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RETORTING OF PULVERIZED OIL SHALE IN FLUIDIZED-BED PILOT PLANT

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This paper describes the research on fluidized-bed retorting of pulverized oil shale on pilot-plant scale (daily processing capacity of 50 tonnes oil shale). Chinese Yi Lan pulverized oil shale 0–3 mm was used as feed. Oil shale is fed and pyrolyzed in a fluidized-bed pyrolyzer, where it is heated to about 500 °C by recycled hot shale ash coming from a fluidized-bed combustor. Nitrogen, coal gas or superheated steam is used as fluidizing agent in the pyrolyzer. The shale char formed from oil shale in the pyrolyzer mixed with shale ash is recirculated to the fluidized-bed combustor, where it is burnt by incoming air (as fluidizing agent) to form shale ash at about 700 °C and is recycled to the pyrolyzer as hot solid heat carrier. The mixture of shale oil vapors and pyrolysis gas, formed from oil shale, mixed with steam or other fluidizing agent coming from the pyrolyzer, comes through two cyclones for removal of entrained dust and is washed by warm water to condense heavy shale oil, then goes to cooler for recovery of light shale oil. The total shale oil yield accounts for 80% of Fischer assay. The properties of shale oil and pyrolysis gas are analyzed.

Introduction

Fluidized-bed retorting of pulverized oil shale has the advantages of fully utilization of mined oil shale (after crushing and grinding), with the shortest pyrolysis reaction time and highest retorting intensity in comparison with ordinary retorting technologies for processing lump oil shale.

Early in the nineteen forties, fluidized-bed retorting of pulverized oil shale was conducted in the USA and Australia. In 1947 US Mobil Research & Development Corp. developed fluidized-bed retorting technology of oil shale, with utilization of air and steam [1].

In the nineteen seventies Chinese Maoming Petroleum Industry Company conducted two-vessel fluidized-bed retorting technology, i.e. fluidized-bed pyrolysis of pulverized oil shale and fluidized-bed combustion of shale char on pilot scale (daily processing 24 t pulverized oil shale), and achieved preliminary success [2].

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In recent years, China National Coal Group Corp. has been supporting Longhua Harbin Coal Chemical Company (former Harbin Coal Gasification Factory) in Yilan, Heilongjiang, to conduct pilot-scale test of two-vessel fluidized-bed retorting for processing Da Lian He oil shale (50 t/d) [3]. Several test runs and improvements have been made, and the unit can now be operated continuously, with shale oil yield higher than 80% of Fischer assay. Besides, shale oil and other pyrolysis products are analyzed in detail.

Process scheme of Longhua pilot-scale fluidized-bed retorting of oil shale

The Longhua pilot-scale unit (50 t/d) consists of two-vessel fluidized-bed reactors, i.e. the fluidized-bed pyrolyzer of pulverized oil shale and the combustor of shale char. The two reaction vessels are set up parallel with each other. Two U tubes between them serve for circulation of solid materials. Figure 1 shows the schematic scheme of the pilot plant. Chinese Yi Lan pulverized oil shale (0–3 mm) was used as feed. Oil shale is fed and pyrolyzed in the fluidized-bed pyrolyzer, where it is heated to about 500 °C by recycled hot shale ash coming from the fluidized-bed combustor through a U tube. Nitrogen, coal gas and superheated steam have been used separately as fluidizing agent in the pyrolyzer. Shale char formed from oil shale in the pyrolyzer mixed with shale ash is recirculated to the fluidized-bed combustor (through another U tube), where it is burnt by incoming air (as fluidizing agent) to form shale ash at about 700 °C and then recycled to the pyrolyzer as hot solid heat carrier. Shale oil vapors and pyrolysis gas mixed with steam or some other fluidizing agent coming through two cyclones, installed on the top of the pyrolyzer for removal of entrained dust, leave the pyrolyzer and are scrubbed by warm water. Part of heavy shale oil condenses and mixes with dust, forming oily sludge. Thus only a small amount of dust is left in the scrubber-off gas, preventing the blockage of the system and ensuring normal operation of the unit. The scrubber-off gas is cooled and shale oil is condensed. The non-condensable gas is of high heating value. The oily sludge is treated by an appropriate separation method to get heavy shale oil and water, with sludge residue left behind.

Operation of Longhua pilot-scale fluidized-bed oil shale retort

The pilot-scale unit for fluidized-bed retorting of oil shale in Longhua was designed by Shanghai Boshen Engineering Technology Company Ltd.**,** by following the heavy oil catalytic cracking technology widely used in petroleum refineries. It was built in 2005 by Longhua Harbin Coal Chemical Industry Co., Ltd.

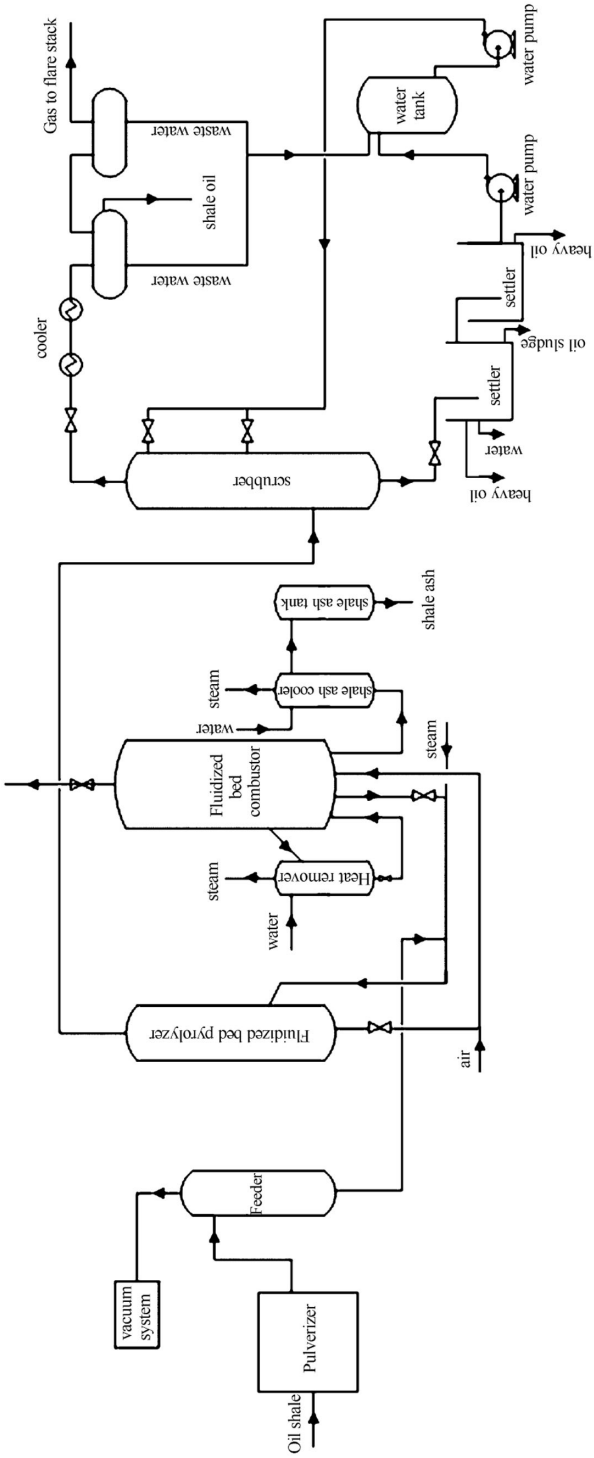

Fig. 1. Schematic sheme of the oil shale fluidized-bed retorting pilot plant.

However, at the beginning of the trial period, many problems were encountered, and the unit could not be normally operated. It was found that oil shale feed with the lamellate characteristics (causing abnormal fluidization) is quite different from the spherical catalyst used in the refinery catalytic cracking unit of heavy oil, and that the shale fines in the retort-off gas are difficult to be removed. From 2005 to 2008, the test group of Longhua Coal Chemical Company has made many improvements: such as enlarging the shale feed size, improving dedusting, changing the retort-off gas scrubbing from oil washing to water washing and utilizing a proper method for separating shale oil from oily sludge. Totally 13 test runs have been made with the total operation time reaching 3000 hours. Finally, the unit could be put into normal operation with the longest continuous running time of 192 hours; the shale oil yield reached about 80%.

Test results of Longhua pilot-scale fluidized-bed retorting of oil shale

(1) The Fischer assay of the oil shale feed is as follows (%, dry basis): shale oil 10.10; water 4.84; shale char 80.31; gas + loss 4.75.
(2) Operating conditions of the pilot unit are the following: reactor pressure – 0.10–0.20 MPa; pyrolyzer temperature – 480–520 °C; combustor temperature – 650–750 °C; oil shale feed (0–3 mm) – 1.2–2.0 t/h.
(3) Shale oil yield and analysis

Three fractions of shale oil are obtained: light oil, middle oil, and heavy oil (recovered from oily sludge), the total oil yield accounts for 80% of Fischer assay.

The analysis of mixed shale oil is shown in Table 1.

(4) Gas analysis

The pyrolysis gas analysis is shown in Table 2.

Table 1. **Analysis of mixed shale oil**

Item	Data	Distillation, %	
Moisture, %	0.062	Initial point, °C	154
Density, g/cm^3	0.9350	179	5
Solid content, %	0.17	189	10
Oil content, %	99.77	212	20
Flash point, °C	170	233	30
Carbon residue, %	2.59	250	40
		278	50
		310	60
		322	70
		285	80
		337	84
		Final point, °C	337

Table 2. **Analysis of pyrolysis gas, %**

Item	Data	Item	Data
O_2	1.00	Propane	0.60
N_2	4.00	Propylene	2.88
CH_4	29.52	*i*-Butane	0.24
CO	6.97	*n*-Butane	0.61
CO_2	20.83	*n*-Butylene	2.09
H_2	25.88	*trans*-Butylene	0.10
Ethane	2.22	*cis*-Butylene	0.24
Ethylene	2.74	Heating value, MJ/kg	26.53

(5) Shale ash analysis

The industrial analysis of shale ash has given the following data, %: moisture – 0.37; ash (dry basis) – 98.54; volatile matter (dry basis) – 1.09; carbon residue (dry basis) – 0.32.

(6) Chemical composition of shale ash is shown in Table 3.

Table 3. **Chemical compositions of shale ash, wt%**

SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O	MnO_2	P_2O_5	SO_3
65.78	24.13	5.41	0.66	0.26	0.55	1.24	0.18	0.05	0.10	0.27

(7) Waste water analysis

Waste water is of a little amount, and comes from the oil shale feed. The waste water analysis has given the following data, g/L: total nitrogen – 0.05; free ammonia – none; fixed ammonia – 0.05 and total phenols – 12.76.

Conclusions

From 2004 to 2008, total thirteen testing runs have been made for retorting pulverized oil shale in a fluidized-bed pilot-scale unit. Many difficulties, especially the dusting and dedusting problems have been encountered and solved. Total running hours of the plant reached 3000 in the five years' trial, with the longest continuous running of 192 hours. It may be considered that this achievement provides a preliminary basis to scale up for the commercial plant designing.

Several detailed conclusions may be drawn as follows:

(1) Shale oil yield from the pilot plant reaches about 80% of Fischer assay. It indicates that the pyrolysis reaction proceeds well.
(2) Carbon residue content of shale ash is very small, only 0.32%. It indicates that the combustion of shale char proceeds nearly completely.
(3) Retort gas is of high heating value, accounting about 26.53 MJ/kg.
(4) Shale oil produced in fluidized-bed pyrolyzer is of lower density and contains much more light fractions.

(5) The data and results obtained from operating this pilot plant indicate that such a proprietary innovation technology is promising.

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