Probing and Controlling Interactions and Assemblies at the Nanoscale

Understanding how to tune molecular assemblies and the properties of surfaces of different materials at the meso- and nanoscales can lead to unique and controllable interactions at interfaces for a variety of applications. In the first part of this talk, we use dipolar forces to control the adsorption and alignment of liquid crystals (LCs), which are highly sensitive to surface interactions. Here, we use carboranethiol and -dithiol isomers, which possess the same geometry and lattice when self-assembled on Au(111), but differ in the magnitude and direction of their dipole moments. Hence, self-assembled monolayers (SAMs) of carboranethiol isomers enable us to deconvolve dipole interactions from other factors that influence LC alignment. We fabricate LC displays using carboranethiol SAMs on transparent gold surfaces, prepared by oblique evaporation, and measure the LC orientation and anchoring energy on surfaces treated with each isomer. Our results suggest that the dipole moment direction strongly influences the LC alignment and anchoring energy. In the second part of the talk, we describe bioinspired omniphobic surface coating for rapid cell-deformation devices to enable high-throughput intracellular cargo delivery. Currently, these devices clog within minutes, rendering them inefficient for sustainable cell processing. We have developed a method for coating polyvinylidene fluoride syringe filters with slippery liquid-infused porous surfaces (SLIPS). We see that without this coating, essentially no cells are recovered from the device, due to clogging. However, with the SLIPS coating, we are able to recover 25% of cells. Additionally, we have successfully delivered CD¬­19 chimeric antigen receptors to Jurkat cells, a model T cell line, while maintaining high cell viability. These SLIPS-functionalized devices will enable new opportunities in the development of gene and cellular therapies for a wide variety of disease treatments, which are currently limited due to toxicity, low throughput, and off-target effects.

Multiple-Patterning Nanosphere Lithography: Top-Down Approaches for High-Throughput Gene Modification

Three-dimensional periodic silicon-based nanostructures have shown useful optical, electrical, and mechanical properties, however these types of structures remain difficult to fabricate in a facile manner. Nanosphere lithography is a simple, high-throughput technique that can be used to form large-area, close-packed monolayer arrays of nanospheres. These arrays can be directly used as etching masks to generate silicon-based nanostructures. Typically, the nanostructures produced are created by single etches of the nanosphere array mask. Here, we report multiple patterning nanosphere lithography, which enables size tuning of the nanostructures in three dimensions utilizing the nanospheres for multilayer masking throughout multiple rounds of etching. By exploiting the degradable nature of polystyrene during reactive ion etching, we have fabricated large-area three-dimensional periodic silicon nanostructures. These hierarchical nanostructures can be precisely tailored to be tiered with independent tunability in height and diameter at each level. More complex tube structures can be produced by combining metal deposition with subsequent reactive ion etching. Using this technique, we fabricate periodic arrays of conical nanoneedles for non-viral delivery in cancer immunotherapy. Targeted gene editing using non-viral methods has significant advantages in terms of safe delivery of cargo and cost. Physical techniques to produce membrane disruption via nanoneedles have the capability to penetrate cell membranes and to deliver gene-editing nucleases. However, the challenges for these systems include inconsistency of membrane penetration and slow processing throughputs, which lead to low transfection efficiency and poor viability of cell products. Here, we use an ordered array silicon nanoneedles fabricated by combining nanosphere lithography with isotropic dry etching of silicon. Gene-encapsulated supramolecular nanoparticles are then tethered on the nanoneedle surfaces via supramolecular recognition. By integrating the nanoneedles with microfluidic devices, we demonstrate rapid, efficient, and non-toxic in vitro gene-delivery to model cell lines. The versatile and scalable nanostructured platform proposed here represent an ideal system for the genetic engineering of a variety of cell types that will empower research focused on gene therapies and enable translation of these cellular therapy approaches.