Oil Shale, 2014, Vol. 31, No. 1, pp. 42–53
doi: 10.3176/oil.2014.1.05
ISSN 0208-189X

COMBINED FLUIDIZED BED RETORTING AND CIRCULATING FLUIDIZED BED COMBUSTION SYSTEM OF OIL SHALE: 1. SYSTEM AND KEY ISSUES

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Abstract. *The uncertainty about the current prices of petroleum, its growing worldwide consumption and limited availability have motivated many countries rich in oil shale resource to explore more efficient retorting technologies to widely produce and use shale oil as an alternative. On the basis of the retorting technology progress and the characteristics of oil shale, this paper recommends a combined system with a fluidized bed (FB) reactor for retorting oil shale and a circulating fluidized bed (CFB) boiler for burning semicoke and fuel gases, realizing the effective and clean use of oil shale. The FB retort and CFB boiler are arranged side-by-side, and hot circulating ash and semicoke will be conveyed from one side to the other. Both CFB circulating ash and fuel gases as fluidizing gases of the FB retort are proposed to be heat carriers together, transferring the combustion heat from the CFB to oil shale in the FB retort. Retorting temperature, heat capacity distribution of heat carriers and parameter optimization of the whole system are further discussed as key issues.*

Keywords: *oil shale, fluidized bed retorting, circulating fluidized bed combustion.*

1. Introduction

This is the age of oil. Petroleum has assumed a very important role in affecting the economic development in the world. However, as a limited non-renewable resource, petroleum cannot continue to meet the ongoing and increasing demand of human being for it. Especially, oil production has become limited by the capacity of extraction technology, causing supply and demand to diverge in recent years. The current fluctuating prices of oil, its

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growing worldwide consumption and limited availability have motivated many countries to explore more alternative oil resources.

Oil shale, one of the most important potential substitutes for petroleum, is widespread in the world. According to the data reported by Qian et al. [1], shale oil (converted from the in-situ oil shale) accounts for about 400 billion tons in the world, higher than that of crude oil (more than 300 billion tons). In the past, during the periods of every world "oil crisis", retorting of oil shale was always initiated largely to produce shale oil as a necessary substitute for oil. Having predicted that oil crises would occur again in the future, many countries, including Estonia, Brazil and China, have begun to investigate more efficient and environmentally friendly methods to retort oil shale to produce shale oil. For example, Estonia uses Galoter and Kiviter retorts [2, 3], Brazil uses Petrosix retorts [4], China uses Fushun-type retorts [5], Australia has scaled up Canadian Taciuk particulate shale retorts called Aostra Taciuk Processing (ATP) [6–8], and Shell Company (USA) is conducting in-situ oil shale retorting pilot testing [5, 9]. However, some technical issues hinder the wide development of these processes. For example, Kiviter and Fushun retorting processes produce a large amount of harmful semicoke waste, and Galoter has a complicated multistage technological scheme. In view of this, a new and compact comprehensive utilization technology of oil shale was recommended for shale oil production, electricity generation by burning semicoke, and ash recycling utilization [10–12]. In this comprehensive utilization process, the semicoke produced in the oil shale retort is directly introduced into a circulating fluidized bed (CFB) as a fuel, and the circulating ash of the CFB is directed into the retort as a high-temperature solid heat carrier. Among the solid heat carrier retorting technologies, the rotating drum retorting, such as ATP and Galoter, has been commercialized and the fluidized bed retorting has been experimentally validated to have a high shale oil yield [13, 14]. It has also been proven that pure semicoke from the Kiviter retort could burn steadily in a 50 kW_{th} circulating fluidized bed without any additional fuel under certain conditions [15]. At its moisture content of over 10% the addition of oil shale in the amount of 10–20% is necessary. The semicoke from the comprehensive utilization system retorts has a higher physical heat (400–500 ^{o}C), higher calorific value and lower moisture content than that from the Kiviter retort. Thus, it will be easier for semicoke to burn steadily in the CFB of the comprehensive utilization system. The oil shale ash discharged from the low-temperature CFB usually has a cementing strength and adsorbent capacity and thus may be used as a building material or an adsorbent [16–18]. Consequently, each subsystem of the comprehensive utilization system of oil shale is able to work well. However, some new issues might have to be considered after these subsystems are incorporated into the comprehensive utilization system of oil shale.

For successfully developing and optimizing the comprehensive utilization process of oil shale, the authors have recently explored a combined fluidized bed (FB) retorting and CFB combustion system of oil shale, and carried out a series of related investigations, including combustion characteristics of the semicoke CFB, retorting characteristics of the FB retort, energy analysis of the whole process, etc. The current paper mainly focused on the process of this system and analysis of several potential key issues.

2. Combined FB retorting and CFB combustion system

Considering problems of contemporary utilization technologies of oil shale, a combined FB retorting and CFB combustion system of oil shale was put forward, expanding the authors' former pertinent studies of oil shale [10]. In this system, a FB is proposed as the retort of oil shale, a CFB is used to burn semicoke and fuel gases, and the hot circulating ash from the CFB and the circulating fuel gases are used together as the heat carriers of the FB retort. The major products include shale oil, fuel gases, etc. Also, with electricity as one final outcome, the combined system contributes to the comprehensive utilization of oil shale.

2.1. System process

The combined system process is shown in Figure 1. Dried oil shale particles of 0–20 mm are fed into the FB retort at a bed temperature between 460–490 ^{o}C. The intense gas-solid two-phase motion and hot bed materials in the dense phase make oil shale heat up fast to the bed temperature and decompose into shale oil vapor, pyrolytic water, fuel gases and semicoke. The formed mixing gases, which are composed of shale oil vapor, steam, fuel gases and fluidizing gases, flow downstream in turn through a gas-solid separator and a gas-liquid separator, producing a shale oil-water mixture, fuel gases and semicoke powder. Shale oil can be obtained in an oil-water separator, and the semicoke powder will return back into the FB retort. The fuel gases will be pumped into a gas storage unit and subsequently divided into three portions: one will be introduced into a gas combustor as a fuel; one will be heated to above 600 ^{o}C by a gas-solid heat exchanger and then fed into the FB retort as fluidizing gases; and the rest will be for sale. The hot semicoke in the FB retort will be discharged directly from the overflow port of the FB into the CFB as a fuel. The CFB furnace temperature is maintained at 800–950 ^{o}C by burning semicoke with an additional heat input from the gas combustor. The bottom slag will be released into the gas-solid heat exchanger. The hot flue gas entraining a large amount of fine ash will flow into the cyclones, separating into the circulating ash and flue gas. The circulating ash will be divided into three parts, respectively the solid heat carrier of the FB retort, the circulating ash of the CFB furnace, and the hot material of the gas-solid heat exchanger. The proportion for each purpose can be adjusted according to the operational need. The flue gas from the cyclone will flow in turn through steam heat exchangers, air preheaters and oil shale driers, etc., and then emit into the atmosphere. The high-temperature steam produced from the heat exchangers will be used to supply heat and generate electricity via a traditional steam-electric power system. The cold ash from the gas-solid heat exchanger may be used as building and chemical material, which depends on the components and content of its inorganic minerals.

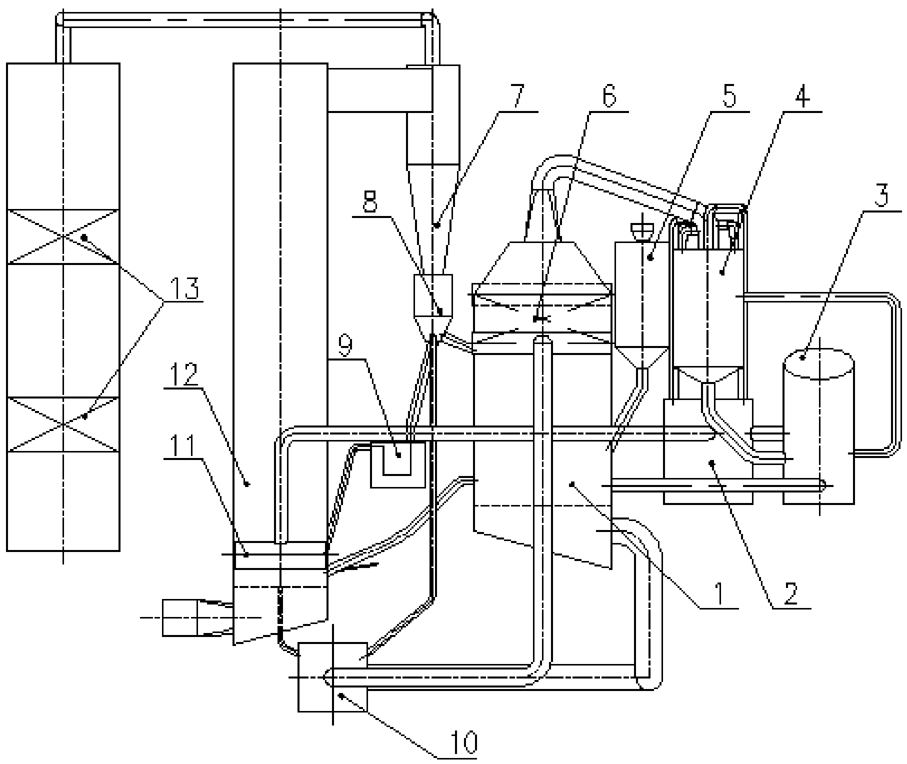

Fig. 1. Combined system of the FB retort and CFB boiler.

1 – fluidized bed retort; 2 – gas tank; 3 – oil-water separator; 4 – gas-liquid separator; 5 – oil shale storage; 6 – gas-solid separator; 7 – cyclone; 8 – oil shale ash storage; 9 – loop seal valve; 10 – gas-solid heat exchanger; 11 – gas combustor; 12 – CFB furnace; 13 – steam heat exchangers.

2.2. System characteristics

Compared with traditional combustion and retorting technologies of oil shale, this combined system possesses the following advantages:

(1) Diversified products, such as shale oil, fuel gases and electricity, can be obtained. The oil shale ash formed in the semicoke CFB boiler has a potential to be used in the chemical and building engineering.

(2) The FB retort and CFB boiler are arranged side-by-side, and hot circulating ash and hot semicoke will be transferred directly from one side to the other, making the system simple and compact, and reducing environmental pollution and energy losses in the transmission process.

(3) The use of both circulating ash and fuel gases as heat carriers contributes to the operational flexibility of the FB retort and the quality improvement of retorting products.

(4) Burning semicoke in the CFB can afford high combustion efficiency and low levels of gaseous pollutants.

3. Analysis of key issues of the combined system

Combined with previous studies of Huadian oil shale and its semicoke [19–21], some key issues will be discussed below to develop the new utilization system of oil shale.

3.1. Retorting temperature

Retorting temperature is a very important parameter affecting the yields and components of pyrolytic products of oil shale. It has been widely proven that for obtaining higher oil yield the optimum retorting temperature is between 500–550 °C. However, some other factors have to be considered before determining the practical retorting temperature of the combined FB retorting and CFB combustion system.

3.1.1. Effect on the yield of pyrolytic products

Figure 2 illustrates the effects of the retorting temperature on the yields of pyrolytic products obtained from the pyrolysis of Huadian oil shale in a lab-scale fixed bed reactor. The formation of shale oil started at the retorting temperature of about 415 °C; upon heating up to 460 °C, the yield of shale oil increased significantly, to 17.97%. However, when the retorting temper-

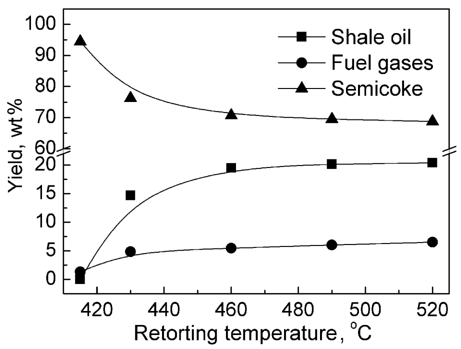

Fig. 2. Effect of the retorting temperature on the yield of pyrolytic products.

ature increased further from 460 to 520 °C, the yield of shale oil increased slightly, by 0.81%. During the whole retorting process, the fuel gas yield increased with increasing retorting temperature, which was attributed to the increasing cracking and coking reactions of shale oil as the retorting temperature was elevated [22–24]. Pakistan oil shales have also shown similar retorting characteristics [25]; at a temperature of 450 °C, the oil yield amounted to 14.1%, i.e. approximately 85.5% of the maximum oil yield. So, shale oil was mainly formed at temperatures below 460 °C. If the residence time at every retorting temperature is long enough, the oil yield will be almost constant in the low temperature range of 450–525 °C [13].

3.1.2. Effect on the components of shale oil

The composition of shale oil depends on the shale from which it was obtained, as well as on the retorting processes by which it was produced, while the retorting temperature plays a significant role. When the retorting temperature was lower, distillation of high boiling point oils was dominant over pyrolysis in the initial stage of retorting; however, the processes took place nearly simultaneously at higher retorting temperature [26]. As a result, it was suggested that the retorting at 450 °C was appropriate for western US oil shale in consideration of additional upgrading process and shale oil properties.

Compared with petroleum, shale oil is high in nitrogen and oxygen compounds, and sometimes high in sulfur. In addition, it has high levels of aromatic compounds. The presence of carcinogenic and mutagenic polycyclic aromatic compounds in the shale oil, especially if present in high concentrations, might limit the use of this fuel as a direct substitute for petroleum-derived fuels, since the oil would result in a health hazard. Organic nitrogen-containing compounds poison catalysts in different catalytic processes, and also have an unfavourable effect on the stability of shale oil during storage. Williams and Nazzal [27] investigated the polycyclic aromatic compounds (PAC) of shale oil formed from the fluidized bed retorting of oil shale, showing that the concentration of PAC in the shale oils increased with increasing retorting temperature. Table 1 shows the elemental analysis of four shale oil samples obtained from Huadian oil shale at different retorting temperatures. It can be seen from the table that the content of N element increased and H/C ratio decreased, similarly to the trend observed for Stuart oil shale in a fluidized bed retort [13].

Table 1. Elemental analysis of shale oil samples

Retorting temperature, °C	N, %	C, %	H, %	O, %	S, %	Others, %	H/C molar ratio
430	0.82	70.34	11.51	15.84	0.41	1.09	1.96
460	0.90	76.08	12.00	8.44	0.37	2.22	1.89
490	0.96	77.84	11.84	4.81	0.37	4.19	1.82
520	0.99	75.59	11.46	9.00	0.39	2.57	1.82

3.1.3. Effect on the combustion characteristics of semicoke

Table 2 presents the ultimate and proximate analysis of Huadian oil shale semicoke samples obtained at different retorting temperatures, Figure 3 shows their ignition temperatures recorded using a Leitz II-A heatable stage microscope with a high definition video camera (Leica Inc., US). These semicoke samples show an increasing ignition temperature and decreasing heating value with increasing retorting temperature, due to the decreasing content of organic matter residues in the semicoke as the retorting temperature was elevated. So, the semicoke obtained at lower retorting temperatures should burn more steadily in the CFB furnace and help increase the thermal capacity of the boiler.

Considering the above-mentioned properties of shale oil and semicoke, the retorting temperature of 460–490 °C was recommended for the FB retort of the combined utilization system of oil shale.

Table 2. Effect of retorting temperature on proximate and ultimate analysis of semicokes

Retorting temperature, °C	Proximate analysis, wt%					Ultimate analysis, wt%				
	M_{ar}	V_{ar}	A_{ar}	FC_{ar}	$Q_{ad.net}$ /kJ/kg	C_{ar}	H_{ar}	O_{ar}	N_{ar}	S_{ar}
Oil shale	11.54	36.21	48.23	4.02	11076.07	27.33	3.59	7.90	0.57	0.84
430	0.91	22.38	67.13	9.58	8277.84	22.43	1.71	6.03	0.71	1.08
460	0.88	17.28	72.89	8.95	5894.86	18.86	1.16	4.48	0.78	0.95
490	1.01	15.40	74.16	9.43	5831.71	17.37	0.90	4.90	0.71	0.95
520	0.21	16.16	73.79	9.84	5052.19	17.88	0.88	5.46	0.70	1.08

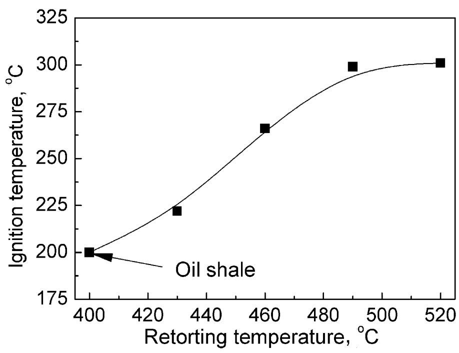

Fig. 3. Effect of the retorting temperature on the ignition temperature of semicoke.

3.2. Heat carriers

Based on the materials used to deliver heat energy to oil shale, heat carriers are usually divided into solid and gas. For the combined system of oil shale, the hot circulating ash from the CFB and fluidizing fuel gases would be considered together to transfer the combustion heat of CFB to oil shale in the

FB retort. One potential issue is the effect of circulating ash on the low-temperature pyrolysis of oil shale. According to the experimental results obtained with a 65 t/h industrial CFB boiler of oil shale [28], the equivalent diameter of circulating ash is 0.237 mm smaller than that of solid carriers used in ATP and Galoter retorts. It has been experimentally demonstrated that the co-pyrolysis of oil shale and ash under standard Fischer Assay conditions influenced pyrolysis product composition and yields, due to the alkaline composition of ash and the resulting chemisorption properties [29]. Also, the rate and extent of adsorption by ash was increased by decreasing its particle size, because the effective surface area increased as the particles size decreased and, as a consequence, the saturation adsorption per unit mass of the adsorbent increased [30]. So, it will have to be considered how to control the quantity of circulating ash as heat carrier and distribute the heat capacities between circulating ash and fluidizing fuel gases.

Figure 4 shows the thermogravimetric (TG) and differential scanning calorimetry (DSC) curves of Huadian oil shale in the low-temperature range. The total input heat amount as high as 2886 kJ/kg can be achieved for the pyrolysis of oil shale at temperatures ranging from 40 to 490 °C. So, if only hot ash is used as heat carrier, the mass of hot ash required for pyrolyzing oil shale in the FB retort is approximately calculated as follows:

$$m_{\text{ash}} = \frac{m_{\text{os}} \cdot Q}{C_{\text{ash}}\left(T_1 - T_r\right)\cdot\left(1-\eta\right)} = \frac{2886 m_{\text{os}}}{3.48\times\left(800-490\right)\cdot\left(1-0.8\%\right)} = 2.7 m_{\text{os}},$$

where m_{ash} and m_{os} are the mass of hot ash and oil shale, respectively, kg; Q is the consumption heat for the pyrolysis of oil shale, kJ/kg; C_{ash} is the average specific heat of hot ash, kJ/(kg·K); T_1 and T_{r} are the initial temperature of hot ash and the retorting temperature of the FB retort, respectively, °C; η is the heat loss of the retort. So, the mass of hot ash is at least 2.7 times as high as that of oil shale for the pyrolysis of oil shale without the fluidizing gases as hot carrier. Figure 5 shows the effect of fine oil shale ash on the yield of pyrolytic products of oil shale obtained by

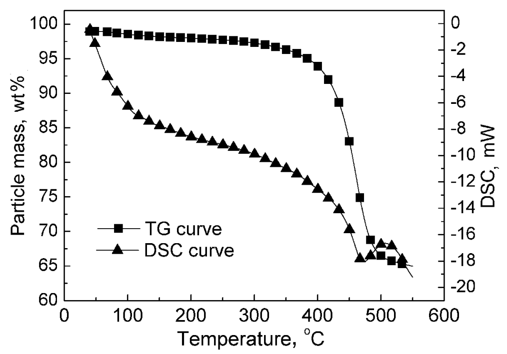

Fig. 4. Pyrolysis TG and DSC curves of Huadian oil shale.

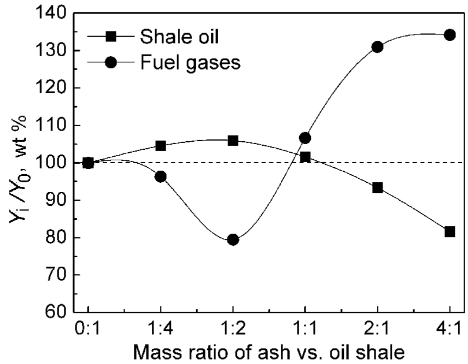

Fig. 5. Effect of oil shale ash on the yield of pyrolytic products.

retorting oil shale and ash mixture at different mass ratios in a fixed bed reactor [31]. The Y-axis represents the ratio of the yield of pyrolytic products from oil shale and ash mixtures (Y_i) compared to oil shale (Y_0). It can be seen that there was an increase in shale oil yield with increasing ash/oil shale ratio from 0:1 to 1:2, after that the shale oil yield obviously decreased as the oil shale ash fraction increased. At the same time, the yield of fuel gases appeared to first decrease and then increase as the fraction of oil shale ash increased. So, if only hot ash is used as the heat carrier for retorting Huadian oil shale, the ash with at least 2.7 times as much oil shale will result in a significant decrease in the yield of shale oil (10%).

The fluidized bed technology features a strong turbulent dense zone, a relatively high fluidizing gas velocity, etc., which can produce a fast heating rate, a uniform temperature distribution and a short residence time of shale oil vapor. However, because of the potential adsorption and catalytic effects of fine circulating ash and bed material on the organic matter from oil shale retorting, the fluidizing gases may be considered to try to supply more heat for retorting oil shale by keeping lower feed rate of fine circulating ash. In addition, increasing the particle size of circulating ash will be helpful for enhancing the yield of shale oil as well.

3.3. Coordination and optimization of process parameters

Coordination and optimization of process parameters of the combined system are very significant to enable the combined system operate stably and economically. These process parameters may be classified as parameters of oil shale FB retort and semicoke CFB boiler. The key parameters of the FB retort include the yield and properties of pyrolytic products, heat capacity distribution ratio of solid and gas heat carriers, retorting temperature, residence time and particle size. Parameters of the CFB boiler are steam capacity, furnace temperature, distribution ratio of circulating ash, and amount of NOx and SO_2. The semicoke samples obtained under different retorting conditions have variable combustion characteristics, affecting the operation of the CFB boiler. On the other hand, the CFB boiler makes use of heat carriers to provide necessary heat for retorting oil shale in the FB retort. The more heat is distributed for the retorting of oil shale, the less steam is produced by the CFB boiler. So, developing the combined system should aim at getting maximum benefit from the whole system, not from a particular subsystem. As a result, mass and energy distribution, oil shale utilization degree and heat efficiency of the whole system will be used to comprehensively evaluate the combined system.

In [32], the authors have attempted to define the energy efficiency η_{system} that describes the percentage of output energy, on the basis of the input energy of the whole system. The output energy includes the energy from all the available products, including shale oil and steam. Another evaluation factor is the total profit. Changing any process parameter will affect the value η_{system} and total profit, according to which a set of optimum process parameters can be obtained with reference to the operation of the combined system of oil shale.

4. Conclusions

Based on the current utilization technologies and characteristics of oil shale, the combined FB retorting and CFB combustion system of oil shale was recommended for an effective and clean use of oil shale. The FB retort features a strong turbulent dense zone and a relatively high fluidizing gas velocity, etc., which can produce a high heating rate, a uniform temperature distribution and a short residence time of shale oil vapor. The CFB boiler can effectively burn semicoke with lower calorific value and supply pyrolysis heat for the FB retort, also reducing gaseous emissions. Considering the properties of shale oil and semicoke, the retorting temperature range of 460–490 °C might be recommended for the FB retort. Both CFB circulating ash and fuel gases as fluidizing gases of the FB retort are proposed to be heat carriers of the FB retort. The fluidizing gases should supply more heat for retorting oil shale, avoiding the potential adsorption and catalytic effect of fine circulating ash on the organic matters from retorting oil shale. Coordination and optimization of the whole system should be conducted further for achieving maximum economic and environmental benefit under the stable operation.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (Grant No. 50906051).

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Presented by A. Ots
Received February 12, 2013