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Title: Autonomous Fluid Flow on Supramolecular Interfaces: A step towards Powerless Microfluidic Device

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Abstract:

Flow control is at the core of a microfluidic device and plays an important role in unit operations. Non-mechanical nano and microfluidic devices that operate without an external power source and can be customized to individual demands define the next generation of smart devices. Due to their promise of combining various features with high throughput and low sample amounts, microfluidic devices have gained substantial attention in both academic and industrial research, particularly for microfluidic applications. We have shown that supramolecular interfaces (such as host functionalized films and gels) were used as scaffolds to devise the pumps which drive fluid flow in the presence of any guest molecule that initiates the "host-guest" molecular recognition. The utilisation of noncovalent interaction-based reactions to construct a self-powered micropump provides specificity, sensitivity, and selectivity while removing a significant barrier in microfluidics: the requirement for external pressure-driven pumps to push fluids through devices. The fluid velocities achieved in these systems are directly proportional to the concentration of the component that triggers "host- quest" molecular recognition. The micropumps made from these noncovalent building blocks will open up new avenues in designing dynamic systems for applications ranging from single-use diagnostics to microanalysis. However, fluid flow generated from supramolecular interactions has mainly remained unexplored, and further research is needed to have a thorough grasp of the fundamental principles of actuation and the parameters that determine the output of the micropump, as well as experimental analysis of some of their possible applications. This work highlights all of the studies conducted with self-powered micropumps, from the invention of the micropump design to the efforts made in understanding the "host-guest" based pump concept as a whole. Self-powered supramolecular micropumps could potentially provide a solution for a powerless microfluidic device where the fluid flow can be manipulated by modulating noncovalent interactions. In our first approach, we attempted to fabricate thin filmbased micropumps by depositing β-cyclodextrin ("host") functionalized polymer on a glass slide via LbL assembly. These supramolecular micropumps turned on fluid flow upon the 1addition of "guest" molecules to the multilayer films. The flow velocity was tuned by the concentration of the "guest" molecules as well as the number of "host" layers inside the multilayer films. The numerical modeling uncovers that the solutal buoyancy originated from "host-guest" complexations is primarily responsible for the fluid flow. In view of potential application in self-powered devices, the micropump was integrated into the microfluidic device to show molecular and colloidal transport over long distances. In the next work, a valveless micropump was designed via dynamic supramolecular interaction between βcyclodextrin (β- CD) and benzimidazole (Bzl). It shows flow reversal in response to the pH change. An L- shaped microchannel was used to demonstrate the flow reversibility over long distances. In another approach, again we have developed thin film-based micropumps by depositing β- cyclodextrin ("host") functionalized polymer and pillar[5]arene on a glass slide, which can be utilized to recognize isomers of tryptophan and nitrophenol, respectively. When "guest" molecules were added, these supramolecular micropumps activated fluid flow, and the difference in fluid velocity of "guest" molecules was further used for their recognition. Such a device could aid in the development of new recognition techniques and lead to a better understanding of chiral and molecular recognition in biological systems. Finally, we explored the self-sorting behaviour of pillar[n]arenes (pillar[5]arenes and pillar[6]arenes) on a substrate based on a principle of geometrical complementarity by shape using a macroscopic fluid-flow strategy. Overall, this work demonstrated the designing of a self-powered supramolecular microfluidic device that utilises different supramolecular interfaces to trigger fluid flow. The recognition of different molecules using fluid flow can pave the way for the development of point-of-care devices for the detection of drugs, insecticides, and physiologically relevant

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