

**COAL LIQUEFACTION TECHNOLOGY DEVELOPMENT IN NEDO**

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456/417

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### 1. Introduction

As coal has much larger amount of deposit than petroleum, it is supposed to be inevitable technology in the future to convert the coal to liquefied fuel. The opportunity to commercialize the coal liquefaction will be come without fail, though it has postponed from the time we imagined before.

Technology of coal liquefaction was industrialized in West Germany for the first time through the World War 2nd era, but it was not so economy to be competitive to abundant petroleum afterward. Research on coal liquefaction has become active again in U.S.A. since 1960s and in West Germany since 1970s, thinking future energy crisis.

In Japan research on coal liquefaction was carried on a little during and after the World War 2nd. But it stopped for a long time. In 1974 Sunshine Project was started by MITI in which research program on liquefaction and gasification of coal, solar energy and geothermal energy were included.

### 2. Coal Liquefaction Program in Sunshine Project

Sunshine Project intends to develop a new clean energy production technology except petroleum and nuclear power. It is the first long term technology development projects in Japan. Coal liquefaction technology takes important position in it

In coal liquefaction project three processes, that is Extractive Coal Liquefaction, Direct Hydrogenation and Solvolysis Coal Liquefaction were taken up at first. Later Australian Brown Coal Liquefaction Project was added. Now four coal liquefaction projects are kept going. These projects are commissioned to the contracted company and at present whole research cost is supported by the Government.

### 3. Establishment of NEDO

Establishment of NEDO caused drastic change for the coal liquefaction technology development in Japan. NEDO was established in October 1980 according to the special law to promote the general development of technology for the purpose of commercializing alternative energy resources. NEDO has succeeded the Sunshine Project and has been promoting the Projects more intensively.

Disclaimer—Reference in this report to any specific commercial product process or service is to facilitate understanding and does not necessarily imply its endorsement or favoring by NEDO.

#### 4. NEDO's Policy for Coal Liquefaction Process

NEDO should be thought that it is not only an administrative organization, but has technical policy for process development and leads the contractors to get the better results.

As our purpose is commercialization of coal liquefaction, we intend to develop the more economical process than other processes in development in other countries. Process conditions to be economical are as follows.

- (1) to use the catalyst cheap but of high efficiency
- (2) to use hydrogen donor solvent
- (3) to adopt the two stage hydrogenation

Our processes are consist of one or some of these conditions. In addition to this, NEDO is developing the new material for high pressure and high temperature vessel and the computer program for the total system analysis of coal liquefaction.

#### 5. Status of Each Coal Liquefaction Process

##### 5.1 Extractive Coal Liquefaction Process

###### a) Description of the Process

Crushed feed coal and disposable catalyst are slurried in hydrogen donor solvent, which is the middle and heavy oil from fractionator and is catalytically hydrogenated. This slurry and hydrogen are sent to the reactor through the preheater. Reaction condition is rather mild, but oil yield is rather high. Products from the reactor is fractionated in both atmospheric and vacuum distillation column. Most part of middle and heavy oil is used as recycle solvent.

Bottoms from vacuum column can be used as (1) material for gasification to get hydrogen, (2) material for solid separation to get more light oil by second hydrogenation, (3) feed stock for various use after solid separation. We are now discussing these three methods of utilizing of bottoms.

As disposable catalyst, noble catalyst, iron ore, red mud and dust from molten iron gasifier were tested. From the results of these tests dust from molten iron gasifier was chosen as most active and cheap catalyst (Table.1). In case that the molten iron gasifier is taken into the process, the catalyst is recovered when bottoms are gasified.

###### b) Status of Process Development

Basic research was started on 1978, and at the end of November 1981, P.D.U. with capacity of 1 t/day of feed coal was completed in Hasaki, Ibaragi. The P.D.U. test will be continued to get the design basis for the pilot plant.

##### 5.2 Direct Hydroliquefaction Process

Crushed feed coal and disposable catalyst are slurried in recycle solvent. This slurry and hydrogen are sent to the reactor through preheater. Though this is an orthodox process, we are trying to reduce the reaction pressure by using highly active iron catalyst which can be produced with such low cost as is disposable.

#### b) Status of Process Development

Basic research of this project started in 1975. Bench Scale Unit with capacity of 0.1 t/day of feed coal was constructed in 1980, and P.D.U. of 2.4 t/day was completed in Kawasaki at the end of April 1982. In B.S.U. now we use red mud as catalyst. The example of results is shown in Table 2. But in autoclave test we recognized the high efficiency of new type catalyst, and at present, we are developing the technology for reducing the production cost of the catalyst.

### 5.3 Solvolysis Coal Liquefaction Process

#### a) Description of the Process

At first this process started as one using asphalt from petroleum as solvent. But later, the process has improved. In the present process, crushed coal is slurried in recycle solvent. Slurry is liquefied in first stage by short contact time reaction. Products were deashed and sent to hydrogenation reactor where they are catalytically hydrogenated. Products were distilled and heavy fraction is used as recycle solvent. Fig.3 shows the flow sheet of Solvolysis Coal Liquefaction Process.

#### b) Status of Process Development

B.S.U. of 0.1 t/day including whole the process of Solvolysis was constructed in Hiroshima in 1980. Example of products yield is shown in Table 3. Now we are studying on condition of first stage and improvement of catalyst and discussion of reactor form in second stage.

### 5.4 Brown Coal Liquefaction

#### a) Description of the Process

This process is developed for the Brown Coal of Latrobe Valley Victoria, Australia. This Brown Coal has little ash and sulfur content, but about 60% of water content ( Table 4 ).

Brown Coal and catalyst are slurried in recycle solvent. Coal is dewatered as slurry and sent to first stage hydrogenation. Products of first stage are deashed by solvent deashing process and catalytically hydrogenated in second stage reactor, producing various product oil. A part of heavy oil is recycled to slurry preparation.

#### b) Status of Process Development

Basic research has been mainly conducted using B.S.U. of 0.1 t/day in Kobe, Japan. According to the results of basic research, the P.D.U. with the capacity of 50 t/day of feed coal has been under construction in Victoria since the end of 1981. Schedule of construction is divided into two steps. First step construction aims to establish first stage liquefaction technology, and will be completed in 1983.

Some test results about the first stage hydrogenation are shown in Table 5. At present, we are making research on optimum share of hydrogenation rate between first and second stage reaction.

### 5.5 Development of Heavy Section Pressure Vessel Steel

The reactor of the commercial coal liquefaction plant will be designed for higher temperature and pressure than usual petroleum refinery

reactor, and the size of the reactor will become gigantic. For the purpose to manufacture the reactor vessel for more efficient reaction condition with lower cost, it is necessary to develop the new material which has higher design stress at high temperature and high hydrogen partial pressure, and to develop the manufacturing technology both in reasonable cost.

By alloy modification of 3Cr-1Mo steel, we are developing the steel which has design stress of 30 % improved at 480°C and enough hardnability at more than 450 mm thick. It has good characteristics in other items, too. We have already ascertained the uniformity of composition in midium size shell made from ingot weight of 80 t. Now we are manufacturing commercial size shell made from ingot weight of 250 t.

We are thinking of codification of modified steel. To facilitate the ASTM and ASME codification, confirmation test with the samples and production of actual three heats will be carried on.

#### 6. Future Programs

Development of coal liquefaction technology in Japan is at the step of P.D.U.. Compared with U.S.A. and West Germany. We are behind by a few years about the size of plants. But we are making effort to improve the process and getting good results.

As the next step, NEDO evaluates the experimental results of Extractive Coal Liquefaction, Direct Hydroliquefaction and Solvolysis Coal Liquefaction Processes, and now NEDO is consulting with the concerned agencies about the construction of a Pilot Plant with the capacity of 250~500 t/day of feed coal in a few years.

Australian Project will complete the second step construction in 1985 in our present schedule. After a few years' operational study, we will establish the most appropriate process for the Brown Coal of Latorobe Valley Victoria.

Table 1 Example of Oil Yields by Extractive Coal Liquefaction Process

Catalyst	Oil Yields Z	
	C <sub>5</sub> ~538°C	C <sub>5</sub> ~180°C
none	37	20
Ni-Mo	56	39
Pyrite+S	45	20
Iron Ore+S	48	18
Red Mud+S	49	21
Sulfur	40	19
Dust from Molten Iron Gasifier+S	50	20

Dry coal basis autoclave test  
 Temperature: 450°C, Pressure: 170 kg/cm<sup>2</sup>  
 Wandoan coal ( Sub-bituminous coal)  
 Reaction time 1 hour

Table 2 Example of Products by Direct Hydroliquefaction Process

Component	Yields wt%
Gas	9.1
H <sub>2</sub> O	17.2
Oil (C <sub>5</sub> ~530°C)	55.6
SRC and Unreacted Coal	22.3
Hydrogen	-4.2
Total	100.0

Dry ash free basis

Table 3 Example of Products by Solvolysis Liquefaction Process

Component	Yields wt%
Gas and H <sub>2</sub> O	7.4
Oil (C <sub>5</sub> ~450°C)	40.5
SRC	40.8
Unreacted Coal	15.0
Hydrogen	-3.7
Total	100.0

1. Maf basis
2. Ash of coal: 8.7%

Table 4 Analysis of Victorian Brown Coal

		Yaliouru	Morwell	Gellion- dale	Loy Yang	Rhein. (Germany)
Moisture	%	66.1	60.3	66.1	62.8	59.0
Ash	%	1.1	3.4	6.5	1.4	5.9
Volatile Matter	%	51.2	47.9	48.1	51.5	53.5
Fixed Carbon	%	47.7	48.7	45.4	47.1	40.6
Elemental Analysis						
(d.a.f.) %						
C		68.0	70.5	66.7	69.5	67.8-68.7
H		4.9	5.0	4.7	5.0	5.0- 5.1
S		0.3	0.4	1.1	0.4	0.4- 0.8
Cl(d.b.)		0.1	0.1	0.3	0.1	-

Table 5 Some Test Results About the Primary Hydrogenation

Products		H <sub>2</sub> Consumption wt%				
		3	4	5	6	
GAS	CO, CO <sub>2</sub> , H <sub>2</sub> O etc.	24.5	26.5	28	30.5	
	C <sub>1</sub> ~ C <sub>4</sub>	5.0	7.0	11	16	
	Total	29.5	33.5	39	46.5	
OIL	Naphtha C <sub>5</sub> ~ 180°C	7	13	18	23	
	HS {	180 ~ 420°C	7	5.5	4	2
		+420°C	6.5	12	12	10.5
	Total	20.5	30.5	34	35.5	
HI	BS-HI	33	26	23	18	
	PS-BI	20	14	9	6	
	Total	53	40	32	24	
TOTAL		103	104	105	106	

HS: hexane soluble      HI: hexane insoluble  
 BS: benzene soluble     BI: benzene insoluble  
 PS-BI: pre-asphalten  
 BS-HI: asphalten



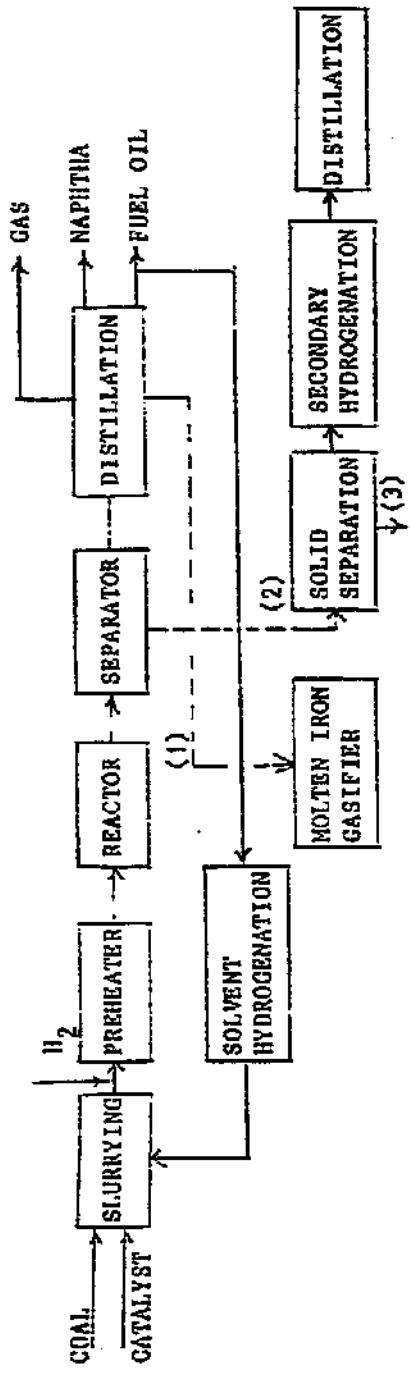


Fig. 1 Extractive Coal Liquefaction Process Flow

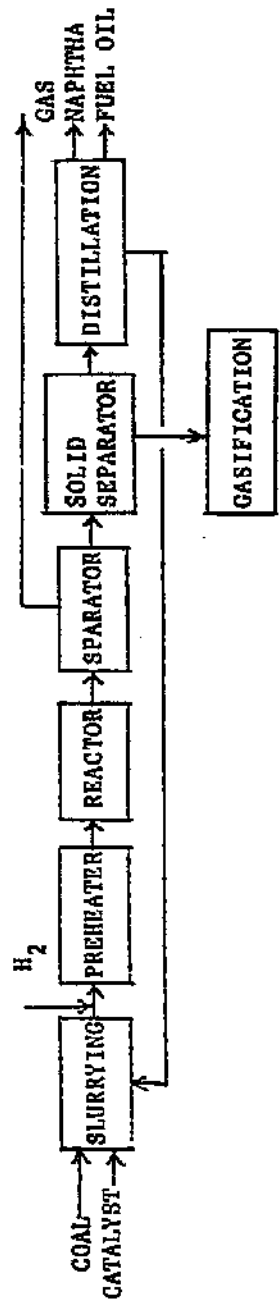


Fig. 2 Direct Hydroliquefaction Process Flow

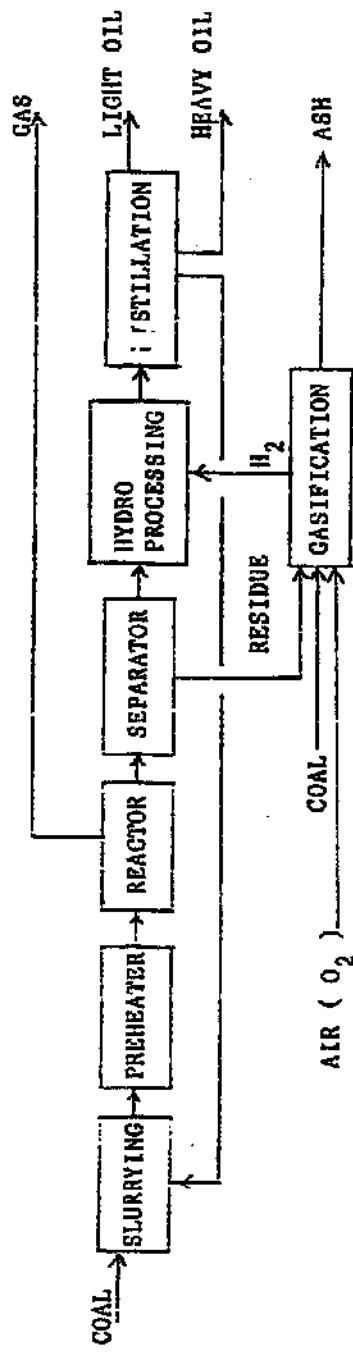


Fig. 3 Solvolysis Coal Liquefaction Process Flow

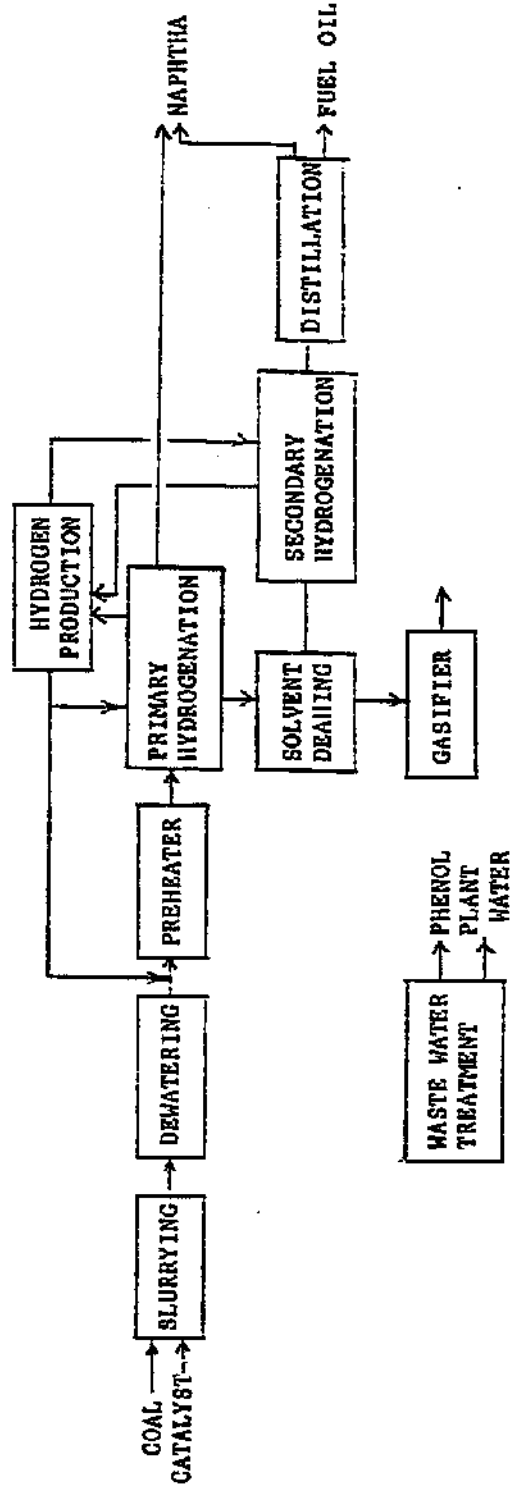


Fig. 4 Brown Coal Liquefaction Process Flow