



## **A Matter of Size: Triennial Review of the National Nanotechnology Initiative**

Committee to Review the National Nanotechnology Initiative, National Research Council

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# 5

## Molecular Self-Assembly

The National Research Council Committee to Review the National Nanotechnology Initiative was asked to “determine the technical feasibility of molecular self-assembly for the manufacture of materials and devices at the molecular scale.”<sup>1</sup> The committee convened a workshop of experts in February 2005 to examine the technical information and discuss the issues. With input from the participants, the committee parsed this task into two parts: to consider the technical feasibility of self-assembly first, for the manufacture of materials, and second, for the manufacture of devices. In this system of nomenclature materials are undifferentiated structures. Devices are more complex, with parts or structures dedicated to particular functions. For instance, a lump of brass is a material, while a brass door hardware set has various functional parts (knob, latch, etc.) and is thus a device. A wafer of silicon is a material, while a silicon transistor has various functional parts (electrical contacts, conducting channel, etc.) and is thus a device. Devices typically are made from various materials.

After further discussion, the committee elected to address in addition a broader question—the feasibility of manufacturing systems capable of building, with molecular precision, complex systems that consist of multiple components. The committee’s discussions are summarized below, and the workshop agenda is given in Appendix C.

## WHAT IS SELF-ASSEMBLY?

In the broadest sense, self-assembly describes the natural tendency of physical systems to exchange energy with their surroundings and assume patterns or structures of reduced free energy. Random thermal motions bring constituent particles together in various configurations, so that stable configurations (those with significant binding energy) form, tend to persist, and eventually become predominant. Through this simple operation of physical law, pattern or structure arises in a bounded system with the input of relatively little information from outside. The information on how to assemble the structure is embodied in the structures of the individual components. A system slowly approaching equilibrium will assume a simple repetitive structure, while a dynamic system may generate structures of great complexity. For example, molecules in a cooling bucket of water will self-assemble as simple ice crystals, while the same molecules in a turbulent cloud with ever-changing temperature and humidity will self-assemble as complex snowflakes in enormous variety. Many fascinating structures in the natural world around us are self-assembled.

Chemists and biologists often use the term self-assembly in a more restricted sense to describe structure formation in a fluid containing various types of molecules, particularly organic molecules that form weak chemical bonds with a strength that depends sensitively on molecular shape and orientation. The strongest bond between such molecules often occurs when the molecules fit together in a “lock and key” fashion. Biological molecules such as proteins are particularly suited to forming complex higher-order structures. For example, the bacterial ribosome—a complex molecular machine consisting of about 55 different protein molecules and several ribosomal RNA molecules—will, under appropriate conditions, self-assemble in a test tube.<sup>2</sup>

## MOLECULAR SELF-ASSEMBLY AS A MANUFACTURING TECHNOLOGY

### For the Manufacture of Materials

Relatively complex materials such as semi-permeable membranes are manufactured every day by processes that exemplify molecular self-assembly. In a broad sense, fabrication and manufacturing processes for many common materials are exercises in self-assembly—quenching, solidification and crystallization, solution- and vapor-phase chemical reactions, and polymerization. The properties of the resulting materials—for example, the strength of metals or the electron mobility of semiconductors—depend exquisitely on the self-assembly of atoms and molecules to form the atomic and molecular structure of the finished material. The trick

for the technologist is to find just the right variation of process conditions—for example, the changes in temperature or the addition of impurities—that result in the desired material properties. Therefore, molecular self-assembly is certainly feasible for the manufacture of materials.

### **For the Manufacture of Devices**

Simple devices such as sensors for medical diagnostics are built every day with the aid of processes that exemplify molecular self-assembly. More complex structures can be generated by more sophisticated self-assembly processes. Processes requiring dynamic steering of process variables are often called “directed” self-assembly. “Templated” self-assembly describes processes requiring control of spatial boundaries such as container material and geometry. Thus, molecular self-assembly is also feasible for the manufacture of devices.

### **Challenges**

As spatial and temporal variations of boundary conditions and process variables become more complex, the emphasis shifts from self-assembly to the flow of information in the control system. However, the committee could not identify a “bright line” distinction between self-assembly and more complex integrated manufacturing processes. For instance, the above-mentioned example of the self-assembly of the bacterial ribosome from its constituent proteins is an elegant biological phenomenon, but it is only one part of the complex process that has evolved to build the ribosome. The various constituent proteins are themselves the product of RNA-driven amino acid catalysis called RNA translation in other functioning ribosomes, and RNA molecules are, in turn, the product of another catalytic process called DNA transcription. This complex assembly process, proceeding in every living cell, involves more than just self-assembly.

Manufacturing processes that can build very complex objects with high yield and repeatability will generally include processes more complex than simple self-assembly. This statement follows primarily from the fact that simple self-assembly does not include a mechanism for error correction.<sup>3</sup> The error rate for assembly of any two constituent parts can often be arranged to be very low, but the total probability of any error will tend to be the sum of the error rates for assembly of all the individual parts. Thus, the probability of a critical error occurring at some point in the assembly process will increase with the complexity of the system and the number of parts that must interoperate. At some level of complexity, the yield of a simple self-assembly process will become negligible.

Practical manufacturing systems solve this problem in a number of ways.

Kinetic constraints on the possible motions of constituents can greatly reduce the error rate in the assembly of constituent parts. Error-correction processes, such as sorting, refining, and purification, can provide a supply of good subcomponents for the next stage in a hierarchical self-assembly. These and other mechanisms are found in engineered manufacturing systems and in the structures and processes of biology.

Thus, the important task before the committee was to assess the feasibility of sophisticated manufacturing processes to produce more complex materials, devices, and, perhaps even entire complex systems from molecular components in a bottom-up fashion. Such processes are not usually considered to be examples of self-assembly.

### CURRENT STATE

The current states of two different technologies that have relevance to “bottom-up” or molecular manufacturing are described below. Lithography and nanobiotechnology are very different fields using vastly different methods, and, through the examples they provide, both enlighten the discussion of technical feasibility and future applications.

#### **Microelectronics Manufacturing: Lithography**

As scientists and engineers have gained confidence in their ability to develop bottom-up manufacturing processes that exploit the principles of biology, their ability to build small structures “top down” has also rapidly improved. Top-down processes are exemplified by machining, where the desired structure is produced by cutting, drilling, grinding, polishing, or otherwise shaping a block of material. For most of human history, machining was limited to structures that were readily visible to the naked eye. The ability to machine smaller structures developed slowly during the industrial revolution and accelerated with the beginnings of information technology as ever smaller components led to ever faster and more affordable computing, information storage, and communication. Today’s microelectronics factories use photolithographic processes to optically project desired structural patterns onto silicon wafers coated with a thin film of photosensitive polymer. After chemical development, the patterned polymer is used as a mask to transfer the desired structure to the silicon, typically by an etching process. In a very real sense, these lithographic systems are the “machine tools” of the information age.

At the time of this writing, the latest generation of microelectronics factories uses 90-nanometer technology, meaning that the lithographic systems and associated etching and deposition processes can routinely build circuits with wires

as narrow as 90 nanometers. Certain critical features, such as the transistor gate length, can be even smaller. Pattern dimensions must be controlled to tight tolerances. For example, if the transistor gate length is 40 nanometers, the allowable variance in this dimension (within three standard deviations) would be a few nanometers. This level of precision is necessary because the behavior of each transistor depends sensitively on the gate dimension, and the millions of transistors on a silicon chip cannot interoperate unless each device operates in nearly the same way as the others.

Currently semiconductor manufacturers are equipping the first 65-nanometer factories. In addition, 45- and 32-nanometer manufacturing processes and tools are already under development. At what point will this progression end? Several classes of commercially available lithographic systems—electron beam writers, various contact printing systems, and scanning probe systems—can define structures as small as 5 to 10 nanometers. These systems cannot yet meet the high-volume demands of microelectronics manufacturing, but they are already used in some “niche” manufacturing applications. At the research frontier, scanning probe systems have shown some ability to pattern matter one atom at a time. Many of the experiments involve the comparatively easy placement of metal atoms on atomically smooth metal surfaces. The resulting structures are weakly bonded and only stable at cryogenic temperatures. A few experiments have demonstrated some control of strong covalent bond interactions. For example, Hla and co-workers were able to induce all elementary steps of a simple organic chemical reaction by using various manipulations with a scanning tunneling microscope.<sup>4</sup> An explicitly lithographic process with atomic site specificity is the “hydrogen passivation resist” pioneered by Lyding’s group at the University of Illinois in the 1990s.<sup>5</sup> The process involves covering (passivating) a silicon wafer with a single layer of hydrogen atoms and removing selected hydrogen atoms with an electrical current from a scanning probe tip. The hydrogen-silicon bond is stable enough that the resulting pattern can be used to mask further chemical reactions on the surface, with atomic site specificity, at room temperature and above. In 2004, scientists associated with the Australian National Quantum Computer project used this method to introduce single atoms of phosphorus into a silicon crystal at selected atomic sites.<sup>6</sup> In order to perform this feat of atomic-resolution lithography, the silicon surface had to be atomically flat with a low density of defects to allow the formation of a nearly perfect hydrogen resist layer with one hydrogen bond for each surface silicon atom. Desorption of single hydrogen atoms required a scanning probe system equipped with an atomically sharp tungsten tip.

It should be noted that the atomically flat and clean silicon surface, the hydrogen layer, and the atomically sharp tungsten tip were each prepared by simple chemical processes—that is, processes that embody the bottom-up concept of

self-assembly. Only the selective removal of single hydrogen atoms embodies the top-down concept of machining. Essentially all of today's practical manufacturing processes mix top-down and bottom-up processes.

These results suggest that there is no fundamental physical barrier to practicing lithography at atomic dimensions. Of course, there is a vast gulf between these slow and very difficult pioneering experiments and today's high-volume, high-yield lithographic manufacturing processes. If the minimum lithographic dimension in large-scale manufacturing continued to be halved roughly every 5 years, atomic-scale lithography would be used in industrial processes in about 40 years. The actual course of technological developments will depend on many factors that cannot be predicted. The silicon transistor, the dominant device of information technology, will not function if shrunk toward atomic dimensions. Economic incentives to continue the current furious pace of research and development in top-down manufacturing might depend on currently unforeseen inventions of new devices that will require or benefit from extreme miniaturization.

Whether or not atomic-resolution lithography can be developed on an industrial scale, there is a rapidly growing body of research results in which lithographic patterns are used as templates to guide the self-assembly of smaller structures. In one recent example, a specially formulated polymer spontaneously forms nanometer-scale patterns that self-align with larger lithographic features, enabling construction of experimental "nanowire" transistors.<sup>7</sup> This is a technique of great interest for fabrication of a variety of next-generation transistor structures. A bit further out, some scientists envision the use of increasingly sophisticated self-assembly processes, including biomolecular processes such as those discussed below, to routinely bridge between the molecular scale and the larger structures that are readily fabricated with the aid of lithography. The result would be the ability to build structures approaching or exceeding biological levels of complexity—a capability that would have enormous implications for information technology, medicine, and energy production, and for endeavors not yet imagined.

### **Structural Chemistry: Nanobiotechnology**

The ability to engineer biological systems has long been a goal of biochemists that has recently been taken up by a new generation of physical scientists. Existing work on recombinant DNA molecules holds promise for the construction and evaluation of new gene arrangements.<sup>8</sup> The new applications of nanotechnological techniques to biological systems hold substantial promise.<sup>9</sup> Today, rudimentary devices have been produced, including sensors and actuators, input and output devices, and genetic circuits to control cells.<sup>10</sup> Future developments in this new structural chemistry will target both the stabilization and the simulation of bio-

molecules for use in a wide range of activities, including various manufacturing processes.

A number of strategies have been demonstrated by which the material properties of biomolecular systems may be moved outside the relatively constrained environment of the living cell. Perhaps the simplest example is the direct substitution of nonbiological organic or inorganic chemistries for bioorganic chemistries. Examples include bacteria grown in extreme environments and enzymes that catalyze reactions at high pressures and temperatures found outside the normal range of conditions for life processes. The protein complex responsible for production of oxygen in photosynthesis does not, in fact, operate in an aqueous environment but in an electrochemical interphase region of complex physical chemistry including an extremely high electric field, greater than 20 megavolts per meter.<sup>11</sup> Once the structure and the function of a specific biomolecule are elucidated, it should be possible in many cases to simply substitute alternative forms of chemistry for selected components. Protein engineering is already addressing this strategy in a rudimentary manner in synthetic amino acid analogs. Preliminary experimental validation that such nanobiotechnology may be useful for manufacturing is found in the ability to design synthetic bis-amino acid oligomers to have specific rigid shapes, which should be useful in constructing complex atomically precise three-dimensional objects.<sup>12</sup>

Fully synthetic analogs can be created today by molecular imprinting, a technique to create template-shaped cavities in polymer matrices with memory of the template molecules. This technique is based on the system used by natural enzymes for substrate recognition, which is called the "lock and key" model. In recent decades, the molecular imprinting technique has been developed for use in receptors, chromatographic separations, catalysis, and fine chemical sensing.<sup>13</sup> Structural biologists can now engineer enzymes to interact with chemicals that do not occur in nature. For example, proteins have been modified to bind poison gas and explosives so that they can be used as single-molecule sensors, and motor proteins have been modified from their natural function and show promise as mechanical components in hybrid nano-engineered systems. In one such system, the cytoplasmic fragment of the F1-ATPase has been integrated into self-assembled nanomechanical systems as a mechanical actuator.<sup>14</sup> Repeated cycles of zinc addition and removal by chelation result in inhibition and restoration, respectively, of motor rotation in the engineered protein. These results demonstrate the ability to engineer single-molecule chemical regulation into a biomolecular motor. Using these methods, synthetic biologists eventually aim to build cells from the ground up rather than tinkering with a handful of genes or tweaking a metabolic pathway or two, as do today's genetic engineers.

A third strategy is the stabilization of biomolecules in biomolecular-materials

composites. For example, nanoporous materials can be specifically tailored to accommodate individual protein catalysts. Such materials could simultaneously protect the bulk protein molecule from destructive physical forces while retaining a channel to the catalytic site. The ability to synthesize biological macromolecules with novel materials components creates both the opportunity to build enzymes that function outside the normal cellular environment and the opportunity to modify the cellular environment by filling it with hybrid biomolecular-materials composites. The synthesis of DNA molecules containing metallo-base pairs creates a molecular structure that can transfer both biological information and an electrical signal. Methodology has recently been developed to genetically encode novel amino acids. This has already been used to create heavy-atom-containing amino acids to facilitate x-ray crystallographic studies; amino acids with novel steric/packing and electronic properties; photocrosslinking amino acids that can be used to probe protein-protein interactions *in vitro* or *in vivo*; and keto- and acetylene-containing amino acids that can be used to selectively introduce a large number of biophysical probes, tags, and novel chemical functions.<sup>15</sup>

As the examples above make clear, the lines between nanotechnology and biotechnology are becoming blurred. Indeed, at the molecular level of structure, the border between living and nonliving materials is also rapidly fading. This reality begins to redefine commonly used definitions and confounds accepted paradigms.

### **Technical Feasibility of Site-Specific Chemistry for Large-Scale Manufacturing**

Prudent extrapolation of the current research results presented above suggests an amazing future for nanotechnology. Indeed, many scientists foresee a long-term future in which a variety of strategies, tools, and processes allow nearly any stable chemical structure to be built atom by atom or molecule by molecule from the bottom up. However, there is still a gulf between this vision and popular images of nanotechnology in which the bottom-up approach is routinely used to manufacture complex, large-scale industrial objects such as computers or buildings at very low cost. The feasibility of such developments would depend on the attainable *efficiency* of the manufacturing processes. The proposed manufacturing systems<sup>16-19</sup> can be viewed as highly miniaturized, highly articulated versions of today's scanning probe systems, or perhaps as engineered ribosome-like systems designed to assemble a wide range of molecular building blocks in two or three dimensions rather than the linear assembly of amino acids by the ribosome. In this approach, reactions are described with both reagent and product as part of extended "handle" structures, which can be moved mechanically.<sup>20</sup> To be practical for the manufacture

of large-scale objects, such mechanisms would have to operate at a very low error rate, a very high speed, and near-perfect thermodynamic efficiency. Technical arguments for the eventual attainability of these attributes have been provided.<sup>21</sup> Design strategies have been outlined that, it is maintained, would allow such systems to greatly exceed the error rates, speed, and average thermodynamic efficiency of naturally evolved biological systems. Proponents of these design and manufacturing strategies foresee the exploitation of exquisitely controlled site-specific chemistry on a vast industrial scale. While scanning probe systems have demonstrated the feasibility of some site-specific reactions, scale-up to manufacturing systems is still a daunting task, and the majority of nanoscale scientists and engineers believe it is too early to try to predict the ultimate capabilities of such systems.

The committee found the evaluation of the feasibility of these ideas to be difficult because of the lack of experimental demonstrations of many of the key underlying concepts. The technical arguments make use of accepted scientific knowledge but constitute a “theoretical analysis demonstrating the possibility of a class of as-yet *unrealizable* devices.”<sup>22</sup> Thus, this work is currently outside the mainstream of both conventional science (designed to seek new knowledge) and conventional engineering (usually concerned with the design of things that can be built more or less immediately). Rather, it may be in the tradition of visionary engineering analysis exemplified by Konstantin Tsiolkovski’s 1903 publication, “The Exploration of Cosmic Space by Means of Reaction Devices,”<sup>23</sup> and today’s studies of “space elevators” based on hypothetical carbon nanotube composite materials.<sup>24</sup>

Construction of extended structures with three-dimensional covalent bonding may be easy to conceive and might be readily accomplished, but only by using tools that do not yet exist.<sup>25</sup> In other words, the tool structures and other components cannot yet be built, but they can be computationally modeled. Modeling the thermodynamic stability of a structure (showing that it can, in principle, exist) does not tell one how to build it, and these arguments do not yet constitute a research strategy or a research plan.

To bring this field forward, meaningful connections are needed between the relevant scientific communities. Examples include:

- Delineating desirable research directions not already being pursued by the biochemistry community;
- Defining and focusing on some basic experimental steps that are critical to advancing long-term goals; and
- Outlining some “proof-of-principle” studies that, if successful, would provide knowledge or engineering demonstrations of key principles or components with immediate value.

## CONCLUSIONS

Materials and devices of moderate complexity can be designed and manufactured by molecular self-assembly. Although self-assembly operates on simple and well-understood scientific principles, understanding of the details is far from complete. The ultimate potential of self-assembly processes in nature and in engineered manufacturing systems remains to be explored.

Proceeding beyond simple self-assembly, there is experimental evidence that biological systems can be modified to operate in conditions far outside those of the living cell, and therefore, many biotechnologists believe that these systems will form the basis for many future manufacturing processes.<sup>26</sup> Manufacturing trends and research directions in information technology and related fields also suggest the eventual development of manufacturing processes with some capability to pattern structures with atomic precision.<sup>27</sup>

Although theoretical calculations can be made today, the eventually attainable range of chemical reaction cycles, error rates, speed of operation, and thermodynamic efficiencies of such bottom-up manufacturing systems cannot be reliably predicted at this time. Thus, the eventually attainable perfection and complexity of manufactured products, while they can be calculated in theory, cannot be predicted with confidence. Finally, the optimum research paths that might lead to systems which greatly exceed the thermodynamic efficiencies and other capabilities of biological systems cannot be reliably predicted at this time. Research funding that is based on the ability of investigators to produce experimental demonstrations that link to abstract models and guide long-term vision is most appropriate to achieve this goal.

## NOTES

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