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VIEWPOINT

Self-Assembly at All Scales

George M. Whitesides* and Bartosz Grzybowski

Self-assembly is the autonomous organization of components into patterns or structures without human intervention. Self-assembling processes are common throughout nature and technology. They involve components from the molecular (crystals) to the planetary (weather systems) scale and many different kinds of interactions. The concept of self-assembly is used increasingly in many disciplines, with a different flavor and emphasis in each.

There are several reasons for interest in self-assembly (*1, 2*). First, humans are attracted by the appearance of order from disorder. Second, living cells self-assemble, and understanding life will therefore require understanding self-assembly. The cell also offers countless examples of functional self-assembly that stimulate the design of non-living systems. Third, self-assembly is one of the few practical strategies for making ensembles of nanostructures. It will therefore be an essential part of nanotechnology. Fourth, manufacturing and robotics will benefit from applications of self-assembly. Fifth, self-assembly is common to many dynamic, multicomponent systems, from smart materials and self-healing structures to netted sensors and computer networks. Finally, the focus on spontaneous development of patterns bridges the study of distinct components and the study of systems with many interacting components. It thereby connects reductionism to complexity and emergence (*3*).

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Is Anything Not Self-Assembly?

"Self-assembly" is not a formalized subject, and definitions of the term "self-assembly" seem to be limitlessly elastic. As a result, the term has been overused to the point of cliché. Processes ranging from the non-covalent association of organic molecules in solution to the growth of semiconductor quantum dots on solid substrates have been called self-assembly. Here, we limit the term to processes that involve pre-existing components (separate or distinct parts of a disordered structure), are reversible, and can be controlled by proper design of the components. "Self-assembly" is thus not synonymous with "formation."

Types of Self-Assembly

There are two main kinds of self-assembly: static and dynamic. Static self-assembly (S) (Table 1; Fig. 1) involves systems that are at global or local equilibrium and do not dissipate energy. For example, molecular crystals (*4, 5*) are formed by static self-assembly; so are most folded, globular proteins. In static self-assembly, formation of the ordered structure may require energy (for example in the form of stirring), but once it is formed, it is stable. Most research in self-assembly has focused on this static type

In dynamic self-assembly (D) (Table 1; Fig. 2), the interactions responsible for the formation of structures or patterns between components only occur if the system is dissipating energy. The patterns formed by competition between reaction and diffusion in oscillating chemical reactions (*6, 7*) are simple examples; biological cells are much more complex ones. The study of dynamic self-assembly is in its infancy.

We define two further variants of self-assembly. In templated self-assembly (T), interactions between the components and regular features in their environment determine the structures that form. Crystallization on surfaces that determine the morphology of the crystal is one example (*8, 9*); crystallization of colloids in three-dimensional optical fields is another (*10*). The characteristic of biological self-assembly (B) is the variety and complexity of the functions that it produces.

Common Features of Self-Assembly

Self-assembly reflects information coded (as shape, surface properties, charge, polarizabil-

ity, magnetic dipole, mass, etc.) in individual components; these characteristics determine the interactions among them. The design of components that organize themselves into desired patterns and functions is the key to applications of self-assembly.

The components must be able to move with respect to one another. Their steady-state positions balance attractions and repulsions. Molecular self-assembly involves noncovalent or weak covalent interactions (van der Waals, electrostatic, and hydrophobic interactions, hydrogen and coordination bonds). In the self-assembly of larger components—meso- or macroscopic objects—interactions can often be selected and tailored, and can include interactions such as gravitational attraction, external electromagnetic fields, and magnetic, capillary, and entropic interactions, which are not important in the case of molecules.

Because self-assembly requires that the components be mobile, it usually takes place in fluid phases or on smooth surfaces. The environment can modify the interactions between the components; the use of boundaries and other templates in self-assembly is particularly important, because templates can reduce defects and control structures.

Equilibration is usually required to reach ordered structures. If components stick together irreversibly when they collide, they form a glass rather than a crystal or other regular structure. Self-assembly requires that the components either equilibrate between aggregated and non-aggregated states, or adjust their positions relative to one another once in an aggregate.

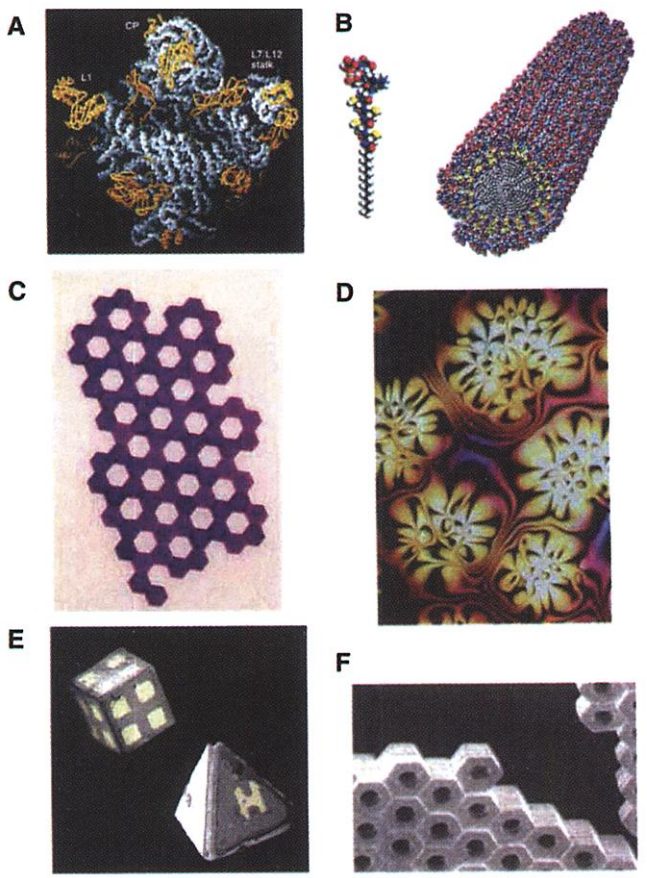

Fig. 1. Examples of static self-assembly. **(A)** Crystal structure of a ribosome. **(B)** Self-assembled peptide-amphiphile nanofibers. **(C)** An array of millimeter-sized polymeric plates assembled at a water/perfluorodecalin interface by capillary interactions. **(D)** Thin film of a nematic liquid crystal on an isotropic substrate. **(E)** Micrometer-sized metallic polyhedra folded from planar substrates. **(F)** A three-dimensional aggregate of micrometer plates assembled by capillary forces. [Image credits: (A) from (*24*); (B) from (*25*); (C) from (*26*); (D) from (*27*); (E) from (*28*); (F) from (*29*)]

Dynamic Self-Assembly

Although much of current understanding of self-assembly comes from the examination of static systems, the greatest challenges, and opportunities, lie in studying dynamic systems. Perhaps the most important justification for studying self-assembly is its central role in life. The components of a cell replicate and assemble into another cell during mitosis; bacteria swarm (*11*); fish school (*12, 13*). Most efforts in biology have focused on static self-assembly. Life is, however, dynamic: stop the flux of energy through the cell and it dies.

We understand that the living cell is a sack that contains a number of reacting chemicals, is studded with environmental sensors, and allows heat and certain chemicals to pass across its walls. We also understand that the cell is a structure that is enclosed, self-replicating, energy dissipating, and adaptive. Yet we have little idea how to connect these two sets of characteristics. How does "life" emerge from a system of chemical reactions? Self-assembly may be one thread that connects the relative simplicity of chemical reactions to the complexity of the dividing cell. At the molecular level, static self-assembly describes formation of the lipid bilayer, pairing of bases, and folding of some proteins. The behavior of critical structures in the

Table 1. Examples of self-assembly (S, static, D, dynamic, T, templated, B, biological).

System	Type	Applications/importance	References
Atomic, ionic, and molecular crystals	S	Materials, optoelectronics	(*1, 4, 5*)
Phase-separated and ionic layered polymers	S		(*19*)
Self-assembled monolayers (SAMs)	S, T	Microfabrication, sensors, nanoelectronics	(*8*)
Lipid bilayers and black lipid films	S	Biomembranes, emulsions	(*20*)
Liquid crystals	S	Displays	(*21*)
Colloidal crystals	S	Band gap materials, molecular sieves	(*9, 18*)
Bubble rafts	S	Models of crack propagation	(*22*)
Macro- and mesoscopic structures (MESA)	S or D, T	Electronic circuits	(*14–16*)
Fluidic self-assembly	S, T	Microfabrication	(*23*)
"Light matter"	D, T		(*10*)
Oscillating and reaction-diffusion reactions	D	Biological oscillations	(*6, 7*)
Bacterial colonies	D, B		(*11*)
Swarms (ants) and schools (fish)	D, B	New models for computation/optimization	(*12, 13*)
Weather patterns	D		(*1*)
Solar systems	D		
Galaxies	D		

cell—including actin filaments, histones and chromatin, and protein aggregates in signaling pathways—involves dynamic self-assembly. The complex processes that occur in mitosis involve every type of self-assembly. A hierarchy of self-assembling processes is thus fundamental to the operation of cell.

Dynamic self-assembly is also common in nonliving systems, although these processes are less studied, and less understood, than those in living systems. Oscillating reactions in solution and on the surface of catalysts, Rayleigh-Bernard convection cells, patterns that form in fluidized beds of particles, and storm cells in the atmosphere are all examples; Table 1 lists others.

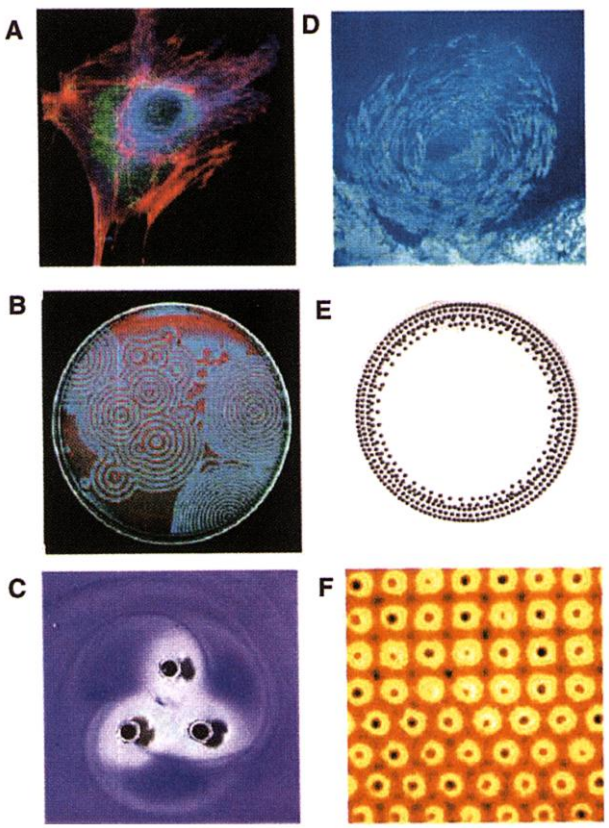

Fig. 2. Examples of dynamic self-assembly. (**A**) An optical micrograph of a cell with fluorescently labeled cytoskeleton and nucleus; microtubules (~24 nm in diameter) are colored red. (**B**) Reaction-diffusion waves in a Belousov-Zabatinski reaction in a 3.5-inch Petri dish. (**C**) A simple aggregate of three millimeter-sized, rotating, magnetized disks interacting with one another via vortex-vortex interactions. (**D**) A school of fish. (**E**) Concentric rings formed by charged metallic beads 1 mm in diameter rolling in circular paths on a dielectric support. (**F**) Convection cells formed above a micropatterned metallic support. The distance between the centers of the cells is ~2 mm. [Image credits: (A) from (*30*); (B) from (*26*); (C) from (*31*)]

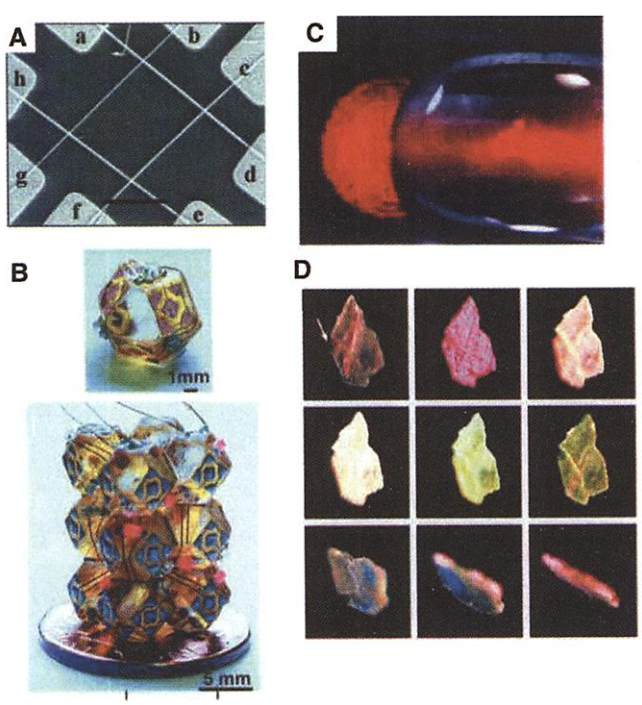

Fig. 3. Applications of self-assembly. (**A**) A 2 by 2 cross array made by sequential assembly of *n*-type InP nanowires with orthogonal flows. (**B**) Diffraction grating formed on the surface of a poly(dimethylsiloxane) sphere ~1 mm in diameter. The sphere was compressed between two glass slides, and its free surface was exposed to oxygen plasma. Upon release of compression, the oxidized surface of the polymer buckled with a uniform wavelength of ~20 μm. (**C**) Three-dimensional electronic circuits self-assembled from millimeter-sized polyhedra with electronic components (LEDs) embossed on their faces. (**D**) An artificial, ferromagnetic opal prepared by templated self-assembly of polymeric microbeads. The optical properties of the aggregate can be adjusted by modifying external magnetic field. [Image credits: (A) from (*32*); (B) from (*16*); (C) from (*26*); (D) from (*33*)]

Self-Assembly in Designed Systems

A difficulty in studying self-assembly in living cells (and in many nonliving systems) is that it is impractical to change many of the parameters that determine the behavior of the systemthe components and the interactions among them—and thus difficult to test hypotheses relating structures and properties of these components and the aggregates that they form. We wished to have available a set of self-assembling components in which these parameters *could* be changed easily, in order to understand (and to be able to manipulate) the processes by which components self-assembled into aggregates. With this objective, we have studied the self-assembly of polyhedral plates or disks—a few millimeters wide and a millimeter high—floating at the interface between water and perfluorodecalin (*14*). These sizes are attractive because the components can be fabricated and observed easily, and because the interactions between these components are under precise experimental control. Static versions of this system depend on capillary interactions (*15*) between menisci at the edges of the plates, and typically produce ordered aggregates with irregular edges. The processes observed at millimeter dimensions scale (with some modification) to submicron dimensions. Templating produces aggregates with defined shape. When drops of liquid are patterned on the faces of components suspended in an immiscible, isodense fluid, three-dimensional structures can be generated. If this liquid is solder, cooling forms interconnections that are mechanically strong and electrically conducting. This type of system points toward functional, self-assembling microelectronic systems (*16*).

An extension of these static systems illustrates *dynamic* self-assembly. Small ferromagnetic disks, floating at the liquid interface, rotate under the influence of a rotating external bar magnet. The average field of this magnet generates a central field that pulls the disks together. As they spin, they generate vortices in the fluid; the vortex-vortex interactions are repulsive. The spinning disks assemble into a variety of stable patterns (*17*).

Learning from One Another

Different fields of science take different roads to understanding; each brings something to self-assembly. Chemists and engineers tend to solve problems by designing

and synthesizing (or fabricating, or building) new systems; physicists observe existing systems; biologists make modifications by mixing preexisting parts. Each style will be important in some aspect of self-assembly.

For self-assembly to generate structures more complex than simple crystals, different components in a mixture must come together in an ordered way. The selective recognition of different molecular components in a mixture is the basis for much of molecular biology and medicinal chemistry. The parameters that control molecular recognition (complementary shapes, complementary forces, and appropriate levels of plasticity) will also be broadly useful in the self-assembly of larger systems.

Dynamic systems are an important part of the future of self-assembly. Biology is replete with examples of dynamic (and static) systems that may stimulate designs for inanimate systems. Because the components in dynamic self-assembling systems interact with one another in complex (and often nonlinear) ways, their behavior will often be complex. Physics now has the most sophisticated understanding of complexity.

Present and Future Applications

Self-assembly is already a widely (if unwittingly) applied strategy in synthesis and fabrication (Table 1; Fig. 3). Can one predict areas where self-assembly will be used in the future? Perhaps. These are possibilities:

(1) Crystallization at All Scales. The formation of regular, crystalline lattices is a fundamental process in self-assembly, and is a method to convert ~100-nm particles into photonic materials (*18*); using micrometer-scale components may lead to new routes to microelectronic devices (*16*).

(2) Robotics and Manufacturing. Robots are indispensable to current systems for manufacturing. As components become smaller, following the trend in miniaturization through microfabrication to nanofabrication, conventional robotic methods will fail because of the difficulty in building robots that can economically manipulate components only micrometers in size. Self-assembly offers a new approach to the assembly of parts with nano- and micrometer dimensions.

(3) Nanoscience and Technology. There are two approaches to the fabrication of nanosystems: bottom-up and top-down. Chemical synthesis is developing a range of methods for making nanostructures—colloids, nanotubes, and wires—to use in bottom-up approaches. Self-assembly offers a route for assembling these components into larger, functional ensembles.

(4) Microelectronics. The fabrication of microelectronic devices is based almost entirely on photolithography, an intrinsically two-dimensional technology. Another computer of great interest—the brain—is three-dimensional. There are no clear strategic paths from two-dimensional to three-dimensional technology (and, of course, no absolute certainty that three-dimensional microelectronic devices will be useful, although the brain is certainly a three-dimensional system, and three dimensionality offers, in principle, the advantages of short interconnects and efficient use of volume). Self-assembly offers a possible route to three-dimensional microsystems.

(5) Netted Systems. At the outer limits of self-assembly, at least as it is currently defined in the physical and biological sciences, are netted systems: computers, sensors, and controllers that interact with one another only through the flow of bits and configure (or self-assemble) themselves based on that flow into functional systems. These netted information systems will be entirely different in their realization from self-assembled aggregates of material components, but will share underlying concepts of design and architecture.

Coda

Self-assembly, as a field, originated in organic chemistry. It has become a rapidly growing part of this field for two reasons. First, it is a concept that is crucial to understand many structures important in biology. Second, it is one solution to the problem of synthesizing structures larger than molecules. The stability of covalent bonds enables the synthesis of almost arbitrary configurations of up to 1000 atoms. Larger molecules, molecular aggregates, and forms of organized matter more extensive than molecules cannot be synthesized bond-by-bond. Self-assembly is one strategy for organizing matter on these larger scales.

Although self-assembly originated in the study of *molecules*, it is a strategy that is, in principle, applicable at all scales. We believe that some of the self-assembling systems that are most amenable to fundamental study, and that are also most readily applied, may involve components that are larger than molecules, interacting by forces (for example, capillarity) that have not commonly been used in synthesis or fabrication. Self-assembly thus provides one solution to the fabrication of ordered aggregates from components with sizes from nanometers to micrometers; these components fall awkwardly between the sizes that can be manipulated by chemistry and those that can be manipulated by conventional manufacturing. This range of sizes will be important for the development of nanotechnology (and the expansion of microtechnology into areas other than microelectronics). It will also be an area in which understanding biological structures and processes, and using this understanding to design nonbiological mimics of them, will offer many opportunities to build systems with new types of function. In the emerging area of dynamic self-assembly, it is unclear whether the study of molecules, or of other types of components, will lead more efficiently to understanding. We understand very little about how dissipation of energy leads to the emergence of ordered structures from disordered components in these systems. But we know that they are vitally important in the cell. That knowledge, by itself, makes it worthwhile to study them.

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