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Editorial

Control of molecular self-assembly

Feedback control enables the robust operation of engineered and biological systems in the presence of uncertainty and disturbances. While biological systems embed feedback across the scales, including at the molecular level to regulate intracellular phenomena, engineers typically employ feedback control only at macroscopic length scales, which limits the functionality of engineered systems in comparison to the robustness and adaptivity of function achieved by biology. A primary barrier to the insertion of control in engineered systems is the lack of real-time measurements at small length scales. Additional challenges for feedback control at the molecular scale include the high dimensionality associated with modeling each atom, nonlinear and stochastic dynamics, fast timescales associated with atomic dynamics, and the limited number and bandwidth of actuation.

The promise of molecular scale control was articulated in 1959 by Richard Feynman in a speech entitled “There’s plenty of room at the bottom:”

> I would like to describe a field, in which little has been done, but in which an enormous amount can be done in principle . . . What I want to talk about is the problem of manipulating and controlling things on a small scale.

With the ability to control molecular structure, hierarchical and modular design beginning at the smallest length scales becomes possible, such that materials and systems can be precisely designed and produced for applications ranging from computing to energy to medicine. Arguably the most complex and hierarchical *engineered* system today is the integrated circuit and the systems that are subsequently enabled by embedded computing. The smallest length scales in integrated circuits are now measured in nanometers, and fabrication is achieved in a top-down manner, largely by patterning features on a two-dimensional surface using light. Extending this approach down to the molecular scale is limited by the wavelengths of light. Methods for top-down manipulation of *individual* atoms and molecules include the use of the atomic force microscope (AFM) and electron beam technologies. In AFM, a sharp tip is directly actuated while interacting with atoms or molecules on a surface, based on optical sensing of the probe position. Such technologies provide precise spatial control, but are limited in their scalability for mass production of materials and systems.

Bottom-up self-assembly is an alternative to top-down fabrication. Self-assembly describes the process in which a large collection of molecules interacts through their intramolecular forces. In the unactuated context, the collection of molecules will eventually achieve a configuration that is consistent with its thermodynamic minimum, although at non-zero temperatures the molecules will continue to fluctuate. Achieving this thermodynamic minimum may take a very long time. Directed self-assembly describes the *actuated* process, in which the drift component of the dynamics is defined by intramolecular forces and the actuation is defined by forces between the molecules and one or more externally applied fields. In practice such systems are generally underactuated, having, for example, one mole of molecules (10^{23}) but less than ten “knobs” on the process that can be adjusted. Common process inputs include temperature, pressure, electric field, and magnetic field. Each of these inputs typically alters the dynamics of *every* molecule in the system, although not always equally. By manipulating these inputs, it is possible to influence the overall evolution of the system. System-level metrics such as density and crystallinity are often used to track the evolution, and may also form the underpinnings for a reduced-order dynamic state.

The first paper in this special section provides a review and perspective on the broad range of applications, progress, and challenges for control of molecular scale assembly (contributed by Joel A. Paulson, Ali Mesbah, Xiaoxiang Zhu, Mark C. Molaro, and Richard D. Braatz).

Three original research contributions are then highlighted in three distinct systems:

- “Analysis and control of heteroepitaxial systems,” by Jacob A. McGill, Nasser M. Abukhdeir, Babatunde A. Ogunnaike, and Dionisios G. Vlachos:

  Multiple system-level metrics are considered as process outputs for control of thin film deposition. Reduction techniques are applied to compute a smaller number of composite metrics. A partial integro-differential equation defines the process dynamics, and a traditional multivariable PID control scheme is employed in simulation to demonstrate the approach. This paper primarily addresses the challenges of high state dimension in directed self-assembly.
- “Controlling assembly of colloidal particles into structured objects: Basic strategy and a case study,” by Michael A. Bevan, David M. Ford, Martha A. Grover, Benjamin Shapiro, Dimitrios Maroudas, Yuguang Yang, Raghuram Thyagarajan, Xun Tang, and Ray M. Sehgal:

  The assembly of micron-size particles is controlled using dynamic programming. While these particles are larger than the molecular scale, they embody similar stochastic and nonlinear dynamics, and provide a surrogate system for understanding molecular assembly. Because the particles can be observed in real-time under a microscope, this system provides new insights

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into the specific possibilities and challenges for molecular scale control.

- “On Control of Transport in Brownian Ratchet Mechanisms,” by Subhrajit Roychowdhury, Govind Saraswat, Srinivasa Salapaka, Murti Salapaka:

  The final research contribution is on controlling Brownian rectifiers. Brownian rectifiers are a class of naturally occurring mechanism in bio-molecular transport, which is well suited to enable nano-scale directed transport in engineered systems. Dynamic programming is used to compute the optimal feedback policy, and is applied to a coarse-grained stochastic representation of the high-dimensional molecular assembly. This feedback policy provides a balance between maximizing velocity and energy efficiency, and shows that a significant tradeoff exists.

All three studies are applied in simulation, and the challenges and possibilities for real-time experimental implementation are discussed.

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