MICROCAPSULES WITH TUNABLE DIMENSIONS AND MECHANICAL PROPERTIES MADE USING MICROFLUIDICS

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ABSTRACT

Filled capsules are featured prominently in self-healing materials as carriers of healing agents, which are released upon crack-induced rupture of the capsules [1]. They are routinely fabricated by emulsification of immiscible liquids and subsequent interfacial polymerization. Deliberately tuning the size, shell thickness and composition of capsules is crucial to control their strength and permeability. These features are important to tailor the capsules to specific matrices, but accurate control is often difficult to achieve using traditional emulsification techniques.

We exploit a recently developed microfluidic technique to form tailored microcapsules from monodisperse double emulsion templates [2]. In this method, a fluid is dripped from an orifice into a second, immiscible fluid in a co-flow configuration within a glass microcapillary device. Both fluids are in turn engulfed by a third fluid, which flow-focuses all phases into a collecting orifice, forming the double emulsions. To stabilize the emulsions, polymeric surfactants such as poly(vinyl alcohol) are added to the inner and outer fluids. The size of each double emulsion phase can be coarsely tuned through the orifice diameters and finely adjusted with the respective fluid flow rates. Using suitable acrylate monomers as the middle fluid and in situ photopolymerization [3], we can produce monodisperse microcapsules of diameters varying from 60 to 250 µm and shell thicknesses in the range of 7 to 50 µm, such as the ones depicted in Fig. 1.

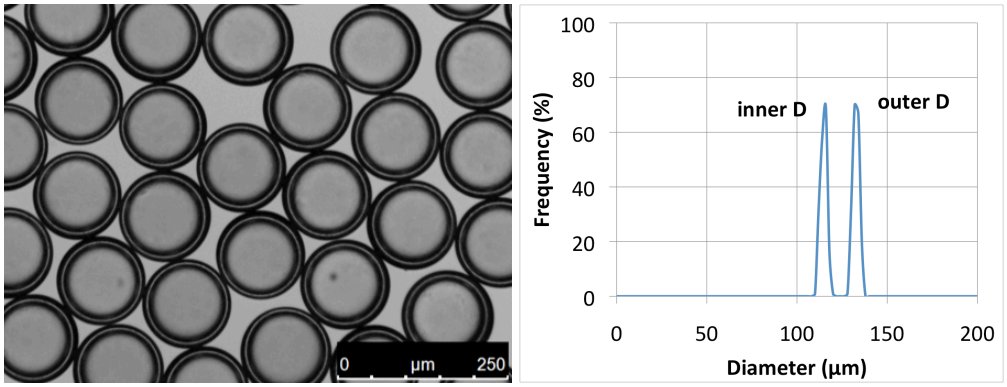

Figure 1: A sample of polymerized microcapsules (left) and its size distribution (right).

To predict our droplet and microcapsule sizes, we developed an analytical model that describes the flow-induced dripping based on a balance between shear forces and interfacial tension. We found that despite the complex flow behavior in a microfluidic device, droplet size can be predicted with established break-up dynamics of isolated droplets. The model accurately describes a wide range of experimental data from both our work and literature without relying on fitting parameters.

Using different chemical compositions for the middle fluid allows for tuning of the permeability and mechanical properties of the capsule shell. Monomers whose homopolymers have low glass transition temperatures (T_g), such as 2-phenoxyethyl acrylate (PEA), yield elastic and ductile capsules. In contrast, high T_g systems like isobornyl acrylate (IBOA) form stiffer and brittle capsules, as indicated in Fig. 2. The spectrum in between these extreme cases can be covered using monomer blends. By adding difunctional monomers, a cross-linked shell network can be formed which decreases its permeability. The shell can be further modified by the addition of reinforcing or functional nanoparticles, thereby creating composite microcapsules.

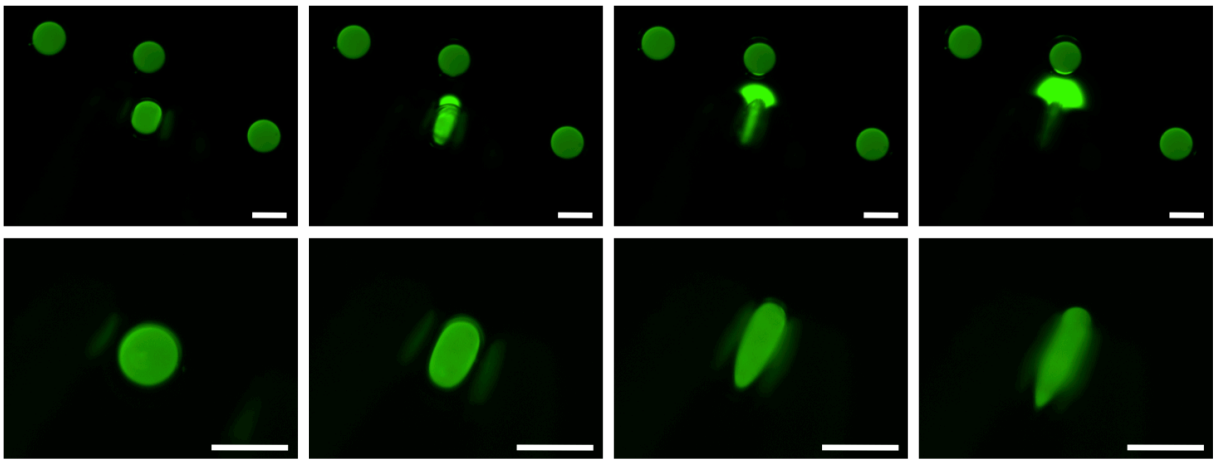

Figure 2: Manual deformation of poly(IBOA) (top row) and poly(PEA) (bottom row) capsules filled with a concentrated fluorescein solution. All scale bars represent 100 µm.

The proposed model to predict droplet sizes combined with the wide spectrum of materials that can be used in this microfluidic approach allow for unprecedented control over both the dimensions and the properties of microcapsules, enabling the design and fabrication of capsules for specific mechanical conditions.

REFERENCES

[1] S. R. White, N. R. Sottos, P. H. Geubelle, J. S. Moore, M. R. Kessler, S. R. Sriram, E. N. Brown, S. Viswanathan, Autonomic healing of polymer composites, *Nature*, **409**, 2001, pp. 794-797.

[2] A. S. Utada, E. Lorenceau, D. R. Link, P. D. Kaplan, H. A. Stone and D. A. Weitz, Monodisperse Double Emulsions Generated from a Microcapillary Device, *Science,* **308**, 2005, pp. 537-541.

[3] H.-J. Oh, S.-H. Kim, J.-Y. Baek, G.-H. Seong and S.-H. Lee, Hydrodynamic micro-encapsulation of aqueous fluids and cells via 'on the fly' photopolymerization, *Journal of Micromechanics and Microengineering*, **16**, 2006, pp. 285-291.