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Comparative study of coal, natural gas, and coke-oven gas based methanol to olefins processes in China

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ABSTRACT

Traditional olefins production mainly depends on oil. In view of the short supply of oil, feedstocks are expanded to coal, natural gas, coke-oven gas, and methanol in China. In this paper, a comparative study of alternative olefins production is conducted from aspects of techno-economic feasibility and environmental friendliness. Results show that coal-to-olefins has a significant cost advantage. However, it suffers from low energy efficiency and serious CO_2 emissions. To address these problems, this study proposes and analyses coal-to-olefins with CO_2 capture, coal and natural gas-to-olefins, and coal and coke-oven gas-to-olefins. The two co-feed systems ensure great reduction of CO_2 emissions and significant improving energy efficiency. They should be actively developed in regions with rich coal and gas. While in regions with rich coal and lean gas, coal-to-olefins with CO_2 capture should be developed in large scale. This paper also provides several suggestions on planning these olefins production routes in China.

1. Introduction

Ethylene, propylene, and butene (olefins) are one of the most important platform chemicals, which drive the development of a variety of organic chemical products. The world olefins production capacity and demand have been steadily growing. Just in China the yields and equivalent demands of olefins increased by 15.6% and 11.2% from 2005 to 2011 (Qu, 2012). However, the self-sufficiency rates of ethylene and propylene in China would be only 53% and 74% by 2015 (Qu, 2012). The situation should be improved by the quick development of olefins industry.

Traditional olefins production mainly depends on oil. With the rapid development of petrochemical industry, Chinese oil-import increases year by year. In 2012, China imported 271 Mt oil, accounting for 56% of the total oil consumption (478 Mt) (Hong, 2013). Chinese oil resources could not satisfy the development of domestic petrochemical industry. The sustainable development of olefins industry requires diverse resources from both domestic and overseas. Alternative olefins production under operation is coal-to-olefins (CTO) and methanol-to-olefins (MTO) in China (CNII, 2013). The present feedstock of methanol production in China is coal – 63.7%, natural gas – 23.0%, and coke-oven gas – 11.3% (Su et al., 2013). Shenhua Group successfully applied DMTO technology to produce olefins by coal based methanol in 2011. Therefore, the successful operation of the MTO process marks that olefins could be produced from coal, natural gas, coke-oven gas, and methanol.

Techno-economic analysis is an essential part for planning Chinese olefins industry. Gao et al. (2008) proposed natural gas-based poly-generation system for methanol production. Luo et al. (2012) conducted a techno-economic study of coke-oven gas-to-methanol. Dong (2013) used CO_2 in coke-oven gas-to-methanol process to improve its utilization. These studies only analyze natural gas-to-methanol and coke-oven gas-to-methanol processes. Ren et al. (2008) analyzed energy use and CO_2 emissions of steam cracking and methane to olefins. Ren and Patel (2009) also analyzed energy use and CO_2 emissions of basic petrochemicals from natural gas, coal and biomass. Fu and Xu (2013) employed dynamic simulations to investigate energy consumption and emissions generation for an ethylene plant under different start-up strategies. They paid more attention on energy utilization and CO_2 emissions of these processes. Yang and Dong (2012) investigated techno-economic performance of light olefins producing from syngas. The authors' previous work also conducted a techno-economic analysis of coal-to-olefins and oil-to-olefins (Xiang et al., 2014a). However, very few authors systematically

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Nomenclature

CCS	carbon capture and storage
CTO	coal-to-olefins
DMR	dry methane reforming
DMTO	dimethyl ether/methanol-to-olefins
MTO	methanol-to-olefins
SMR	steam methane reforming
CGTO	coke-oven gas-to-olefins
NGTO	natural gas-to-olefins
C-NGTO	coal and natural gas-to-olefins
C-CGTO	coal and coke-oven gas-to-olefins
CTOwCCS	coal-to-olefins with CCS

analyze alternative olefins production in China. Natural gas-to-olefins and coke-oven gas-to-olefins is synthesized by integrating methanol synthesis and MTO technology in this paper. Then, this paper makes a comprehensive technical and economic analysis of olefins production from coal, natural gas, coke-oven gas, and methanol. The main indicators for assessment are energy efficiency, CO_2 emissions, capital investment, and product cost. In addition, the study also attempts to identify the techno-economic bottleneck for different olefins routes.

There have been a number of CO_2 mitigation methods developed in terms of huge CO_2 emissions (Markewitz et al., 2012). Physical reduction methods involve storing CO_2 underground. Carbon capture and storage (CCS) technology has received increasing attention because of its high capacity of reducing CO_2 emissions (House et al., 2009). Mantripragada and Rubin (2011) conducted a techno-economic evaluation of coal-to-olefins (CTL) plants with carbon capture and sequestration. Techno-economic performance of China's indirect coal liquefaction projects with different CO_2 capture alternatives was also analyzed in the work of Zhou et al. (2011). By using chemical reduction methods, CO_2 is reused as feedstock in many different applications, such as the production of urea, methanol, and olefins. Besides, co-feed systems of coal with other hydrogen-rich resources to produce chemicals have been attracting more and more attention. Zhou et al. (2009) studied systems based on coal and natural gas for producing dimethyl ether. Adams and Barton (2011) made a techno-economic analysis of a co-feed system of coal and natural gas for production methanol and electricity. They all found that the performance of co-feed system is much better than that of single feed system. Combined dry methane reforming with steam reforming for CO_2 treatment was also investigated in the study of Lim et al. (2012). Based on the situation, this study also proposes and analyses coal-to-olefins with CCS (CTOwCCS), coal and natural gas-to-olefins (C-NGTO), and coal and coke-oven gas-to-olefins (C-CGTO).

2. Methodology

In this paper, major units of alternative olefins production processes with the same capacity of 0.7 Mt/y olefins are modeled in process simulation software Aspen Plus. For a plant with given capacity and specified operating conditions, the model calculates all mass and energy flows. Then, the analysis of energy efficiency, CO_2 emissions, capital investment, and product cost are conducted. Therefore, models of techno-economic analysis and process simulation are introduced in this section.

2.1. Energy efficiency and CO_2 emissions estimation

The energy efficiency is the ratio of product energy (E_{pd}) to total input energy (E_{in}) given in Eq. (1). The product energy contains energy of ethylene, propylene, and butane. The total input energy contains energy of feedstock, steam, and electricity. The energy of olefins and feedstock is calculated on the basis of their lower heating value.

$$\eta = \frac{E_{pd}}{E_{in}} \times 100\% = \frac{E_{Ethylene} + E_{Propylene} + E_{Butane}}{E_{Feedstock} + E_{Electricity} + E_{Steam}} \times 100\% \quad (1)$$

Total CO_2 emissions can be counted as the sum of direct emissions and indirect emissions. Direct emissions are generally from exhaust gas of the production process, while indirect emissions are from upstream, such as resource extraction, processing, and power generation. It is calculated by the sum of product of process energy consumption and the corresponding emissions factor. At the electricity mix of coal 80.8%, oil 1.8%, and NG 0.7% in China, the indirect CO_2 emissions factors of electricity and steam are 248.02 g/MJ and 113.87 g/MJ. The indirect CO_2 emissions of coal, natural gas, and coke-oven gas are 5.73 g/MJ, 16.58 g/MJ, and 6.69 g/MJ, respectively (Ou et al., 2011).

Table 1
Equipment investments of different olefins production routes.

Processes	Capacity	Equipment investment
CTO[a]	0.7 Mt/y	3.17×10^9 RMB
NGTO[b]	0.7 Mt/y	2.05×10^9 RMB
CGTO[c]	0.7 Mt/y	2.33×10^9 RMB
MTO[b]	0.7 Mt/y	1.45×10^9 RMB
CTOwCCS[a]	0.7 Mt/y	3.41×10^9 RMB
C-NGTO[d]	0.7 Mt/y	1.90×10^9 RMB
C-CGTO[e]	0.7 Mt/y	2.23×10^9 RMB

[a] The investments of CTO and CTOwCCS are estimated from Xiang et al. (2014a).
[b] The investments of NGTO and MTO are estimated from Feng et al. (2011) and Xiang et al. (2014b).
[c] The investment of CGTO is estimated from Wang et al. (2009) and Xiang et al. (2014b).
[d] The investment of C-NGTO is estimated from Zhou et al. (2009) and Xiang et al. (2014a).
[e] The investment of C-NGTO is estimated from Man et al. (2014).

2.2. Economic assumption and cost estimation

Before an industrial plant can be put into operation, a large budget need to be allocated to purchase and install the necessary machinery and equipment. The capital needed for manufacturing and plant facilities is fixed-capital investment, while that for the operation of the plant is working capital. The sum of fixed-capital investment and working capital is total capital investment. The equipment investments for olefins production routes are estimated according to references in Table 1, which has been updated to 2013 prices by using the Chemical Engineering Plant Cost Index. The total capital investment can be assessed by these equipment investments, ratio factor in Table 2, and Eq. (2) (Orhan et al., 2008).

$$\text{Total capital investment(TCI)} = I_{EI}\left(1 + \sum RF_i\right) \quad (2)$$

I_{EI} is the investment for main equipment components, RF_i is the ratio factor for direct/indirect and working capital investment i = (1.1)–(1.7), (2.1)–(2.4), and (4) in Table 2.

Product cost is calculated by Eq. (3) and economic assumptions presented in Table 3. The costs of raw materials and utilities are calculated according to corresponding consumption and market price in 2013 of China. The capital investment is involved in the product cost as the form of depreciation cost. A straight-line method is adopted to calculate the depreciation cost with the assumption of 20 year life time and 4% residual value. The rest part of product cost is calculated by ratio estimation (Orhan et al., 2008). The exchange rate between US dollar and RMB is assumed to be 6.2 and