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## **RESEARCH AND DEVELOPMENT OF OIL SHALE IN JAPAN**

### **Abstract**

The Japan National Oil Corporation (JNOC) has promoted research and development of oil shale under the guidance of the Ministry of International Trade and Industry (MITI) in cooperation with 35 Japanese companies in various industrial fields including iron and steel, heavy machinery, mining and so on.

Three types of plants (vertical shaft, circular grate and cross flow type) were constructed to promote oil shale research and development on a 3 ton/day bench scale. These were designed and constructed in 1982 and operated through 1983. On the basis of the experimental results, the process of the 300 ton/day pilot plant for research of commercial plants consisted mainly of a vertical shaft type retort.

In addition to a vertical shaft type retort, as a retorting/gasifying unit, crushing, screening, oil recovery and heat recovery units, etc. were installed. The individual units, installation work and trial runs were completed by the end of 1986. The formal operation for research started in 1987 and totaled 270 days (including a 100-day continuous operation) by the end of 1988. The pilot plant operated smoothly and stably. JNOC dismantled the pilot plant by the end of March, 1989 and the research and development activities of the pilot plant operations were successfully completed as scheduled.

Based on the results of pilot plant operations, JNOC conducted the conceptual study of a 50,000 ton/day commercial plant for processing Condor shale in Australia and Maoming shale in China under certain assumptions, which are described later.

### **Introduction**

In October 1980, JNOC organized a new project department to investigate and study the worldwide distribution, present development situation and future development feasibility of oil shale resources. The research and development on oil shale technology supported by MITI has been conducted mainly by JNOC. Commissioned by JNOC, Japan Oil Shale Engineering Company, Ltd. (JOSECO) has been engaged in the research and development of oil shale retorting process. JOSECO was established in 1981 as a joint venture of 35 Japanese leading companies in various fields, that is, the iron and steel, heavy machinery, mining, plant engineering, cement, oil refining and trading. Japan has no oil shale resources of its own. Therefore, the main objects are to promote the research and development of retorting and other technologies for oil shale and to give technological cooperation to other countries in developing their oil shale. This paper presents research and development of oil shale in Japan.

### **Research and Development Program**

In participating in oil shale research and development, we decided to devise and develop new technologies. In order to realize these new technologies, there were

several factors of research and development in oil shale technologies as follows:

1. High adaptability to various kinds of oil shale
2. High process ability
3. Simple operation
4. Clear scale-up concept
5. Economical process

The 9-year research and development program was divided into two phases.

Phase 1: Research on the basic properties of various kinds of oil shale and research using three types of 3 ton/day bench-scale retorting plants.

Phase 2: Research using a 300 ton/day pilot plant based on the results of Phase 1 and conceptual study of a 50,000 ton/day commercial plant based on the results of the pilot plant operation.

### Outline of Phase 1

**Basic Study of Oil Shale.** To understand the behaviors of oil shale in the retorting process and the properties of shale oil, experiments were conducted on the physical and chemical properties of oil shale from 12 districts in seven countries (United States, Australia, China, Brazil, Jordan, Morocco and Sweden). 91 kinds of oil shale samples were investigated.

**Bench-scale Retorting Process Description.** Three types of 3 ton/day bench-scale retorting plants (vertical shaft, circular grate and cross flow type) were designed and constructed in 1982 and operated through August, 1983 to confirm the basic process conditions and obtain the design data for the pilot plant. The main sources of oil shale for these experiments were Colorado in U.S.A., Timahdit in Morocco, Condor in Australia and Maoming in China. Each bench-scale retorting plant has several features.

#### 1. Vertical shaft type retort:

- (a) Simple furnace structure in which the retorting and gasifying zones are integrated
- (b) Stable operation against external disturbances such as the variations in water content of feed shale, etc.
- (c) High heat efficiency on account of counterflow of gas and shale

#### 2. Circular grate type retort:

- (a) Arbitrary zoning of process functions (preheating, retorting, combustion and cooling)
- (b) Independent control of operational conditions in each zone
- (c) High reduction of size degradation on account of absence of movement in charged layer of shale

#### 3. Cross flow type retort:

- (a) Low pressure loss
- (b) High combustion efficiency of residual carbon on retorted shale by fluidized-bed combustor which is combined with the retort.

There were no major differences among the conclusions of evaluation in three types of retort in terms of the ease of scale-up and the stability to the different shale.



But the vertical shaft type retort was selected for the pilot plant, considering some factors such as simplicity of structure, operational stability and economic feasibility of commercial-scale plant.

## Outline of Phase 2

**Objectives.** The objectives of Phase 2 are to construct a 300 ton/day pilot plant on the basis of the results of bench-scale plant operation, to identify the conditions and specifications for the future commercial plant and to establish original oil shale technology through its operations. The work of Phase 2 has been 75 % funded by MITI through JNOC and 25 % by private companies.

**Composition of Pilot Plant.** The pilot plant was located in Kitakyushu city approximately 1,000 km west of Tokyo. The major elements of the pilot plant were vertical shaft-type retorting and gasifying facilities. Circular grate type drying facilities were installed to study the economics of pre-drying high water content of Maoming shale in China. Fluidized-bed combustion facilities were also juxtaposed to study the high efficiency combustion of carbon in raw powder shale and residual carbon in retorted shale (Fig. 1).

**Pilot Plant Description.** The raw shale delivered from the storage yard is crushed and screened to the size of between 6 and 70 mm. The 6-70 mm sized Maoming shale containing 17 wt% of free moisture is fed to drying facilities and dried to an average free moisture content of 10 wt% by hot gas at 150 °C. The raw or dried shale is screened to remove fines generated during handling. Screened shale is weighed and fed from the top of the retort. The retort is composed of two chambers which are connected with two connecting pipes. The upper chamber is a retorting zone, where the kerogen is pyrolyzed by hot gas at 550-600 °C injected into the

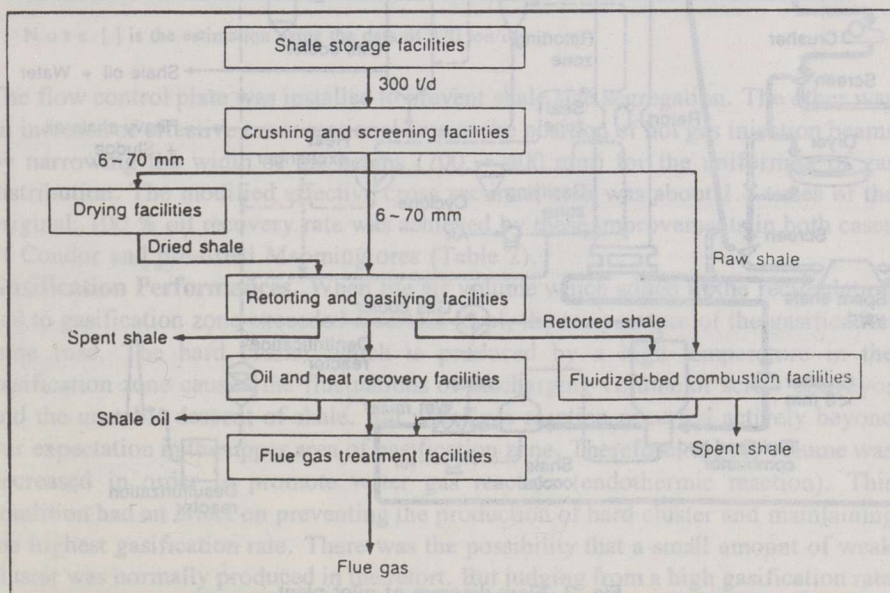


Fig. 1. Block diagram of pilot plant





## Results of Operations

**Retorting Performances.** An important condition of stable operation in the retorting and gasification zone was the smooth descent of shale. When the pressure loss of the retorting zone exceeded 1700 mm Aq by an increase of the hot recirculation gas supply volume to the retorting zone, the descent of shale became unstable. The maximum gas space velocity which maintained the smooth descent of shale in the retorting zone was 0.6-0.7 m/sec. The amount of maximum processing ores for a stable operation was about 220 ton/day in Condor and 240 ton/day in pre-dried Maoming ores by the restriction of the acceptable hot gas supply volume. To prevent Maoming containing high free moisture, pre-drying contributed to the increase of the amount of maximum processing ores in order to suppress the thermal-shock of ores (Table 1). In order to achieve a high oil recovery rate, two parts of the retort were improved. One was the improvement of the charging device for the uniform distribution of ores. Shale size in the cross sectional center of the retort was smaller than the border of the retort before the modification of the charging device. An increase of the permeability resistance by shale size segregation caused a drop in temperature and retorting was insufficient in the cross sectional center of the retort.

Table 1. Maximum Capacity for Stable Operation

Ore	Free moisture (%)	Max. gas volume (Nm <sup>3</sup> /h)	Max. capacity (ton/day)
Condor	8	10,300	220
Maoming	14	7,800	140
Maoming	7-9 (pre-dry)	8,200	170
Maoming	3-6 (pre-dry)	[9,700]	[240]

Note. [ ] is the estimation using the data of 220 ton/day.

The flow control plate was installed to prevent shale size segregation. The other was an increase of effective cross sectional area at the position of hot gas injection beams by narrowing the width of the beams (700 → 400 mm) for the uniformity of gas distribution. The modified effective cross sectional area was about 1.4 times of the original. 100 % oil recovery rate was achieved by these improvements in both cases of Condor and pre-dried Maoming ores (Table 2).

**Gasification Performances.** When the air volume which added to the recirculating gas to gasification zone exceeded a certain level, the temperature of the gasification zone rose. The hard cluster which is produced by a high temperature in the gasification zone caused the fluctuations of discharging volume of screw conveyor and the unstable descent of shale. But water gas reaction occurred actively beyond our expectation in the upper area of gasification zone. Therefore, the air volume was decreased in order to promote water gas reaction (endothermic reaction). This condition had an effect on preventing the production of hard cluster and maintaining the highest gasification rate. There was the possibility that a small amount of weak cluster was normally produced in the retort. But judging from a high gasification rate of 78-85 % in Condor and 65-75 % in Maoming, the straight furnace configuration which has no reduced-sectional area portions and the screw conveyors which have

Table 2. Main Results of Operation

Indices	1987 FY operation			100-day continuous operation
	Ore			
	Condor		Maoming	
	Run No.			
	R-C12*	R-C11	R-A14	R-A
Capacity (ton/day)	200—500	220	170—200	115—220
Oil yield (%)	79—96	99	96—102	100
Gasification rate (%)	78—85	-	65—75	72

\*R-C12 is the operation before modification of charging device and hot gas injection beams.

forced-discharging ability were very effective for the smooth descent of shale (Table 2).

**Heat Recovery Performances.** About 70 % of the heat recovered from the gasifying process was transferred to the recirculating retort gas through two heat exchangers which were installed in series. The remaining heat was recovered by another heat exchanger and released into the atmosphere. Due to a layer of very fine dust clogging to the inside of the heat exchanger tube and the heat exchangeability dropped below the design value during one day's operation or so. Therefore, a steel shot cleaning device was installed and operated periodically to maintain the design value of the heat exchangers (Fig. 3).

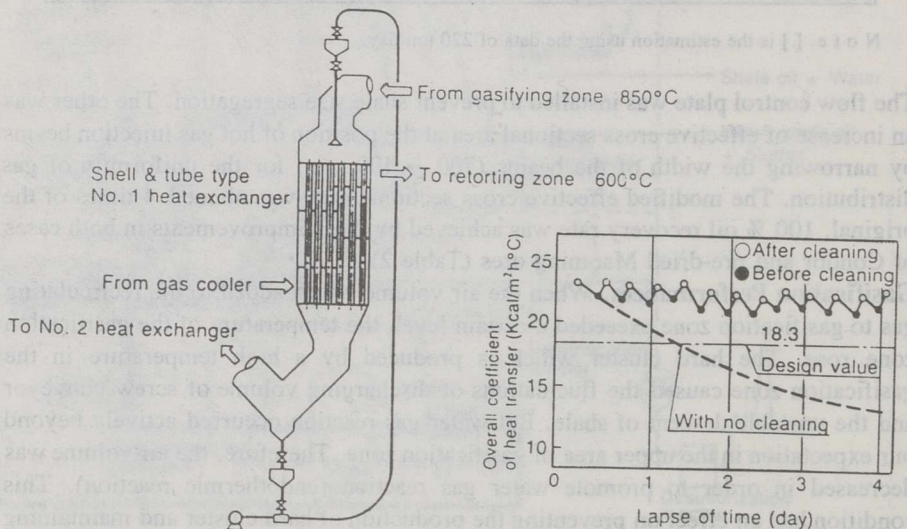


Fig. 3. Changes of heat exchange ability and shot cleaning device



**Fluidized-bed Combustion Performances.** In a combustion test of the retorted shale, where the average feed shale size was 1.5-2.9 mm, the combustion efficiency was 93-96 % in Condor and 90-93 % in Maoming. In a combustion test of raw shale where an average feed shale size was 1.7-8.6 mm, the combustion efficiency was over 99 % in both Condor and Maoming. The stability of shale size that had an effect on temperature distribution was very important for the stable operations. The high density materials such as  $\text{Fe}_2\text{O}_3$  and  $\text{SiO}_2$  settled in the bottom of fluidized-bed. This phenomenon that is called "segregation" caused a drop in temperature and an increase of the permeability resistance. To overcome this problem, the gas space velocity was increased temporarily to blow off the settled material. It is necessary to install a discharge system in the bottom of the fluidized-bed for the prevention of segregation. A considerable amount of ultrafine dust that was produced by the gasification of volatile matter in shale and the collision of shale were generated in the combustion and carried over to the downstream (Table 3).

Table 3. Size Degradation Rate and Dust Carryover Rate

Material	Size degradation rate (%)	Dust carryover rate (%)
Raw shale	60—90	20—55
Retorted shale	0—40	5—20

$$\text{Size degradation rate (\%)} = \left( 1 - \frac{\text{average shale size after combustion}}{\text{average feed shale size}} \right) \times 100$$

The inside wall of the heat exchanger tube which cooled the waste gas was covered with the sticky ultrafine dust. Therefore a steel shot cleaning device was installed and operated periodically (Table 4).

Table 4. Typical Operation Data of Fluidized-Bed Combustor

Indices	Feed ore			
	Condor		Maoming	
	Raw shale	Retorted shale	Raw shale	Retorted shale
Combustion performances:				
1) Feed rate, t/h	2.91	6.3	2.61	4.5
2) Particle size, mm	3.5	2.4	2.9	2.6
3) Supply air vol., $\text{Nm}^3/\text{h}$	5,150	5,620	5,800	6,300
Temperature, $^{\circ}\text{C}$	169	392	97	276
Pressure, mm Aq	450	615	400	570
4) Combustion temperature, $^{\circ}\text{C}$	900—920	870—890	850—900	880—900
5) Combustion efficiency, %	99—100	94—95	99—100	92
6) Dust carryover ratio, %	24	8	44	15

**Circular Grate Drying Performances.** Three types of flow mode (2 pass through, 1 pass through, 1 pass through and partial gas recycle) were tested. Two pass through flow mode that we expected in an effective use of energy had little effect on drying and uniforming the free moisture content of the shale. One pass through and partial gas recycle flow mode was more suitable for economical and operational efficiencies. The downward gas flow caused a drop in gas temperature and a high moisture content of gas in the lower part of shale layer. When the feed shale containing free moisture of 10-16 % was dried to an average free moisture content of 7-10 %, the free moisture of dried shale varied widely in the vertical direction of the shale layer. After the operational condition of free moisture content changed 3-6 %, free moisture of dried shale was within the limit of  $\pm 1$  % against an average. In the drying operation, care had to be taken to prevent the natural burning of dried-shale in the storage hopper, etc. To counteract this, a CO-detector, a sprinkler and an inert gas blowoff were installed in the storage hopper.

### The Conceptual Study of Commercial Plant

On the basis of the results of the pilot plant operations, the following conceptual studies of commercialization were conducted under the assumptions that the plant was to be constructed in Australia or China without restrictions of layout.

- (1) Basic design of the oil shale commercial plant
- (2) Estimation of the shale oil production cost

The plant capacity of conceptual design is 50,000 ton/day (feed capacity of retort). In design of plant, Fluidized-bed boiler unit, Off gas boiler unit, Shale oil hydrotreating unit, Retorting gas treatment unit, Hydrogen unit and Waste water treatment unit are added to the fundamental unit of pilot plant in order to realize the closed-system as much as possible.

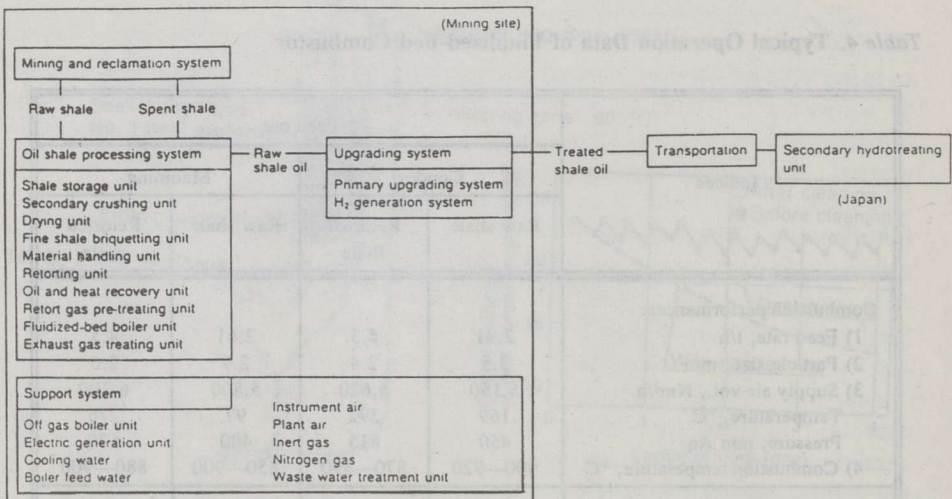


Fig. 4. Scope of estimation



Based on this design, the production cost of shale oil was estimated by simulation computer program. The simulation program was developed to simulate a total system of oil shale plant and put out material balance, utility consumption and economic calculation in 1982-1983 and revised for an accurate simulation on the basis of the results of the pilot plant operations in 1989. This estimation includes mining and reclamation, but does not include infrastructure and land required (Fig. 4). The capital cost is \$1,57 million in Condor and \$1,972 million in Maoming. The shale oil production cost is \$30.28/bbl in Condor and \$31.99/bbl in Maoming under the following assumptions.

- (1) Project life 20 years
- (2) Depreciation 15 years
- (3) Salvage rate 10 % of capital cost
- (4) Long term dept 90 % of capital cost
- (5) No restrictions of plant layout
- (6) Shale oil shall be shipped after hydrogen treatment to the level of 1,000 ppm or less
- (7) Final hydrogen treatment shall be conducted at the existing refineries in Japan
- (8) By-products (sulfur and gypsum) and surplus power shall be sold outside the plant
- (9) Environmental protection outside the plant battery, infrastructure, royalty, etc. shall not be included

Each condition and assumption of estimation may change according to each project.

## Conclusions

Our research and development of oil shale through the pilot plant operations were successfully completed as scheduled. We believe our expected objectives were achieved and we are confident that our results show this process has a high reliability.

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