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DOE Computational Nanoscience Project:
Integrated Multiscale Modeling of Molecular Computing Devices

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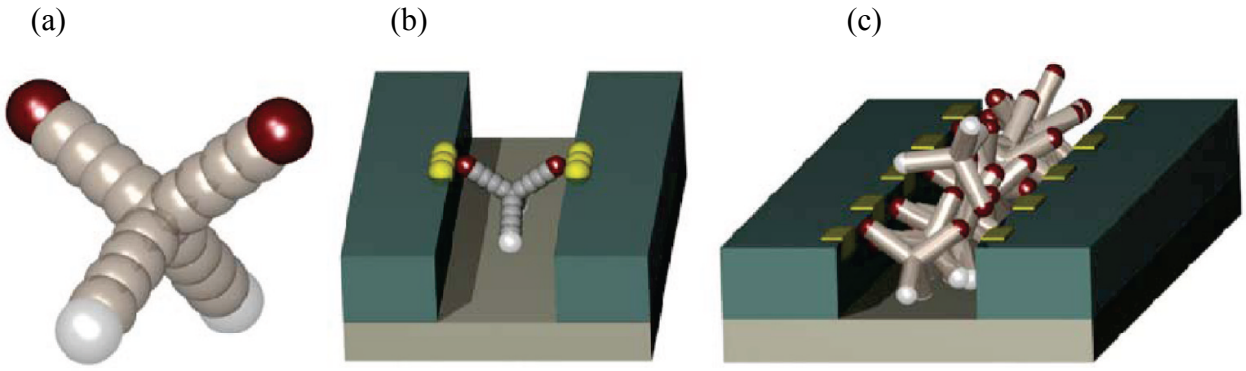
A major challenge in the fabrication of molecular electronics (ME) devices is the nanopatterning of millions of molecular wires necessary in constructing transistors and other devices to realize complex circuitry. Two approaches to ordering molecules into arrays are nanolithography and SA. While nanolithography may ultimately attain pattern sizes near 10 nm, 1-2 nm patterns containing well separated, individual molecular wires are not expected to be achieved with this approach. Instead, self-assembly approaches in which thiolated molecules tightly bind and form well-ordered arrays on gold surfaces is being pursued for fabrication of (ME) devices. The details of the assembly process and the resulting patterns will depend intimately on the interplay between entropic and energetic forces between the molecules and between the molecules and substrate. By appropriately tuning these forces, different nanopatterns that persist over large distances and compartmentalize millions of molecular wires can potentially be obtained. Key to fabrication processes relying on self-assembly is strict control over the assembled structures. ME devices involving chemical assembly in the fabrication process are likely to be imperfect, resulting in potential defects that, if excessive, could cause failure of the device. For this reason, is it important to quantify the degree of order attained by various assembly routes, and the propensity for certain assembly methods to produce defect-free or low-defect ordered structures. As ME devices become increasingly complex, interfacing with arrays of quantum dots and other nanocomponents to yield complex nanoelectronic machines, quantification of order and the development of strategies to control order will become increasingly important.

In collaboration with researchers at Vanderbilt University, North Carolina State University, Princeton and Oakridge National Laboratory we developed multiscale modeling and simulation

methods capable of modeling the synthesis, assembly, and operation of molecular electronics devices. Our role in this project included the development of coarse-grained molecular and mesoscale models and simulation methods capable of simulating the assembly of millions of organic conducting molecules and other molecular components into nanowires, crossbars, and other organized patterns.

Nanotetrapods served as the model nanoscale building block for this study, since they are well studied experimentally. Studies of CdTe nanotetrapods have included fabricating particles with independent control over arm aspect ratio, using capillary interactions to deposit particles into lithographically defined spaces, and analyzing the particle's electrical responses. Our research capitalized on the natural anisotropy of nanotetrapods. Variation in material composition of tetrapod arms allows for a range of current- voltage responses, which can be used to construct working devices. Additionally, material asymmetry can be exploited and material-specific interactions tuned to direct the assembly of nanotetropods into circuits.

Complete results of the computational studies and experimental validation were published in 2009.[1] The use of nanotetrapods as simple logic circuit elements is conceivable, and future focus will shift to more complex logic circuit elements derived from their assemblies. This conclusion is further supported by the results of our directed assembly simulation studies. When material specific interactions are tuned, systems of nanotetrapods can be backgated and aligned between electrodes, thus presenting an avenue to achieve logic circuits. A range of particle parameters lead to assembled structures, and exact material interactions should be determined by the required system tolerance. Future work will center on the electronic modeling of specific tetrapod circuit elements and devising self-assembly schemes for the resulting device.



(a) An individual tetrapod. The red tips are attracted to the electrodes while the white tips are attracted to the floor of the trench. (b) A single tetrapod in a trench. The electrodes are shown in yellow, the floor of the trench in light brown and the substrate surface in green/gray. (c) Multiple tetrapods aligning within a trench with no particle-particle interactions other than excluded volume.

[1] Teich-McGoldrick, S. L., Bellanger, M., Caussanel, M., Tsetseris, L., Pantelides, S. T., Glotzer, S. C. and Schrimpf, R. D., "Design considerations for CdTe nanotetrapods as electronic devices," Nano Letters, 9, 3683-3688 (2009).