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LIQUID-LIQUID ENCAPSULATION: A PARADIGM TOWARDS ROBUST, EFFICIENT, MULTIFUNCTIONAL WRAPPING

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ABSTRACT

Encapsulation of liquid-phase analytes is crucial in industries such as pharmaceuticals, nutraceuticals, cosmetics, and food processing, as it protects sensitive compounds, facilitates controlled release, and enhances stability. In this study, we present a novel and scalable liquid-liquid encapsulation framework that enables controlled wrapping of liquid-phase analytes with thin shell layers of compatible shell-forming liquids. Two approaches are demonstrated: an ultrafast impact-driven approach and an active-actuation-assisted approach.

The impact-driven approach involves the impingement of a core droplet, containing the target analyte(s), from a vertical separation onto thin interfacial layers of shell-forming liquids floating on a host liquid bath. Depending on the kinetic energy of the core droplet, two possibilities arise: the core droplet either penetrates through the interfacial layer(s) ("interfacial penetration") or becomes entrapped within ("interfacial trapping"). Through theoretical analyses and direct experimental evidence, we show that both these outcomes result in encapsulation, provided certain interfacial energy criteria are met. We demonstrate several practical pathways to harness the potential of both outcomes, including robust wrapping of vulnerable core analytes in aggressive environments, control over shell thickness, the development of multifunctional and multicomponent capsules with tunable mechanical robustness, and multilayer wrapping. This method also facilitates the incorporation of stimuli-responsive shell materials. For example, with magneto-responsive ferrofluid as the shell layer, we achieve stable, non-contact manipulation of encapsulated droplets, enabling controlled coalescence and targeted release using an externally applied magnetic field. This is particularly valuable for on-demand release in dynamic environments, such as targeted drug delivery applications.

We also introduce an alternative active actuation-assisted approach, which could be useful when the core droplet is responsive to external stimuli. In this approach, a magneto-responsive core droplet (e.g., polyethylene glycol-based ferrofluid) is dispensed onto a polydimethylsiloxane (PDMS) interfacial layer floating on a host water bath inside a cuvette. By applying a permanent magnet underneath the cuvette, we demonstrate that the core droplet can be pulled through the interfacial layer and successfully encapsulated without the need for impact. This approach circumvents the kinetic energy requirement, providing better control over the process and avoiding issues like splashing or air entrapment. The versatility of this technique is further demonstrated by encapsulating multiple ferrofluid core droplets within a single PDMS shell, with each core compartmentalized to prevent mutual interactions, relevant for micro-reaction or micro-mixing applications.

We believe the approaches presented herein will open up a paradigm for the minimally restrictive fabrication of practically relevant multifunctional encapsulated cargos.