

The catalyst industry of Japan (2)

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The trends of the catalyst industry in Japan are described based on the statistical data published by the Japanese government offices. Three topical items in the recent catalytic technology developments in Japan are described: resid FCC, methylmethacrylate, and emission control for lean-burn engines.

Keywords: shipments of catalysts, supply and demand, import and export, resid FCC catalyst, MMA, lean-burn NO_x

1. Production and shipment of catalysts

The general trend of Japanese catalyst industry from 1992 to 1995 was described in the previous paper [1]. Therefore, as to the production and shipment, the trend from 1995 to 1996 is principally dealt with in this paper. Export and import of catalysts were summarized according to catalyst category in the previous paper. They are summarized by countries as partners in this paper. Besides, the items that were not described in detail in the previous paper are taken up in this paper. Table 1 shows production and shipment of catalysts from 1993 to 1996 in Japan. The distinctive feature of the catalyst industry in Japan in 1996 is the sharp decrease in the production and shipment of petroleum refining catalysts and remarkable increase in those of the pollution control catalysts. The production and the shipment in volume of petroleum refining catalysts in 1996 were only 60.1% and 65.4% of those in 1995, respectively. In the meantime, from 1995 to 1996, the production and the shipment in volume of pollution control catalysts excepting autoexhaust catalysts are 145.8% and 198.6% of those in 1995, respectively. Table 1 also shows that the heavy oil desulfurization market grew considerably from 1995 to 1996.

1.1. FCC catalyst

Petroleum refining catalysts include naphtha reforming catalysts, FCC (fluid catalytic cracking) catalysts and hydrogenation of intermediate fraction catalysts. The principal cause of the decline of the petroleum refining catalyst sales in 1996 is attributable substantially to decreased sales of FCC catalysts. From the global standpoint of view, the growth of FCC demand is sluggish. From 1995 to 1996, the demand decreased from 493500 tons to 470000 tons. Besides, the price of FCC catalysts dropped in the past three years. On the other hand, FCC demand in Asia/Pacific has been growing by 10%/year, and due to this growth, the situation of Japanese FCC catalyst business is not necessarily bad. In fact, FCC catalyst shipment in Japan continuously increased until 1995. All in all, it is unfortunate

that Japan's FCC catalyst sales dropped on the whole, as Japanese manufacture lost the market share in 1996. In the first half of 1997, the production and the shipment of FCC catalyst showed a recovery. The production, the shipment both in volume and the value with regard to the petroleum refining catalysts in the first half of 1997 were 119%, 109% and 78% compared with the first half of 1996, respectively. The production and the shipment in volume increased considerably, while the shipment in value decreased. This results mainly came in a reflection of FCC catalyst business.

1.2. Heavy oil desulfurization

The volume of the shipment of heavy oil desulfurization catalysts increased from 13908 tons in 1995 to 14415 tons in 1996. Heavy oil desulfurization catalysts are classified into the following two categories:

- (1) Direct desulfurization of atmospheric and vacuum residue (AR, VR).
- (2) Indirect desulfurization by desulfurization of vacuum gas oil (VGO) and mixing with residue.

Domestic use of AR and VR hydrodesulfurization catalysts is gradually increasing and has become approximately 7000 tons/year. Formerly, direct desulfurization catalysts were mainly applied to the production of low-sulfur heavy oil, which was used as fuel by various kinds of industries such as electric power industry, steel industry and so on. Recently, this type of catalyst becomes to be applied to the pretreatment of the feedstock for FCC process and because of the more severe operating conditions, its life becomes shorter than before.

For desulfurization of VGO in Japan, the units were applied to the pretreatment of the feedstock before FCC units. Presently, one third of VGO desulfurization units are changing their operating manner to produce middle distillate by mild hydrocracking of VGO. As a result, the operating conditions are getting more severe. Such type of application shortens the catalyst life, which causes increased demand of VGO catalyst.

Table 1
Production and shipment of catalysts (unit: tons; ¥1 million).

	1993			1994		
	Production	Shipment		Production	Shipment	
		Volume	Value		Volume	Value
Petroleum refining	17662	18029	7326	27023	26482	9004
Heavy oil desulfurization	13038	12696	12411	13935	13288	11845
Petrochemical production	11267	10937	22460	11535	10933	22076
Polymerization	4283	4393	4927	5236	5472	6004
Gas production	2909	2629	4658	2834	2810	4495
Oil and fats processing	1506	1566	1888	1265	1329	1585
Drugs, foodstuffs production	695	693	1568	634	661	1647
Pollution control	6938	6578	44375	7142	7236	40951
Autoexhaust	4958	4952	38954	4999	5014	34747
Other pollution control	1980	1626	5421	2143	2222	6204
Inorganic chemicals and others	94	147	235	60	163	180
Total	58392	57668	99848	69664	68374	97787

	1995			1996			1996/1995(%)		
	Production	Shipment		Production	Shipment		Production	Shipment	
		Volume	Value		Volume	Value		Volume	Value
Petroleum refining	23892	22693	8449	14352	14840	7129	60.1	65.4	84.4
Heavy oil desulfurization	12218	13908	11976	15611	14415	12505	127.8	103.6	104.4
Petrochemical production	12691	11746	24146	12505	11637	24361	98.5	99.1	100.9
Polymerization	7082	6912	6203	5875	5643	5930	83.0	81.6	95.6
Gas production	2559	2918	4148	2573	2618	4498	100.5	89.7	108.4
Oil and fats processing	1266	1352	1586	1421	1350	1560	112.2	99.9	98.4
Drugs, foodstuffs production	880	857	1872	787	744	1703	89.4	86.8	91.0
Pollution control	6661	6410	35653	7464	7826	36537	112.1	122.1	102.5
Autoexhaust	5086	5072	31179	5168	5169	29017	101.6	101.9	93.1
Other pollution control	1575	1338	4474	2296	2657	7521	145.8	198.6	168.1
Inorganic chemicals and others	159	75	158	77	97	220	48.4	129.3	139.2
Total	67408	66871	94191	60665	59170	94443	90.0	88.5	100.3

Source: MITI (Ministry of International Trade and Industry) "Annual Report of Chemical Industry".

1.3. Pollution control catalyst

Among pollution control catalysts excepting autoexhaust catalysts, DeNO_x(NO_x removal) catalysts are a main product. In this regard, the remarkable increase in sales amount of the pollution control catalysts is a reflection of the favorable market situation for DeNO_x catalysts. The principal method of NO_x removal is SCR (selective catalytic reduction), using ammonia as a reductant of NO_x. Although the demand of DeNO_x catalysts is mostly for gas turbines of electric power plants at present, the demand for other applications of DeNO_x catalysts, for instance for incinerators, diesel power generators, and chemical plants, are increasing recently. Especially sales of low temperature NO_x removal catalysts have expanded [2]. For example, for the application to the treatment of the waste incinerator flue gas, a low temperature DeNO_x catalyst is strongly required. The reason is as follows. In the case of the waste incinerator, the flue gas, for example, passes through in order of cooling tower, DeSO_x(SO_x removal) equipment, bag filter(dust precipitator), gas heater, DeNO_x equipment, and stack. Recently, bag filters tend to be used as dust precipitators because when an electric dust precipitator is used, there is a danger of generation of a small amount of dioxin. Therefore, the flue gas, which comes to the DeNO_x equip-

ment is cooled down. The temperature of the flue to be treated by the DeNO_x catalyst is as low as 200–250 °C.

1.4. Autoexhaust catalyst

As to autoexhaust catalysts, the catalyst demand will increase globally since the regulations for autoexhaust have extended to many countries in Europe, Asia, Oceania and South America. In the USA, the regulations for hydrocarbon and NO_x are getting stricter gradually. From the view point of catalyst technology, various kinds of requirements have been continuously arising. For example, to improve fuel economy, the lean burn engine, in which high A/F (air fuel ratio) combustion is performed, has been developed. For the lean burn engine, catalysts with excellent NO_x removal activity at low temperature are required. The increase in the demand of autoexhaust catalyst may cause a shortage of the precious metal supplies. Under such circumstances, many efforts aiming at higher activity with smaller amount of precious metal have been made. The production and shipment of autoexhaust catalysts have been increasing steadily. However, shipment has decreased in value because of severe price competition and inflows of low-priced imports. From 1993 to 1996, the value of shipment

Table 2
Imports of catalysts (unit: tons; ¥1 million).

		1993	%	1994	%	1995	%	1996	%	1996/1995(%)
USA	Volume	17829	84.2	18937	77.0	19339	75.8	19754	65.5	102.1
	Value	23849	87.8	24088	83.2	26458	85.8	27165	81.1	102.7
EU Total	Volume	3147	14.9	5314	21.6	6051	23.7	10211	33.8	168.7
	Value	3017	11.1	4477	15.5	4016	13.0	5588	16.7	139.1
Germany	Volume	261	1.2	267	1.1	275	1.1	985	3.3	358.2
	Value	656	2.4	552	1.9	720	2.3	678	2.0	94.2
UK	Volume	266	1.3	162	0.7	296	1.2	460	1.5	155.4
	Value	622	2.3	488	1.7	755	2.4	839	2.5	111.1
France	Volume	421	2.0	532	2.2	107	0.4	282	0.9	263.6
	Value	421	1.5	874	3.0	129	0.4	308	0.9	238.8
Denmark	Volume	59	0.3	148	0.6	92	0.4	376	1.2	408.7
	Value	230	0.8	308	1.1	313	1.0	896	2.7	286.3
The Netherlands	Volume	1822	8.6	3724	15.1	4789	18.8	7670	25.4	160.2
	Value	550	2.0	1080	3.7	1269	4.1	1868	5.6	147.2
Italy	Volume	132	0.6	137	0.6	155	0.6	279	0.9	180.0
	Value	344	1.3	443	1.5	465	1.5	723	2.2	155.5
Others	Volume	187	0.9	345	1.4	121	0.5	206	0.7	170.2
	Value	296	1.1	383	1.3	347	1.1	762	2.3	219.6
Total	Volume	21163	100	24596	100	25511	100	30171	100	118.3
	Value	27162	100	28948	100	30821	100	33515	100	108.7

Source: MOF (Ministry of Finance) "Monthly Report of Japanese Trade".

of autoexhaust catalysts decreased by approximately ¥10 billion.

On the other hand, in the first half of 1997, the production, shipment in volume, and shipment in value of autoexhaust catalysts are 3047 tons, 2980 tons, and ¥18131 million, respectively and these numbers to those for the first half of 1996 are 120%, 117% and 129%, respectively. The total shipment in value for 1997 is estimated ¥36 billion, which is close in figure to the shipment value in 1993. Both the production and the volume of the shipment for the first half of 1997 are the highest in the years since 1990. It seems that the decrease in the value of the shipment of autoexhaust catalysts nearly came to an end. This trend is considered to have been brought about by the increase in total shipment and the sudden rise of Pd and Pt caused by the recent Yen's depression.

2. Export and import of catalysts in Japan

Imports and exports of catalysts from and to each of the countries are summarized in tables 2 and 3, respectively. As is shown in table 2, the major part of imported catalysts comes from the USA and both the volume and value of import have been steadily increasing. However, the ratio of the catalysts from the USA both in volume and value has been decreasing these four years principally because of the increased import from the EU. Especially, in 1996, the import from the EU sharply increased both in volume and value. Consequently, the ratio of volume and value of the imported catalysts from the USA dropped from 75.8% to 65.5% and from 85.8% to 81.1%, respectively. Among the imports from the EU, those from The Netherlands increased remarkably, as a result of the sharp increase in the import of FCC catalysts.

Table 3 shows that Japanese catalysts are mainly exported to Southeast Asia. In this table, the Republic of Korea, Taiwan and Hong Kong are included in the geographical category of Southeast Asia. From 1994 to 1996, however, the volume of the catalysts exported to Southeast Asia decreased. Especially, from 1995 to 1996, the volume of the catalysts exported to Southeast Asia dropped from 14721 tons to 9046 tons, while the value remains almost unchanged. This phenomenon is again caused by the decrease in the export of FCC catalysts.

3. Demand for catalysts in Japan

In table 4, total shipment, export, and import of Japanese catalysts are shown, and from these numbers the catalyst demand in Japan is estimated. The total value of catalyst shipment slightly increased from 1995 to 1996. The difference is an increase of only ¥252 million, i.e., by 0.3%. The total export value increased by 4.8% (¥1834 million) from 1995 to 1996. Meanwhile the total import also increased by 8.7% (¥2694 million). Consequently, the value of domestic demand increased only by 1.3% (¥1112 million).

On the other hand, from 1995 to 1996, the volume of total shipment and export decreased by 11.5% (7701 tons) and 20.7% (5409 tons), respectively. This is caused by the decrease in both shipment and export of FCC catalysts as mentioned above.

4. Several topics of newly developed industrial catalysts and catalytic technology in Japan

Three examples of Japanese catalyst developments are described here; the first is a development of a new resid

Table 3
Exports of catalysts (unit: tons; ¥1 million).

		1993	%	1994	%	1995	%	1996	%	1996/1995(%)
South-east Asia	Volume	5404	29.1	17055	55.0	14721	56.3	9046	43.6	61.4
	Value	12004	42.6	16207	46.5	18272	47.9	18168	45.5	99.4
West Asia	Volume	572	3.1	1963	6.3	1530	5.9	1182	5.7	77.3
	Value	691	2.4	1309	3.8	1469	3.9	1627	4.1	110.8
Europe	Volume	2909	15.7	1781	5.7	2184	8.4	2307	11.1	105.6
	Value	4502	16.0	3940	11.3	4784	12.5	5883	14.7	123.0
North America	Volume	4514	24.3	4567	14.7	2917	11.2	5157	24.9	176.8
	Value	7738	27.4	8971	25.7	9318	24.4	11270	28.2	120.9
Middle & South America	Volume	223	1.2	139	0.4	61	0.2	12	0.1	19.7
	Value	814	2.9	600	1.7	539	1.4	386	1.0	71.6
Oceania	Volume	4314	23.3	4446	14.3	2672	10.2	1836	8.9	68.7
	Value	973	3.4	1027	2.9	594	1.6	739	1.8	124.4
Africa	Volume	310	1.7	593	1.9	316	1.2	659	3.2	208.5
	Value	377	1.3	689	2.0	352	0.9	687	1.7	195.2
Former Communist Countries	Volume	296	1.6	444	1.4	1746	6.7	539	2.6	30.9
	Value	1108	3.9	2148	6.2	2797	7.3	1199	3.0	42.9
Total	Volume	18542	100	30988	100	26147	100	20738	100	79.3
	Value	28207	100	34891	100	38125	100	39959	100	104.8

Source: MOF "Monthly Report of Trade in Japan".

South-east Asia: Republic of Korea, Taiwan, Thailand, Singapore, Malaysia, the Philippines, Indonesia, Hong Kong.

West Asia: Myanmar, India, Pakistan, Bangladesh, Saudi Arabia, Kuwait, Israel, Iran, Bahrain, Qatar, Jordan, Uzbekistan.

Europe: Sweden, UK, The Netherlands, Belgium, France, Germany, Portugal, Italy, Spain, Finland, Turkey, Austria, Switzerland.

North America: USA, Canada.

Middle & South America: Brazil, Mexico, Venezuela.

Oceania: Australia, New Zealand.

Africa: Kenya, S. Africa, Egypt, Algeria.

Former Communist Countries: North Korea, China, Russia, Poland, Romania, Slovak Republic, Czech Republic.

Table 4
Export, import and domestic use of catalysts (unit: tons; ¥1 million).

		1993	1994	1995	1996	1996/1995(%)
Shipment in total (A)						
	Volume	57668	68374	66871	59170	88.5
	Value	99848	97787	94191	94443	100.3
Total export (B)						
	Volume	18542	30988	26147	20738	79.3
	Value	28207	34891	38125	39959	104.8
Total import (C)						
	Volume	21163	24596	25511	30171	118.3
	Value	27162	28948	30821	33515	108.7
Autoexhaust catalyst (included in total import)						
	Volume	2335	3071	2960	2621	88.5
	Value	16254	16549	16703	16123	96.5
Estimated domestic demand (A) - (B) + (C)						
	Volume	60289	61982	66235	68603	103.6
	Value	98803	91844	86887	87999	101.3

Source: MOF "Monthly Report of Japanese Trade".

FCC catalyst, the next is a development of a catalyst for the production of methylmethacrylate using a new process, and the third is developments of autoexhaust catalysts for lean-burn engines.

4.1. Resid FCC (R-FCC) catalyst

Catalytic cracking processes produce LPG, high-octane gasoline and middle distillates from VGO or residual oil.

Recent trends are towards an increase of resid blending in VGO FCC units and the construction of resid FCC units, which means the increase in the ratio of heavier oil with high metal (Ni, V) and high asphaltene contents. Besides, FCC demand especially in Southeast Asia has been increasing. To cope with this situation, Catalysts & Chemicals Ind. Co., Ltd. (CCIC) recently developed new Resid FCC catalysts. Their commercial names are BLC (Bottom Least Catalyst) and STW (Stalwart) [3]. The requirements for residue catalysts are:

- (1) high crackability of bottoms,
- (2) low coke/gas formation,
- (3) high metal-tolerance,
- (4) high hydrothermal resistivity.

In order to meet these requirements, they designed pore size distribution and solid acid features of matrix, and they also selected favorable metal trap species. The FCC catalyst consists of Matrix and Zeolite. In conventional FCC catalysts RE(rare earth)-Y or US(ultra stable)-Y is used as a zeolite species. When resid cracking is necessary, as in the case of R-FCC catalyst, US-Y, Y of which is treated with high temperature steam, is mainly employed. Furthermore, the design of Matrix affects the performance of resid cracking considerably.

The average size of large hydrocarbon molecules contained in resid is said to be 100–150 Å. According to

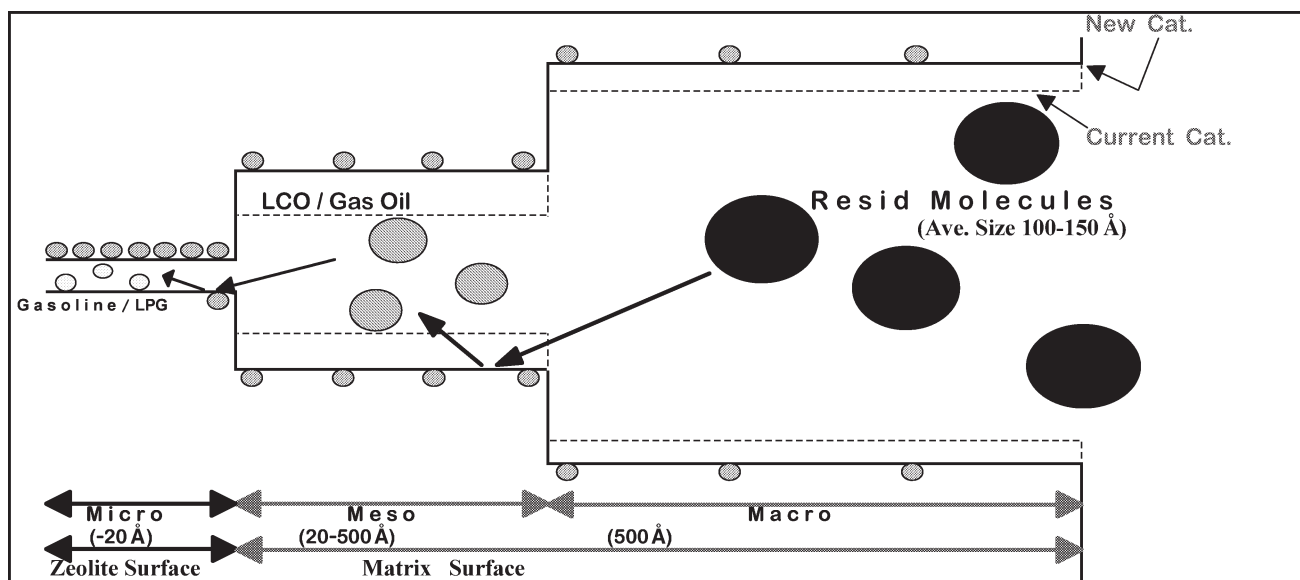


Figure 1. Design for pore distribution of matrix.

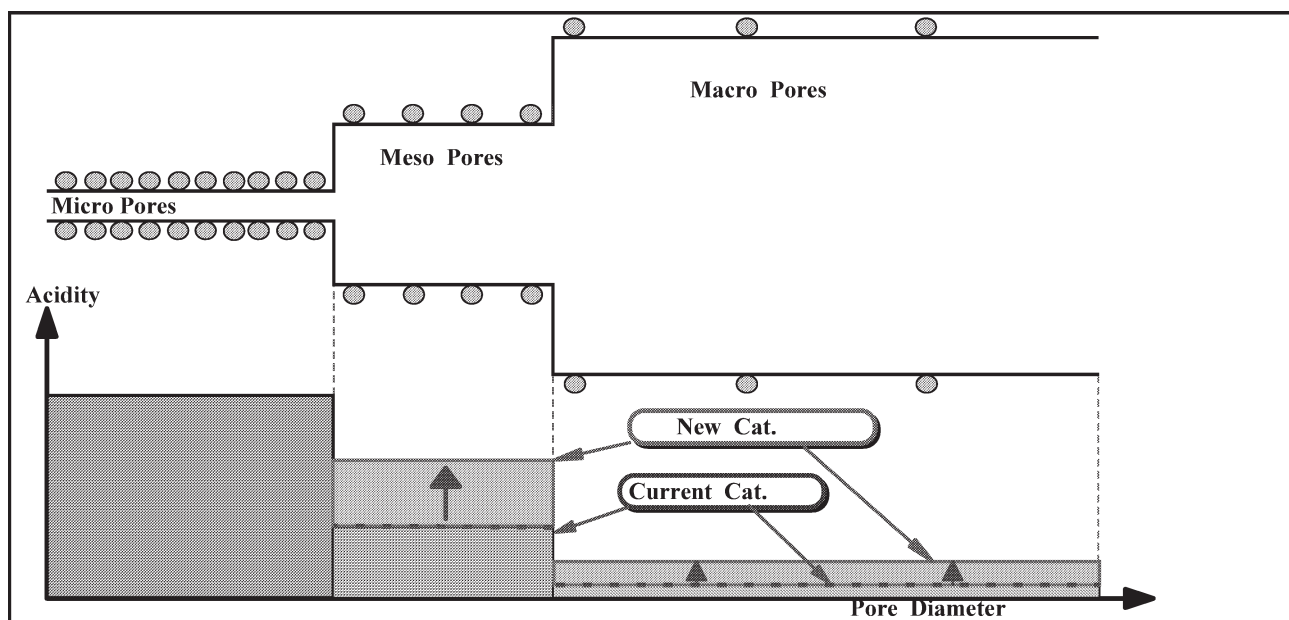


Figure 2. Design for solid acid properties of new catalyst.

the researchers of CCIC, it is important that macropores of nearly 1000 Å are made to facilitate diffusion of large molecules and their pre-cracking. The effectiveness of the resid cracking becomes further better, if the size of macropores is larger than 1000 Å. However, when the size of the macropores is bigger than 1000 Å, crash strength and attrition resistivity of the catalyst become lower. After large molecules are pre-cracked, they go into mesopores and are further cracked to light cycle oil (LCO). The mesopores with diameter of about 150 Å are also important. The more the volume of mesopores is, the more LCO fraction of the product is obtained. Finally, molecules go into the micropores of the zeolite and are cracked to gasoline fraction. The whole concept is summarized in figure 1.

Acidity of the catalyst is another important factor of the performance of FCC catalysts. Since it is important to pre-crack large hydrocarbon molecules in macropores, acid site is necessary. However, cohesion of basic large molecules occurs during cracking when acid site is too strong. Therefore, it is necessary to prepare weak acid sites in macropores. In mesopores, acid strength needs to be stronger so that pre-cracked hydrocarbon molecules can be efficiently cracked to LCO fraction (figure 2). The strength and the amount of acid sites are optimized by adding active alumina or silica.

In resid, there exist a large amount of V and Ni compounds, both of which are poisonous to the catalyst. Vanadium from cracked large hydrocarbon molecules easily

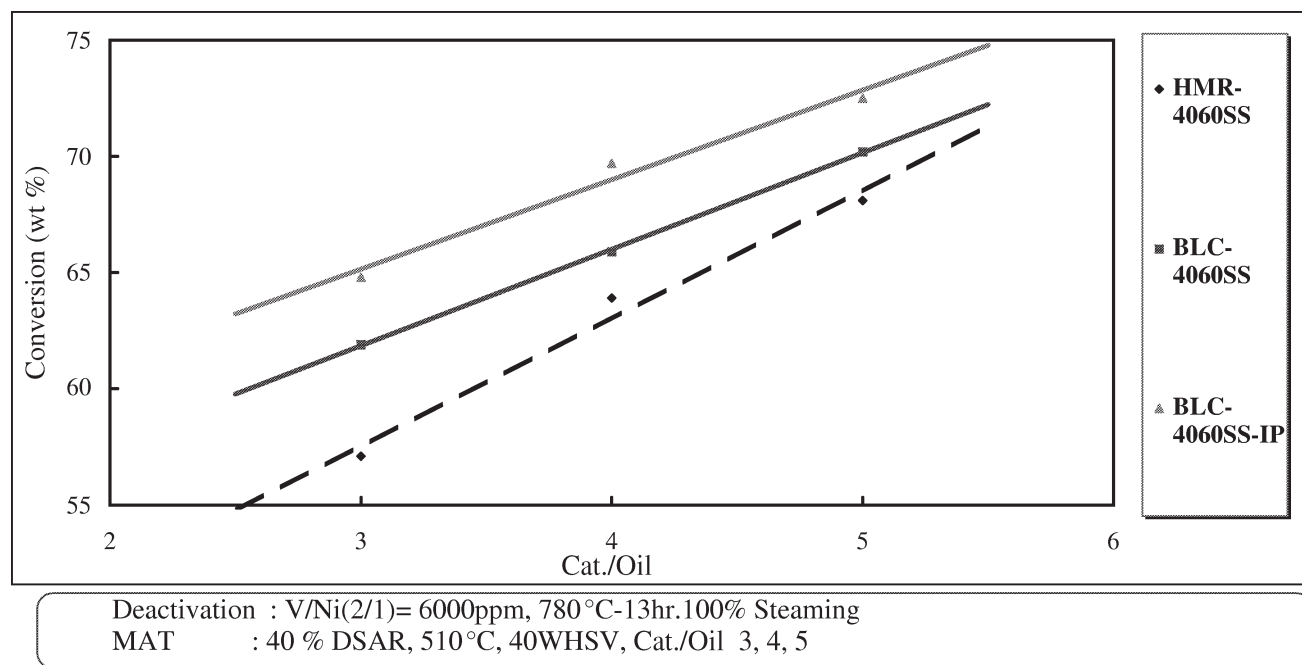


Figure 3. MAT performance of resid cracking test.

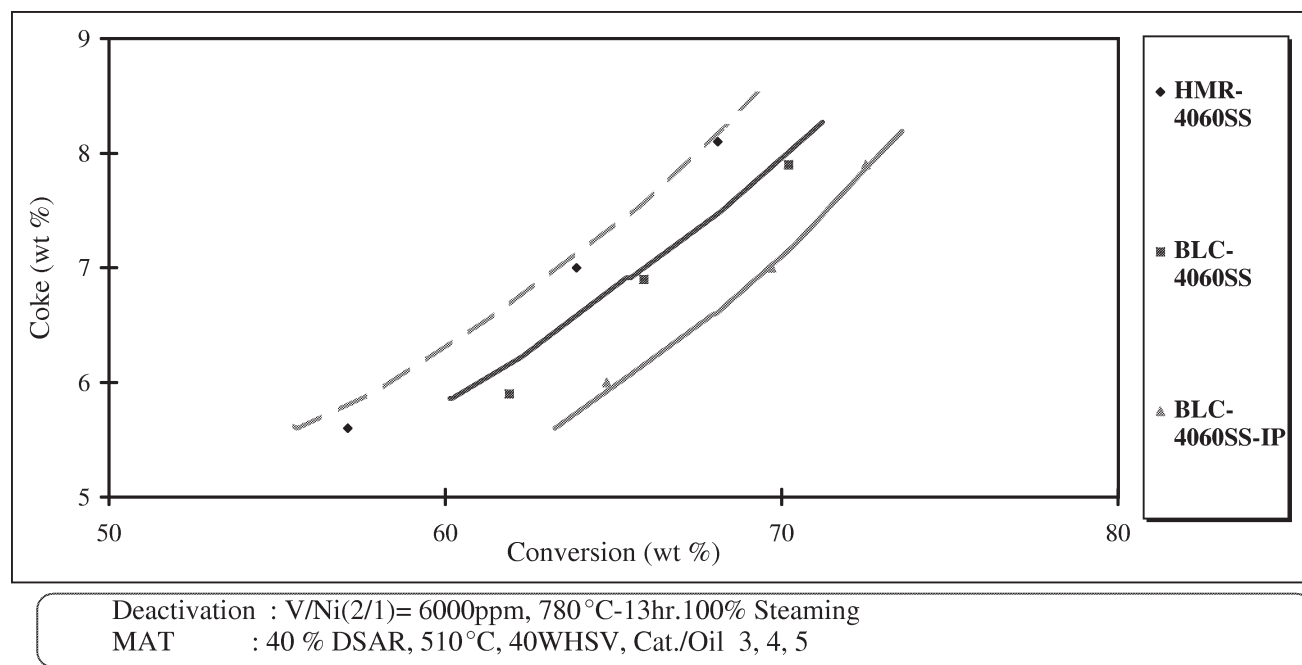


Figure 4. MAT performance of resid cracking test.

plugs the pores of the catalyst. It destroys the zeolite crystal and poisons the active alumina. Nickel causes an increase in hydrogen yield by intensifying dehydrogenation activity. One method to minimize the poisonous influences by these metals is to trap them with other chemical species such as Sb, Sn, or Mg compounds. Since these metal trap materials are generally strong bases, they have the tendency of destroying the acidity balance of the catalyst and causing less bottom crackability. Therefore, CCIC chose materials with high affinity with V or Ni and low basicity.

In the case of their new catalysts, BLC and STW, pore structure and acidity are designed on the basis of the concept mentioned above, and especially, as to STW, a new metal trap compound is incorporated. Some of their results are shown in figures 3–6. These results are obtained by a micro activity test (MAT), which simulates a fluidized bed reaction using small amount of the catalyst. Ranking of conversion level, coke yield, gasoline and LCO yield, and bottom crackability is in the order: (1) BLC-4060SS-IP, (2) BLC-4060SS, and (3) HMR-4060SS. HMR as in

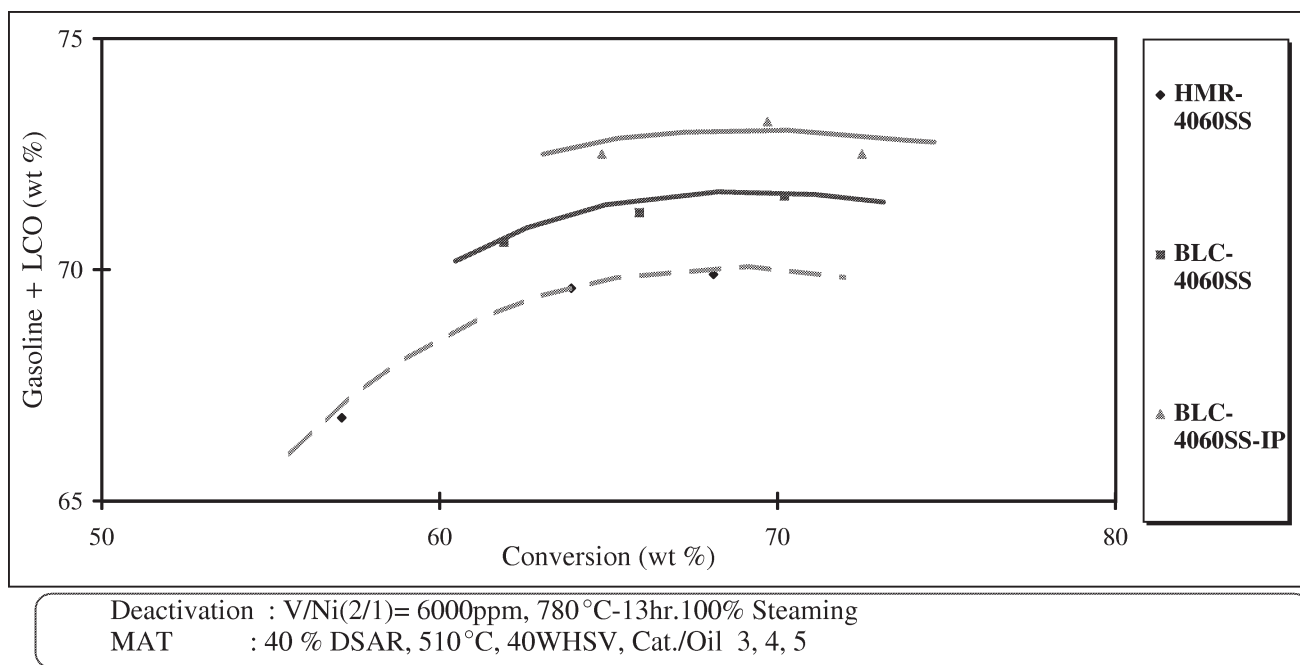


Figure 5. MAT performance of resid cracking test.

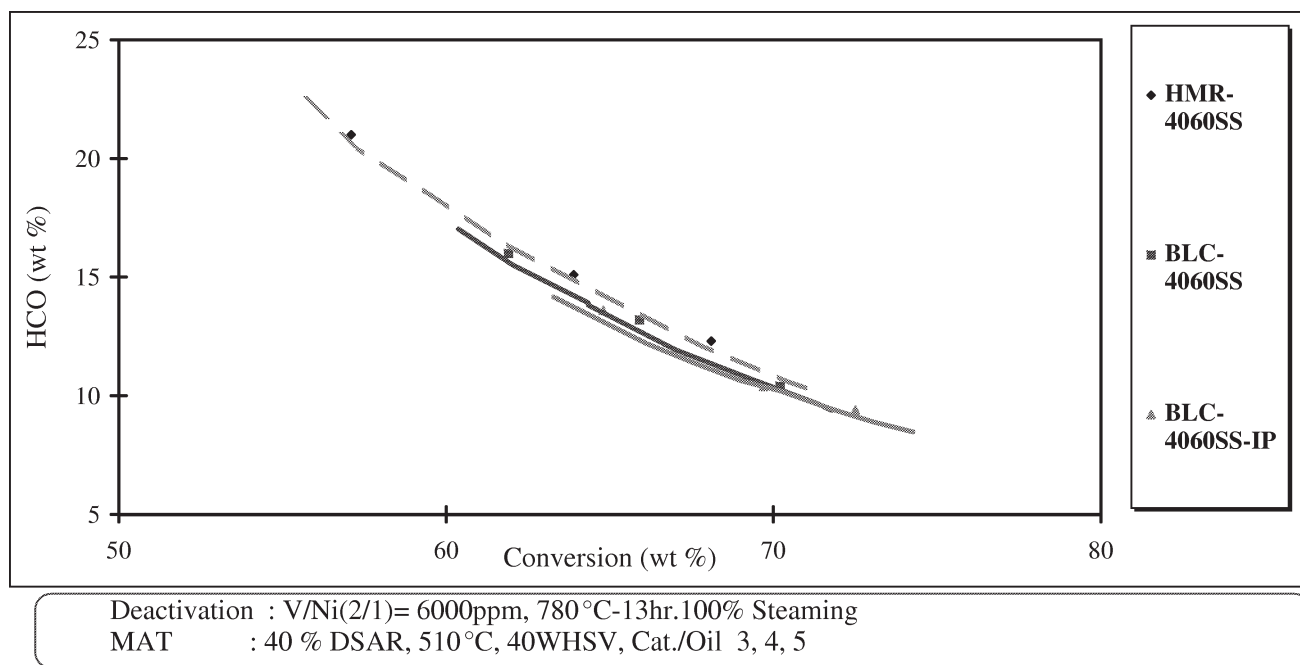


Figure 6. MAT performance of resid cracking test.

(3) is a conventional catalyst, while a final symbol IP in (1) means that it is a catalyst with diffusion of feed being improved by the optimization of the pores of the matrix component.

4.2. New MMA (methylmethacrylate) catalyst

Methylmethacrylate (MMA) is an important monomer of various kinds of polymers. MMA resin is an excellent material for its antiweatherability and transparency, and there

are many applications for construction material, car material, optical materials and others. Recently, Asahi Chemical Industry decided to commercialize a new process of MMA production [4]. The key technology of this process is oxidative esterification of methacrolein (MAL) (figure 7). There are two major problems in this step. One problem is that oxidative decarbonylation does occur as a side reaction and propylene is formed. Another problem is that the reactivity of an unsaturated aldehyde like MAL is not so high as that of a saturated aldehyde. It was known that Pd catalyzes

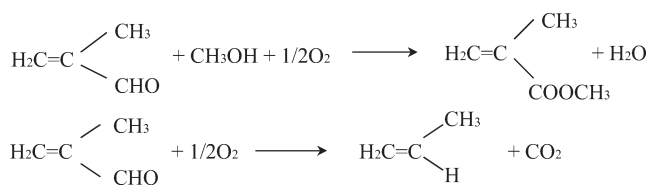


Figure 7. Oxidative esterification of MAL and its side reaction.

the oxidative esterification of MAL, but the selectivity of MMA was as low as 30%. They studied the additives to Pd metal in order to improve the activity and the selectivity, and finally they found Pb to be the best additive. They started to construct a pilot plant in 1993 and completed the whole test for the new process of MMA.

4.3. Development of catalysts for lean-burn engines

The gasoline fuel lean-burn engine is one of the key technologies to improve the fuel efficiency of a passenger car. However, under lean combustion conditions, CO and hydrocarbon preferentially react with oxygen and a conventional three-way catalyst cannot remove NO_x effectively. Consequently, the lean operating conditions have been limited.

Toyota Motor Corporation developed a NO_x storage-reduction type catalyst (NSR catalyst) [5]. On this catalyst, NO_x is oxidized under oxidizing conditions and stored as $\text{Ba}(\text{NO}_3)_2$, and under stoichiometric conditions, it is reduced and desorbs as nitrogen gas. As to the details of the development of this catalyst, they were explained by in this Journal. Later, Matsumoto and his co-workers published the schematic diagram lean-burn system [6]. Because of lean combustion, they paid attention to the stable mixing and stable combustion. Since NO_x is reduced and removed under rich or stoichiometric condition, it is necessary to insert stoichiometric or rich mixture combustion at intervals. However, when fuel concentration becomes higher, the engine torque also becomes higher. Therefore, the timing of the ignition is adjusted so that the torque may not increase. According to their report of NO_x conversion efficiency during Japanese 10.15 mode running in vehicles with the lean-burn engine, in the initial stage of testing the engine exhibited 80% or more conversion efficiency. Even after 100 000 km-equivalent durability running with Japanese regular gasoline, the catalyst retained 60% or more NO_x conversion efficiency [7].

Another type catalyst was developed by Mazda Motor Corporation [8]. Their concepts were as follows: to find out active components, they searched for precious metal assortment that has high NO_x adsorption capability at high oxygen concentration and has high resistivity to sintering. For a carrier, they searched for a substance that can hold large amount of hydrocarbon, which is the reductant of NO_x , and is thermally stable as well.

They found that Pt–Ir–Rh is the most active species for lean NO_x conversion after $1073 \text{ K} \times 6 \text{ h}$ in air (10% H_2O) aging. They observed aged catalyst by TEM and found that Pt–Ir–Rh deposited on zeolite is thermally more stable than Pt deposited on the same carrier.

Table 5
Specifications of developed catalyst for lean-burn engine ([8]).

Active metal:	Pt–Ir–Rh
Support:	H^+ exchanged zeolite
	Structure MFI
	$\text{SiO}_2/\text{Al}_2\text{O}_3$ 80 ± 10
	Contaminations $\text{Na} < 0.10 \text{ wt\%}$
	Crystallinity $> 90\%$
	BET surface area $> 300 \text{ m}^2/\text{g}$
Additives:	ceria, alumina

It is known that active carbon or zeolite can adsorb hydrocarbon molecules. Since zeolite is more stable than active carbon under high temperature, they studied the thermal stability of various types of zeolite. Finally, they concluded that H-MFI is the most stable. They also measured lean NO_x removal activity by changing the silica alumina ratio of MFI and they concluded that $\text{SiO}_2/\text{Al}_2\text{O}_3 = 80$ is the most favorable ratio. Their specifications for developed catalyst for lean-burn engine are summarized in table 5.

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