

Integrated petroleum coke and natural gas polygeneration process with zero carbon emissions



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ABSTRACT

In the present work, a new polygeneration system is developed that uses petcoke and natural gas as feedstocks and coproduces different products such as chemicals, olefins, electricity and transportation fuels. Furthermore, by incorporation of chemical looping combustion and chemical looping gasification technologies, 100% of CO₂ emissions are captured effectively. The particle swarm optimization technique is implemented with Aspen Plus models to determine the optimum product portfolio at different market conditions. Techno-economic optimization results show that this plant can be profitable for a broad range of petcoke consumption (up to 74%) and various feeds and products price changes.

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1. Introduction

As the supply of light crude oils diminishes and the processing of heavier oil increases, crude oil refineries face growing challenges associated with this increased use of heavier crude. For instance, the API (American petroleum institute) gravity of crude produced in the United States has been steadily decreasing [1]. One result of this change is an increasing rate in the production of extremely heavy residues called petroleum coke (or petcoke), even when upgrading units such as delayed coking and fluid coking are implemented [2,3]. The global production of petcoke has been accelerated to about 4% per year [4] due to the invention of new technologies that can extract crude oil from shale oil and oil sand reservoirs.

Among the several emerging technologies available for the upgrading of petcoke, gasification has emerged as an effective approach that can convert petcoke to more valuable products with minimum environmental impacts compared to commercial combustion systems. In this approach, solid petcoke is converted to synthesis gas and, after purification, is used as a feedstock for other chemical units. Although different petcoke gasification mechanisms and technologies have been studied extensively [1,5–7], little research has studied the techno-economic analysis on the poly-generation processes that use them. For example, a conceptual

study of a large scale polygeneration plant that converts petcoke to power, chemicals such as ammonia and MeOH, hydrogen and transportation liquids was performed by Jacob Consultancy [8]. To the best of our knowledge, this report is the only techno-economic analysis of petroleum-based polygeneration systems. However, their preliminary results showed that the proposed system was not profitable in most cases with current technologies.

The incorporation of a natural gas reforming process which produces relatively inexpensive hydrogen rich synthesis gas can improve the performance of polygeneration plants significantly. Comprehensive studies which consider the coupling of pipeline natural gas reforming with commercial coal gasifiers [9,10], as well as shale gas resources [11,12] have shown promising results for a variety of different combinations of products and processing routes. Some of these proposed routes incorporate chemical looping gasification [13–15] and chemical looping combustion [16,17], which can help facilitate low cost CO₂ capture.

Although these studies provide promising ways to produce energy products with low-to-zero process CO₂ emissions with coal and gas, petcoke has not yet been examined as a potential resource to use at the systems level. Even though petcoke and coal have some similarities, the results of the prior studies using coal cannot be readily extrapolated to petcoke. Fundamentally, coal is a more valuable commodity, and petcoke is a low-quality waste product of limited use. As such, petcoke gasification systems produce syngas with a different quality from the coal-based equivalent, which

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significantly affects the design and economics of the polygeneration plant. The primary innovation of this work is development and techno-economic optimization of the first petcoke-based polygeneration process with zero CO₂ emissions. In addition, the incorporation of shale gas steam reforming is another novelty of this work. We focus in the present study on various processes that can upgrade petcoke to chemicals and fuels with minimal environmental impacts. Therefore, the purpose of this work is to determine how petcoke can best be used in combination with natural gas in a polygeneration system which produces different energy products while maintaining zero direct CO₂ emissions.

In the present work, a novel polygeneration process is presented that consumes petcoke and natural gas as the main feedstock, utilizing novel technologies for power generation, hydrogen production and olefin production. Although many kinds of petcoke could be used, Syncrude coke was selected as the example for this work since it is produced and stockpiled in large amounts in Canada. In 2011, the total petcoke production rate in Canada was about 10 million tonnes per year [4], which is equivalent to 3500 MW electricity generated by typical IGCC (integrated gasification combined cycle) plants [7]. Although conventional natural gas could be used, Haynesville shale gas was selected as the example gas feed in order to demonstrate that, if desired, it is possible to use desulfurized shale gas that has not been completely converted to conventional natural gas quality. Conventional pipeline natural gas or other shale gases could be used instead with minor operating changes to the reforming system [11,12].

As illustrated in Fig. 1, the syngas produced from petcoke gasification and shale gas reforming are routed into two parallel trains, the methanol (MeOH) and dimethyl ether (DME) synthesis unit, and the Fischer–Tropsch (FT) synthesis unit. In the MeOH/DME unit, syngas is converted to MeOH which can be stored or optionally routed to the DME reactor section to produce fuel-grade DME. In addition, the MTO (methanol-to-olefin) unit is coupled with the MeOH/DME section to diversify the product portfolio by producing ethylene and propylene from MeOH. In the FT section, the syngas stream produced by the petcoke gasifier is sent to the FT reactor after being mixed with H₂-rich syngas to get the ratio H₂/CO = 2.1 which is ideal for the FT chain reactions. It should be noted that

prior to sending syngas to the FT reactor, it must be purified to remove CO₂. In our proposed system, a nickel-oxide (NiO) CLC (chemical looping combustion) system is used to supply the electricity demand of plant from purge gas streams. Furthermore, the heat, steam and hydrogen demands of the FT unit are provided by using an iron-oxide (Fe₂O₃) chemical looping gasification unit. One key advantage of the CLC system is that it facilitates 100% CO₂ capture from these purge gases. As a result, the proposed polygeneration process features essentially zero direct CO₂ emissions, and is profitable in a wide range of market prices and feedstock blends (different ratios of petcoke and natural gas can be used). In addition, the particle swarm optimization technique was employed to determine the optimum process conditions for a variety of objective functions, such as the maximization of profit or yield.

2. Process simulation

Each polygeneration plant considered in this work was scaled to a combined 1111 MW LHV (lower heating value) energy input of petcoke and shale gas, which is equivalent to around 2000 tonne per day of shale gas. The composition and other key properties of petcoke and shale gas are listed in Table 1. All process unit operation models were simulated in Aspen Plus 2006.5 simulation software, except for the gas turbines for which a custom model in

Table 1
Properties of petcoke and shale gas feedstocks.

Petcoke [5]: Syncrude coke;					
Gross heating value: 13923 Btu/lb (HHV)					
Property proximate (wt. %)					
Moisture	0.25	Ash	4.83		
Volatiles	4.99	Fixed carbon	89.95		
Ultimate (wt. %)					
Carbon	83.74	Chlorine	0.25	Nitrogen	2.03
Hydrogen	1.77	Sulfur	6.52	Oxygen	0.88
Shale gas [21]: Haynesville					
Gas composition (% molar)					
Methane	0.948	CO ₂	0.05		
Ethane	0.001	N ₂	0.001		

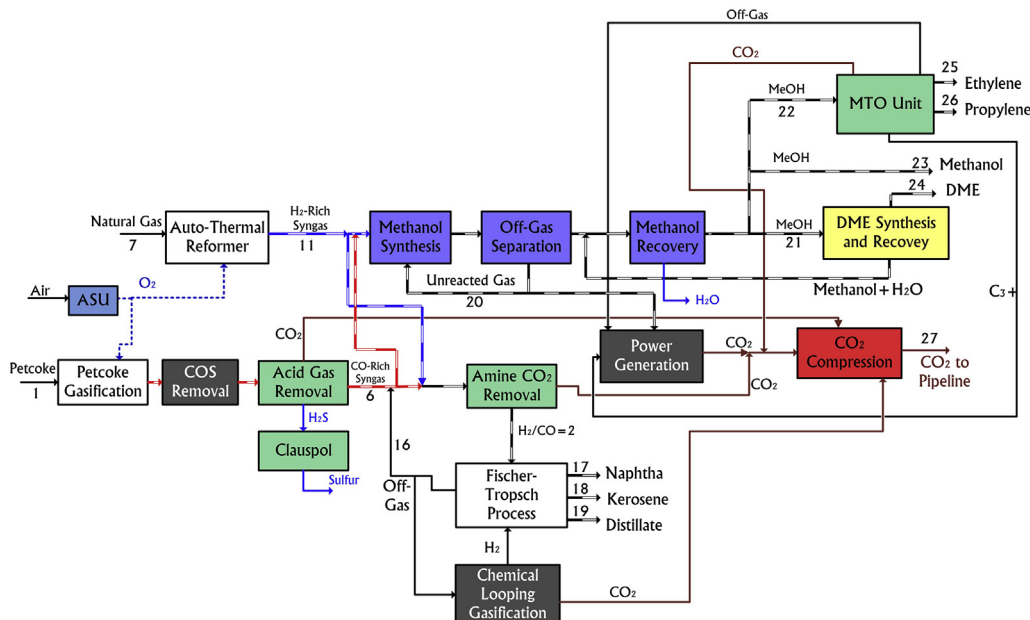


Fig. 1. Scheme of the petcoke-natural gas superstructure. Streams numbers also correspond with those in Figs. 2–4.

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