

A 3D-PRINTED MICROFLUIDIC APPROACH FOR FABRICATING MICROCAPSULES IN AQUEOUS ONE-PHASE SYSTEMS (AOPS)

Mingjie Li ^a, Xiaotong Song ^a, Joseph Berry ^a and Raymond Dagastine ^{a}*

^a Department of Chemical Engineering, University of Melbourne, Parkville, Victoria 3010, Australia

Mingjie2@student.unimelb.edu.au

Fully aqueous microcapsules are attractive for encapsulation applications because they offer mild processing conditions, good compatibility with aqueous formulations, and avoid the use of organic solvents. So far, the most established strategies for water-in-water (w-w) capsule fabrication relied on aqueous two-phase systems (ATPS). These systems enable compartment formation through phase separation, but they also restrict formulation design because capsule generation depends on preserving immiscibility between the two aqueous phases. Here, we propose an alternative route based on aqueous one-phase systems, where water in water capsules are formed directly from miscible polymer and surfactant streams. This approach preserves the benefits of fully aqueous processing while broadening the range of materials and formulations available beyond conventional ATPS based designs.

In this study, we present a needle-assisted 3D-printed microfluidic platform for the direct formation of w-w capsules from polymer-surfactant systems [1, 2]. By bringing the aqueous streams into controlled contact within a tailored flow geometry, the device eliminates the need for a separate templating phase. Its 3D-printed design also allows channel dimensions and device architecture to be readily adapted for different formulation systems, enabling reproducible production of microscale capsules with storage stability of up to six months.

Capsule formation is driven by polymer-surfactant coacervation [3, 4], which generates a shell around the inner aqueous stream and achieves encapsulation. Surfactant addition further improves colloidal stability and reduces aggregation during storage. SEM and AFM results showed that the capsule shell remained continuous and intact across both the microscale and nanoscales. In addition, particle accumulation tests using 1 μm polystyrene particles dispersed in the inner stream showed that, after storage, the particles accumulated along the inner surface of the capsule shell while the surrounding continuous phase remained clear. These observations indicate that the capsules maintained their encapsulation integrity and effectively confined the particles within the shell boundary.

Overall, this work establishes a practical strategy for generating stable and tunable w-w capsules in an aqueous one-phase environment. While further optimisation is required to minimise ancillary build up of PS complex precipitation in the microfluidic channel, the integration of controlled microfluidic assembly and polymer-surfactant complexation provides a versatile platform for developing all-aqueous capsule systems with potential applications in personal care, food, and drug delivery.

Keywords:

microencapsulation, microfluidics, coacervation, microcapsule, aqueous one-phase system (AOPS)

References:

1. Jeyhani, M., et al., Microneedle-assisted microfluidic flow focusing for versatile and high throughput water-in-water droplet generation. *Journal of colloid and interface science*, 2019. 553: p. 382-389.
2. Li, M., et al., Template-Free Microfluidic Fabrication of Water-in-Water Microcapsules for Controlled Release. *ACS Applied Materials & Interfaces*, 2026. 18(7): p. 12243-12254.
3. Keshavarzi, B., et al., Formation of Structured Membranes by Coacervation of Xanthan Gum with C_n TAB Surfactants. *Langmuir*, 2019. 35(42): p. 13624-13635.
4. Guzmán, E., et al., Self-consistent mean field calculations of polyelectrolyte-surfactant mixtures in solution and upon adsorption onto negatively charged surfaces. *Polymers*, 2020. 12(3): p. 624.