5

Trends in Material Simulation, Centering Around Ab Initio Calculation

JUN-ICHIRO TAKANO
Materials and Manufacturing Technology Research Unit

5.1 Introduction

Simulation has increasingly been used in Materials Research. Tens of years ago, instances of applying simulation to Materials Research were rare, and few of them were practically useful to Materials Research. The situation, however, has been changing, due to the rapid improvement in the performance of computers (refer to the special feature, "Trends in Supercomputers," in the October 2001 issue of Science and Technology Trends), advancement in simulation methods, and enhanced experimental technologies in the micro regions as is called Nanotechnologies (nano is equivalent to one-billionth).

In fact, in "Promotional Strategy for Nano Technologies and Materials Field (September 2001)" laid down by Council for Science and Technology Policy, Cabinet Office, "basic technologies, such as instrumentation/evaluation and numerical analysis/simulation, and the fields on which they have impact" is listed as one of the five key areas. Also, the following is listed as one of the performance goals to be achieved in this area in the coming five years:

> Establishment of the utilization of simulation in the development of new materials and devices

In this article, I would like to comment on the current level of Material Simulation as one of the basic technologies of nanotechnologies, and propose the necessity of making efforts towards acceleration of utilization rates of simulation in syntheses for new materials.

5.2 Roles of Simulation in Materials Research

Material Simulation is a "tool" and also a method of Materials Research: "computer experiment (imitative experiment)". Researchers carry out, as required, computer simulations instead of conducting real experiments using precision equipment to "examine (try, verify, experiment) ideas".

Now, some merits of simulations dealing with nano-meter scale materials and their respective instances are listed below:

Merit 1) Speeding up and Cost Reduction in Materials Development

Out of all conceivable experiments, minimum necessary ones are selected after prediction of the structure, physical properties and phenomena of a material by use of simulation. In this way, the number of experiments necessary for research and development can be reduced, thus contributing to speeding up as well as the cost reduction of materials development.

•Example of merit 1)

A phenomenon called "electro-migration," in which metal atoms migrate as high-density current runs through the metal wiring, can be a problem for the preparation of semiconductor devices. A certain electric company, who applied an atomistic simulation to the selection of additive elements to prevent breaking down of metal wiring and also to the selection of substrate material to prevent removing, succeeded in shortening the period for material design down to 4 months from 1 year, which would normally be

required when relying totally on repetition of experiments on a trial-and-error basis. (Source: Professor Takayuki Kitamura, Graduate School of Engineering, Kyoto University)

Merit 2) Electronic / atomistic understanding of phenomena that are difficult or impossible to verify by means of experiment

It is possible to analyze the electronic or atomic mechanism of phenomena by direct atomistic observation the behaviors of the atoms at every moment through simulation. It is also possible to obtain various information such as the inside conditions of the material, and a phenomenon of chemical reaction that takes place in an extremely short period of time.

•Example of merit 2)

What role does the Ziegler-Natta catalyst, which is indispensable for the manufacture of high strength polyethylene and polypropylene for use in various moldings and fibers, play in chemical reactions and how the reaction progresses? Its mechanism has been made clear on the basis of the ab initio calculation (see comments in Section 5.4) made as a result of a joint research program between the Joint Research Center for Atom Technology (JRCAT) and the Max Planck Institute for Solid State Research (Germany). JRCAT is a centralized industry-government-academic joint research organization, with Angstrom Technology Partnership (ATP), technical research partnership commissioned by the New Energy and Industrial Technology Development Organization (NEDO), and the National Institute of Advanced Industrial Science and Technology (AIST) as parent bodies.

It was not possible to directly verify this chemical reaction mechanism by experiment, as it is very fast and progresses in an extremely short time, so that details of the reaction process have hardly been known. It is expected that such results can be utilized for upgrading and improvement of manufacturing technologies.

Since a lot of behaviors of the atoms and molecules in such a complicated chemical reaction cannot be analyzed by experiments alone, material simulation play a big role in analyzing the chemical reaction mechanism.

Next, I would like to review the trends in academic lectures presented at annual academic meetings for an overview as to what extent Material Simulation is used in Materials Research and which features are attracted for utilization. While presentations on study results concerning Material Simulation are made at a lot of meetings, here I would like to introduce the case of the Japan Institute of Metals (JIM) as one of examples showing a cross-sectional view of the current status of Materials Simulation studies.

More than 1,000 presentations are made at each of the spring and fall Annual Meetings of the JIM. "Ten years ago, presentations of studies relating to simulation which numbered several at the most. But it has come to account for about 10% of the total in recent years," says Professor Yoshiyuki Kawazoe of the Institute for Materials Research, Tohoku University.

Also according to an investigation by the Science and Technology Foresight Center, presentations of study results, which have a close relationship with Material Simulation, in the 2001 JIM Fall Meeting of the academic lectures numbered 120 out of 1,388 presentations in total, accounting for 8.6% of the total.

5.3 Material Simulation in the 7th Technology Foresight

The National Institute of Science and Technology Policy, under the Ministry of Education, Culture, Sports, Science and Technology, has continued to conduct a large-scale and exhaustive "Technology Foresight" almost every five years since 1971, to investigate the courses of development of science and technology from a long range perspective. The latest 7th Technology Foresight, which was published in July 2001 and covered more than 1,000 survey topics in 17 different fields, was carried out with the cooperation of almost 4,000 experts in the respective fields.

Out of 103 survey topics in total, in the field of "Materials and Processing" of the above survey, 4 topics relating to Material Simulation are shown in Table 1.

Out of the 4 topics in Table 1, attention was paid to the topic (3) relating to the "Ab Initio

Calculation" (refers to the comments in Section 5.4), which is expected to make a contribution particularly to the area of below nano-meter scale. Results on comparison between the average of responses to the various survey items on all the topics in the Materials and Processing Field and those to the topic (3) are shown in Figure 1 below.

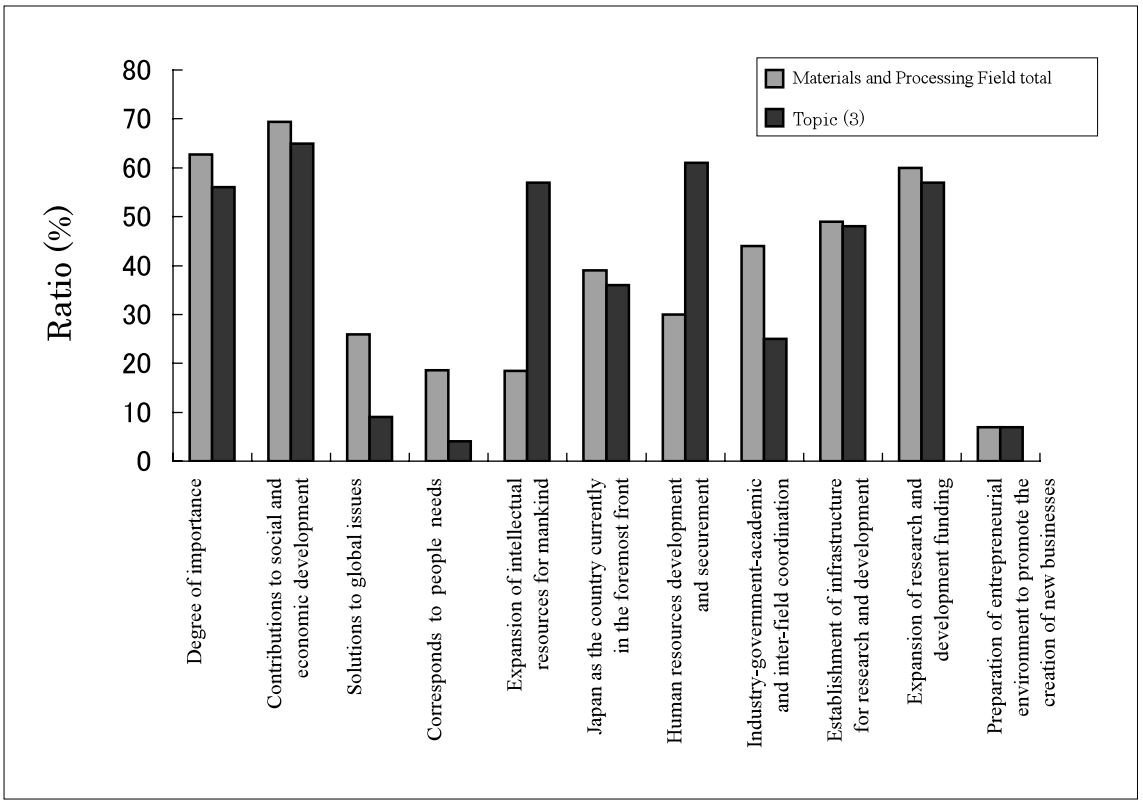
Also, the following survey results were obtained:

- While it is estimated that it will be in the year 2016.6 on the average when all the topics in the Materials and Processing Field are realized, it will be in the year 2018, comparable to the above average, for the topic (3) to be implemented.
- The countries the respondents selected as those that took the lead concerning the topic (3) were the United States (86%), Japan (36%) and EU (26%), in the order of score. (Since the responses are collected as "check all that apply", the total is not 100%.)

Table 1: Topics Related to Material Simulation in the 7th Technology Foresight

Topics related to Material Simulation	Importance index	Forecasted realization time
(1) Theoretical design of performance using computers for metallic materials becomes possible.	55	2016
(2) Computer-aided material designing method is put to practical use for solid catalysts having the necessary composition, structure and physical properties.	54	2017
(3) Technology of designing a material of prescribed features is put to practical use by means of simulation based on the ab initio calculation.	56	2018
(4) Computer simulation technology enables to estimate strict structures and physical properties in thermal equilibrium state of multi-element material, once elementary composition is provided.	49	2018

Figure 1: Comparison between average response to the topics in the Materials and Processing Field in total and that to the topic (3).



5.4 Current Level of Ab Initio Calculation in Materials Research

In Materials Research, it is an important subject of study to clarify how the functions of materials, as aggregates of atoms, are developed through understanding of the electronic states, locations, and interactions of individual atoms and their changes with time, etc., and to create materials of innovative functions by means of positively controlling their mechanisms.

Regarding Material Simulation on the nano-meter scale, studies have now been carried out for establishment of the method called "ab initio calculation" and for its application to real Materials Research.

While it is the basic equation of classic mechanics that describes the world we see routinely, it is the basic equation of quantum mechanics that describes the world of around 30 nano-meter or less. It is known that various physical properties are obtained on the nano-meter scale by changing conditions to solve the basic equation of quantum mechanics. This method represents a study method of performing nonempirical calculations without using experimental results as parameters of calculation to obtain the required information, and it is called "ab initio calculation".

The reason why ab initio calculation is highly attracted among various methods of simulation is that it is considered to be almost the only method enabling to predict quantitatively the structure, electronic state and physical properties of various materials by obtaining their electronic density distribution functions based on the basic equation of quantum mechanics, as different from conventional simulation methods that contain empirical parameters to be determined by experimental results.

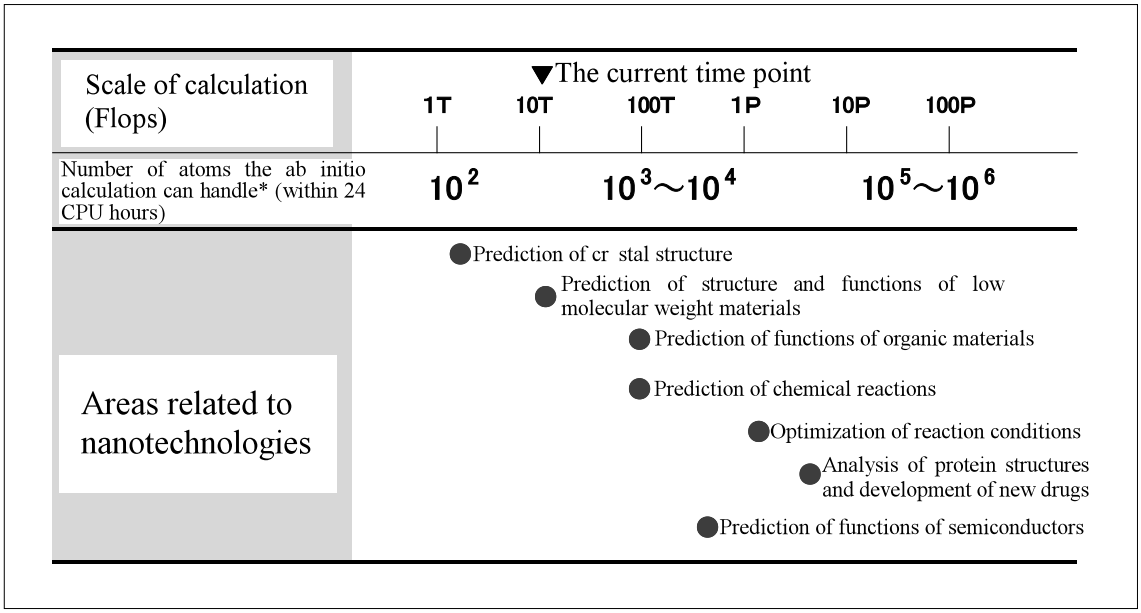
Then, what is the current level of the ab initio calculation?

The ab initio calculation is at such level as is applicable to clarification and analysis of structure and electronic properties of a material having a crystal structure.

For example, with regard to calculation concerning the lattice and elastic constants of silicon, simulation can be done with accuracy of within 1% of the experimental result.

On the other hand, there is an example in which the experimental value came to match the value obtained from simulation as accuracy of experiment has been enhanced and preparation of ideal single crystals and artificial lattice has been made possible, though it was initially said that such magnetic momentum of artificial lattice

Figure 2: Scale of calculation of the ab initio calculation, and examples of industrial applications



* It is assumed here that such high-speed algorism as computing amount is proportional to the number of atoms or its square comes into practical use.

Source: Mechanical Engineering Research Laboratory, Hitachi, Ltd.

made of aluminum and iron, as was obtained from simulation, did not agree with the experimental value.

Figure 2 shows a summary of the scale of computation, the number of atoms that the ab initio calculation can handle, etc., when a supercomputer of the highest performance level currently available is operated for 24 hours.

It is highly difficult for a researcher to exclusively use a large, high-performance computer at a research site. Assuming such exclusive use should be made possible, if it takes one month to carry out a simulation for selection of an optimum material for design of a new device, it necessitates operating such a high-performance computer for one year without interruption only for the purpose of selecting an optimum material out of 12 different materials. Thus, it cannot be said to be realistic. In fact, in view of the performance of the computer assigned to each individual researcher and the limitation in the environment of use, the ab initio calculation could be conducted to handle several hundreds of atoms at the most.

Namely, when a three-dimensional material is considered, one thousand atoms (10 x 10 x 10 = 1000) should be handled, assuming that 10 atoms exist on one side. However, under the current performance of computers and the environment of use, it is not realistic to conduct the ab initio calculation on all of such one thousand atoms, and, in reality, it could be applied only to a material smaller than a cube consisting of 10 atoms on one side.

Therefore, the advent of "Peta Flops Computer (its computing scale is described as 1P in Figure 2)" is awaited, so as to conduct ab initio calculation useful to such real Materials Research as shown in the column of areas related to the ab initio calculation in Figure 2.

(Peta Flops computer is capable of executing 1,000 trillion times per second of calculations.)

As to the time when this Peta Flops computer is appear, it was forecasted in the Information and Communication Field of the 7th Technology Foresight that a "parallel computer system with one million processors connected and having a computing speed of 1 Peta Flops class will be put to practical use" in the year 2013.

Not only improvement in the performance of computers but also development of "the methods of reducing the amount of computation necessary for simulation" are required to cover a larger spatial scale (necessary for research of such structure, physical properties and phenomena, as a larger number of atoms are involved) and a longer time scale (necessary for study of phenomena over a longer time). Thus, studies on such issues are urged.

Now, assuming that the number of atoms to be handled by the ab initio calculation is N, the amount of computation is roughly in proportion to N^m. While m varies depending on simulation method, m = 2 ~ 7 applies. When the number of atoms to be handled is increased ten-fold, the resulting amount of computation increases from 100 times to 10 million times. In reality, the number of atoms to be handled in simulation cannot be easily increased.

Many researchers are therefore engaged in the development of a method called "Order N Method" (a method of curbing the increase in the amount of computation up to around ten times that of the original computation).

Basic theories as prerequisite for simulation, are also important so as to increase the number of atoms the ab initio calculation can handle.

For instance, it is not currently possible to predict structure, constituent elements or physical properties such as superconductive transition temperature of a superconductor. However, once a theory that can handle a strongly correlated electron system is established, there is the possibility of predicting the structure and physical properties of a superconductor on the basis of the ab initio calculation with such a theory incorporated. Study is in progress based on such assumption.

5.5 Conclusion

Concerning study on such ideal systems containing no impurity and are free from disorder in the structure on the atomic scale, as represented by carbon nanotube, theoretical study by use of simulation has preceded due partly to the technical difficulties of experiment.

However, owing to advancement in micro fabrication and other nanotechnologies in recent

years, experimental technologies of handling extremely clean systems where a single impurity atom is found out of 10 billion of silicon atoms have been increasing in number among others in the semiconductor industry. With the development of experimental technologies enabling the preparation of materials extremely free of impurities, it is now becoming possible to compare the result of simulation with the experimental result.

Hereafter, as direct comparisons between the experimental results and the simulation results increase in volume, improvement in quality such as accuracy and reliability of simulation is expected through feedback from such comparisons.

Also as illustrated in the example shown below, support by high-precision, high-reliable simulation is indispensable for upgrading of experimental technologies and establishment of theories, in order to achieve steady progress of nanotechnologies in the future.

- Scanning tunneling microscope (STM) and atom force microscope (AFM) provide important means of experiments in nanotechnologies. Atomic scale images of surface conditions can be obtained with STM and AFM. By comparing the result of high-precision simulation of quantum mechanical interaction between the atom located at pointed top of the probe of STM or AFM and the atom on the substrate with corresponding atomic scale images obtained from real STM or real AFM, more precise information can be obtained on the substrate observed.

  Thus, simulation can play a great role in experimental methods on the nano-meter scale.
- Contributions of Material Simulation to establishment of various theories including clarification of the mechanism of a chemical reaction that can not be observed by current experimental technology as it takes place in an extremely short time, which provide the basis for progress of nanotechnologies, are also highly important.

However, according to Table 1 and Figure 1, which show the result of the 7th Technology Foresight, the topics related to Material Simulation are regarded to be slightly lower in importance than the average of the Materials and Processing Field in total. This fact seems to indicate a situation that even researchers in the Materials and Processing field are not fully aware of the significance of simulation as basic technology to advance nanotechnologies.

In the near future, when material design by simulation becomes practicable due to the realization of the Peta Flops computer, etc., it is suspected to have not just a small effect on the gap in "research and development capability" among various research organizations based on "how well they can utilize Material Simulation".

It is necessary to try from now on to direct efforts toward Material Simulation by increasing the rate of utilization of simulation in research and development of new materials and devices, and on establishing collaborative practices between theoretical and experimental studies for the purpose of maintaining and enhancing research and development capability and competitiveness in the area of nanotechnologies and materials studies in the individual research organizations.

(Original Japanese version: published in December 2001)