Designing droplet microfluidic systems: from chemistry of surfaces, through rheological properties of fluids to geometries of the channels

Author: Ladislav Derzsi

Supervisor: Piotr Garstecki

Abstract

Microfluidic systems provide new vistas for generation of droplets and bubbles with a perfect control over their volume and structure. The ability to form organic droplets in microdevices finds uses (and promises of uses) in a range of applications that divide into two general classes: (i) preparatory technique that utilize the droplets to form particles and capsules and (ii) analytical and synthetic techniques that use micro-droplets as beakers for chemical reactions or for incubation of living cells. These techniques offer a range of attractive characteristics in comparison to conventional processing in chemistry and biology.

Flow in microfluidic systems is dominated by the interfacial effects. Thus the subject of wettability of the walls of the channels, and of the interfacial tension between liquids are of outmost importance. As the Reynolds number is typically small, the value of viscosity coefficient and all non-Newtonian effects have direct impact on the dynamics of microfluidic systems. Finally, the combination of laminar flow and of capillary effects makes it possible to use geometry of the channels to effectively modulate the flow and higher order functions of microfluidic systems.

This thesis discusses all the above effects and presents the results of experimental studies of (i) chemical modification of the surface of the substrate material, (ii) the role of visco-elastic effects in droplet formation, and finally (iii) the application of some specific geometries for passive generation and handling of microdroplets.

Stability of the process of generation of droplets depends critically on the interfacial tension between the liquid phase and the solid substrate of the channels walls. Considering e.g. water-oil two phase flow the preferential wetting of the walls by the continuous phase determines the type of the emulsion. It follows that in hydrophobic devices (as e.g. polycarbonate) produce water-in-oil (W/O) emulsions while for generation of oil-in-water (O/W) emulsions hydrophilic walls are needed. In the first part of the thesis we describe and characterize two novel modification techniques of the microchannels milled in

polycarbonate to render the surface hydrophilic. The first technique is based on a deposition of a polyelectrolyte double layer (poly(allyamine hydrochloride) (PAH) and poly(sodium styrene sulfonate) (PSS), which is referred as layer-by-layer (LbL) polyelectrolyte multilayer (PEM) assembly. This modification technique yields stable hydrophilic surface proper for generation of organic droplets in aqueous continuous media over weeks to months even without the use of any surface active additives. The second technique uses a solution of tin(II)-chloride and renders the surface also hydrophilic. This latter method is compatible with other hydrophobic modification techniques of the polycarbonate and allows the formation of water-in-oil-in-water (W/O/W) double emulsion.

The overwhelming majority of droplet generator studies are dealing with solely Newtonian flow and only a small fraction of studies consider non-Newtonian fluids. Furthermore, these rare studies use the non-Newtonian (usually viscoelastic) liquids only as the droplets phase, while the effect of the non-Newtonian behavior of the continuous phase liquid remained undiscovered.

In the second part of the thesis we investigate the effect of the elasticity of the outer phase liquid on formation of Newtonian droplets in flow focusing droplet generators. We use a constant viscosity elastic liquid (also known as Boger fluid) along with the Newtonian liquid of the same shear viscosity, which makes possible the separation of the elastic effect from the viscous effects.

Finally, in the third part of the thesis we present passive methods of generation and handling of microdroplets based solely on the geometry of the device. These microfluidic modules allows us (i) to produce droplets with fixed volumes regardless of the rate of flow, and thus significantly reduce the fluctuation of the size/volume of the droplet caused by the pumps, (ii) synchronize the droplet merging (iii) or perform fast on-chip titration using the module as a microfluidic dilutor.