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# Development of an Integrated Oil Shale Refinery with Retorting Gas Steam Reforming for Hydrogen Production

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**ABSTRACT:** The conventional oil shale refinery suffers from inefficiency in resource use and poor economic performance. This is because that detrital oil shale and retorting gas are not properly used. Hydrogenation of crude shale oil is not cost-efficient with outsourcing hydrogen. Proposed in this paper is an integrated oil shale refinery with reforming of the retorting gas. The retorting gas is used to produce hydrogen, which is then supplied for the hydrogenation. Crude shale oil is upgraded to higher valued products, such as naphtha, diesel, and liquefied petroleum gas. The detrital oil shale is combusted to provide heat for the regeneration of CaO. Economic analysis is conducted by comparing to the conventional refinery process. Results show that the return on investment of the proposed process is 18.89%, much higher than the previous 10.53% of the conventional refinery process.

## 1. INTRODUCTION

With the limited reserve of oil and gas in China, various alternative energy resources have to be explored to meet the increasing demand for energy. Oil shale is one of the most promising alternatives, attracting increasing attention because of its rich reserve.<sup>10</sup> According to the statistics, the total reserve of oil shale in China is about 500 000 billion t. It is distributed mainly in Fushun, Liaoning province, Huadian, Jilin province, and Maoming, Guangdong province.<sup>32</sup> The gradual decrease in the reserves of conventional energy resources has motivated many countries, especially China, Estonia, and Brazil, to develop efficient and environmentally friendly processing technologies of oil shale.<sup>13,20</sup>

*Ex situ* retorting and *in situ* retorting are two promising technologies for oil shale exploitation. The *in situ* retorting technology is under investigation but not likely to be implemented at scale in the near future.<sup>9</sup> The *ex situ* retorting technology is mainly applied in Fushun-type retort in China, Petrosix-type retort in Brazil, ATP-type retort in Australia, and Kiviter- and Galoter-type retort in Estonia.<sup>12,18,23</sup>

The conventional oil shale refinery technology suffers from three major problems: (i) The retorting gas is used only as fuel for boilers because of its relatively low heating value ranging from 3.3 to 4.0 MJ N<sup>-1</sup> m<sup>-3</sup>.<sup>18</sup> (ii) The detrital oil shale (<10 mm) is abandoned, leaving only 75% of oil shale processed.<sup>10</sup> A number of studies<sup>7,24,33</sup> proposed the use of the detrital oil shale or ash for building material production (BMP). This measure could relieve solid waste pollution and, meanwhile, improve economic performance to a certain degree.<sup>19</sup> However, this improvement was limited, with only a reported 2.6% increase on the return on investment.<sup>31</sup> (iii) The shale oil contains a considerable amount of heteroatomic compounds, especially unsaturated hydrocarbons and sulfur, which might cause many troubles, such as instability of fuel in transportation or storage.<sup>8,32</sup> To solve this problem, a few shale oil processing techniques have been developed, such as the catalytic hydrogenation technique,<sup>6,32,34</sup> which is characterized with high adaptability to different raw feedstocks, stable, and long

operating experience.<sup>34</sup> However, expensive hydrogen has to be outsourced, leading to high production cost.

Proposed in this paper is an integrated oil shale refinery with retorting gas steam reforming for hydrogen production to solve the problems mentioned above. The sorption-enhanced reforming reaction is introduced to produce hydrogen.<sup>11,14,26</sup> This hydrogen not only satisfies the need in the hydrogenation process but is also partially sold to improve the economic benefit. In addition, the heat needed for the CaCO<sub>3</sub> calcination reaction is supplied by the combustion of the detrital oil shale. This heat integration raises energy efficiency of the proposed process. Followed in this paper, the process structure will be elaborated in section 2. It will be modeled and simulated in section 3. Economic analysis of the process will be conducted in section 4 in a comparison to the conventional refinery process.

## 2. INTEGRATED OIL SHALE REFINERY PROCESS

In the conventional oil shale refinery process, oil shale is converted into shale oil accompanied by retorting gas and ash. The ash is used for BMP, such as ceramsite and brick. The flow sheet of a conventional oil shale refinery (OSR) is shown in Figure 1.

In this paper, we propose an integrated oil shale refinery with retorting gas steam reforming (OSR-RGSR) process. The retorting

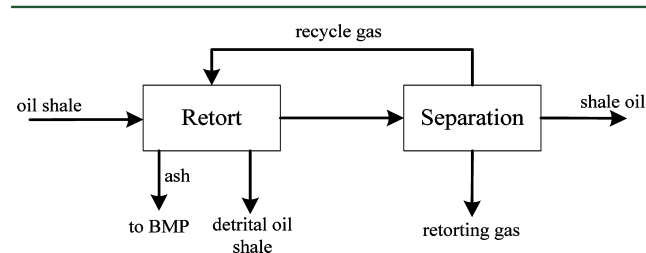


Figure 1. Schematic diagram of the conventional oil shale refinery.

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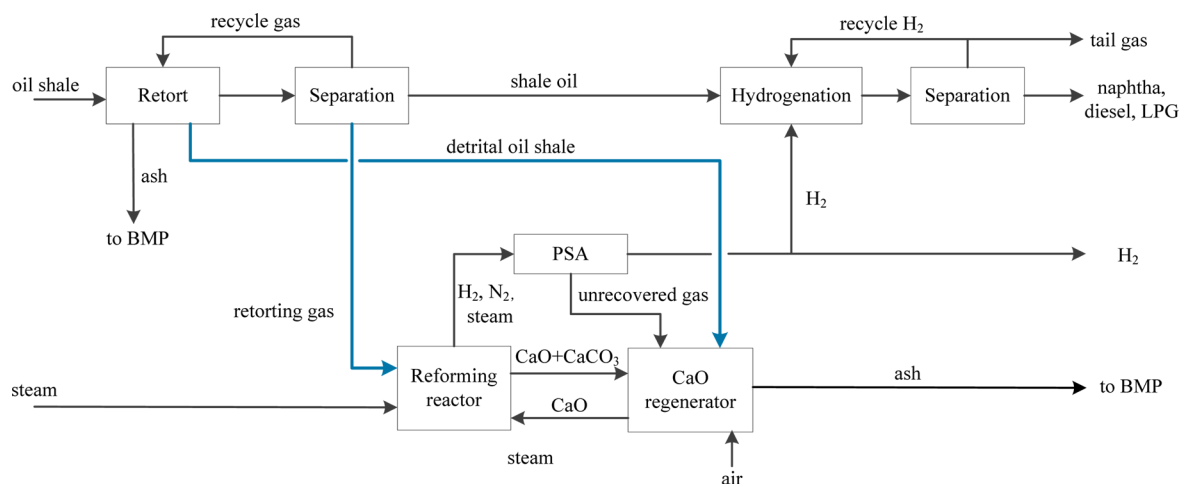


Figure 2. Schematic diagram of the proposed OSR–RGSR process.

Table 1. Proximate and Ultimate Analyses of Oil Shale

	proximate analysis (wt %, ar)				ultimate analysis (wt %, ar)				
	M	FC	V	A	C	H	O	N	S
oil shale	5.00	3.69	18.56	72.79	79.07	9.93	7.02	2.12	1.86

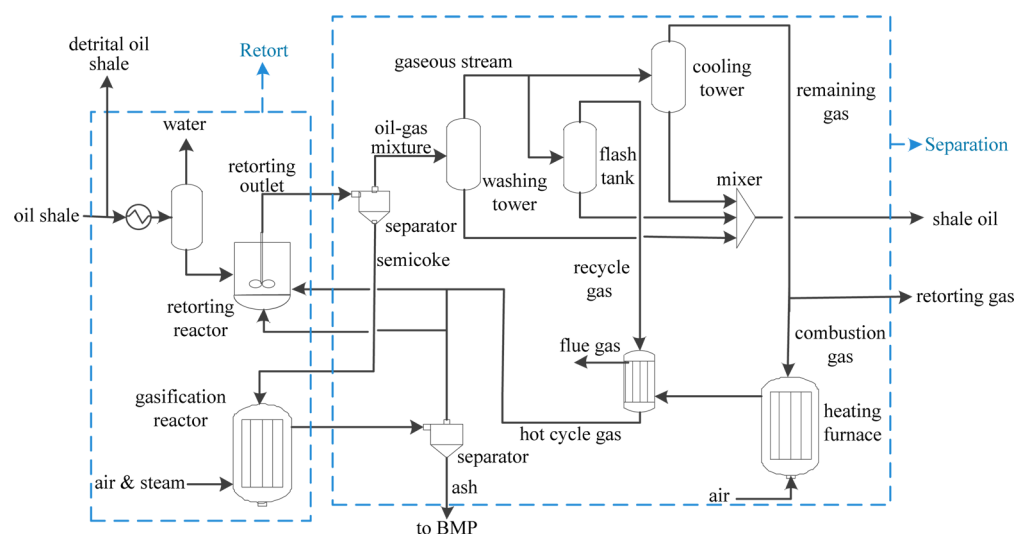
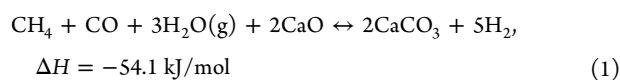


Figure 3. Flow sheet of the oil shale retorting unit.

unit as the basis is integrated with a retorting gas steam reforming unit and shale oil hydrogenation unit. The reforming unit consists of a reforming reactor that applies the sorption enhanced methane steam reforming technology for hydrogen production.<sup>11,14,26</sup> In this reactor,  $\text{CH}_4$  and  $\text{CO}$  react with steam and are converted to  $\text{H}_2$ . Produced  $\text{CO}_2$  is absorbed by  $\text{CaO}$ , which promotes conversion of the reforming reaction and increases the content of  $\text{H}_2$ . In addition, the reaction between  $\text{CaO}$  and  $\text{CO}_2$  as an exothermal reaction can provide a part of heat for the reforming reaction. The general reaction, including the two above reactions, can be written as follows:



The gaseous product from the reforming process is hydrogen-rich gas mixed with  $\text{N}_2$ .  $\text{H}_2$  is separated by the pressure swing adsorption (PSA) device. The solid products  $\text{CaCO}_3$  and unreacted  $\text{CaO}$  are fed into the regenerator, in which  $\text{CaCO}_3$  is decomposed back to  $\text{CaO}$  and  $\text{CO}_2$ . The heat for  $\text{CaCO}_3$  calcination is supplied by combustion of the

detrital oil shale and the unseparated gas from the PSA process.  $\text{H}_2$  is then fed into the hydrogenation unit to upgrade the crude shale oil to higher valued products, such as naphtha, diesel, and liquefied petroleum gas (LPG). Remaining  $\text{H}_2$  is sold as a product. A flow sheet of the OSR–RGSR process is shown as Figure 2.

The mathematical model of the OSR–RGSR process will be discussed in the next section. Both the proposed process and the conventional process will be simulated for comparative analysis from the economic point of view in the last section.

### 3. MODELING AND SIMULATION OF THE OSR–RGSR PROCESS

The OSR–RGSR process is modeled and simulated in Aspen Plus simulation software (version 7.2). It consists of the retorting, the reforming, the hydrogenation, and the BMP units. The modeling and simulation of the first two units will be discussed in detail. The hydrogenation unit and the BMP unit are simulated referring to the relevant literature. In this paper,

	mole fraction (%)						
	N <sub>2</sub>	O <sub>2</sub>	H <sub>2</sub>	CO	CO <sub>2</sub>	CH <sub>4</sub>	oil productivity (%)
simulation data	54.92	0.40	11.91	4.55	20.79	7.43	6.83
industrial data	54.91	0.40	11.90	4.43	20.84	7.52	6.75

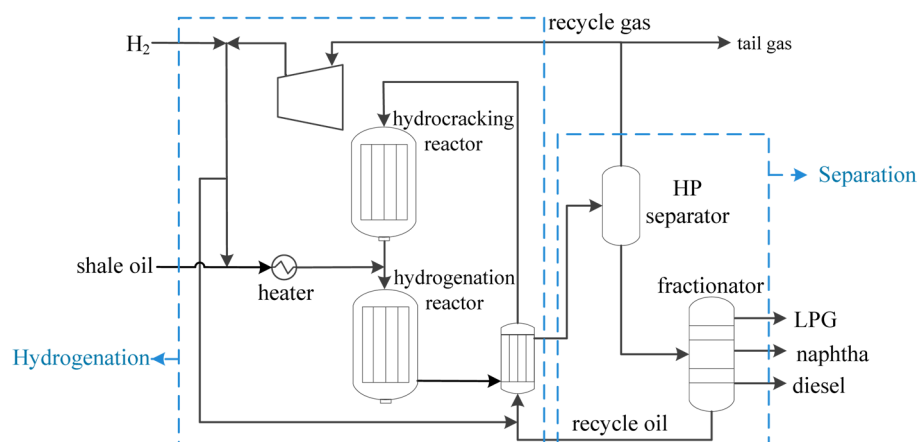
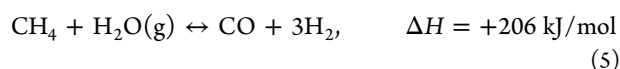


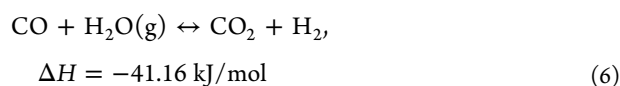
Figure 5. Flow sheet of the shale oil hydrogenation process.

reaction; and CaO works as a CO<sub>2</sub> acceptor to remove CO<sub>2</sub> in a carbonation reaction. These reactions are formulated as follows:

methane steam reforming reaction



water–gas shift reaction



carbonation reaction



CaCO<sub>3</sub> generated by the reforming reactor is fed into the CaO regenerator for recycle.<sup>11</sup> The heat for CaCO<sub>3</sub> calcination in the regenerator is provided by combustion of the detrital oil shale and the unrecovered gas from the PSA process. Ash from the combustion is used for BMP. The mixture gas (mainly H<sub>2</sub>, N<sub>2</sub>, and steam) from the reforming reactor is fed into the PSA device to separate. N<sub>2</sub> is included in the retorting gas coming from the air. It does not participate in the reforming reaction with low reaction activity. H<sub>2</sub> is used in the hydrogenation process.

According to the literature,<sup>11,26,27,29</sup> the model is established for the reforming process coupled with the CaO carbonation. RCSTR in Aspen Plus is used to model the reforming reactor. A FORTRAN subroutine is developed for simulating the kinetics of the methane steam reforming reaction, the water–gas shift reaction, and the CaO carbonation reaction. According to the sensitivity analysis, the optimal operational condition is selected. Simulated by ADSIM in Aspen Plus, the recovery rate of H<sub>2</sub> in the PSA process is set to 72%. The concentration of H<sub>2</sub> is 99.95%. These results are highly consistent with the industrial data.<sup>21</sup>

**3.3. Hydrogenation and BMP Units.** The single-stage reverse sequencing combination of hydrocracking technology is applied in the hydrogenation unit.<sup>34</sup> The flow sheet is shown in Figure 5. The shale oil from the retorting process is mixed with H<sub>2</sub>. It is sent to the hydrogenation reactor, in which a desulfurization reaction, a denitrogenation reaction, and a hydrogenation reaction occur at the same time. The reaction products from the hydrogenation reactor are then sent into the high-pressure separator to separate the gaseous phase and the

liquid phase. The gas is recycled to the hydrogenation reactor, while the liquid is separated in a fractionator to diesel, naphtha, and LPG. Residual oil extracted from the bottom of the fractionator is mixed with H<sub>2</sub> and sent to a hydrocracking reactor, in which high-molecular-weight hydrocarbons are cracked into lower molecular weight hydrocarbons and then recycled to the hydrogenation reactor. According to Zhao's work,<sup>34</sup> the operating temperature in the hydrogenation unit is set to 400 °C and the mass ratio of H<sub>2</sub>/shale oil is 0.34.

Ash from the retorting unit and the reforming unit is used to make ceramsite and bricks in the BMP unit. This is beneficial to reduction of ash pollution and, meanwhile, improves the economic performance of the oil shale refinery.<sup>7</sup> The simulation of the BMP unit refers to the previous work of the authors.<sup>28</sup>

**3.4. Selection of Key Operating Parameters.** There are several key operating parameters studied in the simulation. They are the retorting temperature in the retorting unit, the operating temperature, the pressure, the mole ratio of CaO/carbon (including CH<sub>4</sub>, CO, and CO<sub>2</sub>), and the mole ratio of steam/carbon in the reforming unit.

The retorting temperature affects the oil productivity and the production of the remaining gas. According to the simulation, both of the two indexes rise as the retorting temperature increases, as shown in Figure 6. The increasing rate is kept at a high level from 400 to 525 °C because of the quick decomposition rate of oil shale in this temperature range. However, as the temperature increases further above 525 °C,

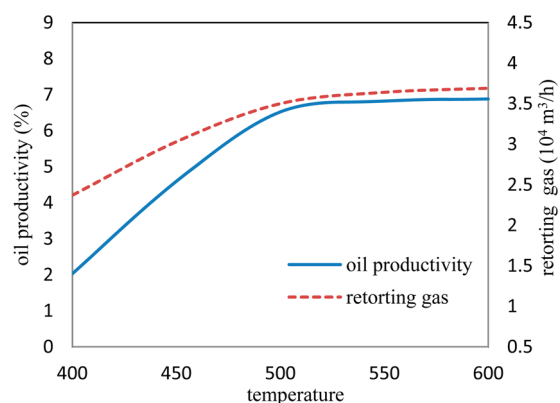
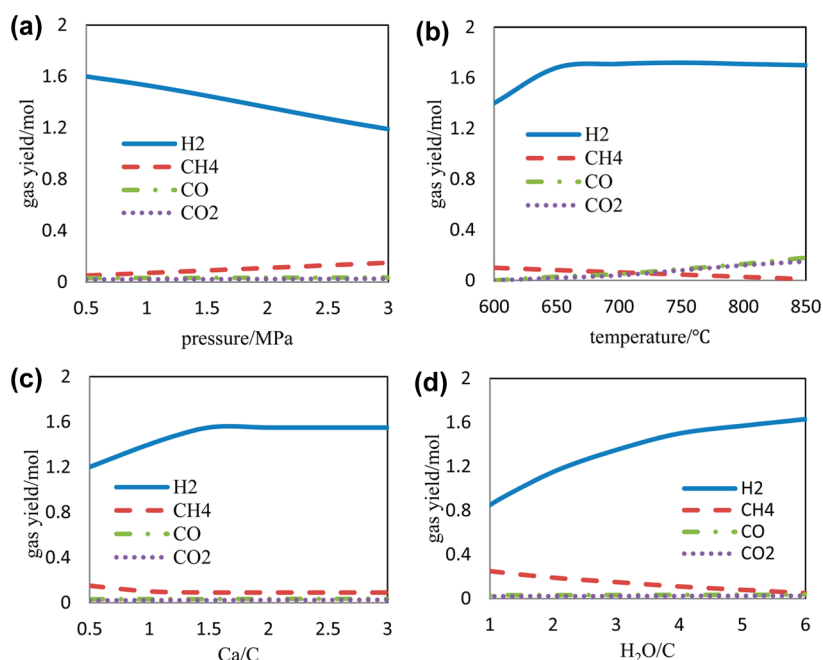


Figure 6. Effect of the temperature on oil productivity and the retorting gas production.



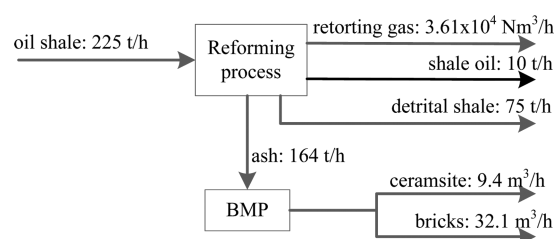


**Figure 7.** Effect of the (a) pressure on the gas yield (temperature, 650 °C; CaO/C, 1.5; and steam/C, 4), (b) temperature on the gas yield (pressure, 1.5 MPa; CaO/C, 1.5; and steam/C, 4), (c) CaO/C on the gas yield (temperature, 650 °C; pressure, 1.5 MPa; and steam/C, 4), and (d) steam on the gas yield (temperature, 650 °C; pressure, 1.5 MPa; and CaO/C, 1.5).

the productivity and the production are kept nearly the same. This is because the small amount of the remaining organics can then be decomposed. According to the analysis, 525 °C is selected as the appropriate retorting temperature for simulation.

The key parameters in the reforming unit are studied. As the pressure increases, the production of H<sub>2</sub> and the conversion of CH<sub>4</sub> decrease, as shown in Figure 7a. However, with a very low pressure, the size of the reactor will be overlarge. This will lead to high equipment investment. Thus, 1.5 MPa is selected as the operating pressure for simulation. The operating temperature in this reactor is analyzed, and the results are shown in Figure 7b. The production of H<sub>2</sub> increases quickly from 600 to 650 °C. Then, the production tends to be smooth above 650 °C. The productions of CO and CO<sub>2</sub> will, however, rise at high temperatures. The temperature of 650 °C is selected as the operating temperature for simulation. CaO/C is also an important parameter. A larger production of H<sub>2</sub> can be obtained with a higher CaO/C in the range between 0.5 and 1.5, as shown in Figure 7c. The production then tends to be smooth as CaO/C increases from 1 to 3. CaO/C is set to 1.5 in the following study. The last important parameter is steam/C, of which the analysis is shown in Figure 7d. In general, the production of H<sub>2</sub> rises as the ratio increases. However, with a overlarge steam/C, excessive energy will be consumed for steam generation, increasing the product cost. In this simulation, we set steam/C to 4 for heat balance in the reforming unit.

**3.5. Simulation Results.** The OSR process: in the retorting unit, 225 t/h oil shale is processed at 525 °C to produce 10 t/h shale oil,  $3.61 \times 10^4$  N m<sup>3</sup> h<sup>-1</sup> retorting gas, and 75 t/h detrital oil shale. The retorting unit discharges ashes at the flow rate of 164 t/h, which produce 9.4 m<sup>3</sup>/h ceramsite and 32.1 m<sup>3</sup>/h bricks in the BMP unit. The input–output data of the material flows of the conventional OSR process are shown in Figure 8.



**Figure 8.** Input–output material flow of the conventional OSR process.

The OSR–RGS process: same as in the above retorting unit, 225 t/h oil shale is processed to produce 10 t/h shale oil,  $3.61 \times 10^4$  N m<sup>3</sup> h<sup>-1</sup> retorting gas, and 75 t/h detrital oil shale. Then, in the reforming unit, the retorting gas is reformed at 650 °C and 1.5 MPa with a water steam of 38.02 t/h. In the reforming unit, 1.06 t/h H<sub>2</sub> is produced. In the hydrogenation unit, 10 t/h shale oil and 0.34 t/h H<sub>2</sub> react and produce 1.56 t/h naphtha, 8.13 t/h diesel, and 0.31 t/h LPG. The remaining H<sub>2</sub> of 0.72 t/h is output as a product. The retorting and reforming units discharge ashes of 164 and 55 t/h, respectively. This brings 219 t/h ashes to produce 12.5 m<sup>3</sup>/h ceramsite and 42.8 m<sup>3</sup>/h bricks. The input–output material flow of the proposed OSR–RGS process is shown in Figure 9.

#### 4. ECONOMIC ANALYSIS

In this section, the economic analysis of the OSR–RGS process will be conducted and compared to that of the conventional OSR process. The total capital investment, the total product cost, and the return on investment are selected as the comparison indexes.

**4.1. Total Capital Investment.** Total capital investment mainly includes the investment for fixed and working capitals. The fixed capitals consist of direct investment and indirect investment. We estimate the equipment investment of these

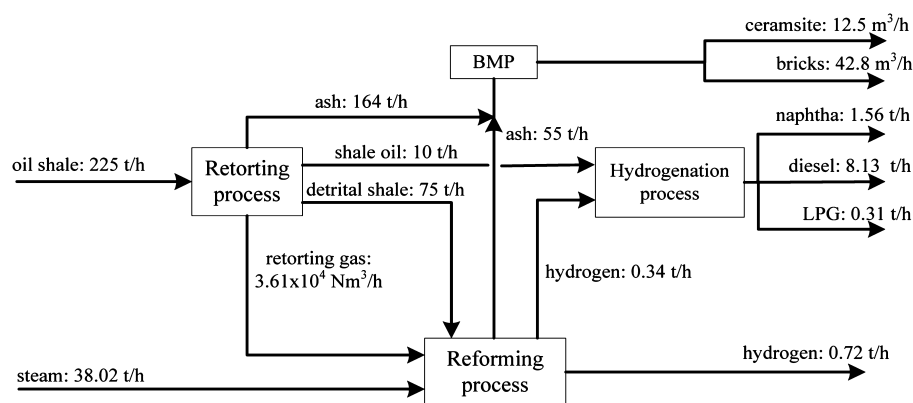


Figure 9. Input–output data of material flow of the proposed OSR–RGSR process.

two processes based on the exponential coefficient method. The calculating equation is expressed as follows:<sup>2,30</sup>

$$I_2 = \theta I_1 \left( \frac{Q_2}{Q_1} \right)^{sf} \quad (8)$$

where  $I_1$  and  $I_2$  are the reference and practical investments,  $Q_1$  and  $Q_2$  indicate the reference and practical scales,  $\theta$  is the domestic made factor, and exponent  $sf$  is the production scale factor. The range of this factor is set between 0.6 and 0.7 if the scale is expanded only by increasing the size of equipment, while it is set between 0.8 and 1.0 if the scale is expanded by increasing the quantity of equipment. In this paper, the first method is applied for expansion and  $sf$  is set to 0.6.

The equipment investment benchmark of the retorting unit, the reforming unit, the hydrogenation unit, and the BMP unit are calculated on the basis of the literature.<sup>3,25,28,34</sup> The equipment investments of these units are estimated by eq 8. Thus, those for the OSR process and the OSR–RGSR process are 173 million and 249 million. According to the work by Peters and Timmerhaus and Orhan et al.,<sup>15,17</sup> the equipment investment is proportional to other capital costs, such as buildings and land costs, engineering and supervision costs, and construction and contractor fees, with specific percentages of the total capital investment as shown in Table 3. Thus, these different costs can be estimated according to the percentages

Table 3. Percentages of Components in the Investment

component	range (%)	basis (%)
(1) direct investment		
(1.1) equipment	15–40	22
(1.2) installation	6–14	8
(1.3) instruments and controls	2–8	5
(1.4) piping	3–20	12
(1.5) electrical	2–10	6
(1.6) buildings (including services)	3–18	15
(1.7) land	1–2	2
(2) indirect investment		
(2.1) engineering and supervision	4–21	10
(2.2) construction expenses	4–16	9
(2.3) contractor's fee	2–6	4
(2.4) contingency	5–15	7
(3) fixed capital	(1) + (2)	100
(4) working capital	15–20	20
(5) total capital investment	(3) + (4)	120

and the equipment investment cost. In addition, the working capital and contingency is set to be 15–20% of the total capital investment.<sup>35</sup> After calculation, the total capital investments of the OSR–RGSR and the OSR processes are 1114 million and 783 million renminbi (RMB). The total capital investment distributions of the new proposed and conventional processes are shown in Figure 10. The total capital investment of the

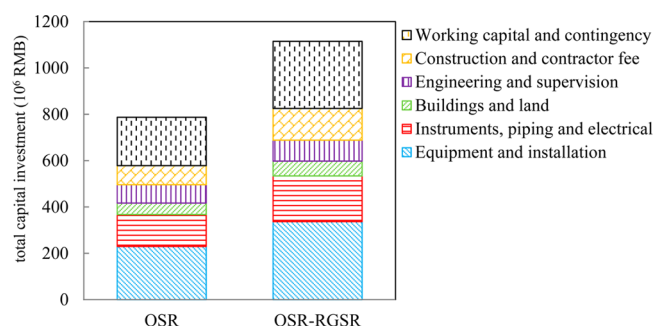


Figure 10. Investment breakdown of the proposed and conventional processes.

OSR–RGSR process is 331 million RMB higher than that of the OSR process. It is not surprising because those additional capitals are expended by introducing the reforming and hydrogenation units.

**4.2. Total Product Cost.** The total product cost consists of the operating cost and the general cost. Estimation of the product cost is also based on the work by Peters and Timmerhaus and Orhan et al.<sup>15,17</sup> The estimation coefficient of the total product cost is shown in Table 4. The second column lists the ranges of percentages of different components in the product cost. The selected percentages in this paper are listed in the third column.

The cost for the raw materials and utilities are estimated according to the simulation. The unit price of the raw materials and utilities are shown in Table 5. According to the literature,<sup>3,25,28,34</sup> we calculate the operating labor cost. Therefore, the total product cost of the OSR–RGSR and OSR processes are 626 million and 412 million RMB/year. The total product cost distributions of these two processes are shown in Figure 11. The main increase of the proposed process compared to the conventional process is found to be the operating and maintenance cost, the general cost, and the utility cost, which are 75 million, 51 million, and 35 million RMB/year, respectively.

Table 4. Estimation Coefficients of the Total Product Cost

component	range	basis
(1) operating cost		
(1.1) direct production costs		
(1.1.1) raw material	10–50% of the total product cost	calculate result
(1.1.2) operating labor	10–20% of the total product cost	reference
(1.1.3) direct supervisory and clerical labor	10–25% of operating labor	12% of operating labor
(1.1.4) utilities	10–20% of the total product cost	calculate result
(1.1.5) maintenance and repairs	2–10% of fixed capital investment	2% of fixed capital investment
(1.1.6) operating supplies	0.5–1% of fixed capital investment	0.5% of fixed capital investment
(1.1.7) laboratory charge	10–20% of operating labor	10% of operating labor
(1.2) fixed charges		
(1.2.1) depreciation	depends upon life period, salvage value	life period, 20 years; salvage value, 4%
(1.2.2) local taxes	1–4% of fixed capital investment	1% of fixed capital investment
(1.2.3) insurance	0.4–1% of fixed capital investment	0.4% of fixed capital investment
(1.3) plant overhead costs	50–70% of cost for operating labor, supervision, and maintenance	50% of cost for operating labor, supervision, and maintenance
(2) general expenses		
(2.1) administrative costs	2–6% of the total product cost	2% of product cost
(2.2) distribution and selling costs	2–20% of the total product cost	2% of product cost
(3) product cost	(1) + (2)	(1) + (2)

Table 5. Unit Prices of Raw Materials and Utilities

items	price
oil shale (RMB/t)	55.00
H <sub>2</sub> (RMB/t)	23000
water (RMB/m <sup>3</sup> )	2.75
heat cost (RMB/GJ)	36.73
electricity (RMB/kWh)	0.65

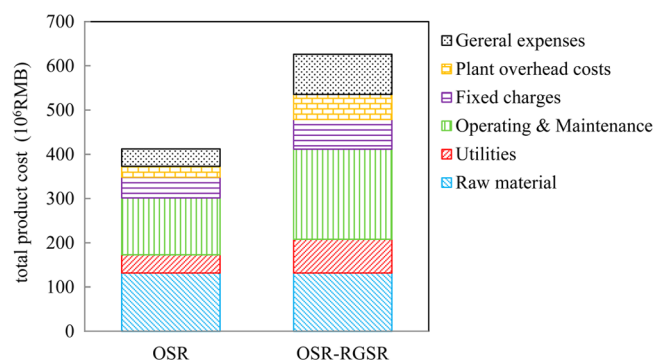


Figure 11. Product cost breakdown of the proposed and conventional processes.

**4.3. Return on Investment.** According to the current market prices of the products, the incomes of the OSR–RGSR and OSR processes are calculated as shown in Table 6. The income of the OSR–RGSR process increases by 385 million RMB/year compared to that of the OSR process. It is obvious that comprehensive use of the oil shale in this proposed process raises the income remarkably. This is because the detrital shale and the retorting gas can increase the economic benefit little because of their low unit prices. Produced hydrogen can give a big increase in the income. In general, it is found that upgrading of the oil product increases the income of 241 million RMB/year, while producing H<sub>2</sub> increases 166 more million RMB/year.

To estimate the benefit of the proposed process, the returns on investment of the two processes are analyzed. It is defined as

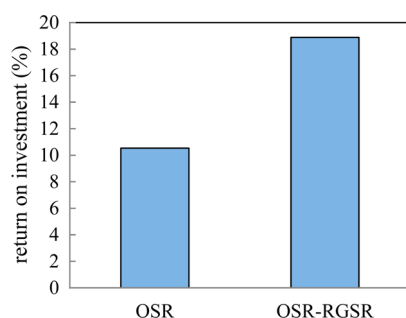
Table 6. Economic Comparison between the Proposed and Conventional Processes

item	price (RMB)	OSR		OSR–RGSR	
		production	income <sup>a</sup>	production	income <sup>a</sup>
shale oil (t)	5300	$8.0 \times 10^4$	424.00		
naphtha (t)	7100			$1.27 \times 10^4$	90.17
diesel (t)	8495			$6.53 \times 10^4$	554.72
LPG (t)	6100			$0.34 \times 10^4$	20.74
detrital shale (t)	35.40	$60.0 \times 10^4$	21.24		
retorting gas (m <sup>3</sup> )	0.07	$2.88 \times 10^8$	20.16		
H <sub>2</sub> (t)	23000			$0.72 \times 10^4$	165.60
ceramsite (m <sup>3</sup> )	160	$0.75 \times 10^5$	12.00	$1.00 \times 10^5$	16.00
brick (m <sup>3</sup> )	175	$2.57 \times 10^5$	44.98	$3.42 \times 10^5$	59.85
total income			522.38		907.08

<sup>a</sup>The unit of income is  $\times 10^6$  RMB/year.

the ratio of the total net profit to the total investment in a certain period of time.<sup>4</sup> The project construction period is assumed to be 1 year, while the production period is 20 years. The total profit of the OSR–RGSR and OSR processes are 261 million and 110 million RMB/year. The income tax rate is set to 25%. The total net profits of the two processes will be 211 million and 83 million RMB/year, respectively. Therefore, the returns on investment of these two processes are 18.89 and 10.53%, respectively, as shown in Figure 12. The return on investment of the OSR–RGSR process increases by 8.46% compared to that of the OSR process. In the proposed process, hydrogen is no longer outsourced but self-produced by the reforming reaction. In addition, the detrital shale is combusted for energy provision, which enhances the heat integration of the whole system and improves the economic benefit. Thus, the new proposed process can obtain much more economic benefit compared to the conventional oil shale refinery process.





**Figure 12.** Return on investment of the proposed and conventional processes.

## 5. CONCLUSION

In an aim to raise the economic performance of the oil shale refinery process, we proposed an integrated oil shale refinery with reforming of retorting gas for the hydrogen production process. This process integrates the oil shale retorting, the retorting gas steam reforming, and the hydrogenation techniques. Hydrogen produced in the reforming unit is supplied to the hydrogenation unit; besides, the remaining hydrogen is sold as revenue. Heat integration was implemented between the combustion of the detrital oil shale and the CaO carbonation reaction. The return on investment of the proposed process is 18.89%, which is 8.46% higher than 10.53% of the conventional refinery process.

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### Notes

The authors declare no competing financial interest.

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## NOMENCLATURE

- $A$  = pre-exponential factor
- $E_a$  = activation energy (kJ/kmol)
- $I$  = project investment (RMB)
- $k$  = reaction rate constant ( $\text{kmol m}^{-3} \text{s}^{-1} \text{Pa}^{-1}$ )
- $Q$  = production capacity (t/h)
- $R$  = gas constant ( $\text{kJ kmol}^{-1} \text{K}^{-1}$ )
- $T$  = temperature (K)

### Abbreviations

- BMP = building material production
- LPG = liquefied petroleum gas
- OSR = conventional oil shale refinery
- OSR-RGSR = integrated oil shale refinery with retorting gas steam reforming
- PSA = pressure swing absorption

### Subscripts

- $n$  = reaction order
- $sf$  = scale factor
- $t$  = time (s)
- $x$  = conversion ratio (%)

## Greek Letters

$\theta$  = domestic made factor

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