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Integration of Catalyst Activity Pattern (INCAP) Artificial Intelligence Approach in Catalyst Design

Tadashi HATTORI,* Shigeharu KITO,+ and Yuichi MURAKAMI
Department of Synthetic Chemistry, Faculty of Engineering,
Nagoya University, Chikusa, Nagoya 464-01
+Department of Industrial Engineering, Aichi Institute of Technology,
Yagusa, Toyota 470-03

A novel knowledge representation scheme for the utilization of activity pattern was developed as a part of a prototype expert system for catalyst design. The result given by the system agreed well with published results, indicating the feasibility of the artificial intelligence approach in the design of catalysts.

An Artificial Intelligence approach has attracted much attention, because it can give a solution of the problem which can not be solved by a logical approach.[1] The design of catalysts[2,3] is one of such problems, and, therefore, it can be expected that an expert system, one of applications of artificial intelligence, will be a great help to the design of catalysts.[4]

In the present study, we have developed a prototype catalyst design expert system, INCAP (Integration of Catalyst Activity Pattern), consisting of conventional knowledge based system and a novel knowledge representation scheme which also has been developed in the present study. The oxidative dehydrogenation of ethylbenzene is taken up as a model target reaction, because the result given by the system can be easily verified by the works on catalyst design conducted by us.[5-7] The object of design is set up as the selection of a catalyst component. In the present letter, the system is briefly introduced laying emphasis on the novel knowledge representation scheme.

The basic strategy of catalyst design in the present system resembles to those of Dowden,[8] Trimm,[3] and Murakami.[2] As illustrated in Fig. 1, the INCAP recommends catalyst components through the estimations of (1) reaction mechanism of target reaction, (2) catalytic functions required for the target reaction, (3) plausible side reactions, (4) unfavorable catalytic functions leading to the plausible side reactions, and (5) catalyst components by integrating activity pattern data.

The knowledge required for the first four estimations could be encoded in a knowledge base by using the conventional production system and frame system.[1] For the estimation of reaction mechanism of target reaction, i.e., (1) in Fig. 1, the adsorbed intermediates of reactant and oxygen and the surface reactions of 20 catalytic oxidations were encoded in the frame system. For example, "π-benzyl", "O^- ion", and "abstraction of β-hydrogen of π-benzyl by O^- ion"[9] were given for

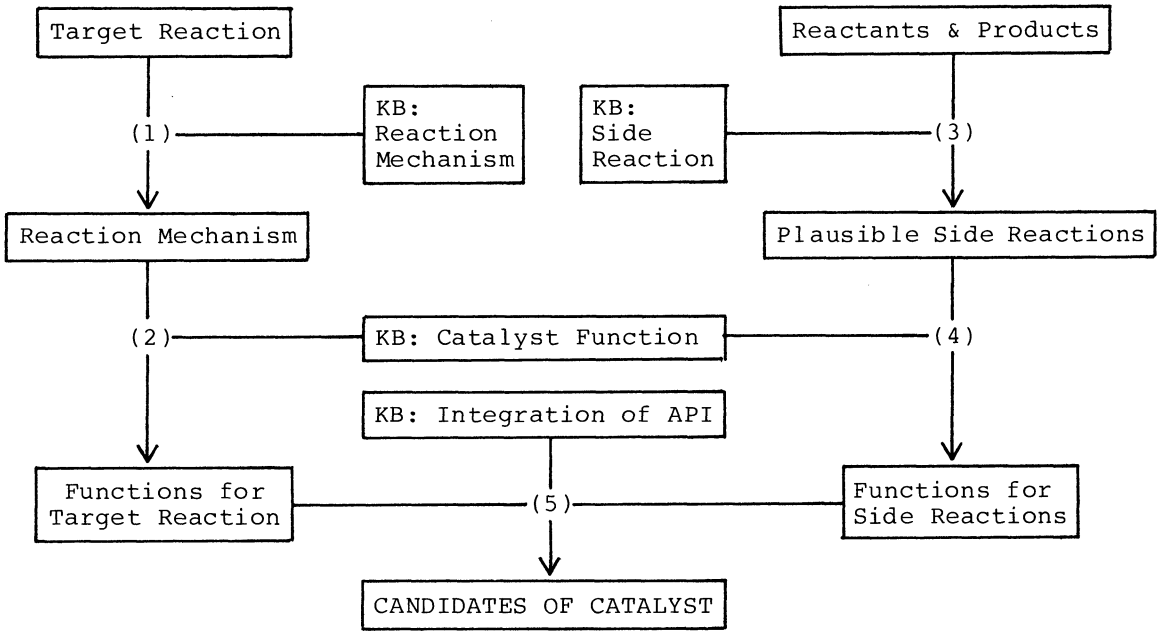

Fig. 1. Overview of INCAP. KB, Knowledge base.

values of the adsorbed intermediates of reactant and oxygen and the surface reaction, respectively, in a frame of the oxidative dehydrogenation of ethylbenzene.

The estimation of plausible side reactions, (3) in Fig. 1, was conducted by using production rules concerning the conditions of reactants leading to the catalytic reactions of alkylbenzenes, such as "If the reactant is hydrocarbon consisting of a benzene ring and an alkyl group with more than two carbon atoms, then the dealkylation is plausible", "If the reactants are hydrocarbon and oxygen, then the complete oxidation is plausible", and so on.

The catalytic functions required for each step of target reaction and for the plausible side reactions were estimated in (2) and (4) in Fig. 1 by using production rules regarding the catalytic functions such as "If the reactant is adsorbed as π-benzyl, then the acid property of medium strength and the basic property are required", "If the reaction is dealkylation of alkylbenzene, then the strong acid property is required", and so on. It should be remarked that the knowledge for the estimations (3) and (4) are not specific to the oxidative dehydrogenation of ethylbenzene, and can be applied for the estimation of the plausible side reactions accompanying the reactions of aromatic hydrocarbons.

Although the knowledge required for these estimations could be conventionally represented, the conventional system could not be used for the representation of knowledge required for the selection of catalyst components on the basis of the estimated catalytic functions. One can easily imagine that the oxides which have favorable functions but not unfavorable functions may be good catalyst components. If the oxides could be clearly divided into the two groups, the knowledge could be conventionally represented as "If the catalytic function is the acid property, then the followings are recommended as the catalyst component: A, B, ...".

However, it seems difficult to distinguish the oxides having a certain function from another group of oxides, because the strength of the catalytic function changes continuously from oxide to oxide rather than discontinuously, as frequently expressed by the activity order or activity pattern, such as "Co_3O_4 > CuO > MnO_2 > NiO > Fe_2O_3 > ZnO > Cr_2O_3 > V_2O_5 > TiO_2" for the oxidation of hydrogen. In the present system, a newly developed knowledge representation scheme was used to estimate overall performance of oxide as a catalyst component by integrating various activity pattern data.

The activity pattern was divided into ten sections, and a numerical value from ten to one named API (activity pattern index) are allocated for each section. 20 sets of activity patterns were collected for the activity of complete oxidation, the activity of partial oxidation, the acid property, and the basic property. Some physico-chemical properties, such as the heat of formation of oxide divided by the number of oxygen and the electronegativity of cation, were also used as the activity pattern data. Thus quantified activity patterns can be widely used beyond the scope of oxidative dehydrogenation of ethylbenzene.

The API data were integrated as shown in Fig. 2. The individual API data was multiplied by an evaluation factor (Ef). The Ef expresses the empirical knowledge about the utilization of activity pattern. The numerical value from ten to one also was allocated for the Ef of each set of API by using production rules regarding the similarity of object reaction (target reaction or side reaction) to the reaction giving activity pattern data, the difference between observed value and estimated value, and so on. Further, the negative value was allocated for the Ef of the unfavorable function also by using production rules. Then, an averaged API was calculated for each function, and it can be multiplied by a weight factor for each function if necessary. Finally, the summation of the averaged API's for all the functions is calculated to give an integrated API expressing the estimated overall performance of the oxide.

The whole system was applied to the selection of promoter oxide to the primary component, SnO_2, for the oxidative dehydrogenation of ethylbenzene. It should be noted that, generally speaking, the selection of secondary component is more difficult than that of primary component.

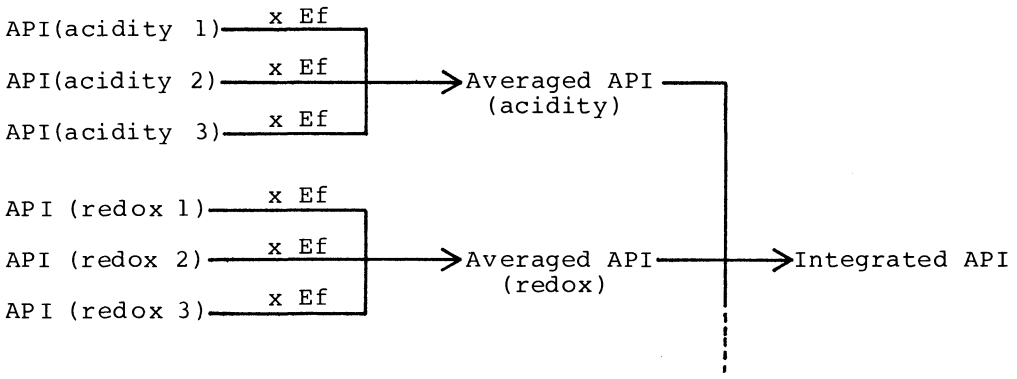

Fig. 2. Schematic diagram of integration of catalyst activity pattern. API, activity pattern index; Ef, evaluation factor.

The following catalytic functions were estimated through (1) to (4) in Fig. 1: Positive functions leading to the target reactions are the acid property of medium strength, the basic property, and the oxidation activity. Negative functions leading to the plausible side reactions are the strong acid property, strong oxidation activity, and partial oxidation activity. The estimated functions agreed well with those described in the literature.[5,7,9]

Then, the promoters to SnO_2 as a primary component were selected through (5) in Fig. 1 on the basis of thus estimated catalytic functions by using 20 sets of API data. The INCAP selected the following promoters out of 50 oxides: P_2O_5, Nb_2O_5, MoO_3, ZrO_2, and SiO_2. Among these, SnO_2-P_2O_5[5] and SnO_2/SiO_2[7] catalysts have been experimentally verified to be selective catalysts for the present reaction. SnO_2-MoO_3 also gave fairly high selectivity.[10] The good agreement with the experimental results indicates the feasibility of newly developed knowledge representation scheme for the utilization of activity pattern. Further, it should be stressed that the INCAP selected some unexamined oxides, such as Nb_2O_5 and ZrO_2, indicating the possibility to give the solution on an extrapolation of the established knowledge. The performances of these promoters are going to be examined experimentally.

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References

1) "Handbook of Artificial Intelligence, I and II," ed by A. Barr and E.A. Feigenbaum, William Kaufmann, Los Altos (1981, 1982).
2) Y. Murakami, "Shokubai Sekkei," ed by the Soc. Chem. Eng. Japan, Maki-Shoten, Tokyo (1981) p.1; "Shokubai Sekkei," ed by Catal. Soc. Japan, Kodansha, Tokyo (1985), p.1.
3) D.L. Trimm, "Design of Industrial Catalysts," Elsevier, Amsterdam (1980).
4) R.B. Alcantara, A.W. Westerberg, E.I. Ko, and M.D. Rychener, Comput. Chem. Engng., 11, 265 (1987).
5) Y. Murakami, K. Iwayama, H. Uchida, T. Hattori, and T. Tagawa, J. Catal., 71, 257 (1981); Appl. Catal., 2, 67 (1982).
6) T. Tagawa, K. Iwayama, T. Hattori, and Y. Murakami, J. Catal., 79, 47 (1983).
7) T. Tagawa, S. Kataoka, T. Hattori, and Y. Murakami, Appl. Catal., 4, 1 (1982); T. Hattori, S. Itoh, T. Tagawa, and Y. Murakami, "Preparation of Catalysts IV," Elsevier, Amsterdam (1987), p.113.
8) D.A. Dowden, Chem. Eng. Progr., Symposium Series, 63(73), 90 (1967); D.A. Dowden, C.R. Schnell, and G.T. Walker, Proc. IV Int. Congr. Catal., 1968, 201.
9) T. Tagawa, T. Hattori, and Y. Murakami, J. Catal., 75, 56, 66 (1982)
10) M. Araki, I. Nishimura, T. Hayakawa, K. Takehira, and T. Ishikawa, Shokubai, 14, 47p (1972).

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