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A short note on the work of Professor Yukio Yoneda in the application of computer chemistry to studies of catalysis

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A unit for research and education (chair or 'Koza') on industrial catalytic chemistry was started in 1962 in the Department of Synthetic Chemistry of the Faculty of Engineering, The University of Tokyo. The department was one of many new ones created in Japan according to the new policy of the government to promote the rapid growth of industry and the economy. Professor Yoneda was the first professor of that Koza, and was succeeded by Misono in 1983 after his retirement. The 60's were a very prosperous period for the research and development of catalysis in Japan; many new Koza's on catalysis were launched in universities during this period.

Being keenly aware of the necessity of 'designability' in engineering sciences, Professor Yoneda emphasized from the beginning of his study on heterogeneous catalysis the importance of catalyst design, and attempted to establish the methodology for it based on the quantitative correlations between the chemical properties of catalysts (and reactants) and catalyst performance. Early studies were devoted to the measurement of the distribution of acid strength and oxidizing ability on the catalyst surface and to correlating these measurements quantitatively with the catalyst performance. The concept and achievements which he made in this direction were given as a plenary lecture at the fourth ICC meeting held in Moscow (1968) [1] and published in a series of papers under the general title of Linear Free Energy Relationships (LFER) in Heterogeneous Catalysis (1965–1978) [2]. This is based on the assumption that the free energy of activation, $\Delta G^{\ddagger}$, is linearly correlated with the free energy of reaction, ΔG, (kinetics vs. thermodynamic correlations). The changes of free energies consist of those of catalytic properties (δ_c) and the reactivities of reactant molecules (δ_R).

$$\delta \log k(=\delta\Delta G^{\ddagger}) = \delta\Delta G \qquad (1)$$

He expected that these correlations would predict quantitatively the optimum catalysts for desired reactions, as well as the rate and selectivity of the reactions. For this purpose, he utilized computers to provide a numerical analysis of experimental data, e.g., regional analysis for LFER, and quantum chemical calculation of reactivity indices.

From the late 60's, he started intensively working with computers in chemistry. Table 1 lists the possible applications of information chemistry for studies of catalytic chemistry, according to Professor Yoneda [3]. His work covers A–D in Table 1. He placed more stress on the computerization of 'chemical logic' than on 'mere' computation.

The plenary lecture of Dr. Dowden presented at the fourth ICC meeting motivated Professor Yoneda to extend his ideas further and he proposed a flow chart for catalyst design in an essay on "the System of Catalyst Design" in October,

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Table 1
Information chemistry for studies of catalytic chemistry

A	Information retrieval (data retrieval; database)
B	Computational chemistry; simulation
C	Laboratory automation
D	Computer graphics
E	Molecular computation; reaction design; catalyst design

Table 2
Computer programs for catalyst design

HYPO [4] (1972): Given the raw materials and products, all possible reaction routes are automatically generated based on the stoichiometry of the reactions, together with equilibrium conversions and values added (optional)

THEDIC [5] (1972): (EMPRIC/EROICA (Version 4, 1984)): Retrieval and estimation of thermodynamic data of organic compounds as well as several physical properties such as vapor pressure and critical constants

GRACE [6] (1979): All possible reaction routes (networks of elementary steps) are generated, given the reaction equations and active sites

STERIC (Version 4, 1983): Atomic coordinates are retrieved and/or estimated from the molecular formulae (CHEMO input)

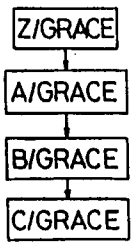

Fig. 1. Flow chart of GRACE.

1968. The chart comprises: (1) input of target molecule, raw materials, etc., (2) derivation of virtual mechanisms, (3) estimation of reaction rates, (4) experimental tests, and (5) evaluations. Since then he has published several computer programs which constitute the elements of the catalyst design flow chart listed in Table 2. He wrote a 600 page book entitled "CHEMOGRAM" in which the 'CHEMO input' system together with THEDIC is fully described (Vol. 1, Maruzen, 1972). The CHEMO input was invented for unambiguous and obvious representation of molecules for computer manipulation. The 'CHEMOGRAM' is the computer program package for chemical logic, according to his expression.

The flow chart of GRACE is shown in Fig. 1. An example of the output from A/GRACE is illustrated in Fig. 2. The associative and dissociative mechanisms are derived. A program for finding the structure of activated complexes of simple reactions by using quantum chemistry and linear programming [7], and ordinary quantum chemical analysis of catalyst structure and catalytic reactions were also attempted in this period.

In 1984–85, he organized a study group for the construction of a prototype database for C_1 chemistry, named CATD. After this, he became a general coordinator of a national study group (1985, Innovative Chemical Industry by Computerization; MITI) and for a national project on knowledge databases (1986–1990, Knowledge Base System for Chemical Design; Science and Technology Agency). One branch of the project was about catalysts, of which he was also the leader. Professors Hattori (Nagoya University), Kito (Aichi Institute of Technology), Miyamoto (Tohoku University) and Okada (Kwansei Gakuin University) were among the active members.

In addition to the study of the use of computers in chemistry, his research group also carried out experimental work on heterogeneous as well as homogeneous catalysis. He further attempted to construct an automated reactor for catalytic research [8]. Professor Y. Saito (Science Uni-

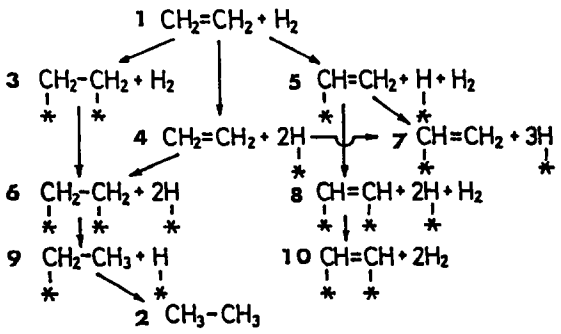

Fig. 2. Reaction routes of catalytic hydrogenation of ethylene.

versity of Tokyo, Prof. Emeritus of the University of Tokyo), Professor T. Uchijima (Tsukuba University), Professor H. Arai and Professor I. Mochida (Kyushu University), Professor N. Mizuno, Dr. J. Take, Professor T. Okuhara and Professor M. Misono (University of Tokyo) are among his former students and staff.

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