Motility of active droplets in lipid systems

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SUMMARY

In nature, microorganisms and cells have evolved sophisticated protein machinery that enables them to move effectively towards favorable conditions that ensure their survival. To engineer artificial compartments mimicking motile behavior observed in nature requires these compartments to move autonomously, discern their environment, and adapt their movement in response to external triggers such as light and chemicals. This thesis shows that microscopic oil droplets can develop complex motile functions through the interplay between molecular reactivity and physical processes occurring at the microscopic scale. By coupling chemical reactions to interfacial tension gradients, we show that droplets evolve adaptive motility where the motile speed and trajectory alter in response to external triggers such as light and chemical fuel. Such adaptiveness results in complex motile patterns, enhancement of lipid-producing reaction through chemo-motile coupling, and light-triggered shape morphogenesis.

Chapter 1 outlines this thesis, and it introduces the challenge of building artificial motility in microscopic compartments.

Chapter 2 reviews the potential of microscopic oil droplets in lipid solutions as minimalistic motile compartments and discusses how complex motile functionalities can arise by combining chemical reactions with physicochemical phenomena.

Chapter 3 shows that a chemical reaction can be coupled reversibly to the motility of microscopic droplets. Our system consists of a lipid-producing chemical reaction and oil droplets that can move spontaneously through the establishment of Marangoni flows. As lipids are produced in the system, the oil droplets remain stationary, but once the concentration of lipids surpasses the critical micelle concentration, the micelles induce Marangoni flows in the fatty alcohol droplets in the system, which sets them in motion. The motile droplets that are chemotactic can move towards regions populated by the precursors. We show that this results in an increase in the production of lipids in the system as the droplets create additional interfaces for the self-producing chemical reaction to form lipids. Hence, we establish a chemo-motile coupling where lipids produced by the self-producing chemical

reaction induce Marangoni motility in droplets. In return, the chemotactically moving droplets enhance the production of lipids.

In Chapter 4, we study the motility of chemically distinct oil droplets and narrow down the parameters that contribute to the efficiency of motility. We show that the physicochemical parameter such as viscosity and solubility of the oil in the micelle solution determines how fit a motile oil droplet is. This can be studied by looking at the influence of advection and diffusive forces (through Peclet number) in the system. Taking this a step ahead, we demonstrate that by playing with the precursors of a lipid-producing reaction, we can promote selective pre-programmed motility in a population consisting of two distinct oil droplet populations.

After establishing that droplets can propel autonomously and be coupled to chemical systems, in Chapter 5, we show that motile droplets can be chemically encoded to have a predesigned functionality. We use photoswitches as a mediator to make motile droplets responsive to light. The micelles that drive Marangoni propulsion is composed of lipids containing an azobenzene photoswitch. Upon activation by light, a change in the shape and dipole moment of azobenzene moiety alters the properties of micelles. As micelles drive the motion of droplets, this translates into a modification in the motile behavior of droplets.

Building complex systems where functional motile compartments are part of it requires a better understanding and control over the motile characteristics and insights into how these characteristics can be modified on demand. Chapter 6 shows that motile droplets with reconfigurable motile speed and trajectory can be chemically pre-programmed. The oil droplets contain an azobenzene oil component which, upon light irradiation, switches between liquid crystal or isotropic states. With this phase transition, the droplet either accelerates or decelerates depending on the lipid that initiates Marangoni motility. Through selective irradiation, we create spatially segregated droplet populations with different motile characteristics.

Several swimming microorganisms use appendages such as cilia or flagella to propel them forward. So far, autonomous movement of compartments has been created in artificial systems by either catalytic reactions that produce bubbles or through interfacial mechanisms such as the Marangoni effect. This thesis focuses on the latter. In Chapter 7, we show that interfacial mechanisms can also result in the formation of appendages on liquid crystal droplets in addition to motility. Molecular

photoswitches induce the driving interfacial effects by amplifying a change in molecular properties occurring at the nanoscale to larger length scales.