Experimental and Computational Study of Mass Transport in Novel Emulsion Systems: Strategies for Reaction Engineering and Microparticle Preparation

by

Tonghan Gu

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Emulsions are complex fluids with interesting physiochemical properties, which have been widely used in health and personal care, food, coating, and manufacturing, *etc.*. Rather than considering emulsions as passive materials, they can also be used as active blocks for material preparation and chemical synthesis. This thesis presents a study of mass transport phenomena in two specific types of emulsion systems: microfluidic emulsions and concentrated food emulsions.

For microfluidic emulsion systems, the first mass transport phenomenon studied is the exchange of chemicals between microfluidic droplets, which are 10-100 μm in size, and nanodroplets, which are dispersed as a nano- or mini-emulsion. Chemically, thermally, or electrically induced coalescence and micelle activity control the mass exchange between micro- and nano-droplets, leading to applications in reaction engineering and microparticle preparation. Microdroplets function as micro-reactors that receive chemical from nanodroplets with both the addition rate and dosage well-controlled. The microdroplets could also function as micro-reservoirs that steadily supply chemical to the nanodroplets. For microparticle preparation, microdroplets function as templates to be solidified by reagents carried by the nanodroplets.

The second mass transport phenomenon in microfluidic emulsions is the evaporation of droplet solvents or the exchange of solvents between droplets and the continuous phase, which leads to solid precipitation. In particular, this thesis focuses on the formation of drug crystalline particles. A novel solvent/anti-solvent exchange method with a hydrogel binder was developed to prepare highly monodisperse microparticles of either hydrophilic or hydrophobic drugs from microdroplet templates. In addition, we also improved a previously developed spherical crystallization method based on droplet solvent evaporation. We used the same hydrogel but as a temporary immobilization media to prevent droplet coalescence and to extend the applicable solvent library of this method for industrial applications.

For concentrated food emulsions, the mass transport phenomenon studied is the fast removal of the continuous phase and the microencapsulation of lipids into microparticles. With the spray drying technology, “powdered oil” containing up to 55 wt% (dry mass basis) of liquid oil was successfully prepared from concentrated milk protein stabilized emulsions. We discovered that pre-evaporation of raw milk not only offers energy cost savings, but also reduces fat loss. With additional carbohydrates, the surface extractable fat was reduced and powder wettability was improved. This product will serve as the main ingredient of an instant powder ready-to-use therapeutic food for treating child malnutrition in India.

Thesis supervisor: T. Alan Hatton
Title: Ralph Landau Professor of Chemical Engineering and Director of David H. Koch School of Chemical Engineering Practice
Massachusetts Institute of Technology

Thesis supervisor: Saif A. Khan
Title: Associated Professor of Department of Chemical and Biomolecular Engineering
National University of Singapore