中国炼油技术新进展

侯祥麟 主编

ADVANCES OF REFINING TECHNOLOGY IN CHINA

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ZY02/105

China Petrochemical Press

1997

图书在版编目(CIP)数据

中国炼油技术新进展: 英文版/侯祥麟主编. —北京: 中国石化出版社, 1997.10
ISBN 7-80043-714-0

Ⅰ.中… Ⅱ.侯… Ⅲ.石油炼制-进展-中国-英文 Ⅳ.TE62

中国版本图书馆 CIP 数据核字(97)第 16114 号

中国炼油技术新进展

（英文版）

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Advances of Refining Technology in China

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No.58 Anwai Street, Beijing 100011, China

Tel: (010)64241850

中国石化出版社出版发行
地址: 北京市东城区安定门外大街 58 号
邮编: 100011　电话: (010)64241850
纺织出版社印刷厂印刷
新华书店北京发行所经销
*
787 × 1092 毫米 16 开本 15 印张 320 千字 印 1—5000
1997 年 9 月第 1 版　1997 年 9 月第 1 次印刷
定价:200.00 元

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PREFACE

The progress of refining technology in China was greatly accelerated in the 1960s after Daqing oil field was discovered and hence large quantity of crude oil was produced. The overall embargo on China became a driving force of R&D to develop petroleum technology based on our own efforts and subsequently fluid catalytic cracking, catalytic reforming, delayed coking and hydrocracking with all relevant catalysts were developed and commercialized during that decade.

Thereafter, R&D works kept pace with the progress world wide and Y zeolite cracking catalyst, bimetallic reforming catalyst and riser FCC process were developed.

Entering into the 1970s, embargo began to slacken and since then, some refining technologies were licensed such as HF alkylation, resid FCC and VGO hydrocracking. Nevertheless, the policy of relying upon our own R&D was maintained and activities kept which led to the innovation of the existing technologies as well as the development of new ones.

Because of the highly paraffinic nature of Daqing crude, R&D work was focused on the development of atmospheric residue FCC process and a commercial unit was constructed in the early 1980s which was being operated successfully. In the meantime, the accumulation of operation and designing experiences as well as R&D achievements led to innovations of the resid FCC process and a number of patents approved.

By end of 1980s, a hydrocracking unit for VGO of 0.8 million tons per annum capacity was designed and constructed with most of the equipment including the high pressure reactor manufactured domestically. In line with the processing developed, a spectrum of catalysts was also successfully exploited and manufactured which enabled the hydrocracking products to meet the various market requirements.

The status of refining technology in China up to that period was fully described in a book 《Refining Technology in China》 published by China Petrochemical Press in 1991.

With decades of R&D and commercialization works, the research institutes, engineering firms, equipment manufacturers and refineries had accumulated much experiences so that since early 1990s, Chinese petroleum industry not only was equipped with modern refining processes and technologies, but also had the capability to develop and commercialize new ones by our own efforts.

Pursuing new generation catalysts with high quality was of primary concern because its commercialization usually brought about profit immediately to the refineries. Priority was given to FCC catalyst owing to the large quantity employed. As a result, the properties of most FCC catalyst series in China have reached such a level comparable to those manufactured elsewhere.

High quality catalysts for continuous catalytic reforming were also developed and approved by the licenser and have been used in the licensed units successfully. Many other refining catalysts such as those for hydrotreating and hydrocracking, have also reached the quality level comparable to those used worldwide.

Because of the high nitrogen content in most Chinese crudes, an active hydro-denitrogenation catalyst was developed. A batch of this new catalyst has been used in an European refinery yielding good results both for denitrogenation and desulfurization.

Some other catalysts developed, such as those used for isomerization, dispro-portionation, transalkylation of aromatics displayed very good properties and have practically monopolized the domestic market. At the same time, these catalysts have also attracted interests in the international market.

New processes have also started to probe their way to nativity. The first major instance was the development of DCC (Deep Catalytic Cracking) process. With the knowledge accumulated in FCC technology and catalyst and with the incentive of market demand of propylene, R&D work was directed to develop a new process based on FCC to obtain propylene as the major product with heavy hydrocarbons instead of light hydrocarbons as feedstock. This process has been commercialized not only in China but also licensed to a petrochemical complex in Thailand through Stone & Webster Engineering Co. of USA.

In semi-regeneration catalytic reforming, a process of staged loading was developed, in which two kinds of catalysts with different reforming performances were packed in series resulting in enhancing the yield of reformate and octane number of gasoline.

Another instance is the alkylation of benzene with dilute ethylene of FCC dry gas. Key element of the process is the innovation of catalytic material which can resist the poisoning impurities in the dry gas. This process has been successfully commercialized in two plants by using dry gas without pretreatment from FCC units which used feedstock from Daqing crude.

Although there has been only a short span of time since the last book was published in 1991, the major achievements in this period was considered to be worthwhile to compile into a book to present to people interested in the refining technology in China. Because the 15th World Petroleum Congress is to be held in Beijing the coming October, it is decided to publish an English version of this book with the title of 《Advances of Refining Technology in China》.

Through this book we hope that the readers may be able to get a better understanding of the R&D works and present status of Chinese petroleum refining technology.

Hou Xianglin

Honorary President of Chinese Peroleum Socity

August, 1997, Beijing

CONTENTS

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PROGRESS OF CHINA'S PETROLEUM REFINING TECHNOLOGY

Hou Fusheng

ABSTRACT

China's petroleum refining industry has reached a considerable scale with a sound foundation. The crude processing capacity has come up to 200 Mt/a in 1995, meanwhile significant progress has been made in the field of refining technology. Many new processes have been developed, such as resid catalytic cracking, deep catalytic cracking (DCC), catalytic cracking for maximum gasoline plus gas production (MGG), catalytic cracking for maximum isoolefin production (MIO), medium pressure hydro-upgrading (MHUG), etc., and new catalysts have also been commercialized for the related new technologies.

Along with the rapid growing demands of petroleum products for domestic market, the Chinese refining industry will further develop in the next decade and the processing capacity will be increased to 300 Mt/a by the year 2010. Great attention will continuously be paid to advanced technology in petroleum refining. The key projects will be heavy oil upgrading and sour crude processing, improvement of the product quality to meet the needs of environmental protection, development of new routes for light olefins production, increasing raw materials for chemical fibre production, and speeding up the application of computer in process control and energy conservation.

INTRODUCTION

China's petroleum refining industry has reached a considerable scale through almost half a century's hard work, and has become one of the most important industries in China.

A brief review of the history will help to know the advance of Chinese refining industry. Prior to the establishment of the People's Republic of China in 1949, there was only a small amount of crude produced in Yumen oil field, Gansu Province, and almost all oil products were imported. Since the P.R.C. was founded, she has devoted major efforts not only to the exploration and exploitation of crude oil, but also to the development of syncrude production.

In the 1950s, the highest output of shale oil was 700 kt/a in Fushun, Liaoning Province. Through distillation, cracking (both thermo and hydro), dewaxing, and other processes, many petroleum products were produced, such as gasoline, kerosine, diesel fuel, and wax. During the same period, the syncrude industry also employed low temperature retorting and Fischer-Tropsch process using coal as raw material to produce synthetic fuels.

The successful exploitation of the oil field in Yumen, Gansu Province and in Keramayi, Xinjiang Autonomous Region, brought a surge in crude oil production to over 1 Mt/a. In 1958, a fuel/lube type refinery with a capacity 1 Mt/a was built in Lanzhou, Gansu Province, along with several other refineries in Xinjiang Autonomous Region. In addition, some old refineries in coastal cities of Dalian, Liaoning Province and Shanghai were revamped.

In early 1960s, the discovery of Daqing oil field in Helongjiang Province made a big step in crude oil production. The oil output exceeded 10 Mt in 1965, and in 1978 exceeded 100 Mt. The fast growth of crude oil production greatly accelerated the development of the refining industry. China's processing capacity was increased rapidly during that period.

Simultaneously, efforts were concentrated on research and development of processing technologies and new petroleum products, resulting in some of the comparatively advanced technologies such as fluid catalytic cracking, catalytic reforming, and delayed coking as well as related catalysts and additives were mastered and plants constructed. Commercialization and wide application of these technologies in China laid a solid foundation for further development of the refining industry.

The establishment of China Petrochemical Corporation (SINOPEC) in 1983 was a significant step for promoting the refining and petrochemical industry in China. SINOPEC is an economic entity which engages in petroleum refining and petrochemical manufacture. It has brought vigorous vitality to the Chinese refining and petrochemical industry. A number of modern refineries and petrochemical complexes were constructed during the past fourteen years.

Now there are 50 main refineries scattered throughout China with a total capacity of 200 Mt/a by the end of 1995, of which SINOPEC accounts for 82.8 %. In the same year, 135 Mt of crude oil were processed and 75 Mt of gasoline, kerosine, diesel fuel, and lube oil were produced to meet the domestic need.

It is well known that most Chinese crudes are sweet, with less heavy metals, but they are rather heavy, with relatively small percentage of light fractions, naphtha about 6% ~ 10 %, middle distillate 20 % ~ 24 %. The typical properties of some major Chinese crudes are shown in Table 1.

To fit in with this crude situation, a policy of deep refining processing has been adopted and a series of modern bottoms upgrading technologies have been developed. FCC as a major catalytic conversion technology developed rapidly. China has a 56 Mt/a catalytic cracking capacity in 1995, 28 % of the total crude distillation capacity (Table 2).

Table 1 Typical Properties of Some Major Chinese Crudes

	Properties			
Crude	API	Sulfur / w%	Ni / wppm	V / wppm
Daqing	32	0.07	3 .9	0.10
Shengli	25.4	0.73	30	1.80
Liaohe	24.3	0.18	32.5	0.6
Renqiu	27.9	0.31	15	0.7
Xinjiang	33.4	0.05	5.6	0.07
Zhongyuan	34.8	0.52	3.3	2.40

Table2 Major Heavy Oil Conversion Technologies in China

Item	Capacity / (Mt/a)	%
Crude processing	200	100
FCC	56.0	28.0
Delayed coking	12.5	6.3
Visbreaking	8.2	4.1
Hyrocracking	9.2	4.6

The rapid growing of the refining industry greatly enhances the development of petrochemical industry in China. The production capacity of ethylene reached 3 Mt/a at the end of 1995, and the downstream processing technologies for petrochemicals developed correspondingly along with the increasing capacity of ethylene production. In 1995, 4.3 Mt of synthetic resins, 0.45 Mt of synthetic rubbers and 2.8 Mt of synthetic fibres were produced in China.

1. ADVANCES OF TECHNOLOGY

The progress of science and technology has played a significant role as the driving force of development for China's refining industry. China has attached great importance to the scientific and technology work in the area of petroleum processing through R&D in the past decades.

A number of significant achievements have been made and quickly applied to commercial purposes, which have raised the technical competence of the whole refining industry. The main technological advances in recent years are briefly described as follows:

1.1 Resid Fluid Catalytic Cracking

The first resid FCC unit with a capacity of 1.0 Mt/a using Daqing AR as Feedsock was put on stream at SINOPEC Shijiazuang Refinery in 1983. Since then, significant progress has been made in the field of resid FCC. Many new technologies and equipments have been employed, such as single and two stage regenerators, internal and external catalyst coolers, new feed injection nozzle system, PV type cyclone and third stage separator, and flue gas expander for energy recovery, etc.. Simultaneously, a series of new zeolite type FCC catalysts, with good coke selectivity for resid FCC have been developed and commercialized by SINOPEC.

The residues of most Chinese crudes because of their low levels of sulfur and heavy metals content are good feedstocks for resid FCC, with high yield of light products. There are 35 FCC units using AR or VR blending as feedstocks in SINOPEC, and more than 10 Mt of residues were processed in 1995. The typical operating data is shown in Table 3.

1.2 New Family Technologies Derived from FCC

Based on the FCC technology, some new processes, such as DCC, MGG, and MIO have been developed by SINOPEC aimed at producing different types of light olefins using heavy oil as feedstocks.

1.2.1 Deep catalytic cracking (DCC)

DCC is a new process aimed at maximizing propylene production. An existing FCC unit was revamped into a 60 kt/a demonstration plant in 1990 at the SINOPEC Jinan Refinery. The test of DCC in the demonstration plant with a shape-selective zeolite catalyst, using Linpan (an intermediate base crude) VGO as the feedstock, at a temperature below 560℃, gave an ethylene yield of 4 % ~ 5 %, propylene 19 %, and butylene 14 %. This technology has been used by some refineries. A 400 kt/a commercial unit was built at Rrefinery of SINOPEC Anqing General Petrochemical Works and was put on stream in 1995, the propylene from this unit has been used as the raw material for production of acryl fibre.

Table 3 Typical Data of a Commercial Resid FCCU

Item	Data
Feedstock	Daqing AR
Specific gravity	0.89
Conradson carbon / w%	4.6
Ni+V / wppm	6.4
Sulfur / w%	0.17
Operating conditions	
Riser outlet temp./ °C	519
Recycle ratio	0.25
Reactor pressure / MPa	0.12
Catalyst	REHY
Catalyst/oil	6.8
Yield / w%	
Dry gas	4.0
LPG ($C_3 + C_4$)	9.0
Gasoline	46.1
Light cycle oil	25.1
Slurry oil	6.9
Coke	7.8
Gasoline RON	88.0

1.2.2 Maximum gasoline plus gas production (MGG)

MGG is another novel process which is aimed at producing maximum gasoline with high octane and LPG. The first commercial unit with a capacity of 500 kt/a went on stream in 1992 in SINOPEC Lanzhou Petroleum Processing and Chemical Complex. The total yield of gasoline plus LPG can be as high as 80 %, in which the ratio of gasoline to LPG is around 4 : 3.

1.2.3 Maximum isoolefin production (MIO)

MIO is also a new process used for maximizing the yield of isobutene and isoamylene, using a new catalytic material for manufacturing MIO catalyst. A succesful commercial trial was carried out on a revamped FCC unit in SINOPEC Lanzhou Petroleum Processing and Chemical Complex in 1995. The total yield of isobutene plus isoamylene was more than 10 %, and gasoline 45 % with RON 93.

1.3 Hydrocracking

The development of hydrocracking process and catalysts also has been carried out by SINOPEC. Two commercial hydrocracking units with a capacity of 800 kt/a and 1 Mt/a respectively have been built and put on stream in Zhenhai Refining & Chemical Co., Ltd. and SINOPEC Liaoyang Petrochemical Fibre Co..

A new catalyst 3825 was successfully used in a high pressure hydrocracking unit at the Shanghai Petrochemical Co. Ltd., in 1991 with 52 % yield of heavy naphtha and 13.7 % jet fuel. Catalyst 3903, for maximum middle distillate production, has been proved to have good activity, selectivity and stability through long time operation in a commercial unit.

A series of residue hydrotreating catalysts, including those for hydrodemetalization, hydrodenitrogenation and hydrodesulfurization were systemically developed by SINOPEC. The test in a commercial unit showed good results by using Gudao (a naphthenic base sour crude with high nitrogen content) VR as feedstock. The metal removal was 80 %, sulfur removal 83 %, nitrogen removal 50 % and Conradson carbon removal 55 %. A SRHT (SINOPEC Residue Hydrotreating process) commercial unit is under construction.

1.4 Medium Pressure Hydrotreating

1.4.1 A mild hydrocracking process has been successfully developed in recent years. This process set up a new route for production of high quality feedstock for steam cracking from VGO. A demonstration unit was built and put into operation in SINOPEC Qilu Petrochemical Co.. By using Shengli VGO as feedstock and under a hydrogen partial pressure of 6.4 MPa and temperature of 325 ~ 410°C. About 32 % of light fraction was obtained. The 60 % yield of unconverted oil with a low BMCI value of 17.3 was used as feedstock for steam cracking, with an ethylene yield of 26.6 % (single pass), higher than the ethylene yield of Shengli AGO, which is only 22 %.

1.4.2 Medium pressure hydroupgrading process (MHUG) is also a new process for improving the quality of light cycle oil from FCC. Under the medium hydrogen partial pressure, a diesel fuel with low sulfur and high cetane number or a jet fuel could be obtained, the naphtha is a good feedstock for catalytic reforming, and the unconverted oil may also be used as a good feedstock for steam cracking.

1.4.3 Hydrotreating process for AGO, VGO, LCO and CGO at medium hydrogen partial pressure was developed rapidly. The total capacity of various hydrotreating units reached 19 Mt/a in 1995. RN-1, a catalyst with good activity both in HDN and HDS, developed by SINOPEC, has been applied in 33 hydrotreating units in China.

1.5 Catalytic Reforming

Research on catalytic reforming and related catalysts has been carried out in China since early 1950s. The first commercial catalytic reforming unit was built in 1965 at the Daqing Refinery. By 1995, the total capacity of catalytic reforming reached 8 Mt/a.

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