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Prospect, rather than retrospect, on the impact of computers in catalytic research and development

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Abstract

A major issue in the application of computers to future catalytic research and development will be constructing user-friendly systems for computer-aided design of catalytic reactions and catalysts. Only the integration of the following technical elements can construct them: (1) obtaining and processing of knowledge from literature; (2) computational chemistry, simulation and neural network; (3) laboratory automation and laboratory office automation; (4) image processing and computer graphics; (5) expert systems for designing catalytic reactions and catalysts; (6) knowledge-based systems and artificial intelligence. Important in catalysis design is the development of new algorithms that give their probabilistic solutions in very short time rather than the conventional deterministic ones in computational chemistry such as molecular orbitals, molecular mechanics and molecular dynamics. Application of neural networks will be useful if it gives a suggestion of the correlation among variables in the future. Four steps will constitute the thus constructed systems: (step 1) selection of appropriate reactions to cope with request; (2) selection of catalyst components; (3) development of industrial catalysts; (4) improvement of commercial catalysts.

1. Introduction

The design of catalysts may be the common goal of catalysis researchers. Many researchers have devoted their own endeavor, passion and talents to arrive at this goal. The vast and diversified achievements in catalysis, and even know-how, have been accumulated in a large number of academic reports, monographs and patent literature. Although this accumulation certainly stores many scientific and engineering facts, it rarely contains knowledge immediately applicable to designing catalysts. Now, it is the computer's turn to go on stage; we can expect the computer, by searching, rearranging and analyzing the accumulation of information to obtain fairly well reconstructed knowledge which is immediately applicable to catalyst design. The application of information science and technology to catalysis is essential to research and development.

On the other hand, we can also expect to utilize the computer's still rapidly increasing capability in digital computation to develop tools for catalyst design. Description around reaction centers over catalysts has been elucidated by appropriate quantum chemistry or molecular mechanics, and even simulation of reaction paths has been estimated by molecular dynamics.

The daily processing of data in the laboratory, such as adsorption, reaction and analysis, has been carried out by laboratory automation and mathematical analysis by using computers. Graphic displays of catalysts can stimulate chemists to imagine what may happen in catalysis in the

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future; this technique may introduce us to virtual reality of the catalyst world.

All the information science, knowledge science, information processing and hardware for them will certainly progress through the 21st century at a tremendous speed responding to social needs, irrespective of the needs of the catalyst researchers. The only thing what we have to do is to watch the trend in the issues close to us and to adopt the progress to the design of catalysts.

My career has been described by Professor M. Misono [1] elsewhere. The digital calculation methods employed in those days, however, were very primitive, although they were among the best ones used in the computer environment at that time. From this background, I dare to state that it is my dream and belief that we will attain a variety of computer-aided catalyst design systems that will be enthusiastically accepted by those working in catalysis. This dream, I believe, will be realized in the near future. The integration of the components of design systems, from Sections 2–7 shown below, will lead to growth in computer-aided catalyst design systems in the true sense, as will be illustrated at the end of this paper.

2. Obtaining and processing of knowledge from literature

Knowledge in this paper denotes a set of well defined pieces of facts and rules, while *information* means just a set of scattered pieces of them. Up to today, we used to collect knowledge, by the intellectual sweat of our brain and brow, from scattered sources such as original papers, monographs, handbooks and patents. In the near future, we will obtain knowledge, with aid of semantic analysis, from full text databases of original papers, monographs, handbooks, patents and specified databases devoted to catalysis. The last databases, bibliographic and factual, however, should be constructed by us, because no one else will take on this task. The construction of databases of a specified discipline such as catalysis consumes an unexpected amount of time and labor. The only possible, or easier and more economical way of construction should rely upon the semi-automatic editing from full-text databases. In the same manner, the accumulation of knowledge and facts and the consequent preparation of knowledge bases and fact databases on catalysis will be achieved with the aid of computer processing. The preparation of a thesaurus on catalysis and catalysts in advance is inevitable as it is requested in semantic analysis; it is worthwhile to note that only catalytic chemists can prepare it.

3. Computational chemistry, simulation and neural network

(1) Computational chemistry: As is well recognized, these calculations usually cost tremendously large cpu time on an extremely high speed computer with a huge memory in order to obtain a deterministic and a precise numerical output. Furthermore, we know that the selections of algorithms or basis sets of orbitals used in even the ab initio quantum chemical calculation very often gives us contradicting results, whereas strangely ab initio implies *without assumption.* For purely scientific interest, this expense may be allowed. From the viewpoint of designing catalysts, however, we should develop a new digital methodology that can supply not only the detailed results that are required in natural science but also the probabilistic information that is essential to design, in expense of minimum cpu time and cost. As another example, researchers of molecular mechanics used to spend a huge cpu time by millions of iterations to obtain the *true* minimum energy structure of simple molecules, under the rather coarse algorithm of molecular mechanics. If we consider the thermal motion of molecules, detection of only a few structures that probabilistically have almost the lowest energy will suffice for design; design is a discipline in *artificial science* in contrast to *natural science.* In general, the computation time required in calculating N molecular orbitals in ab initio MO is reportedly proportional to N^5. The desire of chemists to solve larger

systems has usually exceeded the increase in the capacity of the hardware. Therefore, catalytic chemists themselves should develop new computer programs for catalysis, because we need only the crucial data of enough large systems involving the catalyst surface within shorter time, overcoming the oath of magnitude.

(2) Simulation: Dynamic behavior of molecules in caves of zeolitic catalysts is a beautiful example of artificial science elucidated by simulation. The requirement of higher computation speed is severe in this case, too, if a real-time representation is expected. The behavior of molecules and surface atoms is more probabilistic than that of static molecular models. Progress in the approximate calculation algorithm seems to be inevitable.

(3) Neural networks: Neural networks can supply excellent answers to the query that is hardly solved by multivariant regressions [2]. But, at present it is still an art. Development of the analysis of the network to convert it from a black box to correlation equations is desired. The correlation may derive new concepts or theories concerned with the query.

4. Laboratory automation and laboratory office automation

(1) Laboratory automation: In infrared spectro analysis, it has been popular to assign automatically the observed wave numbers and consequently to determine the structure of given compounds with the aid of an expert system. In the same way, screening of new catalysts including optimization of reaction conditions may be carried out by an automated reaction system. Further, automated systems for the preparation of a series of new relevant catalysts, which has never been reported, may be developed.

(2) Office automation: Laboratory notebook systems will become popular in the catalysis laboratory very soon. More fascinating tools are awaiting our use in catalysis laboratories. These are multimedia systems. Today, it is usual for us to consult with dictionaries or encyclopedias on a CRT that are stored in a CD-ROM during preparation of a manuscript with a word processor on the same personal computer. In the near future, we can expect to put the multimedia to practical use when we are obtaining new knowledge, thinking about novel hypotheses, planning efficient experiments and writing a report. In your office, it will become a common occurrence to collect and look around the related materials in wide windows on a large screen display simultaneously. The materials may consist of your laboratory note book, the condensed description of relevant literature from a catalysis-specified database, some sections of related literature from full-text databases and so on. If requested, a computer graphic may appear at the corner window, prepared under the conditions which you desire. Thus you can continue your thinking without interruptions caused by finding out appropriate materials from scattered sources. It might be a daydream to you; but, if we are continuing to make preparations for the multimedia age, it can be a real dream.

5. Image processing and computer graphics

(1) Computer graphics in catalysis: Computer graphics has already been demonstrated to be an important tool in the study of catalysis, as it can help us to realize what is happening over the catalyst. Two problems, however, remains: the soundness of assumptions and algorithms to prepare it and the cpu time to draw a frame of graphics. We are responsible for the former problem, whereas the progress made in the relevant hardware and software will solve the latter one. The only way to verify the soundness is to compare the conclusions derived from graphics with those of experimental results. A further difficult issue, under these circumstances, is the description of the reaction over non-crystalline or non-zeolitic catalysts that are more frequently used as a catalyst.

(2) Virtual reality of the catalyst world: This might be a daydream even in the near future,

because an extremely quick algorithm as well as a nearly fictional hardware with a large battery of parallel processors are indispensable to realize it. The fiction is as follows: the development appropriate manual manipulation of reacting molecules over a catalyst guided by the feeling of the chemist's fingers, that communicates the affinity between molecules and the catalyst surface, may result in better configuration over the catalyst. The system has to calculate some reasonable reactivity index of the reaction in real-time. This device would give us a better understanding of catalysts and very helpful to design new catalysis.

6. Expert system for designing catalytic reaction and catalysts

An expert system denotes a computer program package that solves the problems on diagnosis, analysis or design. There exist two major distinct principles in constructing expert systems. Case- or information-oriented systems will induce the solutions to the query from the relevant information, mainly depending on the database of facts, whereas logic-oriented systems will deduce the solution from databases and empirically obtained knowledge. The former are liable to give realistic solutions as they are derived from actually observed facts, although novel solutions are hardly never obtained as the processing generally relies on realistic data. In contrast, the latter sometimes give novel solutions because they employ the deduction of facts by logical processing of chemical logic, that is, the scattered pieces of empirical or theoretical rules, although the solutions very often contain unrealistic ones. A recent trend of development in expert systems in chemistry aims at the integration of logic-oriented and case-oriented systems.

In addition, expert systems have been classified into two sections from the viewpoint of the algorithm used: one is written by a procedure language such as FORTRAN, while the other is carried out by predicative logic with associated knowledge bases as stated in Section 7. In the fields of chemistry and related materials, computer-aided molecular design systems, such as the design of medicines, and the reaction design system of organic syntheses have been extensively developed with procedure languages and have been appraised to some extent. These systems in general, however, do not treat the design of even homogeneous catalysts. The design of heterogeneous catalysts and catalytic reactions has thus been left almost undeveloped.

7. Knowledge-base systems and artificial intelligence

Artificial intelligence (AI) has a rather vague definition, whereas knowledge-base (KB) systems may be defined as expert systems that have knowledge bases storing rules and facts and are processed by predicative logic with an ad-hoc sequence of processing. Expert system are rarely constructed for design with the use of only knowledge-base systems. Rather, KB systems may be merged in a program package written in procedure languages, and the former solves only the appropriate issues in the whole system. An example of the appropriate issues may be the determination of the reaction mechanism of a query reaction by interpolation or extrapolation or by bold analogy both from verified mechanisms of the existing reaction. We can add or revise knowledge, that is, rules and facts, to a KB system more easily as compared to a program written in procedure languages; from this viewpoint, the KB system is suitable for designing catalysis and catalysts.

We are forced, however, to include ambiguous and fuzzy knowledge to a KB on catalysis, which is unavoidable in this field. Nevertheless, the processing of knowledge of this kind is inefficient at the present stage of progress of KB systems. In addition, the collection of information from which we expect to find knowledge, and the rearrangement of this knowledge in a proper description form in a KB was revealed in our experience to need unexpectedly large efforts and time of expert chemists, because the information in the literature

has never been described with any thought of using it in a KB. Therefore, I must confess that collaboration among chemists to share the burden, including the preparation of a thesaurus, among experts, and progress in more versatile shells, or editing and executing programs, of KB systems are requested to construct a widely applicable KB system for designing heterogeneous catalysts. Completion of a KB system in catalysis, however, allows us to think that we will find the missing links in the logical explanation of catalysis, as the procedure in a KB system can strictly pinpoint ambiguous or logically-lacking steps in the logic.

8. Computer-aided catalyst design systems

8.1. Step 1: selection of a catalytic reaction to produce a requested compound, to suppress emission or to cope with environmental problems

In the field of medicine syntheses, many systems have been developed to design antithesis or retrosysthesis paths from the target molecule to some combination of starting molecules. In the field of catalytic process chemistry, however, we need to construct another approach. Our very old report [3] may be a reference, where a large number of reaction paths are logically proposed that satisfy given thermodynamic limitations and crude profit by selecting combinations of raw materials, or molecules, from the given material list.

8.2. Step 2: selection of a combination of the main catalyst substance, promoters and support

This step has usually been designed using the case-oriented way of thinking. As much information, however, has been accumulated in this step, logic-oriented design systems can be constructed to find novel combinations of the main catalyst, promoter and support. I proposed an outline of a computer program package to design a catalyst in 1972 [4]. The idea was dependent on the virtual mechanism proposed by Dowden [5,6] and then followed by Trimm [7]. Hattori and Kito [2] have realized through their collaboration with me a novel system INCAP, combining a procedure-type language with KB-type programs. At first the system logically distinguishes the target reaction and parasitic reactions or side reactions from the constitution of the starting and the product molecule. An example is the partial oxidation of ethyl benzene to produce styrene. At first, the reaction mechanisms of all constituting reactions are determined by case-oriented logic. Next, the characteristics of the proposed catalysts, their acidity and acid strength, and their tendency to partial oxidation and deep oxidation, are determined with a logic-oriented KB system, to accelerate the target reaction whereas the parasitic reactions are suppressed. Combinations of catalyst components are so selected as to satisfy the required characteristics. In this phase, the acid strength of an unknown mixture of the main catalyst, SnO_2, and a promoter was estimated by mathematical calculation at an earlier stage of development and was carried out with a neural network at a later stage. This approach may be applicable to other areas of catalysts. However, the work and time needed to construct this KB system far exceeded our expectations.

8.3. Step 3: development of industrial catalysts

After some candidate catalysts were found by logic and their characteristics were verified experimentally, Step 3 begins. The key jobs in this step are the preparation of catalysts bearing satisfactory activity and selectivity, the determination of reaction conditions with the aid of reaction engineering, and finally the estimation of catalyst lifetimes. At present, these endeavors rely upon the expertise and experience of researchers, as well as human wave tactics. Practically, the behavior of research and development in this step will hardly undergo any significant change in the future. The laboratory office automation mentioned above, however, may accelerate experimental design, and the updated laboratory automation will save time

and labor. An expert system for the diagnosis of subtle problems in experiments may be consulted during experiments. It will be desirable to develop an expert system for catalyst preparation that is case-oriented rather than logic-oriented judging by the present situation of the study on preparation. The goal of the system is that it will be able to suggest to us, by some logical consideration, the selection of raw materials including an appropriate support, and methods of preparation of the slurry, drying, calcining and manufacturing. Note, however, that constructing fact databases on preparation procedures is a prerequisite of the system.

8.4. Step 4: improvement of commercial catalysts

This step has the character of severely challenging patents taking up the accumulated expertise as arms. One of the modest suggestions is the utilization of case-oriented expert systems, the details of which are out of my scope. The other is trials of neuro networks to find out the roles of catalyst components. Possibly, it may elucidate the roles of a variety of constituents of catalysts in patents.

9. Conclusion

This paper describes, in a general way, my dreams for the construction of computer-aided design systems for catalysis and catalysts. These dreams were initiated around a quarter of a century ago but still remain in their infancy. I believe the essential part of the ideas described here will certainly be realized in the near future; rapid progress in the computer environment in a wider sense and the expected benefits that this will bring to catalytic chemists will make my dreams come true.

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