7

Trend of Self-Organization in Materials Research

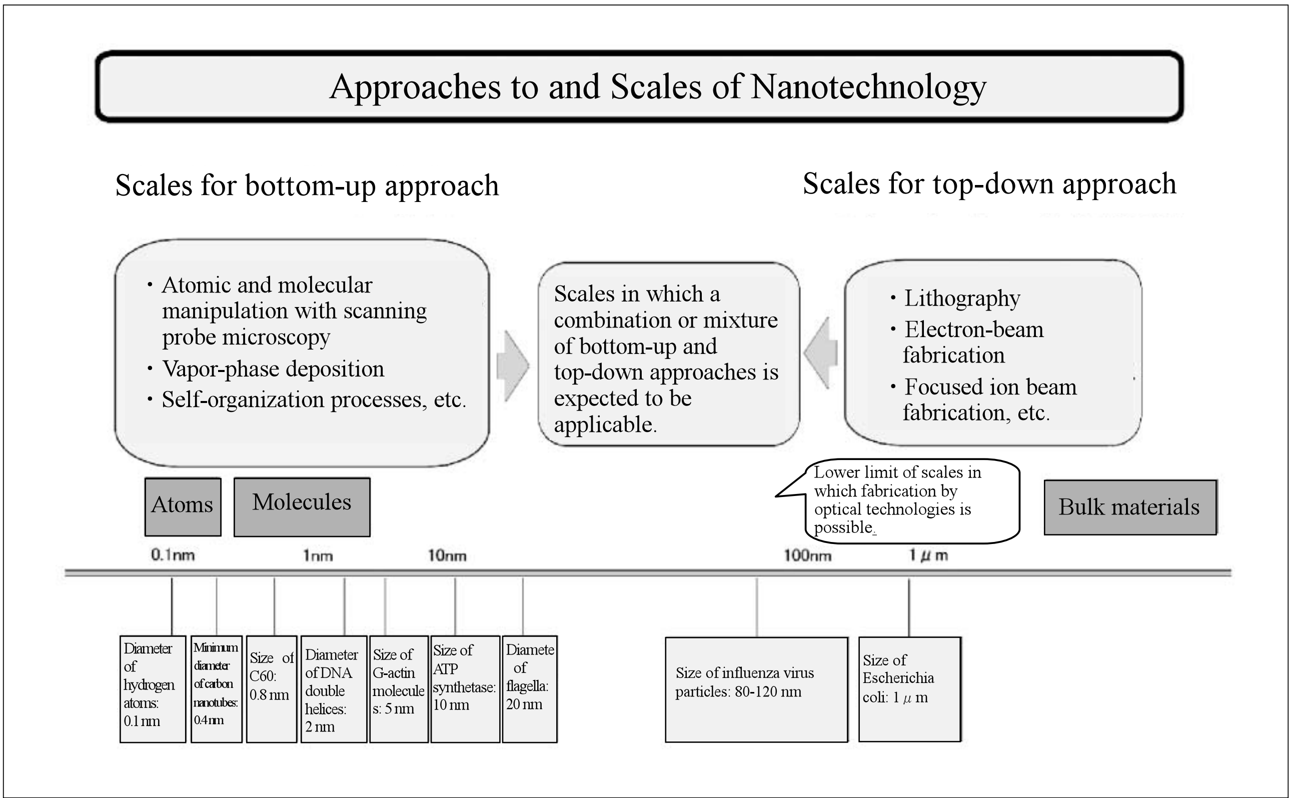
JUN-ICHIRO TAKANO AND NOBUYUKI KOGUCHI (*Affiliated Fellow*)
Materials and Processing Unit

7.1 Introduction

In general, miniaturization of devices provides the advantages of "speeding up," "low power consumption" and "higher integration." Therefore, research on materials on the scale of nanometers (one nanometer is one billionth of a meter: nm) has attracted people's attention. The approach for dealing with such nanometer scale materials can be roughly divided into two categories: One is the "top-down" approach, which carves out a surface as in the case of miniaturization of semiconductors; the other is the "bottom-up" approach, which builds up atoms or molecules into nanometer scale structures (nanostructure). (See Figure 1)

Until recently, nanotechnology has been targeted at semiconductor devices and mainly developed by the "top-down" approach. But researchers in various fields point out that preparation for nanometer scale structures through the top-down approach will face greater difficulties in the near future, i.e., in a few to a few tens of years (these are difficulties associated with technological limitations, physical limitations and economical limitations, but we had to omit the detailed explanation due to space limitations). The bottom-

Figure 1: Approaches to and Scales of Nanotechnology



Source: Authors' compilation by making reference to a report from the Japan Patent Office [http://www.jpo.go.jp/indexj.htm] on investigation into trends in technologies "Related to Nanotechnology and Materials" for which patent applications have been filed up approach has gained the spotlight as a complement or alternative approach to the top-down approach on that account.

This report covers the self-organizing method, which has drawn keen attention among bottom-up approaches, and summarizes goals, present state of self-organized materials researches, and issues to achieve goals.

7.2 Definition and goals of self-organization in materials research

7.2.1 The self-organization method in bottom-up approaches

Among the various bottom-up approaches, which placement should the self-organization method take? In this section, we will explain from which viewpoint the self-organizing method has attracted people's attention by giving an outline of the "atomic or molecular manipulation technique by using the scanning probe microscopy (SPM)".

The atomic or molecular manipulation technique by using SPM refers to the "method of creating artificial structures by pinching atoms or molecules one-by-one with a minute SPM probe and aligning those atoms or molecules at intended positions." Many reports have already been made about the realization, with the use of the manipulation technique, of various characters wrote by aligning atoms (atomic-scale writing) and nanostructure created with atoms. In theory, by applying the manipulation technique, nanostructure with 3 dimensions of a few nanometers (quantum dots) can be produced. And materials with innovative functions realized by quantum dots can be expected to become available.

In practice, however, we may face great difficulties when we try to prepare quantum dots with SPM. Even if the present atomic or molecular manipulation technology (which necessitates about an hour to put together one character by atomic-scale writing) advances, and even if we become able to pick up one atom and place it at an intended position in 1/1000 of a second, we will still need to move about 8,000 atoms in order to assemble a quantum dot measuring about 5 nanometers in diameter. In addition to this, work for about 90 days without a break to align quantum dots on a plane measuring 1/100 mm square. Therefore, preparation of quantum dots with SPM simply cannot be said to be realistic.

Moreover, although atoms can be picked up and moved on a one-by-one basis in the case of substances containing only one kind of element such as silicon, it can be easily expected that greater technological difficulties will arise in the case of manipulation with SPM of compound semiconductor materials composed of two or more kinds of elements such as gallium arsenide (GaAs), which demonstrates superior properties to silicon when used for high-speed transistors. (However, this estimation was developed based on very simplified trial calculations. In reality, high-value-added nanostructure may possibly be prepared by combinations of two or more techniques in the form of, for example, SPM for very small structures and different techniques for larger structures.)

In this section we took manipulation with SPM as an example of the bottom-up approach, it is expected that many problems including considerable time and energy consumption requirements will turn up in other bottom-up approaches, too. In such circumstances, among bottom-up approaches, the "self-organization" method has received attention as the approach that possibly enables the reduction of time and energy needed for the preparation of nanostructure.

7.2.2 What is self-organization?

As mentioned in "Trends in Nanobiology" in the fifth issue (Jan. 2003), no scientific consensus about the concept of "self-organization" itself has been reached by researchers, and there has been no clear definition about it. In this report, we will discuss self-organization by defining it as the "process utilized in the preparation of materials or devices, in which components of materials or devices assemble by themselves to form specific structures (self-assembly), or the process in which components spontaneously form specific patterns (dissipative structures) through energy and matter diffusion."

We give one example of self-assembly here. The formation of a globular structure called micelle, which occurs when soap molecules, having both the hydrophilic group with higher affinity for water and the hydrophobic group with lower affinity for water, are in water with the hydrophilic unit being on the surface and the hydrophobic unit being sequestered inside. On the other hand, we quote "wind patterns on the sand" as one example of dissipative structures. Patterns of the sand on the surface of land exposed to external forces such as wind without being subjected to artificial processes.

So far in this report, we intentionally discussed self-organization in the filed of nanotechnology for the purpose of explaining the reasons for which research on self-organized materials has become a focus of people's attention. However, the concept of self-organization covers processes that vary in their length, from those on very small scales such as the formation of nanostructure to those on very large scales such as the formation of swirling cloud streams associated with overlaying temperature inversion (Karman vortex). Therefore, in the following sections, we will discuss the goals, present state of self-organized materials researches, and issues to achieve goals (bridge the gap between the goals and the present state) without special regard to the length scales of materials.

7.2.3 Goals of self-organized materials research

We authors have reached the conclusion that the goals of self-organized material research at the present stage are the following three (we took into account discussions with some of the researchers named in the "Acknowledgments"):

Goal A:
Precision synthesis of molecular clusters

Goal A sets out to establish technologies that enable the precision synthesis of clusters of atoms or molecules (which can be sometimes components of systems), especially the clusters of molecules and nanostructure that are difficult to prepare when using conventional synthesis methods, by utilizing the functions of atoms or molecules themselves (in a resource-saving and energy-saving manner) as well as to induce innovative functions in materials synthesized in such a manner.

Goal B:
Establishment of patterning and self-aligning technologies

Goal B sets out to establish, irrespective of the physical states (gaseous, liquid and solid state) or scales (nanometer [nm], micrometer [μm], millimeter [mm] , centimeter[cm], and meter [m]) of the components of targeted systems, preparation processes through which useful patterns in the targeted systems can be formed in large quantity at one time by utilizing the functions of components of the systems themselves (in a resource-saving and energy-saving manner). And the preparation of desired structures by self-alignment of components at intended positions with high precision.

Goal C:
Preparation of materials and devices through the self-organization method

Goal C sets out to realize smart materials (also called "intelligent materials," which exercise their functions in response to changes in ambient conditions such as temperature and exposure to light) and molecular devices by creating hierarchical structures in two or more scales by means of, for example, combining the technologies mentioned as Goal A and Goal B and inducing functions characteristic to each hierarchy.

In the rest of this report, we will give, in Chapter 7.3, some examples of research aiming to achieve Goals A to C in order to enhance the readers' understanding of such research, and will discuss, in Chapter 7.4, issues to bridge the gap between Goals A to C (mentioned in this section) and the present state (mentioned in the next section).

7.3 Present state of self-organized material research

In this Chapter, in order to enhance the readers' understanding of the present state of self-organized material research, we will present interesting comments by researchers in Japan who are actively studying such materials, as well as examples of organizational research for self-organized materials, and will compare the different methods for creating the same nanostructure in terms of equipment to be used, the number of steps to be followed, etc. Then, we will cite some examples of research aiming to achieve Goals A to C.

7.3.1 Present state of research in Japan and other countries

Dr. Tomohiko Yamaguchi, chief researcher at the Nanotechnology Research Institute of the National Institute of Advanced Industrial Science and Technology (AIST), an Independent Administrative Institution (IAI) under the Ministry of Economy, Trade and Industry (METI), made the following comment about the "levels of self-organized material research in Japan" and the "alienation between theories and experiments in front-line research."

(1) Levels of self-organized materials research in Japan

While there has been a growing international trend toward application of the processes of dissipative structure formation to material sciences, it was the polymer (macromolecule) research groups in Japan that set the trend in the mid 1990s.

In addition, with regard to research on metallic nanoparticles, the staff on the Hayashi Ultrafine Particle Project in Japan (a project pursued during the period from 1981 to 1986 within the Exploratory Research for Advanced Technology [ERATO] program administered by the Japan Science and Technology Corporation (JST)) established the method for gas-phase synthesis ahead of all other countries. It has been reported that the Clinton administration in the United States closely examined the fruits of the Hayashi Ultrafine Particle Project, etc., when they investigated the state of nanotechnology research in Japan before announcing the U.S. National Nanotechnology Initiative (NNI) in January 2000. We feel that Japan has been ahead of the rest of the world when it comes to preparation technologies by the mechanism of dissipative structure formation.

(2) Alienation between theories and experiments in front-line research

Examples of collaborative work in Europe done by both theoreticians and experimentalists include close collaborative relationship between the theoretician group lead by Mikhailov, a mathematician, at the Fritz-Haber-Institut der Max-Planck-Gesellschaft, which was established in honor of Haber, who accomplished great achievements in research in chemical engineering including the fixation of atmospheric nitrogen. And the group of many experimentalists lead by Ertl, director of the institute, who is a world authority on solid surface reaction and has received the Kyoto Prize, which leads to theoretical demonstration of the formation of periodic nanostructure in the reaction-diffusion-advection system.

In the United States, a group lead by Karim, who is famous for his research on polymer materials, at the National Institute of Standards and Technology (NIST) has succeeded, in collaboration with theoreticians, in inducing spatially-periodic structures on the surfaces of polymer materials by applying external forces such as an electric field. Moreover, Professor Swinney at the University of Texas in Austin and Professor Showalter at West Virginia University, both of whom are well-known as experimentalists well acquainted with theories in the research fields of self-organization of patterns and non-linear dynamics, seem to frequently exchange ideas with theoreticians.

On the other hand, turning our eyes to situations in Japan, no large trend in research on self-organized materials has been created in Japan in spite of the facts that some researchers in Japan have earned excellent reputation from foreign countries and that such researchers have exchanged ideas with theoreticians for several years. In Japan, theoreticians and experimentalists still pursue self-organized material research separately.

7.3.2 Examples of organizational research for self-organized materials in Japan

In Japan, after the Kunitake Molecular Architecture research project within the Exploratory Research for Advance Technology (ERATO) program, administered by the Research

Development Corporation of Japan (JRDC) (research term: 1987-1992), research on self-organized material has been pursued in the form of projects such as the New Strategic Sectors Decided for Core Research for Evolutional Science and Technology (CREST) "Construction and Functions of Molecular Complex Systems (Total Construction of Energy Conversion and Signal Transduction Systems in Biology)" (research leader: Professor Yoshiaki Kobuke at the Graduate School of Materials Science, Nara Institute of Science and Technology)/ Japan Science and Technology Corporation (JST) (research term: 1998-2003), the Yokoyama Nano-structured Liquid Crystal Project within the ERATO program/JST (research term: 1999-2004), etc.

Furthermore, good results have been obtained including the creation of nanostructure with the self-organization method in collaborative research organizations such as the Joint Research Center for Atom Technology [JRCAT] (centralized joint research organization for the collaboration of industry, academia and government, whose parent organizations include the Angstrom Technology Partnership [ATP] and the Agency of Industrial Science and Technology [AIST]), which has completed a ten-year project begun in April 1992 and ended in March 2002.

Recent movements toward the facilitation of self-organized material research include the establishment of the Nanotechnology Research Center at the Research Institute for Electronic Science, Hokkaido University (director of the center: Professor Masatsugu Shimomura) in April 2002. The center is deemed to be a "facility to develop innovative nanoscience technologies integrating the top-down approach for semiconductor technologies into the bottom-up approach utilizing molecular or atomic self-organization, through interdisciplinary and multidisciplinary research activities, and to play a part in the nanotechnology network in Japan." Expectations are now placed on future achievements produced through research activities at the center.

7.3.3 Comparison between the micro-nano fabrication (miniaturization) technique and the self-organization method in the preparation of materials — Examples of the preparation of honeycomb films

In this section, we will compare two methods for preparation of the same honeycomb film: One is the lithography method, a representative "miniaturization technique"; the other is the "self-organization method" (see Figures 2 and 3).

As you can see from Figures 2 and 3, when you prepare the nanostructure such as honeycomb films, it will be more helpful to utilize the self-organization process for the following reasons:

(i) The self-organization method is more energy- and cost-saving (In the lithography method, it does matter what kind of devices you use, while there is no need for the use of large or high-priced devices when you utilize the self-organization process).

(ii) The self-organization method requires fewer steps and a shorter time (You cannot finish the entire process in one hour by using the lithography technique, but you can finish the entire process in about 30 minutes when utilizing the self-organization method).

(iii) The self-organization method enables you to prepare continuous patterns in a large area.

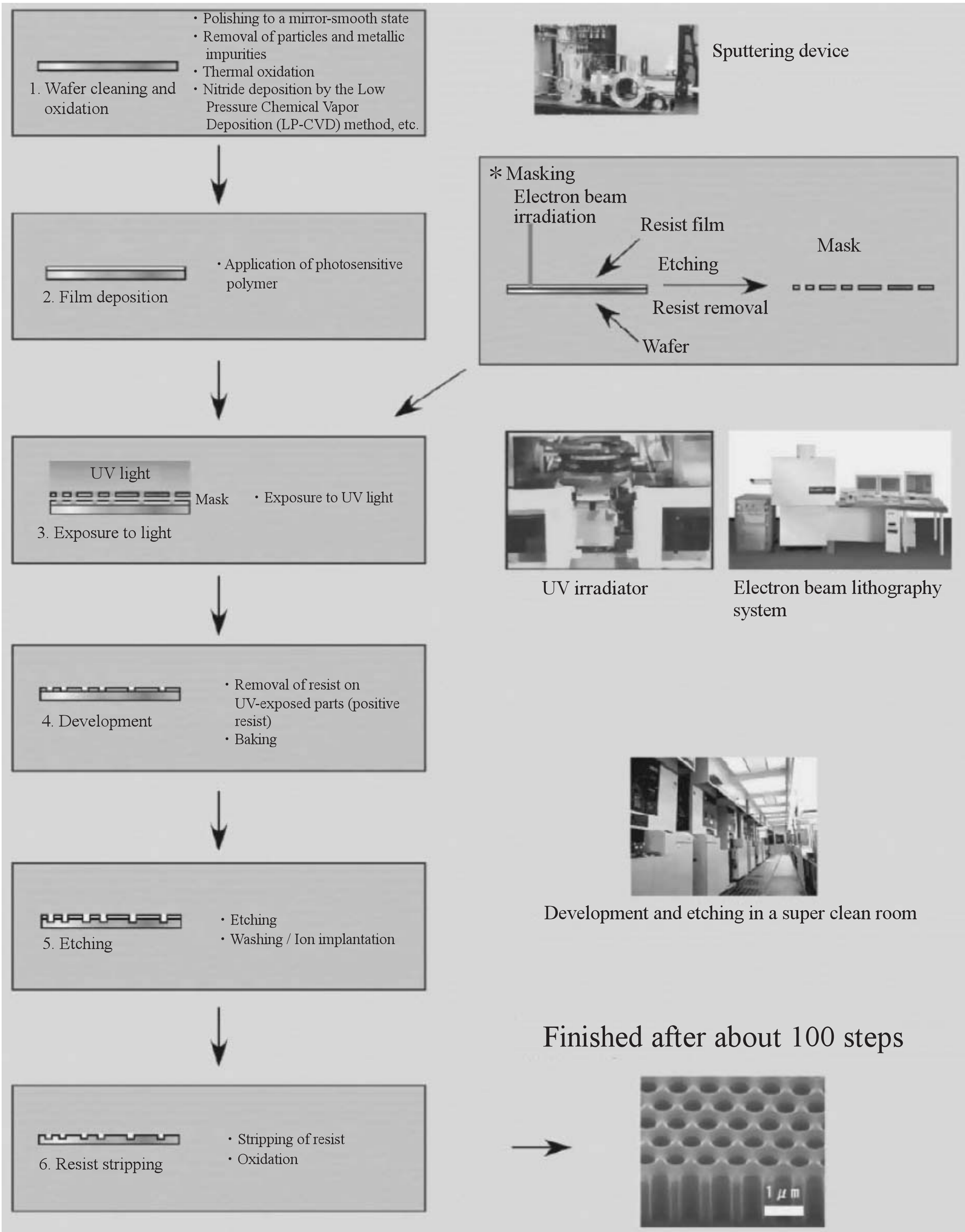
(iv) The self-organization method permits a wider selection of materials to be prepared (Lithography permits only a limited selection of materials like silicon substrate. However you can choose either inorganic or organic substances as materials to be prepared with the self-organization method).

(v) The self-organization method does not require a high degree of engineering skill when you handle devices to be used in the relevant process (The steps of preparation by the self-organization method can be automated).

However, you should take notice that it cannot always be said that preparation with the self-organization method is more advantageous in the production of some structures.

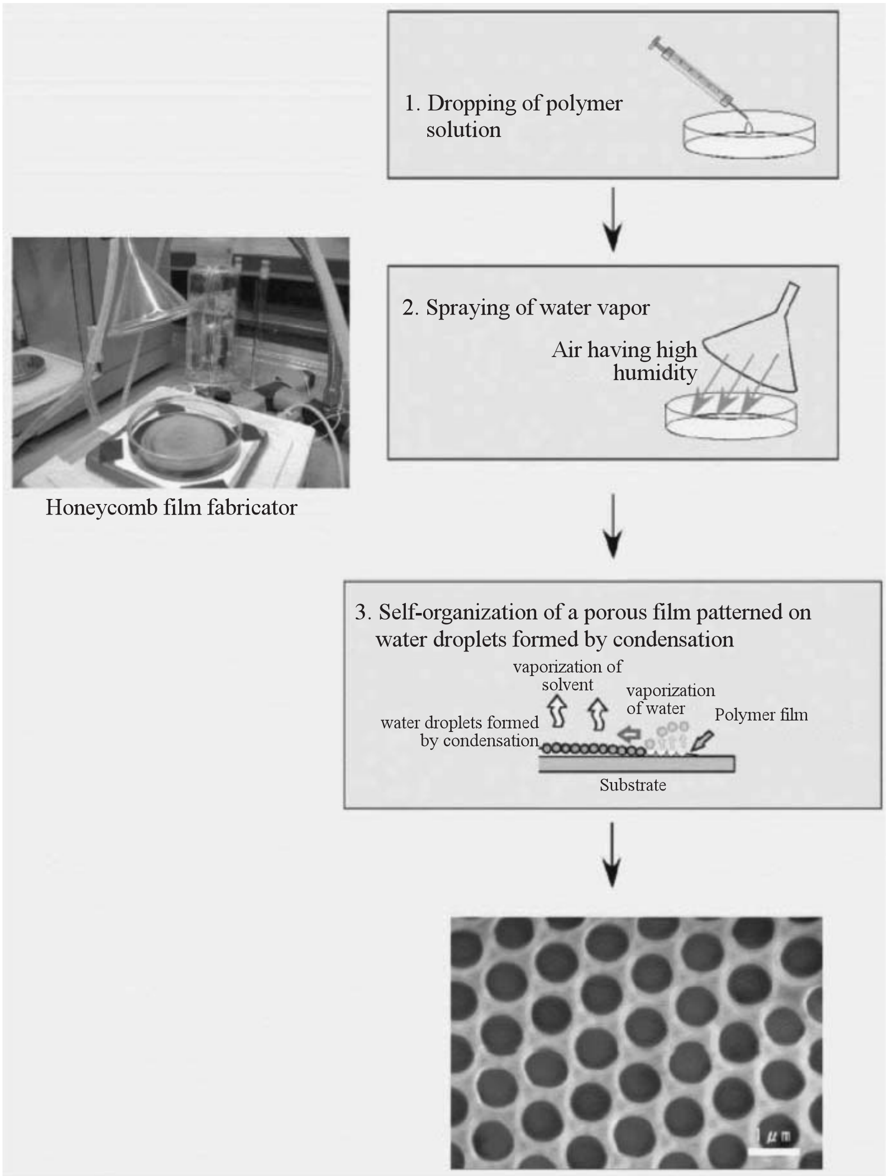
The self-organization process has the following drawbacks: First, it produces "a large environmental load because the method necessitates the use of organic solvents" (but this drawback can be removed if you operate devices in a completely closed system); and second, "it has poor reproducibility" (i.e., it produces errors in places) as opposed to lithography, which allows the production of desired 2-dimensional structures without failure (However, while honeycomb pore sizes show statistical distribution, they are so uniform and regular that they produce a

Figure 2: Preparation Processes for honeycomb films using microfabrication technology.



Source: Authors' compilation by making reference to materials provided by Professor Masatsugu Shimomura, director of the Nanotechnology Research Center, Research Institute for Electronic Science, Hokkaido University

Figure 3: Preparation for honeycomb films through the self-organization process.



Source: Authors' compilation by making reference to materials provided by Professor Masatsugu Shimomura, director of the Nanotechnology Research Center, Research Institute for Electronic Science, Hokkaido University

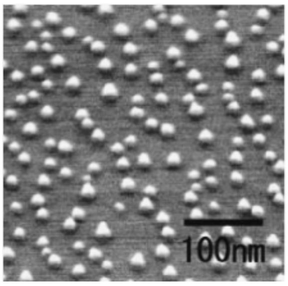
considerable number of high-order diffraction gratings under scattered light. Therefore, under appropriate conditions, almost uniform structures can be fabricated with excellent reproducibility).

7.3.4 Examples of research aiming to achieve Goal A (Precision synthesis of molecular clusters)

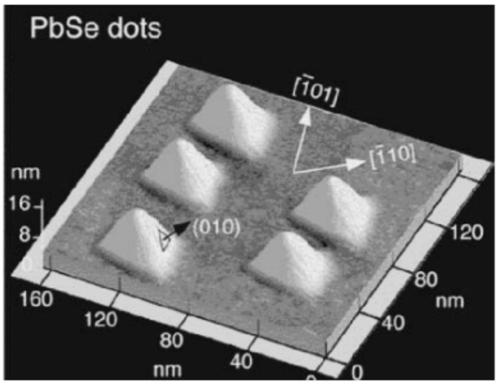
(1) Preparation of Compound Semiconductor Quantum Dots

It is expected that quantum-dot-based devices with novel structures will exert innovative functions that cannot be expected from conventional bulk-type semiconductor devices. So far, such devices including power-saving quantum dot lasers that emit strong light by the passage of only a small current have been prepared at the laboratory level, and research on compound semiconductor quantum dots has been pursued with the aim of applying the findings from research to the production of high-density memory devices and advanced telecommunication devices (see Figure 4).

Figure 4: A scanning electron micrograph (SEM) of GaAs quantum dots fabricated on the surface of a GaAs substrate (left); An atomic force micrograph (AFM) of lead selenide (PbSe) quantum dots with pyramidal shape fabricated on the surface of a lead telluride (PbTe) substrate (right).



Source: provided by Nobuyuki Koguchi, affiliated Fellow



Source: October 23 issue of the journal Science [1998; Vol. 282, pp. 734-737].)

(2) Synthesis of molecular clusters, which are extremely difficult to prepare by conventional chemical synthesis

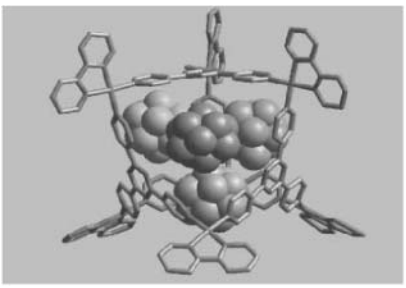
In the biological system, hydrogen bonds are skillfully utilized as the driving force for self-organization. While Professor Makoto Fujita of the Department of Applied Chemistry, Graduate School of Engineering, University of Tokyo, has been spontaneously and quantitatively preparing molecular clusters by utilizing coordinate bond as a driving force. In addition, Professor Fujita found that materials with a 3-dimensionally closed structure as shown in Figure 5 have unique inner space inside their molecular skeletons that is isolated from the outside world, and he considers that molecules incorporated in such materials are expected to have novel properties or chemical reactivity. Therefore, Professor Fujita is now pursuing research on such materials (see Figure 5).

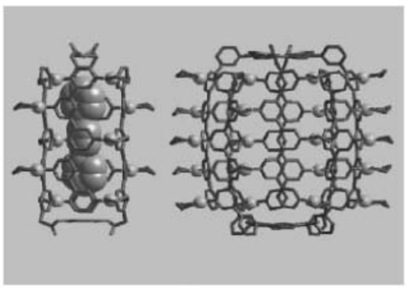
Professor Fujita has analyzed the current state of research on the organization of molecular clusters through self-assembly as follows:

— Since around 1990, marked progress has been seen in research on self-assembly in systems utilizing hydrogen bond or coordinate bond. Unique nanostructure based on ingenious molecular designs have been prepared and reported, including the "tennis ball molecule" that was formed by binding, at the seam of hydrogen bonds, molecules resembling developed tennis balls in shape as well as a "double helical complex" that was formed as two or more metal ions with two molecular strings coiling around them.

— Triggered by our research on compounds such as the "molecular square" (1990) and

Figure 5: Cage structure (measuring about 2 nm in diameter: In this figure, four molecules of carborane (boron-carbon cluster measuring 0.08 nm in diameter) are confined in the cage structure) (left); Tube-shaped capsule structure (measuring about 3 nm in length) and barrel structure (measuring 2 nm and 3 nm in diameter and height, respectively) (right).



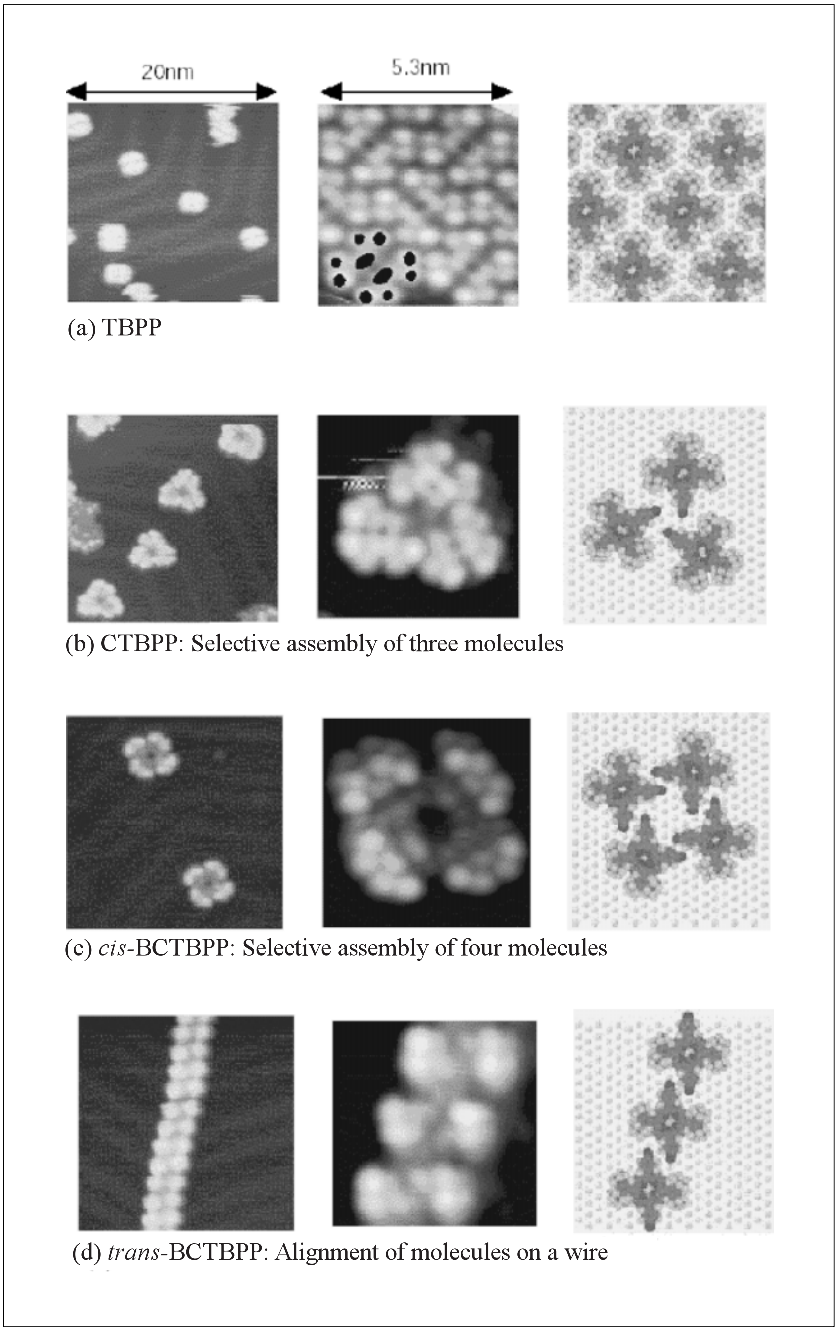


Source: provided by Professor Makoto Fujita of the Department of Applied Chemistry, Graduate School of Engineering, University of Tokyo "molecular regular octahedron" (1995), which we have successfully prepared through self-assembly, research has been actively conducted with the aim of building up various two- or three-dimensional structures into polygons or polyhedrons through self-assembly.

— In our research, we can uniquely and quantitatively self-organize the requested structure by integrating different manners of bonding (directions, forces, and numbers) and variety of molecular designs of organic molecules.

— On the scales of about several nanometers or less, the principles of self-assembly, in which the intermolecular force associated with specific directionality and appropriate binding strength is to be strategically utilized, are about to be established. However, on the scales of several to several tens of

Figure 6: Scanning tunneling micrographs of a porphyrin aggregate (left two columns) and corresponding simulated models used in the consideration before the actual assembly of molecules (right one column).



Source: Website of the National Institute for Materials Science [independent administrative institution]; http://www.nims.go.jp/nims/former/info/press12.pdf nanometers, the principles of self-assembly are far from the establishment.

(3) Control of molecular nanostructure on solid surfaces

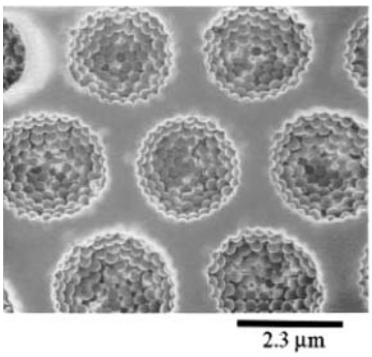
Takashi Yokoyama, a researcher in the Nano-Device Research Group at the Nanomaterials Laboratory, National Institute for Materials Science (an independent administrative institution) has shown that direct binding of porphyrin molecules onto solid surfaces is hindered by the addition of the insulating butyl group ($-CH_2CH_2CH_2CH_3$) as a "foot" to those molecules, which is known to be a functional molecule, and has also shown that the addition of the cyano group (–CN) as a "hand" enables selective and spontaneous molecule-to-molecule binding. Extensive studies have been conducted on technologies for assembling molecules by utilizing molecular hands, but most of such studies have been done in liquid. Since self-assembly on solid surfaces is a prerequisite for the practical use of such technologies, development by Dr. Yokoyama of technologies for assembling molecules into molecular clusters on solid surfaces has great significance (see Figure 6).

7.3.5 Examples of research aiming to achieve Goal B (Establishment of patterning and self-aligning technologies)

(1) Preparation of dots, wires, lattices, honeycomb structure, etc..., by using polymers and fine nanoparticles

Professor Shimomura, et al. mentioned above (Figures 2 and 3) have succeeded in preparing nanostructure as shown in Figures 7 and 8, by utilizing the "polymer-solution-casting process" in which they cast polymer solution onto the surfaces of substrates and dry them to prepare thin films (A photograph of the honeycomb structure is shown in Figure 3). At present, research is being pursued on technologies for preparing honeycomb films with the aim of applying the technologies to the preparation of photonic crystals having periodic alignments of

Figure 7: Fine particles measuring about 100 nm in diameter embedded into the pores (measuring a few micrometers in diameter) on an e-caprolactone (a biodegradable polymer) honeycomb film as the substrate (left); Liquid crystalline polyacetylene deposited on the mica substrate (provided by Professor Kazuo Akagi, Tsukuba University), which was patterned by Professor Masatsugu Shimomura et al., Hokkaido University (measuring 4μm and 50 nm in width and height, respectively)(right).



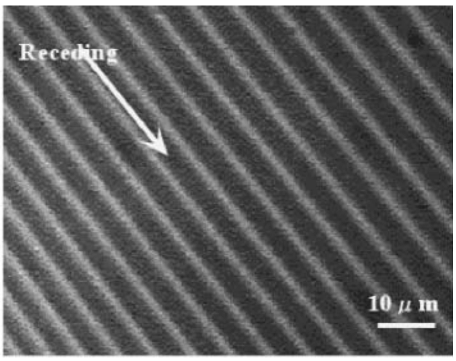
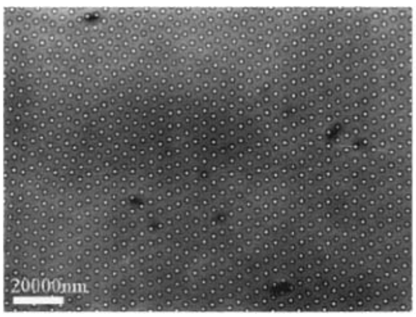
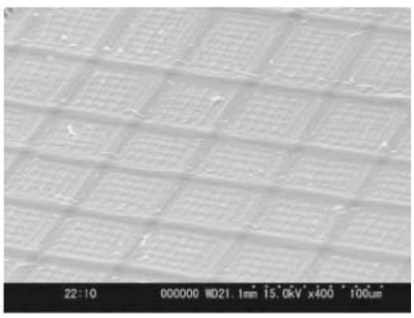


Figure 8: Polystyrene dots measuring 1-2μm and 30 nm in diameter and height, respectively, fabricated on the mica substrate (left); Polystyrene lattice fabricated on a slide (line widths are 5μm and 1μm, and heights are 500 nm and 100 nm.) (right)





unit structure on the scale of light wavelengths (photonic crystals are utilized for the artificial control of light emission and light propagation). In addition, it has been demonstrated that honeycomb films themselves can be used as, for example, the substrate for cell culture.

The "polymer-solution-casting process" is a versatile technique that can be utilized for the patterning of structure independent of the types of materials. Therefore, it is expected that quantum dots and quantum wires (an artificial structure that confines particles carrying electricity on a one-dimensional space [along the anteroposterior axis]) prepared by the technique will contribute to the development of liquid crystal displays and electronic paper (ultra-thin displays that are portable in the rolled-up form) with novel properties (See Figures 7 and 8).

7.3.6 Examples of research aiming to achieve Goal C (Preparation of materials and devices through the self-organization method)

Until now, research aiming to achieve Goal A (Precision synthesis of molecular clusters) and Goal B (Establishment of patterning and self-aligning technologies) has been pursued to some degree. In contrast, research aiming to achieve Goal C (Preparation of materials and devices through the self-organization method) has been conducted with relatively limited success as compared to that of Goals A and B. Therefore, future progress in research for Goal C is being awaited.

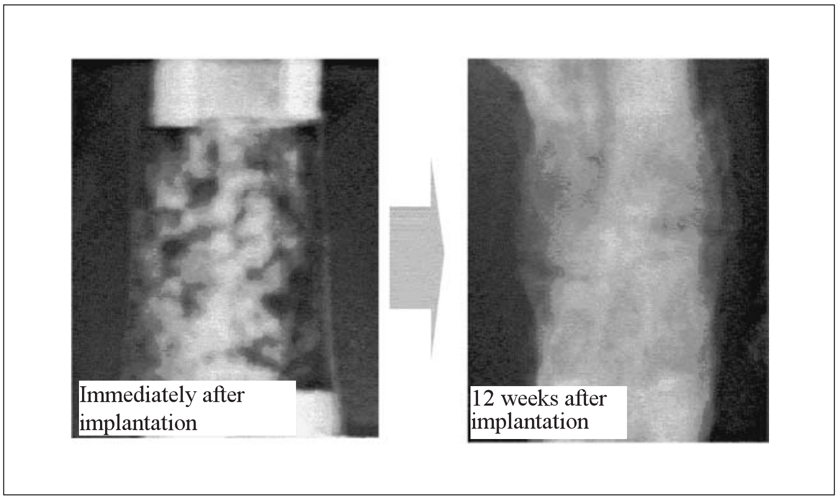
(1) Development of bone-like materials that are to be integrated into the bone metabolism system in vivo

Professor Kenichi Shinomiya at the Graduate School of Tokyo Medical and Dental University and Junzo Tanaka, director and chief researcher at the Biomaterials Center, National Institute for Materials Science (an independent administrative institution) have synthesized bone-like hydroxyapatite/collagen nanocomposites that have a bone-like structure and chemical composition under near-biological conditions (pH 8-9, temperature 40°C). They confirmed that hydroxyapatite crystals (30 nm) and collagen molecules (300 nm) self-organized to form fibers with an overall length of 20μm or more under near-biological conditions (See Figure 9).

(2) Alignment of many nanodevice components on flexible or curved substrates

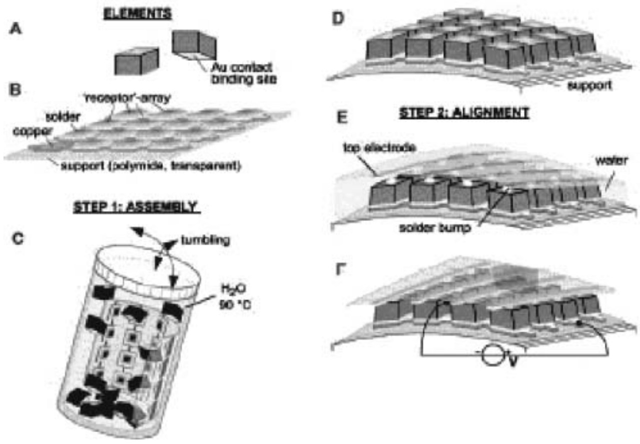
Professor George M. Whitesides has prepared, by utilizing the self-assembly mechanism, a cylindrical display with 113 light-emitting diodes (LED) with a dimension of about 300μm in length. In

Figure 9: Particulate bone-like composite was transplanted into the tibia of a dog (defective part of the tibia measuring 20 mm in length was covered with a bioabsorbable film in order to fix the particles). Twelve weeks after the transplantation, bone tissue at the defective part was almost completely regenerated and the dog regained free mobility.

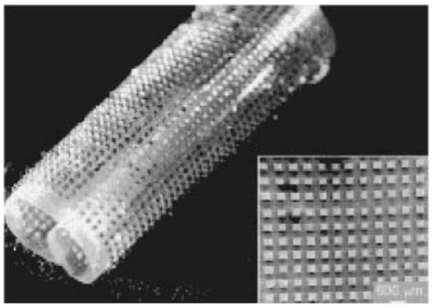


Source: Website of the National Institute for Materials Science [independent administrative institution]; http://www.nims.go.jp/nims/former/info/press11.pdf

Figure 10: When light-emitting diodes (LEDs) covered with gold and copper substrate having solder arrays on its surface were suspended in water having a temperature exceeding the melting point of solder, in such a way as to minimize the free energy of the solder-water interface (A-F); About 1,560 silicon cubes were successfully aligned on the surface of a flexible and curved substrate within 3 minutes (right).



Source: April 12 issue of the journal Science [2002; Vol. 296, pp. 323-325]



Source: April 12 issue of the journal Science [2002; Vol. 296, pp. 323-325], as with the figures on the left

addition, he has succeeded in generating an array containing 1,500 small silicon cubes on an area of 5 square centimeters in less than 3 minutes (See Figure 10).

7.4 Issues to achieve goals (in order to bridge the gap between the goals and the present state)

Primary requirements for the accomplishment of Goal A (Precision synthesis of molecular clusters), Goal B (Establishment of patterning and self-aligning technologies) and Goal C (Preparation of materials and devices through the self-organization method) may need for a deeper understanding of self-assembly, the formation of dissipative structures, and the combination of self-assembly and dissipative structure formation, respectively. And further research for the preparation of practical materials should be promoted through the achievement of Goal C.

7.4.1 Issues to achieve Goal A (Precision synthesis of molecular clusters)

"Intermolecular interaction plays an important role in the self-assembly process, so success in realizing self-organization through self-assembly depends on molecular designs."

The process of self-organization through self-assembly has the advantages that it allows efficient preparation of three-dimensional structure, which is several tens of nanometers or less in size and are difficult to be prepared with the top-down method, and that it permits, once molecular design is properly developed, the precise preparation of three-dimensional structure with minimum energy. However, even when you want to apply a combination of some principles of self-assembly to the process of materials with intended structure, almost no such principles have been discovered as yet. In order to develop the self-organization method through the self-assembly, it is imperative to deepen the understanding of the principles in self-assembly by pursuing research on the control of intermolecular forces such as hydrogen bonds and coordinate bonds and to establish technologies for preparing desired materials for practical use by applying a combination of some of those principles.

Professor Nobuo Kimizuka at the Department of Applied Chemistry, Graduate School of Engineering, Kyushu University said, "From now on, simple preparation of nanostructure will not be enough, and it will be increasingly significant to design molecular structure with electronic states that may realize the exertion of innovative functions and to prepare such structures through the self-assembly mechanisms."

7.4.2 Issues to achieve Goal B (Establishment of patterning and self-aligning technologies)

"When you intend to create specific dissipative structures, you need to take into account not only molecular designs but also the environment around the molecules (external systems). In designing the external systems, you should establish methods for quantitatively evaluating the flows of energy and entropy through the relevant molecules and associated external systems, then discover principles in the designing of appropriate external systems that permit the desired patterning and periodicity of the relevant molecules, and develop technologies that allow the application of those principles to the relevant molecules and external systems."

In order to enhance the readers' understanding of flows of energy and entropy through systems, here we cite crystal growth as an example. Crystal growth is the process during which crystalline substances in solution (raw material) is transformed into solid crystals, and has very great significance in the preparation of various materials. In order to acquire crystals with intended thickness, composition, shape and quality (in terms of defect density, etc.), it is necessary to control; the temperature during the growth process, supply system for raw materials, degree of supercooling or oversaturation, etc. While the necessity for control of these factors has been empirically well recognized, such control exactly corresponds to the external system control for the control of the flows of energy and entropy through the system. In light of the case of crystal growth cited here, in order to establish a method for self-organization through the mechanism of dissipative structure formation, it may be necessary to pursue research on elementary processes (in cases where complex phenomena occurring in the system containing a large number of particles can be seen as an assembly of simpler phenomena occurring among a smaller number of particles, the phenomena among the smaller number of particles is called the "elementary process") from the viewpoint of the flows of energy and entropy.

7.4.3 Issues to achieve Goal C (Preparation of materials and devices through the self-organization method)

Issues to achieve Goal C are "to prepare desired materials and devices through the formation of dissipative structures by utilizing structure and parts (components) prepared through the self-assembly mechanism and to prepare desired materials and devices by inducing the self-organization of the components prepared through the self-assembly mechanism."

As mentioned in section 7.3.4-(1), power-saving quantum dot lasers that emit strong light by the passage of only a small current have been prepared at the laboratory level, and great achievements have been yielded in various research fields of nanotechnology. However, as of now, it is not possible to prepare devices with complex structure for practical use through the self-organization mechanism. The most realistic approach as of now may be as follows: First, create clusters of atoms or molecules through the self-organization of those atoms or molecules; and then, build up various atomic or molecular clusters into hierarchical structure in accordance with their scales to create the desired materials or devices.

7.5 Conclusions

So far in this report, we have summarized and described our views regarding the goals and present state of self-organized material research as well as issues to achieve such goals. As stated, in order to prepare various nanostructure, it is highly significant to develop not only conventional nanopreparation techniques but also techniques utilizing the self-organization method.

Furthermore, if the self-organization method becomes applicable to the preparation of materials and devices, which are now created by other methods, it may greatly contribute to resource saving and energy conservation and, further, to environmental protection. In this context, progress in self-organized material research is expected by researchers not only in the filed of nanotechnology but also from a wide variety of areas. We propose in the following paragraphs two issues to facilitate progress in self-organized material research.

(1) Collaboration between theoreticians and experimentalists targeting the establishment of technologies for molecular designing, etc.

In order to develop the self-organization method from this day forward, it will be necessary to further promote research activities aiming to attain the above-mentioned Goal A (Precision synthesis of molecular clusters) and Goal B (Establishment of patterning and self-aligning Technologies).

In self-organized material research conducted in the United States and European countries, good results have been yielded through collaboration between theoreticians and experimentalists. In contrast, no such research environment has been created in Japan. Improvement in this respect may be important for Japan to make further progress and to constantly stay ahead of other countries in self-organized material research.

With regard to the clarification of principles of self-organization, participation of theoreticians in various fields is necessary because dominating laws of systems are scale-dependent. (For example, quantum effects become obvious on the scales of 30 nm or less.) The same holds true for experimentalists. One of the specific measures enables collaboration between theoreticians and experimentalists is to secure human resources to serve as an interface between such theoreticians and experimentalists. Here, human resources as an interface mean people who act as an intermediary between theoreticians and experimentalists by, for example, analyzing the relationship between theories that have been elaborated after the simplification of various conditions and experiments, which have been actually conducted to verify the theories, and by notifying researchers about the findings from such analyses in the form of feedback. Therefore, the people to serve as an interface should have the peculiar aptitude for such work.

It is not necessarily easy to secure human resources having such special aptitude. One possible practical measure to secure such human resources may be, when organizing a project of research through the approaches proposed in the following sections, the specification of functions to be served by people as an interface and to assign the role as an interface to researchers participating in the relevant research project.

(2) Promotion of multidisciplinary and comprehensive research with clear objectives

In order to realize the application of the self-organization method to techniques for the mass production of materials, devices, etc., we must attain Goal C (Preparation of materials and devices through the self-organization method) mentioned earlier in this report. In order to develop technologies for preparing complex three-dimensional structures by hierarchically building up atomic or molecular clusters, cooperation from researchers in a wide variety of fields including mathematics, physics, chemistry, biology, material engineering and mechanical engineering is required in addition to collaboration between theoreticians and experimentalists discussed in the preceding section. Moreover, with an eye toward practical application of technologies developed, it is imperative for researchers and engineers in the industrial community to participate in such research activities.

As of now, however, concepts regarding self-organization vary in accordance with the field, and the sense of mission in research activities is not always shared among researchers. One possible means of tapping into the collective wisdom of researchers in various fields or organizations to promote research on self-organization may be the implementation of a "multidisciplinary and comprehensive research project with clear objectives."

As the first step to implement such a project, you need to set forth such an objective that gives a concrete image to everyone and for which increase in technological levels is essential, as "preparation of a computer in a beaker," and gather experts whose involvement is necessary for the achievement of the objective as well as researchers in a wide variety of fields who are interested in achieving the objective. As the second step, you need to specify issues to achieve the objective. Specifically speaking, issues to be handled to achieve the above objective may, as an example, include the development and preparation of (i) structures that are analogous to transistors, (ii) structure analogous to circuitry, and (iii) structure analogous to a central processing unit (CPU) (architecture). Moreover, in promoting a research project, it may be necessary to clarify the roles and responsibilities to be taken by research groups and researchers intending to address these issues.

Acknowledgments

We are deeply grateful to Dr. Tomohiko Yamaguchi, chief researcher at the Nanotechnology Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), as well as Professor Masatsugu Shimomura, leader at the Dissipative Hierarchy Structures Laboratory of the Spatio-Temporal Function Materials Research Group, Frontier Research System, Institute of Physical and Chemical Research (RIKEN) and director of the Nanotechnology Research Center, Research Institute for Electronic Science, Hokkaido University, who, on the occasion of the making of this report, provided us with explanations about self-organization and highly valuable suggestions.

In addition, our profound thanks also go out to all those who provided us with the wide variety of valuable information, mainly including Professor Makoto Fujita of the Department of Applied Chemistry, Graduate School of Engineering, University of Tokyo; Professor Toshimi Shimizu, director of the Nanoarchitectonics Research Center, National Institute of Advanced Industrial Science and Technology (AIST) and professor at the Department of Chemistry, Tsukuba University (cooperative graduate school); Mr. Makoto Uchigasaki, graduate student in the first year of the doctoral course, Ohdomari Laboratory, Department of Electronics, Information and Communication Engineering, Graduate School of Science and Engineering, Waseda University; Professor Toshiro Hiramoto of the Department of Material and Life Science, Institute of Industrial Science, University of Tokyo; Professor Nobuo Kimizuka of the Department of Applied Chemistry, Graduate School of Engineering, Kyushu University, and sub-director Dr. Atsushi Suzuki of the Nanotechnology Researchers Network Center of Japan, National Institute for Materials Science (an independent administrative institution); Mr. Junichi Hoshi of Department of Research Project Office of Basic Research, Japan Science and Technology Corporation (JST); and Mr. Hiroyuki Kaneko of the First Research Promotion Division, Department of Research Promotion Office of Basic Research, JST.

(Original Japanese version: published in July 2002)