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OIL SHALE ACTIVITIES IN CHINA

ПЕН ДЭ-ХУН, ЦЯНЬ ЦЗЯ-ЛИНЬ

ИСПОЛЬЗОВАНИЕ ГОРЮЧИХ СЛАНЦЕВ В КИТАЕ

Geology of Oil Shale

In China, oil shale deposits are widespread and discovered in many regions, but so far, only a few have been explored and for the others no detailed geological survey has been carried out. Proved reserves amount to about 32 billion tonnes, while estimated resources reach 700 billion t, which is equivalent to 40 billion t of shale oil. It means, that oil shale is an important potential energy source of China.

In China, oil shale deposits are generally related with the regions of weak tectonic activities, such as the edges and basins of platforms, the intermount basins in geosynclinales and they are always deposited in submerging areas under calm and shallow water conditions.

As for the composition, then besides organic matter oil shale comprises sandy-clayey material, a little calcium and sometimes rare and rare-earth elements. Oil shale is a stratified rock of black or brown colour, its ash content is generally in the range of 70—80 %, the oil yield varies from 4 to 10 %, averaging 6 %. The calorific value ranges from 5400 to 6300 kJ/kg. At present, only three deposits are mined for retorting or for fuel [1].

Oil shales are from the Early Silurian to Neogene in age, being, for the most part related with the Tertiary period.

In SE China, sapanthracite, the so-called stone coal, is widespread. It is a kind of shale with high organic matter content and abundant algal relics. Some geologists consider it as a product of oil shale's metamorphism. The sapanthracite is of the Silurian to Cambrian age, with calorific value ranging from 3350 to 5000 kJ/kg [1].

The types and features of some typical Chinese oil shales are as follows [2, 3].

Fushun oil shale. Age: Tertiary. Occurrence: Fushun basin. Origin: swamp, according to some researchers lacustrine. Features: brown oil shale with underlying coal and overlying green shale.

Maoming oil shale. Age: Tertiary. Occurrence: Maoming basin. Origin: lacustrine. Features: brown oil shale interbedded with carboniferous shale, fish fossils are present, high quality kaolinite overlies the oil shale layer.

Northeast oil shale. Age: Early Cretaceous. Occurrence: Hailaer basin. Origin: lacustrine. Features: dark gray mudstone and black shale interbedded with oil shale.

Liupanshan oil shale. Age: Cretaceous. Occurrence: Liupanshan area. Origin: lacustrine. Features: gray bluish mudstone with thin black oil shale interbeds in the lower and middle parts.

Yenchang Series. Age: Triassic. Occurrence: Southern part of Ordos basin. Origin: lacustrine. Features: black oil shale interbedded with argillaceous siltstone, oil-bearing sandstone, fish and ostracod fossils are present.

Anding Series. Age: Jurassic. Occurrence: Eastern part of Ordos basin. Origin: lacustrine. Features: black oil shale interbedded with varied gray mudstone.

Yili oil shale. Age: Permian. Occurrence: Southern slope of Afular Mountain, Yili basin. Origin: lacustrine. Features: black oil shale interbedded with carbonaceous shale, fish fossils are present.

South Shaanxi oil shale. Age: Silurian. Occurrence: Hanzhong, Southern Shaanxi. Origin: marine. Features: oil-bearing siliceous layers interbedded with argillaceous oil shale in lower part; siliceous oil shale containing disseminated pyrite and dendroid fossils as well as traces of oil and asphalt in upper part.

Major Oil Shale Mining Areas [4]

The proved oil shale reserves in the mining areas in Fushun, Maoming and Huadian amount to more than 10 billion t. Promising oil shale deposits can also be found in some other areas, such as Nong-an of Jilin Province, Dongsheng of Inner Mongolia Autonomous Region, Tanshanlin and Yaojie of Gansu Province, the northern foot of Bogeda Mountain of Xinjiang Uygur Autonomous Regions, Ordos platform of northern Shaanxi, and Zhanxian County of Hai-nan Province.

Major oil shale mining areas are Fushun, Maoming, Huadian and Huang counties.

Fushun, Liaoning Province. The Fushun mining area is located to the east from the Liaoning provincial capital Shenyang. It measures 18 km from east to west and 2—3 km from north to south, and has been exploited for 60 years.

The Fushun oil shale belongs to the Tertiary period of the Cenozoic era. The oil shale seam with coal interlayers is located between the Quaternary and Cretaceous formations. Granitic gneiss under the Cretaceous formation forms the basement of the coal bed. The oil shale seam with green shale on its top overlies the coal bed. The Fischer assay of oil shale in this area varies between 2 and 10 %, averaging 5.5 %. The thickness of the oil shale seam varies from 48 to 190 m, the interbedded coal seams are 0.5—0.8 m thick. The total proved reserves of oil shale with Fischer assay above 4.7 % amount to 3.6 billion t.

The Fushun oil shale deposit is of shallow bedding and has a gentle dip. On the open-pit mining of coal, the overlying oil shale layer is stripped off. In spite of the relatively low Fischer assay, the retorting of the oil shale, a byproduct on coal mining, is profitable because of its low production cost. The shale ash can be used as backfill in Fushun underground coal mines.

Maoming, Guangdong Province. The Maoming mining area is located in the southwest of Guangdong Province. In the east, it reaches Yangjiao township of Dainbai County, extends westward through Jintangyu and Dishan of Maoming City and Shiguyu of Gaozhou County to Lianjieyi of Huazhou County. The mining area takes the shape of crescent from northwest to southeast, 50 km in length and 3—10 km in width, with a total area of 360 km^2. According to its geological conditions and structural features, the Maoming mining area is divided into 6 districts: Yangjiao, Jingtang, Shigu, Shatian, Xinyu and Dishan.

Maoming oil shale is referred to the upper Youganwo formation of Maoming system of the Lower Tertiary period and the Lower Shancun formation of Yongning system of the Upper Tertiary period. The shale deposit has a simple structure and a gentle monoclinal dip. The oil shale is characterized by a dense structure and distinct lamination. The original habitat was possibly a littoral, swampy basin.

Maoming mining area is one of the major oil shale deposits discovered after the funding of new China, with wide acreage, rich resources and clearly known geological conditions. The average Fischer assay of oil shale is 6—8 %. The proved recoverable reserves are estimated at 4.17 billion t. The deposit is of shallow bedding and has been subject to open-pit mining for more than 20 years (Jintang Mine). Recently, high quality kaolinite was discovered in the upper part of the oil shale.

Huadian, Jilin Province. The Huadian mining area is located in Huadian County to the southeast of the provincial capital Changchun. The oil shale comes from the Lower Tertiary formation in the Huadian, Jingozi and Miaoling basins. The formation is 65—244 m thick, and it contains 6—26 oil shale layers.

The total reserves in this area are estimated at 1.3 billion t, the Fischer assay being 6—12 %. Oil shale mining in Huadian began in 1943 and was completed in 1961 because of the high cost of underground mining.

Huang County, Shangdong Province. The Huang mining area is located in Huang County and Penglai County of Shangdong Province. The oil shale, belonging to the Lower Tertiary period, coexists with brown coal. The oil shale is found in an area of about 200 km^2 at a depth of 0—1000 m. The oil shale has a Fischer assay of 9—22 % and an average calorific value of 12 000 kJ/kg. It is a fairly highgrade deposit found in recent years, and is now mined underground along with brown coal, and used as fuel for power generation.

Properties of Oil Shales [5]

Properties of Fushun and Maoming oil shale are shown in Tables 1—3.

Table 1. **The main technological characteristics**
Таблица 1. **Основные технологические показатели**

Oil shale	Water W^a, %	CO_2, %	Ash A^d, %	Low calorific value Q^a, kJ/kg
Fushun	2.76	4.36	72.6	4800
Maoming	3.03	0.76	71.7	6800*

* In general, Maoming oil shale contains more than 17 % water, the calorific value is only about 4200 kJ/kg and less.

Table 2. **Fischer Assay, %**
Таблица 2. **Выход по Фишеру, %**

Oil shale	Shale oil	Water	Retorted shale	Gas + loss
Fushun	6.69	3.88	86.13	3.30
Maoming	8.28	10.78	76.46	4.46

Table 3. **Shale ash analysis, %**
Таблица 3. **Состав минеральной части, %**

Oil shale	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO
Fushun	62.23	23.45	9.7	1.41	1.78
Maoming	64.37	22.37	8.17	1.51	0.85

Production of Shale Oil

The shale oil industry in China has existed for 60 years. The highest record of annual shale oil production in the fifties was 780,000 t. Gasoline, kerosene, diesel fuel, wax and synthetic lubricating oil were produced from shale oil. With the development of Daqing Crude Oil Field starting from 1962, more emphasis was paid to crude oil, the development of shale oil industry was slowed down.

One shale oil plant in Refinery No. 2 in Fushun, has an annual shale oil production of 100,000 t. The Maoming Petroleum Industry Corporation has a yearly production of 100,000 t of shale oil. Therefore the total annual shale oil production in China amounts to about 200,000 t.

Oil shale in Fushun is produced as a byproduct on open-pit mining of coal in the West Open-Pit Mine of Fushun Bureau of Mines. Oil shale is crushed and screened to the size of 8—75 mm. Then the raw shale is fed into the Fushun type retort. In Fushun type retort, drying and pyrolysis of oil shale take place in its upper part, gasification and combustion of shale coke — in the lower part. Oil containing gaseous product evolves from the top of the retort, and is cooled and condensed to form shale oil and to give low calorific combustible gas. Shale oil is now directly used as liquid fuel in the power plant boilers. In addition to the recovery of shale oil, the ammonia in the retorting off gas is absorbed by sulphuric acid, 200 kg of ammonium sulphate can be obtained as a byproduct along with each tonne of shale oil produced. The Fushun shale ash is removed from the bottom of the Fushun type retort and used mainly for the backfilling of underground coal mining space.

The scheme of retorting plant in Maoming is similar to that in Fushun. The oil shale in Maoming is also open-pit mined, but without coal. Fushun and Maoming oil shales are of low grade. 30 tonnes of oil shale are needed for producing 1 t of shale oil. The production cost of shale oil in China is lower than the international market price of crude oil.

The Fushun type retort has the capacity of 200 t of mil shale per day. It is suitable for use in the shale oil plant with small or medium capacity.

Shale oil from Fushun and Maoming has a high wax content: 20 and 13 %, respectively, high pour point of about 30—33 °C, nitrogen content 1.1—1.2 %, sulphur content 0.54 and 0.48 %, respectively, with H/C atomic ratio of 1.7—1.6.

Shale ash in Fushun is mainly used for backfilling of underground coal mines, and also for brick making. A small-scale cement plant in Maoming Petroleum Industry Corporation uses shale ash to produce cement.

Direct Combustion of Oil Shale [4, 5]

In Huang County of Shangdong Province, oil shale and brown coal are mined underground and used as solid fuels for boilers and small-scale power generation.

In Maoming Petroleum Industry Corporation, a small-scale fluid bed boiler has been built with the capacity of 15 t of steam per hour. Shale particles less than 8 mm in size which cannot be processed in Fushun type retort, are used as solid fuel in boilers.

In recent years, both in Fushun and Maoming, fluidized combustion boilers with the capacity of 35 t of steam per hour have been built and put into use for power generation.

Achievements in the Eighties

In the seventies when the world crude oil price rose sharply, many countries began to pay more attention to the development of synfuels, including shale oil. At the beginning of the eighties, it was also recognized in China, that shale oil research and development should not be neglected in spite of the abundant petroleum resources. Several shale oil meetings were convened to sum up experience, work out a policy for the steady development of shale oil industry, draft a program for the comprehensive utilization of oil shale, e. i. for retorting (synfuel production)—power generation—building materials production—chemical production. Decisions were made to enforce research and technology innovation with the aim of higher shale oil yield, higher output of retort, higher automation level, lower energy consumption, lower environmental pollution.

In the eighties, greater success was made owing to the economic reform and open-door policy.

— A book entitled *Oil Shale Industry in China* was written by Chinese specialists and professors to sum up the experience of oil shale development in China in the past decades. It was published in Chinese in 1984 and in English in 1986 [6].

— In 1988, an International Conference on Oil Shale and Shale Oil in conjunction with Colorado School of Mines 21st Oil Shale Symposium was held in Beijing, sponsored by China Energy Research Society, China Petroleum Refining Society, Colorado School of Mines of U.S.A. and U.S. Department of Energy. There were 200 participants from 15 countries. Proceedings of the Conference, containing 89 papers were published [7].

— In 1986, in cooperation with Jordan the research into Jordanian Lajjun oil shale was carried out. Jordanian oil shale differs from the Chinese Fushun oil shale. Its Fischer assay reaches 10 %, sulphur content is about 3 %; it abounds in carbonates, calcium oxide content in ash accounts for 38 %. The research into the Jordanian oil shale showed that the latter can be well processed in the Chinese Fushun type retort [8]. There arose no difficulties in connection with ash removal or separation of water from shale oil. The oil yield from the retort reaches 80—84 % of Fischer assay. It means that Fushun type retorts are suitable for different kinds of oil shale, which makes them economically profitable, and especially suitable for small- and medium-scale shale oil production.

— Scientific and technological cooperation was held between China and the U.S.S.R. The cooperation program (1988—1990) was conducted successfully and fruitful results were obtained in the field of pyrolysis of oil shale, chemical utilization of shale oil and utilization of shale ash for cement.

— International scientific and technological exchange took place between the related institutions of China and the U.S.A., Japan, FRG, Canada.

— New drying and retorting technologies were in development. For example, rotating cylindrical drying and fluidized bed retorting of Maoming oil shale were conducted; fluidized bed combustion of low calorific value oil shale (4000 kJ/kg) for power generation with the capacity of 35 t per hour were studied both in Fushun and Maoming [9]. The size of oil shale particles for fluid combustion is below 8 mm, which cannot be used in Fushun type retorts.

— For the utilization of shale ash, it was found that shale ash can be used as raw material for production of cement with the proportion of 40—50 % [10]. Research was also conducted for using shale ash as raw material for cement blocks, ceramsite, plastic and rubber filler.

— For the purpose of environmental protection, measures were taken for closed circulation of retorting waste water (e. g. injecting water into retorts). Biological treatment was also adopted before the discharge of waste water [11].

— In the eighties, in the field of fundamental research of oil shale, more than 120 papers were published or presented in the international or local periodicals and symposiums, including the drying, pyrolysis and combustion mechanism and kinetics, as well as the chemistry and structure of kerogen, and the composition and constituents of shale oils.

— Doctoral and post graduate students have been trained in the field of oil shale in universities.

Looking Forward to the Nineties

In the nineties, the rise of the world market price of crude oil will stimulate the production of shale oil in the world, as a whole, China inclusive. It is expected, that in the nineties there will be the following developments in oil shale in China.

— Some regional governments are going to promote the development of shale oil industry and to reduce the taxes.

— A new shale oil plant is being built in the coal mining area, for retorting of oil shale which is the byproduct on open-pit mining of coal.

— In energy-deficient regions, it is planned to build fluid-bed combustion boiler for power generation by using particulate oil shale as fuel.

— The existing retorting plant will be modified by using new technology for drying Maoming oil shale with high water content (18 %) and new type of retort will be developed instead of the older one.

— More attention will be paid to the chemical utilization of shale oil, such as production of wax and anticorrosive reagent.

Conclusions

1. China has abundant oil shale resources, of the Early Silurian to Neogene age, the most important being the Tertiary period. The proved oil shale reserves in Fushun amount to 3.6 billion t, in Maoming 4.1 billion t. In Fushun, oil shale is produced by open-pit mining as a byproduct of coal, in Maoming it is also mined in open pits, but without coal.

2. In China, shale oil has been produced from oil shale for 60 years. Annual production of crude shale oil amounts to about 200,000 t. The production costs of shale oil are lower than the price of crude petroleum on the world market.

3. China has accumulated the experience and technologies of oil shale retorting. The Fushun type retort has been elaborated, in which the latent and sensible heat of shale coke is well utilized. But the capacity of such retort is relatively small, therefore it is suitable for use in small or medium oil plants.

4. China has a policy of steadily developing shale oil industry. China is conducting oil shale research and developing oil shale processing technology. Much attention is being payed to the comprehensive utilization of oil shale, shale oil, and to environmental problems.

5. In China, oil shale is mostly used for producing shale by retorting, attention will also be paid to direct combustion for power generation.

6. Great achievements in oil shale research have been made in the eighties, and there will be a further development in the nineties.

РЕЗЮМЕ

Запасы и добыча горючих сланцев

Месторождения горючих сланцев в Китае широко распространены и имеются во многих регионах страны, но только для немногих из них выполнены детальные геологические исследования. Доказанные запасы горючих сланцев составляют 32 млрд. т, а по приближенным оценкам они достигают 700 млрд. т, что эквивалентно 40 млрд. т смолы. Горючие сланцы являются значительным резервом энергии в Китае.

Компоненты горючих сланцев, кроме органической массы, представлены главным образом песчано-глинистым материалом с небольшим содержанием кальция и редкоземельных элементов. Сланцы черного или коричневого цвета с тонкими структурными пропластами, зольная часть составляет в них в основном 70—80 %, а содержание смолы по Фишеру колеблется в пределах 4—10 % (в среднем 6 %). Удельная теплота сгорания сланцев изменяется от 5400 до 6300 кДж/кг.

Возраст горючих сланцев Китая — от раннего силура до неогена, наибольшее значение имеют сланцы третичного периода. В статье дается краткое описание Фушуньского, Маоминского, Хайларского и Люпанчанского месторождений горючих сланцев, северо-восточных сланцев, сланцев аньдунской и йенчанской свиты и других.

Основные районы добычи сланца следующие: Фушунь, Маомин, Хуадянь и Гуандун.

Фушунь, провинция Ляонин. Добыча сланца осуществляется уже более 60 лет. Выход смолы по Фишеру 2—10 %, в среднем около 5,5 %; толщина пласта в пределах 48—190 м; доказанные запасы горючих сланцев с выходом смолы по Фишеру 4,7 % около 3,6 млрд. т. Несмотря на небольшое содержание в сланце смолы, стоимость его добычи низкая, так как сланец, лежащий над углем, при добыче последнего является побочным продуктом. Уголь и сланец добываются открытым способом.

Маомин, провинция Гуандун. Площадь месторождения 360 км^2; содержание смолы по Фишеру в среднем 6—8 %; доказанные запасы горючих сланцев 4,17 млрд. т.

Хуадянь, провинция Гирин. Содержание смолы по Фишеру 6—12 %; общие запасы горючих сланцев 1,3 млрд. т. Добыча сланца была начата в 1943 г. и прекращена в 1961 г. из-за высокой стоимости подземной добычи.

Округ Хуансянь, провинция Шаньдун. Площадь месторождения 200 км^2; содержание смолы по Фишеру 9—22 %; средняя удельная теплота сгорания 12 тыс. кДж/кг. Горючие сланцы высокого качества.

Переработка горючих сланцев

В таблицах 1—3 охарактеризованы горючие сланцы Фушуня и Маомина. Их рабочая влага около 3 и 17 % соответственно; в минеральной части преобладают соединения кремния.

Сланцеперерабатывающая промышленность в Китае существует около 60 лет. В 50-х гг. производство сланцевой смолы в Китае достигло максимального уровня — 780 тыс. т. В этот период на основе сланцевой смолы производились бензин, керосин, дизельное топливо, смазочные масла и другие продукты. В настоящее время завод № 2 в Фушуне ежегодно выдает всего лишь около 100 тыс. т смолы. Столько же смолы получают и на заводе Маоминской нефтепромышленной корпорации. Общее количество производимой в Китае сланцевой смолы составляет таким образом 200 тыс. т.

В Фушуне сланец с пределами крупности 8—75 мм перерабатывают в ретортах фушуньского типа: в верхней их части происходит полукоксование сланца, а в нижней — газификация полукокса. Парогазовая смесь выводится из реторты в конденсационную систему для охлаждения и выделения смолы, после чего остается низкокалорийный газ. Схема добычи и полукоксования сланца в Маомине аналогична применяемой в Фушуне. Поскольку фушуньские и маоминские сланцы низкокачественные, для получения одной тонны смолы расходуется примерно 30 т сланца. Реторты в Фушуне имеют пропускную способность по сланцу 200 т, в Маомине — 150 т в сутки.

Фушуньская и маоминская смолы содержат соответственно 20 и 13 % парафина и имеют высокую температуру застывания — 30—33 °С. Азота содержится 1,1 и 1,2 %, серы — 0,54 и 0,48 % соответственно, соотношение $(H/C)_{ат}$ 1,7—1,6.

В Фушуне зольный остаток используется как материал для заполнения выработанных карьеров, а также для изготовления кирпича. В Маомине небольшая часть зольного остатка используется при производстве цемента.

Непосредственное сжигание сланца

Маоминской нефтепромышленной корпорацией построена установка для сжигания сланца в псевдоожиженном слое с целью получения 15 т пара в час. В Фушуне мелкий сланец крупностью менее 8 мм используется в качестве котельного топлива. В последние годы в Фушуне и Маомине сооружены установки для сжигания мелкого сланца в псевдоожиженном слое с целью получения 35 т пара в час.

Планы на будущее

Ожидается, что цены на жидкое топливо в Китае будут стимулировать развитие производства сланцевой смолы. Благодаря расширению объема добычи угля намечается увеличение добычи сланца как побочного продукта и строительство новых сланцеперерабатывающих заводов.

В Китае в районах с дефицитом энергии планируется сооружение установок по сжиганию мелкого сланца в псевдоожиженном слое для производства водяного пара. Намечается создание новых реторт для полукоксования маоминских сланцев, включая и решение такой сложной задачи, как подсушка этих сланцев, имеющих высокую влажность. Большое внимание будет уделено расширению работ по химическому использованию сланцевых смол, в частности получению антикоррозионных мастик.

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