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Vision Statement: Materials in Motion

Ben L. Feringa

At the dawn of a new era of interactive and responsive materials and dynamic molecular systems there is ample opportunity to raise the level of complexity enabling mechanical motion, autonomous behavior, or interactive self-regulatory functions. Some of the major challenges and opportunities are discussed with a brief perspective toward future developments on responsive materials.

Taking lessons from Nature on our next journey into uncharted territories of molecular materials, we cannot avoid to dream about the astonishing way living systems sense, function autonomously, and respond to their environment. Touching a material, we can feel the soft or stiff nature or act quickly upon thermal or mechanical stimuli. Via the olfactory system, we can sense and distinguish unpleasant molecules among a large collection around us and adapt autonomously, moving away through the collective motion of molecular motors. The self-regulatory behavior of distinct cells, their membranes, and protein channels to differentiate, build tissues and organs, and to enable transport and communication, among others, allows us to marvel about the endless opportunities to design future functional materials. Humbled by Nature but encouraged by many fascinating responsive materials introduced in recent years, one realizes that we are at the dawn of a new era of interactive materials and dynamic molecular systems. We also should not forget that “our chemical space to design” is not facing the restrictions due to the limited set of building blocks used in living systems; a privilege already indicated by Leonardo da Vinci: “Where Nature finishes producing his own species, man begins, with the help of Nature, to create an infinity of species.”

The creating power of molecular chemistry has brought us amazing successes in materials, evident from the wealth of polymers that sustain daily life, ranging from food packaging to medicine and aircraft. Exploring precise molecular interactions has provided supramolecular materials, self-assembled monolayers, sensors, metal–organic frameworks (MOFs), and covalent organic frameworks (COF), just to mention a few fascinating developments, where molecular recognition, assembly, and organization are essential to access novel material properties. More recently, the introduction of responsive functions, in particular molecular photoswitches, showed the potential

of noninvasive triggering and adapting of materials by external (light-) signals.

In the broader context, how to move from molecules to dynamic molecular systems, akin to materials in living systems, we face the multifaceted challenge of molecular structure, organization, responsive functions, multicomponent assemblies, interfaces, hierarchical levels, length scales, etc. This will force us to reconsider our

way to design materials well beyond the level of the individual (macro-)molecule. But in order to make materials that sense, act autonomously, adapt, and are self-regulatory, another level of complexity needs to be tackled.

Motion, the ability to move in response to changes in the environment, is arguably a highly desirable property and responsive systems that undergo mechanical motion might guide us in the design of interactive self-regulatory materials. In **Figure 1**, several key features are illustrated with the recent example of a supramolecular muscle.^[1] Although still primitive in its operation, the muscle can amplify a motion from the molecular scale to achieve a macroscopic mechanical function. The task which it can perform depends on the nature of aqueous environment in which the muscle material is assembled.

Although several polymers, macromolecular gels, crystalline, and liquid-crystalline materials have been introduced mimicking biological muscles and function as potential actuators, the fundamental question was: Can the exclusive use of noncovalent interactions sustain a macroscopic muscle type function? A major advantage of supramolecular assembled materials might be their propensity to adapt, re-configure, and change function. In brief, a light-driven rotary molecular motor was decorated with hydrophobic and hydrophilic groups to allow self-assembly in nanofibers in water. Using calcium ion binding and sheer flow, alignment of the fibers in muscle-type bundles was achieved and the resulting free-standing muscle can perform light-powered actuation and mechanical work. The actuation speed depends on the counter-ions (i.e., the nature of the aqueous salt solution) and one can envision a next generation of muscle-like materials that sense the environment, adapt the assembly based on change in supramolecular interaction, and as a consequence its macroscopic actuator function. Programming molecular structure and function, amplification of molecular motion, the hierarchical organization along different length scales, and the sensitivity of both assembly and function to the aqueous environment provide key insights for the design of next generations of interactive and mechanical materials.

Again taking Nature as our example, it is evident that providing a perspective on future interactive molecular materials is as multifaceted as the materials itself. Only a few of the pertinent challenges will be addressed, but let me emphasize that in

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going beyond the molecular frontiers, great insight and inspiration can be gained from recent advances in molecular biology and biophysics, surface science, robotics, artificial intelligence (AI), computational science, etc. As indicated above we will have to think in terms of functional molecular systems; how to bring the various crucial molecular components together to build a robust functional system that performs the functions that one anticipates. Interactive and self-regulatory materials require also that we learn how to design nonequilibrium systems. Intrinsic to Nature is the propensity to form and adapt metastable materials and systems that demonstrate far from equilibrium behavior, which they will provide tremendous opportunities to achieve self-regulation and autonomous operation. A few of the challenges are outlined:

Synthesis of Function: Instead of synthesis of structure, the focus should shift to synthesis of function; most likely multifunctional and multicomponent molecular systems. This implies not only multiple properties like assembly, sensing, responsive behavior but also maintaining robustness once being part of a more complex interactive material or device. Simultaneously, robustness and responsiveness to external interactions, signals, or triggers, e.g., interfaces, redox, light, pH, temperature, moisture, chemicals will be needed. Basically the integration of structure and interactive function is key, i.e., taking advantage of information-rich molecules for the programming of tasks.

Catalysis: In biological systems, autonomous and self-regulatory functions including assembly, transport, and motion are mainly governed by catalysis. Despite the tremendous success of catalysis in building materials, most prominent in

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catalytic polymerization, the use of catalysis to regulate materials assembly and properties has hardly been explored. Driving assembly in gel-type materials by coupling self-organization to catalytic modification of the components, using catalytic propulsion to induce nanoparticle swarming behavior or, in a reverse manner, light-induced aggregation of nanoparticles to regulate catalytic function are just a few examples of fascinating developments. The question is, how to couple the rich world of catalysis to the design of smart materials?

Assembly: This is a key aspect of any interactive material as the hierarchical assembly of functional molecules is crucial to

Figure 1. A supramolecular muscle based on a light-powered molecular motor, the actuation of which depends on the aqueous environment in which it is assembled. Reproduced with permission.^[1] Copyright 2018, American Chemical Society.

cooperative effects and ultimately the properties of the material. 2D assembly on nanostructures surfaces (self assembled monolayers (SAMs), etc.), 3D assembly (COFs, MOFs), artificial membranes, the detailed understanding how interface phenomena (organic–inorganic) effect the function (or can be used advantageously), multicomponent assembly, multi-valency, and ensemble dynamics continue to offer major challenges for the materials designer. Arguably these are also crucial to be able to interface to the macro-world as well as for device fabrication.

Amplification: Sensing and triggering a materials response usually requires a cooperative effect amplifying the signal. Responsive and interactive surfaces will equally depend on amplification and cooperative effects. Similarly, converting molecular motion into a macroscopic actuator function (see Figure 1) demands for amplification along many length scales. Here, dynamic molecular structure and functions (compare protein folding), precise balance of molecular interactions, and supramolecular assembly and amplification through cooperativity in liquid crystals and macromolecular materials offer tremendous opportunities. Also, the power of catalysis to strongly amplify molecular signals (including catalytic cascades) and integrate in situ functionalization/de-functionalization in order to alter materials properties deserves broader exploration.

Triggering, Addressing, and Sensing: Taking advantage of the major advances in, e.g., supramolecular chemistry, molecular switches, and responsive functional molecules in recent decades, there is ample opportunity to design novel interactive materials to sense, communicate with the outside world or adapt properties. Here, systems that are robust (fatigue resistant) over longer periods, operate properly when incorporated in (hard and soft) materials (or with red light), function on surfaces or in aqueous environment for biomedical application are highly desirable.

Information Processing: One can argue that future interactive materials that have to sense, respond to external stimuli, act autonomously, and are self-regulating have to process information at various levels. Considering molecular materials and molecular processes as information rich and information processing systems might provide a new dimension to the materials designer. Here, AI methodology and computational methods might guide the molecular info-explorer.

Feedback Loops and Repair Mechanisms: Intrinsic to biological systems that interact with and respond upon their environment, are feedback loops as part of complex molecular networks. Designing such network, feedback functions offer fascinating

challenges where advantage can be taken from the recent progress in auto-replicating and oscillating systems designed in the context of chemiogenesis/origin of life studies. Self-repairing materials, another area of significant recent progress, provides ample opportunity to integrate interactive and self-regulatory functions.

Emerging and Adaptive Properties: It should be emphasized again that ultimately we need to learn how to integrate several of the features mentioned above to arrive at adaptive behavior in materials and in particular emerging properties. Imagine the system can learn, memorize, and not only adapt but also develop new properties autonomously, taking advantage of a set of initial interactions.

Moving via responsive to interactive materials, the road to the future for the molecular designer is particularly bright. Addressing some of the fundamental issues associated with dynamic molecular systems as discussed above will offer ample opportunities to bring sensing, motion, adaptive, and autonomous properties to materials. Self-cleaning windows and solar panels, self-healing coatings and polymers, smart biomedical materials or delivery devices sensing the “environmental needs” in the human body and acting autonomously are not too far-fetched. When self-regulatory functions and actuator materials can be integrated, sophisticated soft robotics will likely come closer to reality. These are fascinating times for the explorer of novel materials and the prospects and excitement is reflected in the various contributions in this special issue.

Conflict of Interest

The author declares no conflict of interest.

Keywords

dynamic molecular systems, interactive materials, molecular chemistry, molecular design, smart materials

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