafine double emulsions in femto- to atto-liter scale with unprecedented simplicity and controllability. The structural evolution from single to double emulsions occurs through spontaneous diffusive phase separation. In the present study, we capture the internal structure by solidifying the double emulsion droplets with silica precursor through sol-gel chemistry. The structural transition can be characterized on nanometre scale by electron microscopy analysis of the resulting silica particles. This microfluidic approach enabled facile control over droplet size, internal/external droplet diameter ratio, and internal morphology.