Applied Thermal Engineering 140 (2018) 102–111

Contents lists available at ScienceDirect

Applied Thermal Engineering

journal homepage: www.elsevier.com/locate/apthermeng

Research Paper

Simulation, exergy analysis and optimization of a shale oil hydrogenation process for clean fuels production

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HIGHLIGHTS

- A complete rigorous mathematics model of the shale oil hydrogenation process is proposed.
- The energy bottleneck of the shale oil hydrogenation process is identified and improved using exergy analysis.
- The preferable reaction temperature of hydrorefining and hydrocracking reactors is 350–375 °C and 375–400 °C.
- The reaction pressure and the catalyst grading ratio are 16–17 MPa and 1:3:3 for maximizing products yield.

ARTICLE INFO

Keywords:
Shale oil upgradation
Hydrorefining
Hydrocracking
Exergy analysis
Process simulation
Optimization

ABSTRACT

Because of high contents of unsaturated hydrocarbons, sulfur, nitrogen and other impurities in shale oil, its potential use as an alternative fuel is limited. Shale oil hydrogenation technology is an efficient technique for upgrading shale oil to remove heteroatomic compounds. The whole shale oil hydrogenation process is modeled and simulated firstly in this paper, including embedding the kinetic models of hydrogenation reactions. The corresponding-state group contribution method is applied to estimate the physical properties of the components of shale oil. The performance of the shale oil hydrogenation process is analyzed by exergy analysis. The results indicate that the exergy efficiency of the shale oil hydrogenation process is about 69.20%. The energy bottleneck is the furnaces and compressors because the energy consumption of these devices is 59% and 24% of that of the shale oil hydrogenation process. Furthermore, to optimize the performance of this process, the key parameters of the process are investigated and optimized. The preferable reaction temperature of the hydrorefining and the hydrocracking reactors is 350–375 °C and 375–400 °C. The reaction pressure and catalyst grading ratio are suggested to 16–17 MPa and 1:3:3 for maximizing the yield of products with ultra-low nitrogen and heteroatomic compounds.

1. Introduction

Oil shale, an unconventional fossil fuel, is considered as one of the most promising crude oil alternatives because of its globally abundant reserves of over 4.7 billion tons [1]. Its exploration and exploitation can effectively alleviate the shortage of energy resources. Thus, it has received widespread attention in recent years. Many countries, including the United States and China, are focusing on developing efficient, economic, and environmental deep-processing technologies of oil shale resources [2].

There are a variety of ways to utilize oil shale, such as retorting oil shale for shale oil production, burning oil shale for power generation or heating, and building materials production [3]. In China, oil shale resources are mainly retorted to produce shale oil as shown in Fig. 1(a). More than 60% of oil shale resources are retorted to produce shale oil. In recent years, oil shale industry has rapidly developed as shown in Fig. 1(b). As it shown, the annual production of shale oil has exceeded one million tons in 2014. It is expected to reach 1.2 million tons by 2020 [4]. Furthermore, China has built more than ten oil shale plants since 2015 [5]. These indicate that the oil shale industry has been on a certain scale in China.

However, most of Chinese oil shale retorting plants is simple and extensive. Its product, shale oil, is criticized by ‘three-high-and-one-low properties’, namely high content of wax, high content of unsaturated hydrocarbons, high content of nitrogen, and low asphaltene [6]. The heteroatom compounds can cause many problems in using shale oil,

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https://doi.org/10.1016/j.applthermaleng.2018.05.012
Received 14 December 2017; Received in revised form 12 April 2018; Accepted 5 May 2018

Nomenclature

k	the apparent reaction rate constant
r	the reaction rate
w	the mass fraction
η	exergy efficiency
Ex	exergy
FHC-FHT	single-stage reverse sequenced combination of hydrocracking and hydrorefining
HC	hydrocracking
HDS	hydrodesulfurization
HDN	hydrodenitrification
HeatX	heat exchanger model
HF	hydrofining
HP	hydroprotect
LHSV	liquid hourly space velocity
RadFrac	rectification column
RGibbs	minimum Gibbs energy change rector
RPlug	plug flow reactor
RStoic	stoichiometric reactor
RYield	yield reactor
SOH	shale oil hydrogenation

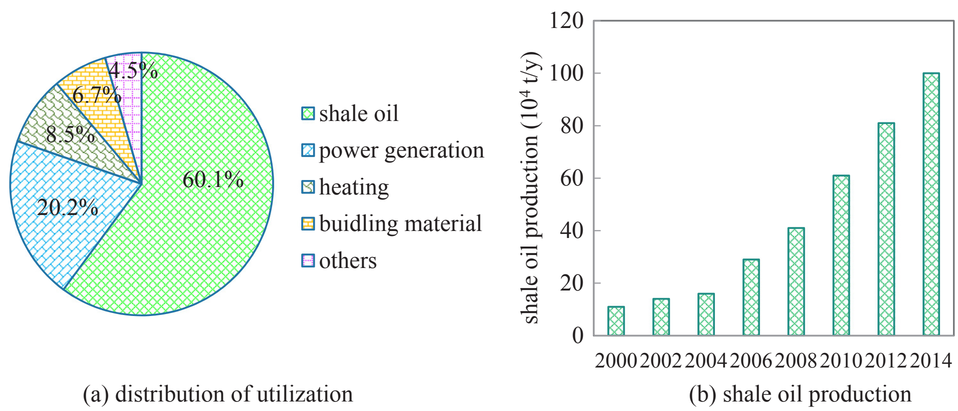

Fig. 1. Development of Chinese oil shale resources in recent years.

including instability of fuel in transportation or storage, catalyst poisoning in downstream treatment, and increase of the content of glial [7]. As a result, shale oil is used as the low-end fuel [8]. However, if the shale oil was directly burnt as fuel without any deep processing process, it would release the organic heterocyclic and unsaturated hydrocarbon compounds contained in the shale oil and pollute the surrounding environment [9]. Furthermore, in terms of energy demand, the demand trends of China are to increase the demand of gasoline and diesel but decrease that of heavy fuel oil. For example, from 2007 to 2015, the average annual growth rate of gasoline and diesel consumption was 8.8% and 8.3%; while the demand for heavy fuel oil decreased from 10.73% to 8.66% [1]. Thus, many researchers [10–12] suggest to upgradation shale oil to produce more high-valued clean fuels (such as, naphtha and diesel), which can improve the economic performance of shale oil plants and have a better social and environmental performance.

To upgrade shale oil, many shale oil upgradation technologies are developed to remove S, N, and O species from shale oil [12–14]. Hydrogenation of shale oil is considered as a common and efficient technique to upgrade shale oil to produce naphtha and diesel products [9]. It is to make a saturation of the aromatics, glial and asphaltenes in shale oil. Moreover, it obtains more saturated hydrocarbons with lower molecular weight through cleavage of open ring along with removing the S, N, O and other impurities [15]. Because of its good economic performance and high-quality products, the shale oil hydrogenation process is receiving more and more attention. For example, a 4×10^5 t/y shale oil hydrogenation plant is built in Fushun city, China.

Many literatures have been reported to investigate the shale oil hydrogenation process from the experimental point of view, including a number of studies on catalysts [12,16] and kinetic models [13,17]. Landau et al. [18] developed a novel catalyst for Israeli shale oil hydrogenation, which successfully reduced sulfur and nitrogen concentration in hydrogenation products and met the requirement for further hydrogenation. Luik et al. [12] systematically investigated the catalytic hydrotreatment of different fractions of Kukersite oil in a laboratory batch autoclave. The experimental results show that the shale oil hydrogenation process can greatly improve the density, heteroatoms, and unsaturation degree of the hydrogenated distillate. Johannes et al. [13] proposed a kinetic model for batchwise hydrogenation of shale oil. Dai et al. [17] developed a lumping kinetic model of hydrodesulfurization, hydrodenitrification, and hydrocracking based on catalysts grading. The results showed that the kinetic model can be used for optimization of catalyst stacking to meet the requirements of hydrogenation products.

To date, however, little work has been published to model and simulate the whole shale oil hydrogenation process, and there is a lack of research analyzing and optimizing the system performance of the shale oil hydrogenation process from the process scales. But many researches [19–21] have been reported to model, simulate and analyze the coal tar hydrogenation process, which is very similar to the shale oil hydrogenation process. For example, Gao et al. [19] modeled the typical coal tar distillation process. On the basis of the models, they analyzed the effects of the main parameters (such as plate count and reflux ratio) on the yields of products and identified the optimal conditions. Tang et al. [20] built the multi-scale models of the coal tar hydrogenation process as well as investigated the effect of reaction temperature and pressure on the quality of hydrogenation products and system energy consumption. Ci et al. [21] simulated a coal tar hydrogenation process by using several typical model compounds. Comparing with the experimental and industrial data, it validated that the whole simulation model is reliable. These studies have greatly shown that it is feasible to apply the existing research tools to multi-scale simulation, analysis and optimization of the shale oil hydrogenation process.

Based on the above discussion, the main research problems discussed in this paper are: (1) The material and energy flow of the whole shale oil hydrogenation (SOH) process has not yet been clearly understood; (2) The energy bottleneck of the SOH process has not yet been identified; and (3) How to reasonably select the key operating parameters of the SOH process to achieve overall optimization. Solving these problems are important to improve the technical, economic and