



Tech-economic assessment of a coproduction system integrated with lignite pyrolysis and Fischer–Tropsch synthesis to produce liquid fuels and electricity

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ABSTRACT

A novel coproduction system of lignite pyrolysis integrated with Fischer–Tropsch synthesis and coal-tar hydrogenation to produce liquid fuels and chemicals is proposed. The moisture content of the lignite, the gasifier adopted and the choice of electricity generation are the critical factors that make a big difference in the assessment of the system. Sensitivity analysis of different moisture contents and gasifier types are investigated and comparisons are done between processes with and without electricity generation. The results showed that the system could benefit a lot from the decrease of moisture content of feedstock, and that although requiring higher financial investment, Shell-gasifier-based process would produce more liquid. Moreover, applying the electricity generation system would enhance the economic performance and make an impact on the energy efficiency. By the energy and economic assessments, nearly 40% of the low heat value and over 15% of the internal rate of return in the best case are obtained at an annual handling scale of 5 million tons of lignite. Meanwhile, over half a million tons of liquid are obtained and the CO₂ emission is within the range of 7.5–9.5 t/t-oil.

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1. Introduction

Energy is a key element of the sustainable development of human society. This is especially true for developing countries. Although crude oil prices have been dropping in the recent half year from July of 2014, future oil prices are still notoriously difficult to predict [1], and no one is certain that it won't rise again. Therefore, the world still has to seek for potential substitutes to crude oil for the sake of the possible rebound of the price. Although rich in coal resources, China is facing increasing dependence on foreign oil in the last two decades [2]. In order to keep the high developing pace and guarantee energy security in the long run, China is now looking for ways to convert its relatively abundant coal resources, such as via Fischer–Tropsch synthesis (FTS), methanol to olefins (MTO) and other advanced coal-based synthesis technologies that are led by gasification. Now, the high-rank and high-quality coal supplies are limited whereas lignite, which counts for more than 10% of the total coal reserves, has not been put into efficient utilization due to its high moisture and ash content and low heat value [3].

Nowadays, many proposals have been put forward about lignite upgrading and conversion. Some studies focus on drying and dewatering aiming at improving the heat value of lignite [4], others are devoted to the improvement of quality of pyrolysis products. Furthermore, lignite is also directly used for gasification in some researches [5]. As for the pyrolysis

approaches, the tar obtained in a multi-stage fluidized bed by Zhou et al. [6] contained more light oil and the char showed higher thermal stability. Xu's [7] study illustrated the relationship between temperature and the distribution of specific components such as CO₂, CO and C₂H₄. As one of the useful byproducts of coal pyrolysis, millions of tons of coal-tar are produced all over the country [8], and the studies and demonstrations of its utilization have been widely carried out, one aspect is to make liquid fuels by hydroprocessing. Tang et al. [9] hydrotreated low-temperature coal-tar under mild conditions to determine the composition of the products. Li et al. [10] analyzed the properties of liquid oil from low-temperature tar hydrotreatment in a trickle bed reactor and found that the oil contained a small amount of sulfur and the diesel fraction could be used as motor fuels without upgrading.

However, previous studies seldom gave an integrated process that could demonstrate how lignite was treated step by step to form the final products, such as oil or liquid fuels. Based on previous work on lignite pyrolysis by solid heat carrier and char gasification, which aimed at improving the tar output and char quality [11], a liquid fuels aimed coproduction system was designed in this work with the consideration of the further usage of those intermediate products. Compared with the previous work, the new system added the coal tar hydrogenation and FTS process to increase the oil output. The crude oil from these two sources both have low sulfur and nitrogen contents, and can act as clean additives for motor fuels in stand-alone plant, respectively. In fact, liquid fuels are a mixture of hydrocarbon compounds that share the same range of boiling points. Therefore, the two streams are designed

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to become mixed for distillation. The combustible gases are either sold to be used for generating electricity or for further processing. The simulation of the coproduction system was established by Aspen Plus. In the end, the coproduction system was evaluated by energy, economy and environmental effects.

2. System description

2.1. System setting-up

The oil-aimed coproduction system is mainly composed of a lignite-drying pyrolysis unit, a gasification unit, a gas clean-up unit, a FTS unit, a tar hydrogenation unit, and a separation unit. The raw lignite is dried and pyrolyzed by solid heat carriers, and then tar, char, and pyrolysis gases are obtained. The used heat carriers are then mixed and reheated in the combustion unit. The lignite tar from the low-temperature pyrolysis unit is rich in phenols, which are highly valuable, so they are first separated and the remaining part that is full of saturated hydrocarbons and olefins goes to hydrogenation. The whole pyrolysis gas and part of the char are burned for heating the solid heat carriers. The rest of the char is gasified to produce syngas, which is then used for FTS to make liquid fuels. The description of the following processes are similar to the stand-alone coal to liquid system. A part of the syngas is used in the water–gas shift reaction (WGS) unit to provide enough H₂ for the FTS unit and the hydrogenation unit. The H₂ in the tail gas is also separated by pressure swing adsorption (PSA) to hydrocrack the heavy fraction from FTS. Finally, all the crude oil is rectified to obtain the target fractions, namely, gasoline and diesel.

Compared with the stand-alone coal-to-liquid system or tar-hydrogenation system, such a coupled coproduction system is able to share some common facilities, and consume less fresh water by guiding the steam from drying unit to gasifier or to WGS reactor.

As shown in Fig. 1, the whole system was simulated with the process simulation software Aspen plus at an annual handling scale of 5 million tons of lignite. In the simulation, as in the previous work [11], HulunBeir lignite was adopted, and its proximate analysis and ultimate analysis were listed in Table 1.

2.2. Simulation assumptions

The crude oil from FTS, tar hydrogenation and hydrocracking have different components and contents. There is no way to identify all those thousands of components in coal tar. Considering that the main target products are gasoline and diesel fractions, several model compounds were selected according to the element content, component

Table 1
Proximate and ultimate analysis of samples [11].

	Proximate analysis, wt.%, ad				Ultimate analysis, wt.%, ad				
	M	A	V	FC	C	H	O ^a	N	S
Coal	9.69	11.01	33.6	45.7	58.56	3.51	15.83	1.02	0.38
Char	2.47	21.44	15.95	60.14	65.06	3.31	5.98	0.95	0.79

Note: ^a- By difference.

category, functional groups and boiling points for the tar, gasoline and diesel. For example, methylphenol and naphthol were model compounds for phenols in different fractions, while quinoline was chosen for nitrogen compounds.

In this simulation, a Fe-based low-temperature Fischer–Tropsch (LTFT) process, which is suitable for the production of high-quality diesel fuel, is adopted. Compared with high-temperature Fischer–Tropsch (HTFT), the LTFT process produces more diesel fraction but less olefins and aromatics [12,13]. Hence, some alkanes, alkenes and alcohols with different carbon number were chosen as model compounds. The Fe-based catalyst is able to catalyze the WGS, so the H₂/CO ratio needed is about 1, while this value can be over 2 for Co-based catalyst [14,15]. Overall, the distribution of FTS products follows the Anderson–Schultz–Flory (ASF) law [16]. The simulated distribution is shown as Table 2.

Kusy et al. [17] found that the mixture of hydrocarbons obtained from tar hydrogenation was similar to the naphtha cut and virgin diesel cut produced in crude oil processing. Moreover, results from Li et al. [10] showed that about 100 peaks and more than 150 peaks were detected in the GC–MS analysis of gasoline fraction and diesel fraction, respectively. A majority of the gasoline fraction was composed of paraffin and substituted cyclohexane, while the diesel fractions were substituted with saturated ring-containing hydrocarbons and straight-chain paraffins. Although the crude products from FTS mainly contain linear hydrocarbons, those from tar hydrogenation are rich in cyclic hydrocarbons, both of them contain tiny sulfur and nitrogen, which are good additives for clean gasoline and diesel. In particular, the diesel fraction of FTS has a very high cetane value, as high as 75, which can be either used directly as a vehicle fuel or as an additive [18,19]. The possible drawback of the diesel from tar hydrogenation might be the relatively low cetane value, but this does not keep it from being a good additive for diesel [10]. The waxes, when being hydrocracked, will become another qualified additive that is characterized by low-temperature performance. Therefore, based on the similarities in components and application, these crude liquids should reduce capital investments if they go into one set of separation system.

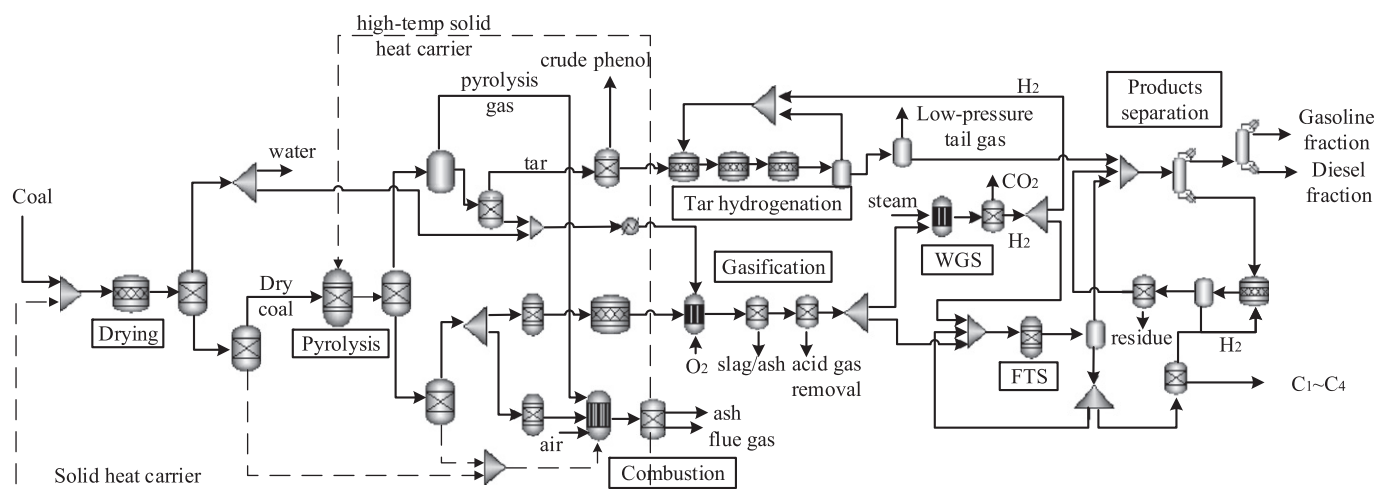


Fig. 1. Process diagram of the new coproduction system.

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