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Recent advance in catalysis for solving energy and environmental problems

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Abstract

Recent advances in catalysis for solving the energy and environmental problems are summarized. For these purposes, rapid conversion and selective reaction even under conditions deviating extremely from reaction stoichiometry must be indispensable requisites. In order to realize these requisites, changes in the state of catalyst surface during the reaction were studied, and the catalyst structures on which the optimum reaction performance occurs were determined. An ultra-rapid reforming of methane to syngas with a space–time yield (STY) of 25 000 mol/l h was achieved by using a Rh-modified Ni–Ce_2O_3–Pt catalyst in which the Rh played the role of portholes for hydrogen spillover and prevents coke deposition on the catalyst surface. As a result, a stable state of the catalyst and the high reaction rate were exhibited. A new catalyst composed of Cu–Zn–Cr–Al–Ga oxides modified with supported Pd exerted a high activity with a high STY of methanol, 6700 g/l h. The catalyst components, Pd and Ga, controlled the reduction state of the catalyst surface by their role on normal and inverse spillover of hydrogen, respectively. The methanol thus produced was then totally converted selectively on a metallosilicate catalyst containing Ga or Fe into an aromatics-lean gasoline using an STY of 1860 g/l h. Finally, non-linear reaction mechanism is used to explain the elimination of NO on metallosilicate catalysts under O_2-excess conditions. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Rapid methane reforming; Catalytic combustion; Highly effective methanol synthesis; Highly effective gasoline synthesis; Rh-modified Ni-based composite catalyst; Uniform gelation method; Metallosilicate catalyst

1. Introduction

Adsorption and catalysis attracted particular attention from the last century to the beginning of this century as the most essential relationship to understand catalytic phenomena. In the early part of this century, adsorption phenomena could be systematized at the beginning by mono-layer adsorption theory proposed by Langmuir. The Langmuir–Hinshelwood reaction mechanism then occupied a central part in the understanding and analysis of catalytic reaction kinetics. This theory is applicable under conditions which afford linear phenomena, in which adsorption rates of reactants are always greater than surface reaction rates or consumption rate of those reactants.

However, in recent years, never previously faced problems have appeared to be solved by the applications of catalysts. These include the elimination of toxic materials contained in exhaust gases at extraordinary low concentrations, mitigation of carbon

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dioxide come from fossil fuel combustion, and new routes for the syntheses of energy sources and raw materials for chemical industries from alternative resources. These tasks need much higher conversion rates than the conventional conversion rates for the production of desired compounds in common industrial process, and the selective elimination of very dilute undesirable compounds in the presence of high concentrates retarding compounds such as oxygen and water in the exhaust gas from diesel engines and lean-burn facilities of large factories.

Merely the use of conventional linear phenomena in catalysis cannot respond to these unusual demands, and in order to realize novel phases in catalytic reactions, it should be necessary to understand and control non-linear phenomena which can be observed in catalytic reactions under these special circumstances in both the catalyst structure and the reaction conditions. In this paper, some recent advances in catalysis for solving energy and environmental problems are presented using typical rapid and effective catalytic reactions as examples.

2. Dynamic behaviors of catalytic surfaces and its control by applying spillover effects of adsorbed species

2.1. *Changes in the oxidation states of metal oxide catalysts and their effects on the reaction rate*

The oxidation–reduction state of a metal and/or metal oxide catalyst may be changed by the reaction environment, such as the reaction-gas composition, temperature, pressure, conversion, and retardation by the reactants and products. However, in conventional analysis of catalytic surface reactions, these practical factors, which often belong to non-linear phenomena, are commonly avoided or neglected for the sake of simplicity. The state of a catalyst surface during reaction under differential reaction conditions and the low temperature conditions such as those used in a circulating reactor system, do not reflect directly the real state of the catalyst surface under the practical reaction conditions in which the reaction of interest progresses actively. In catalytic combustion, exhaust gas purification, reforming of methane, decomposition of methane to syngas, and methanation of carbon dioxide for example, only the total conversion of the target reactants is significant, and intermediate conversions are of almost no significance. Moreover, if the oxidation state of the catalyst surface changes very easily, it is rather difficult to maintain the intermediate conversion level stably.

As a typical example, the combustion of methane on a supported Pt catalyst was investigated. A ceramic foam prepared by Bridgestone was used as the catalyst support; this ceramic foam was prepared by impregnation of a cordierite slurry into urethane, followed by burning off the urethane and calcination at a high temperature. The open channels of foam shape were connected three-dimensionally. The dimension of the foam was about 0.25 mm. Since the surface area of the ceramic foam was very small (0.10 m^2/g), this was coated with alumina-sol before supporting the catalyst material. The support was cut into an appropriate size and dipped into an aqueous suspension of 9.9 wt% alumina-sol. The alumina-sol was produced by Nissan and had a BET surface area of 305 m^2/g in the dry state. The excess of this solution was separated by centrifuging at 2000 rpm. This sol-supporting procedure was repeated four times to obtain a support with 3.3 wt% of alumina loading. The increase in BET surface area was 12 m^2/g. The alumina-precoated support was immersed in an impregnating aqueous solution of H_2PtCl_6 with a concentration of 8.7 wt%, and it was put in a centrifuge (2000 rpm) for 30 s. After drying at room temperature for overnight, the salt was decomposed thermally. The catalyst was then reduced at 400°C for 30 min in a flow of 20% H_2 in N_2.

A continuous flow reaction apparatus was used under atmospheric pressure. The catalyst, cut into 10 mm diameter and 50 mm length, was packed in a quartz tube of 11.0 mm inner diameter. The reaction gas mixture of methane, oxygen and nitrogen was allowed to flow with a space velocity (SV) of 1400 h^{-1}.

The composition of reaction gas mixture was varied as follows: (a) excess oxygen condition (CH_4 6.5%, O_2 19.6%, N_2 balance), (b) stoichiometric condition (CH_4 6.5%, O_2 13.0%, N_2 balance), and (c) excess methane condition (CH_4 38%, O_2 13.0%, N_2 balance) [1]. The hysteresis of conversion was measured by elevating the reaction temperature with a constant heating rate and then decreasing it in the same way. The temperature was raised at a constant rate of 10°C/min until the

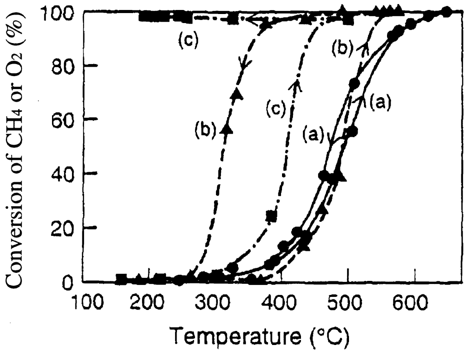
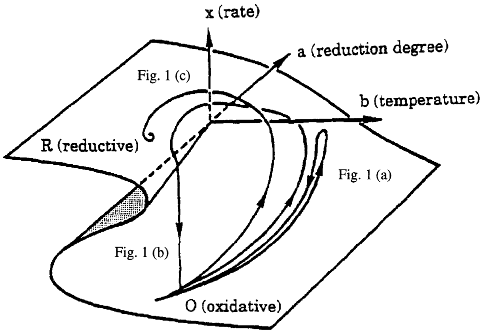


Fig. 1. Different hysteresis behavior of the catalytic activity observed by the forced-oscillating reaction method. Catalyst: 0.33 wt% Pt supported on a wash-coated ceramic foam carrier; SV: 1400 h^{-1}. (a) Oxidative catalytic conditions; (b) stoichiometric conditions for complete combustion; (c) reductive catalytic conditions. In the case of (a) and (b), the y-axis expresses the conversion of methane. In the case of (c), the y-axis expresses the conversion of oxygen.



Curved surface corresponding
to the cusp catastrophe
$f \equiv x^3 + ax + b = 0$

Fig. 2. Cusp catastrophe depiction of the hysteresis appearing in the catalytic combustion of methane with different reaction conditions corresponding to Fig. 1(a)–(c).

methane or oxygen conversion reached 100%, and the temperature was then lowered. The temperature was measured by thermocouples contacted at the centers of both the inlet and outlet of the catalyst. The reaction gas was analyzed by gas chromatographs, and/or an infrared analyzer for continuous analysis of CO_2. This measurement method was named by us as the forced-oscillating reaction method, by which hysteresis of oxidation–reduction state of the catalyst surface can generally be evaluated irrespective of the type of the catalyst. With an oxygen excess, a minor hysteresis was observed as shown in Fig. 1(a). At stoichiometric conditions, the descending line was shifted significantly to the lower temperature side (Fig. 1(b)), and this tendency increased with increasing methane in the feed. With CH_4 excess catalytic combustion of methane maintained an even conversion at about 150°C totally consuming the oxygen (Fig. 1(c)), even when the heat supply from the electronic furnace was cut off. This kind of hysteresis reflects on the oxidation–reduction states.

In order to understand quantitatively these non-linear phenomena, the lines shown in Fig. 1(a)–(c) were fit to the curved surface corresponding to the cusp catastrophe, as shown in Fig. 2. It is well described in this figure that the hysteresis does not occur under oxidative catalytic conditions; the sudden decrease of activity occurs at intermediate reaction condition, and a highly active level can be maintained under the reductive catalytic conditions.

This highly active level can also be created by the aid of catalytic structure. Fig. 3 shows a typical example which could be realized by a combination of a very diluted Rh catalyst with a Cu catalyst supported on an alumina wash-coated ceramic fiber shaped to a plate shape of 1 mm thickness. A single ceramic fiber has a diameter of about 5 μm and the void space of the

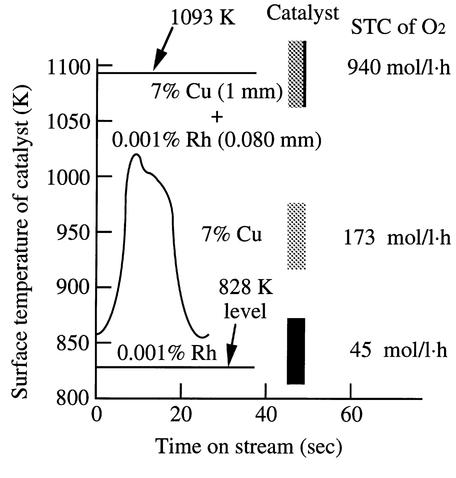


Fig. 3. The effect of the contact of a Rh catalyst with a Cu catalyst on combustion of a city gas. Gas composition: city gas/air=1/1.56.

support was 88% [2]. The surface temperature of the catalyst was measured by a Willamson Infrared thermometer M-1236. The focal point at 0.76 mm was adjusted to be at the center of the circular catalyst plate, and the temperature was recorded for every few seconds. The reading accuracy was ±1.5°C. The composition of city gas was 46% H_2, 22% CH_4, 5% CO, 5% C_2–C_4 hydrocarbons, 12% CO_2, 2% O_2 and 8% N_2. This oscillatory phenomenon in the catalytic activity is ascribed to the role of the added Rh as a porthole for hydrogen spillover towards the main catalyst component, Cu oxide, and maintains the surface of Cu oxide in a fully reduced state.

By the application of this principle to accelerate catalytic activity, extraordinarily highly active catalysts for catalytic combustion, methanation of carbon oxides [3], catalytic decomposition of methanol [4], and catalytic reforming of methane [5] have been developed.

As shown in Fig. 4 for the CO_2-reforming of methane an Rh-modified Ni–Ce_2O_3–Pt catalyst exhibited a very high activity, and the conversion approached near to the reaction equilibrium even at a high space velocity (SV) of 730 000 h^{-1}, corresponding to a very short contact time of the reaction gas of 4.93 m s. The activity of the Rh-modified Ni–Ce_2O_3–Pt catalyst was much larger than the sum of the activities of the component materials, an Rh catalyst and an Ni–Ce_2O_3–Pt catalyst [5]. This means that the modified catalyst involved two-step spillover, i.e., the hydrogen formed was adsorbed on the Rh part very rapidly, faster than on the Pt part, and then the spilt-over hydrogen is abstracted by the Pt part and this is followed by its diffusion towards the major catalyst component, the Ni part. Consequently, the Ni part can maintain a reduced surface state and the rapid reaction can be progressed on it. The role of Ce_2O_3 is not only as the dispersing medium for the Ni component, but also as the transporting medium for spiltover hydrogen.

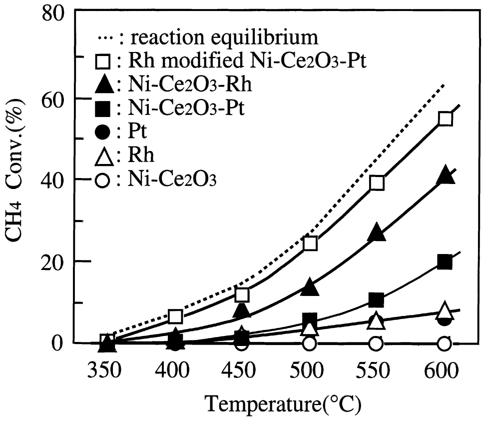


Fig. 4. Effect of Rh modification to a Ni–Ce_2O_3–Pt catalyst used for the CO_2-reforming of methane. Composition of four-component catalyst: 10 wt% Ni–5.6 wt% Ce_2O_3–10 wt% Pt–0.2 wt% Rh wash coated on a ceramic fiber support of plate shape with a 1 mm thickness, involving 86.3% void fraction. The loading of each component in each catalyst was the same as shown in the four-component catalyst. Feed gas: 10% CH_4–10% CO_2–80% N_2, SV (based on the catalyst net volume, excluding the void space of the plate shape) 730 000 h^{-1} or contact time 4.93 m s.

Hydrogen and CO were obtained equivalently by the reaction, and the space–time yields of H_2 and CO obtained at 600°C were 7170 mol/l h at a methane conversion of 55%. Such an extraordinarily high space–time yield had never been realized until joining the spillover effect in the dynamic state of the reactions.

In order to overcome the restriction in conversion of the reaction equilibrium, the combustion of more easily combustible hydrocarbons such as ethane or propane, which are involved in the natural gas, was then combined by adding these hydrocarbons and oxygen while maintaining the concentrations of both CH_4 and CO_2 at 10%, and at the same contact time. The aimed-for reactions to produce only syngas are expressed in reactions Eq. (1) and Eq. (2), which include the generation and consumption of heat by combustion and reforming, respectively.

$$\begin{aligned}&(x+5y)\mathrm{CH_4}+x\mathrm{CO_2}+y\mathrm{C_2H_6}+3.5y\mathrm{O_2}\\&\quad\rightarrow(2x+7y)\mathrm{CO}+(2x+13y)\mathrm{H_2},\\&\Delta H=259x-240y\,\mathrm{kJ/mol}\end{aligned}\tag{1}$$

$$\begin{aligned}&(x+7z)\mathrm{CH_4}+x\mathrm{CO_2}+z\mathrm{C_3H_8}+5z\mathrm{O_2}\\&\quad\rightarrow(2x+10z)\mathrm{CO}+(2x+18z)\mathrm{H_2},\\&\Delta H=259x-375y\,\mathrm{kJ/mol}\end{aligned}\tag{2}$$

In the case of ethane or propane addition to the CO_2-reforming of methane, the catalyst temperature rose abruptly at a very lower furnace temperature (below 400°C), and was maintained at a much higher temperature than the furnace temperature, indicating that by the in situ heat supply due to the combustion, the

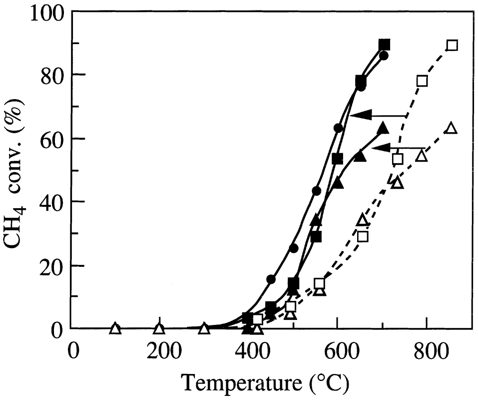


Fig. 5. Change in CH_4 conversion as a function of temperature in CO_2-reforming; influence of reaction gas concentration. Catalyst: Ni–Ce_2O_3–Pt–Rh; SV: 730 000 h^{-1}; CT: 4.93 m s. Reaction gas: (●): 10% CH_4–10% CO_2–80% N_2; (■): 35% CH_4–35% CO_2–30% N_2; (▲): 50% CH_4–50% CO_2. Open symbols and dotted curves: furnace temperature; solid symbols and curves: catalyst-bed temperature.

methane conversion was eventually induced in a very much lower temperature range. Figs. 5 and 6 show the difference between the simple CO_2-reforming of methane and the combination of catalytic combustion of ethane or propane. In the case of the simple CO_2-reforming of methane, the catalyst temperature was much lower than the furnace temperature owing to the large endothermic heat of the reaction. On the other hand, by a combination with combustion, the catalyst temperature was much higher than the furnace temperature due to the in situ heat supply by the heat of combustion. As a result, an extraordinarily high space–time yield of syngas, as high as 25 000 mol/l h was obtained at a very low furnace temperature around 400°C [6,7].

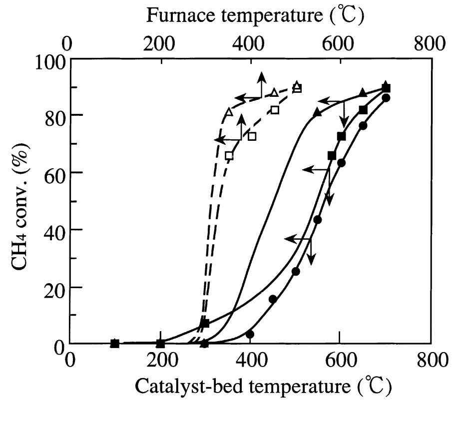


Fig. 6. Influence of the combination of ethane and propane combustion on CO_2-reforming of methane. SV: 730 000 h^{-1}. (○): without addition, 10% CH_4–10% CO_2/N_2; (□): ethane addition, 10% CH_4–10% CO_2–5% C_2H_6–17.5% O_2/N_2; (△): propane addition, 10% CH_4–10% CO_2–3.3% C_3H_8–16.5% O_2/N_2. Open symbols and dotted curves: furnace temperature; solid symbols and solid curves: catalyst-bed temperature; in the case of the simple CH_4–CO_2 reaction, both temperatures coincided.

2.2. *Maintenance of optimum states of catalyst surface under the high reaction rates by a good balance between normal and inverse spillover effects*

As mentioned above, the spillover effect is most effective in extreme reactions such as complete combustion, complete hydrogenation, and decomposition reactions. On the other hand, reactions in which the purpose is to obtain selectively the intermediate oxidation or hydrogenation products, the optimum state of catalyst surface, in case of metal oxide catalyst for example, is an intermediate oxidation state. In order to increase the reaction rate, if the reaction conditions are shifted to more radical ones, the spillover rate of adsorbed species is also markedly enhanced, and the catalyst surface inclines excessively to an undesirable reduced or oxidative state, resulting in a situation in which the selectivity of desired product will decrease and/or the catalyst will be deactivated due to the segregation of catalyst components. In order to maintain the optimum oxidation state of catalyst even under the condition of high reaction rates, a catalyst component having an inverse spillover effect should therefore be combined with a catalyst involving a component exerting a normal spillover effect, anticipating that the rates of reduction and oxidation of the metal oxides of the catalyst show a good balance and a steady state. A typical example [8] is shown below. A Cu–Zn–Cr–Al mixed oxide catalyst was prepared by the uniform gelation method and Ga oxide component, which has the inverse spillover effect was mixed with it. The catalyst was used for conversion of CO_2-rich syngas, and the yield of methanol was found to increase while the formation of CO as a by-product was suppressed. These reaction features could be ascribed to retardation of excessive reduction of the catalyst surface. This was confirmed by a temperature programmed reduction (TPR) experiment. As shown

Table 1
Effect of Ga and Pd combination on Cu–Zn-based catalysts for methanol synthesis from a CO_2–H_2 mixture

Catalyst	Calcination temperature (°C)	Surface area (m^2/g)	Cu dispersion (%)[a]	Reaction temperature (°C)	CO_2 conversion to (%)			MeOH STY (g/l h)
					MeOH	CO	HC	
1	350	114	5.4	250	13.7	2.7	0.0	810
				270	19.6	2.7	0.1	1158
2	600	46	4.3	250	12.8	3.3	0.0	756
				270	21.3	3.3	0.0	1261
3	Pd-modified catalyst 2	38	3.5	250	19.2	3.0	0.1	1230
				270	22.0	3.9	0.2	1410

Composition of catalysts 1 and 2: Cu:ZnO:Cr_2O_3:Al_2O_3:Ga_2O_3=38.1:29.4:1.6:13.1:17.8 (wt%); catalyst 3: modified with 1 wt% Pd to catalyst 2; 22% CO_2–3% CO–68% H_2; 80 atm; SV 18 000 h^{-1}.

[a] Calculated based on N_2O adsorption.

in Table 1, when the catalyst was calcined at a high temperature, the BET surface area decreased to 40% of its original value; however, the reaction activity increased significantly. Furthermore, when a Pd component, which has a hydrogen spillover effect, was added, the activity increased, particularly in the lower temperature range. This effect was also confirmed by TPR measurements. Thus, the Pd-modified catalyst (Catalyst 3) showed a high performance for methanol synthesis from CO_2 and H_2. The space–time yield (STY) of methanol was as high as 1410 g/l h at 270°C. The catalyst was then applied to syngas conversion, and as shown in Table 2, when unprecedented high synthesis rates were achieved. For example, the STY of methanol amounted to 6730 g/l h and the summation of the STY for methanol and dimethylether was 6930 g/l h at 270°C at a pressure of 80 atm.

3. Autocatalysis and its application to rapid catalytic reactions

Catalytic reaction processes, such as the conversion of methanol to hydrocarbons, and the catalytic oligomerization of light olefins involve autocatalysis. The coexistence of a very low concentration of alcohols and higher concentrations of light olefins above certain critical concentrations which markedly enhance the reactions. As a result, an extraordinarily high reaction rate was observed. For example, on an H–Fe–silicate catalyst, propylene fed with a gaseous space velocity 4500 h^{-1}, was almost completely converted to gasoline-range liquid hydrocarbons, and the space–time conversion was as high as 8.09 kg/l h [9]. Moreover, when highly active methanol synthesis catalysts and metallosilicate catalysts

Table 2
Effect of space velocity on the performance of a Ga_2O_3 and Pd-combined catalyst for methanol synthesis from CO-rich syngas

SV (h^{-1})	Temperature (°C)	CO convention (%)	Selectivity to (C-mol%)				STY (g/l h)	
			MeOH	DME	HC	CO_2	MeOH	MeOH+DME
20 400	250	55.0	94.2	6.2	0.4	−0.8	4530	4750
	270	56.3	81.4	15.3	0.9	2.4	4000	4540
	290	52.9	59.6	29.8	1.9	8.7	2750	3750
39 900	250	31.8	99.0	1.3	0.1	−0.4	5380	5430
	270	41.1	95.8	4.1	0.4	−0.3	6730	6930
	290	44.3	87.3	10.5	0.9	1.3	6600	7180

Reaction gas: 30% CO–3% CO_2–67% H_2; pressure 80 atm.

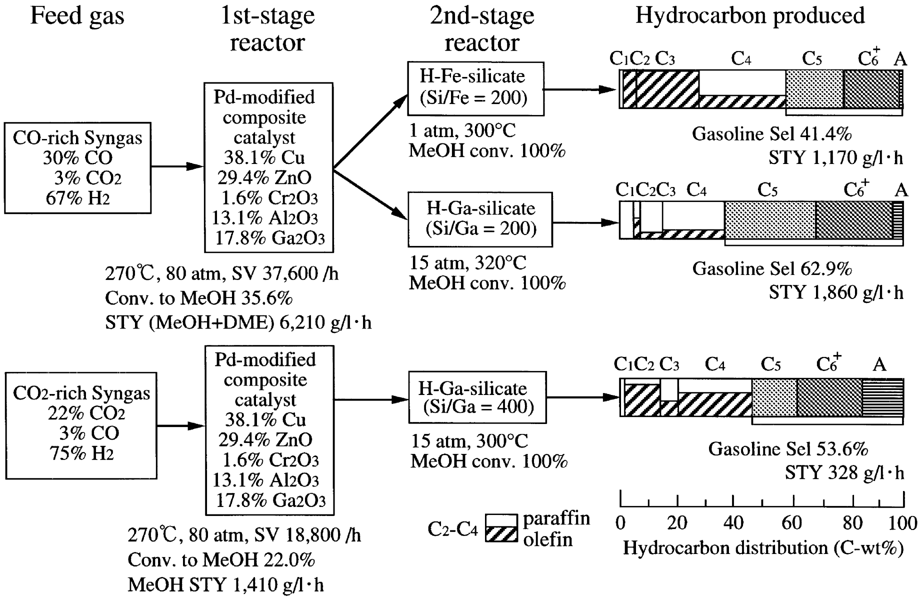


Fig. 7. Gasoline synthesis from a CO-rich and a CO_2-rich syngas via methanol by using two-stage serially connected reactor packed with differently functioning catalysts.

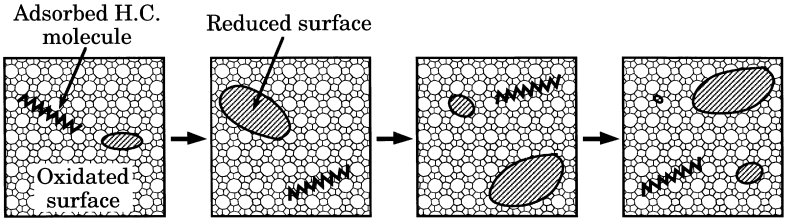
(such as H–Fe–silicate and H–Ga–silicate having MFI-type structure), active for methanol to gasoline conversion, were packed into reactors connected in series, as shown in Fig. 7, gasoline was obtained effectively from either CO-rich or CO_2-rich syngases [10].

4. Application of non-linear catalytic phenomena to the elimination of NO under the extremely deviated conditions like a diesel exhaust gas

The catalytic reduction of NO in diesel exhaust gas has recently been investigated extensively. The key points of this technology are the addition of a small concentration of an appropriate kind of hydrocarbon or oxygen-involving compounds such as methanol and the use of well crystallized silica-rich zeolitic materials as the catalyst support.

Exhaust gases from diesel engine contain components such as O_2, H_2O, and CO_2 which retard the reaction and exist in large excess; on the other hand, both the NO to be eliminated and the hydrocarbon to be added are present in extremely low concentrations compared with the O_2 and H_2O. It seems unlikely that the NO can react with the hydrocarbons selectively by a bimolecular surface reaction as usually considered by adopting a linear reaction mechanism such as a Langmuir–Hinshelwood bimolecular reaction mechanism. In contrast the following non-linear reaction mechanism can rationally explain the experimental observation. Since the catalytic combustion of hydrocarbon molecules can easily cause the local reduction of part of the surface of a zeolitic catalyst, reductive part NO decomposition take place on this part of the surface before it is oxidized by the excess oxygen in an appropriate temperature range. Furthermore, the general experimental observation that at above that temperature, the NO conversion decreases whereas the hydrocarbon can be combusted, can be explained reasonably as follows; in the higher temperature range, the local reduced part which is formed during the combustion of the hydrocarbon is immediately oxidized by excess oxygen and there is no more chance to decompose the NO. This kind of real heterogeneous reaction mechanism, expressed by the sequential reaction equations in Fig. 8, has been proposed by us as the microscopic sequential reaction mechanism (MSR mechanism) [11].

The real heterogeneous reaction mechanism enables us to design a suitable catalyst for practical use from the points view of both hydrothermal stabi-



Oxidised surface

$$\mathrm{CxHy} + \mathrm{nOad}\,[S] \longrightarrow \mathrm{CxHyad}\,(\mathrm{nOad}\,[S]) \tag{1-1}$$

$$\mathrm{CxHyad}\,(\mathrm{nOad}\,[S]) + \mathrm{mOad}\,[S] \longrightarrow \mathrm{xCO_2} + (y/2)\mathrm{H_2O} + (2x+y/2)\,[S] + [(n+m) - (2x+y/2)]\,\mathrm{O\,ad}\,[S] \tag{1-2}$$

Reduced surface

$$\mathrm{NO} + [S] \longrightarrow \mathrm{NOad}\,[S] \tag{2-1}$$

$$2\mathrm{NOad}\,[S] \longrightarrow \mathrm{N_2} + \mathrm{O_2} + 2[S] \tag{2-2}$$

$$\mathrm{O_2} + 2[S] \longrightarrow 2\mathrm{Oad}\,[S] \tag{3}$$

Fig. 8. Conceptional illustration of the microscopic sequential reaction mechanism for the deNO$_x$ reaction under conditions of excess oxygen.

lity and catalytic activity and to adjust the temperature range to around 400°C [12].

5. Conclusions

The dynamic states of a catalyst surface during a reaction were demonstrated using the catalytic combustion reaction as a typical case. Oscillating reactions occurring during this rapid catalytic reaction were controlled by a combination of the catalytic components which can play the role of a porthole for hydrogen spillover. This controlling principle could be applied to many significant reactions related to solving energy and environmental problems. The important function of autocatalysis to realize rapid catalytic reactions was shown in the case of the conversion of carbon oxides to gasoline. The nonlinear reaction mechanism enables one to understand the unique behaviors of reactions occurring in extremely deviating reaction condition such as the elimination of NO in the exhaust gases from a diesel engine.

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