

# Integrated process for synthetic natural gas production from coal and coke-oven gas with high energy efficiency and low emission

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## ABSTRACT

There was a rapid development of coal to synthetic natural gas (SNG) projects in the last few years in China. The research from our previous work and some other researchers have found coal based SNG production process has the problems of environmental pollution and emission transfer, including CO<sub>2</sub> emission, effluent discharge, and high energy consumption. This paper proposes a novel co-feed process of coal and coke-oven gas to SNG process by using a dry methane reforming unit to reduce CO<sub>2</sub> emissions, more hydrogen elements are introduced to improve resource efficiency. It is shown that the energy efficiency of the co-feed process increases by 4%, CO<sub>2</sub> emission and effluent discharge is reduced by 60% and 72%, whereas the production cost decreases by 16.7%, in comparison to the conventional coal to SNG process. As coke-oven gas is a waste gas in most of the coking plant, this process also allows to optimize the allocation of resources.

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## 1. Introduction

In China, coal accounts for 67.4% of the total energy consumption [1]. Too much coal consumption, insufficiently clean processing and burning have brought about serious haze problem in China. Using natural gas to replace coal is one of the key measures to tackle with the air pollution caused by coal combustion [2]. However, the production of natural gas is only  $1.57 \times 10^{11} \text{ m}^3$  (including the production of shale gas) while the natural gas consumption is about  $2.5 \times 10^{11} \text{ m}^3$  in 2015 [3]. China is facing the problem of supply shortage of natural gas.

Aiming at the promising market opportunity, Chinese industry made a great efforts to push forward coal to synthetic natural gas (CtSNG) process in recent years. Till now, there have been  $8.73 \times 10^{10} \text{ m}^3/\text{y}$  production capacity of CtSNG projects in commercial operation in China [4].

A schematic diagram of CtSNG process is shown in Fig. 1. Feedstock raw coal is gasified to crude syngas. The crude syngas is sent into the water gas shift (WGS) unit to adjust the hydrogen-to-carbon (H/C) ratio. Next the syngas is sent into the acid gas removal (AGR) unit to remove CO<sub>2</sub> and sulfide. Finally, the clean syngas is sent into methanation unit for producing SNG [5]. Li et al. [6] made a comparison of techno-economic and environmen-

tal performance between coal-based SNG and coal directly burning. The results showed that the coal based SNG using in residential sector could relieve urban haze pollution caused by the direct coal burning. However, coal based SNG has nearly two times higher consumption of raw material and 1.65 times higher CO<sub>2</sub> emission than coal burning.

CtSNG process uses lignite as the feedstock due to the limitation of resource allocation. That means the crude syngas from gasification would contain large amount of CO<sub>2</sub>. H/C ratio of the crude syngas is around 2.3–2.7, while the required H/C ratio for methanation is about 3.1–3.3. The syngas is lack of hydrogen source. CO of the syngas needs to be converted to H<sub>2</sub> in WGS unit. It results in a high emission of CO<sub>2</sub> and waste of carbon resource, as well as very high consumption of energy in the WGS unit. Meanwhile, the crude syngas from Lurgi gasification needs to be washed with water to remove the phenol and ammonia. This process generates almost 98% waste water of the CtSNG process [7]. To treat the effluent with high concentration of phenol and ammonia would consume large amount of energy together with high production cost. Discharge of the effluent without fully treatment would cause serious impact to the environment [8].

Coke-oven gas (COG) is generated in the coking process as a by-product of coal carbonization. The annual COG production is estimated at  $7 \times 10^{10} \text{ m}^3$  in China [9]. However, most of the COG is directly discharged into the atmosphere. It causes considerable energy waste and serious environmental consequences [10]. COG is a kind of hydrogen-rich gas, which contains H<sub>2</sub> (55–60%), CH<sub>4</sub>

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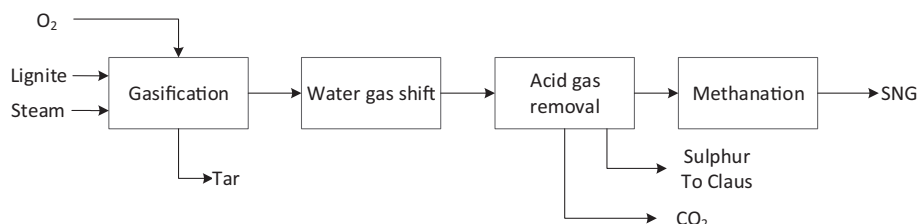


Fig. 1. Schematic diagram of CtSNG.

(23–27%), CO (5–8%), N<sub>2</sub> (3–6%), CO<sub>2</sub> (less than 2%). Co-feed system could take advantage of coal and COG to produce syngas with suitable H/C ratio. Authors' previous work [11] on COG assisted coal to olefins process found that the energy efficiency of co-feed system improved by 10% and the CO<sub>2</sub> emission decreased by 85% when compared with single coal feed. The co-feed process of coal and COG to SNG is a promising alternatives in dealing with the problems of high CO<sub>2</sub> emission and energy consumption. In China, most of CtSNG projects are surrounded by coal coking plants. Plentiful COG source makes assurance for feedstock supply for the co-feed system of coal and COG to SNG process.

Following the above ideas, this paper proposes a novel co-feed process of coal and COG to SNG with high energy efficiency and low CO<sub>2</sub> emission. The co-feed process integrates a dry methane reforming unit to reduce CO<sub>2</sub> emission, and uses the hydrogen resource in COG to raise the H/C ratio of the syngas. A conceptual design and a techno-economic analysis for the co-feed process is put forward in this paper.

## 2. Coal and coke-oven gas to SNG process

The schematic diagram of the proposed co-feed process of coal and COG to produce SNG (CGtSNG) is shown in Fig. 2. CGtSNG process contains coal gasification unit, acid gas removal unit, and methanation unit. However, it removes the WGS unit, while adds a COG separation unit and a dry methane reforming (DMR) unit compared with CtSNG process. Part of methane from COG is sent into DMR unit to react with CO<sub>2</sub> from the coal gasification unit. DMR unit produces clean syngas and reduces CO<sub>2</sub> emission. The syngas with suitable H/C ratio for methanation is obtained by adjusting the proportion of DMR syngas, coal gasification syngas, and H<sub>2</sub> from COG.

CGtSNG process is composed of three subsystems: (1) coal gasification and syngas clean; (2) COG separation and dry methane reforming; (3) methanation. The subsystems are introduced as

follows. Feedstock composition of CGtSNG is shown in Table 1. The proximate and ultimate analysis of lignite is shown in Table 2. The data is referred to the lignite from Shengli coalfield of Xilinhote, China. To facilitate comparison between CGtSNG and CtSNG, the production capacity of SNG in CGtSNG is set to 4.0 billion m<sup>3</sup>/y, same as industrial CtSNG project of Datang Corporation in Chifeng, China [7].

### 2.1. Coal gasification and syngas clean

Due to the resource allocation limitation, all the coal to SNG processes in China use lignite as the raw material. That means only Lurgi and BGL gasification technologies could be used in CtSNG. The syngas from Lurgi gasification contains 39% H<sub>2</sub>, 14% CO, and 32% CO<sub>2</sub>. The syngas from BGL gasification contains 28% H<sub>2</sub>, 55% CO, and 6% CO<sub>2</sub>. BGL has advantage in low CO<sub>2</sub> content. However, H/C ratio of BGL gasification syngas is as low as 0.5, while the H/C ratio of Lurgi gasification syngas is 2.7. For methanation, the H/C ratio is required above 3.05 at least. That means a large scale of water gas shift unit is needed to convert CO into CO<sub>2</sub> and H<sub>2</sub> to raise the H/C ratio. The total amount CO<sub>2</sub> for CtSNG process of BGL is 18.8% higher than that of Lurgi. That also means that the handling scale of coal gasification unit and acid gas removal unit should be enlarge which causes the increasing of capital investment and operation cost. In China, the existent CtSNG projects in commercial operation uses Lurgi gasification technology.

The subsystem of coal gasification and syngas clean selects Lurgi pressurized coal gasification technology and Rectisol methanol washing technology as the simulation model referred to the CtSNG project of Datang Corporation in Chifeng, China. The flow-sheet of this subsystem is shown in Fig. 3.

In coal gasification process, lignite is sent into gasifier together with oxygen from air separation unit (ASU) and steam. This process begins with lignite drying and pyrolysis, and continues with combustion and steam gasification. Main reactions in gasification

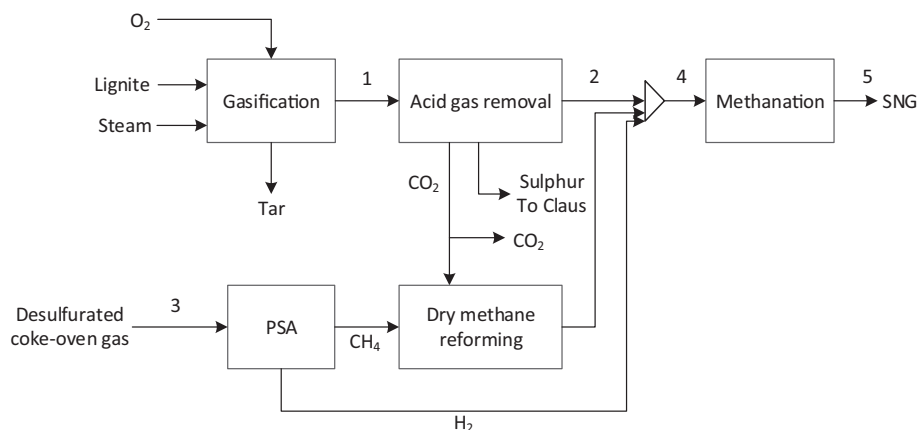


Fig. 2. Proposed schematic diagram of CGtSNG process.

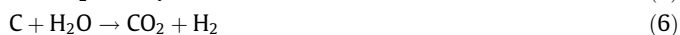
**Table 1**  
Feedstock for CGtSNG process.

Raw material	Temperature, °C	Pressure, MPa	Mass flow, t/h
Lignite	25	4.0	558
COG	25	2.5	470
Water	25	0.4	1670
Oxygen	40	4.0	94.5

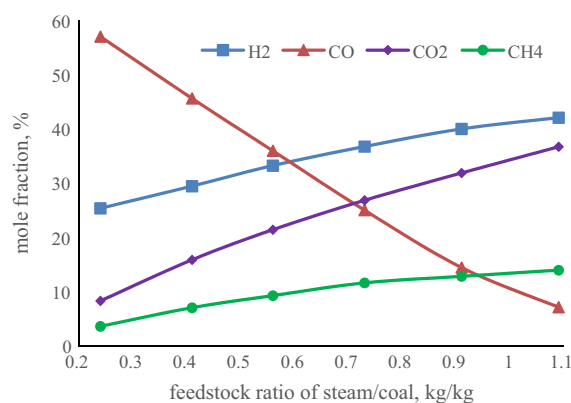
**Table 2**  
Proximate and ultimate analysis of lignite.

	Proximate analysis (wt.%, ar)				Ultimate analysis (wt.%, ar)				
	M <sub>t</sub>	M <sub>ad</sub>	V <sub>daf</sub>	A	C	H	O	N	S
Lignite	36.9	17.8	42.1	9.23	40.1	2.5	9.6	0.5	1.2

process is shown in Eqs. (1)–(9)[12]. The product crude syngas of gasification mainly includes CO, H<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>O, and CO<sub>2</sub>, together with some of sulfide, hydrocarbon, tar, phenol, and ammonia, etc. The gasification process is modeled by RGibbs reactor model and Peng–Robinson state equation is used as the equilibrium properties method [12]. All reactions are simulated based on chemical equilibrium by the minimum of Gibbs free energy.



CtSNG process uses Lurgi type MARK-IV gasifier. The crude syngas contains 39% H<sub>2</sub>, 14% CO, 32% CO<sub>2</sub> and 15% CH<sub>4</sub>. The high content of CO<sub>2</sub> is because of the high feedstock of the steam. The impact of steam/coal feedstock ratio on mole fraction of H<sub>2</sub>, CO, CO<sub>2</sub>, and CH<sub>4</sub> of the syngas is plotted in Fig. 4. With the increasing of steam/coal feedstock ratio, the H<sub>2</sub>, CH<sub>4</sub>, and CO<sub>2</sub> content of the syngas increases, while CO decreases. When the feedstock ratio is around 0.9, the composition of the crude syngas is very close to the industrial data of the CtSNG project of Datang Corporation.



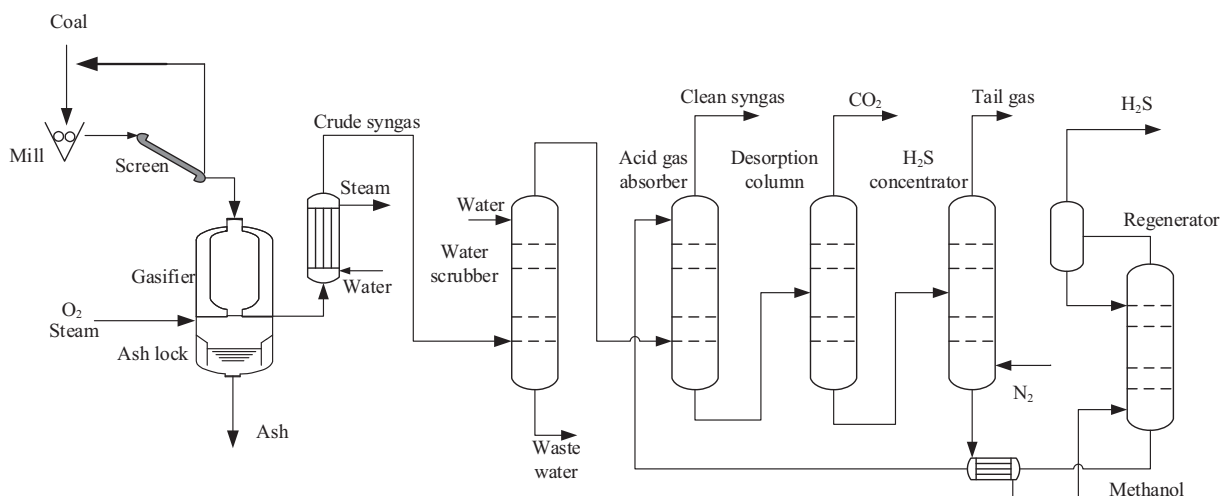
**Fig. 4.** Mole fraction of the H<sub>2</sub>, CO, CO<sub>2</sub>, and CH<sub>4</sub> with different steam/coal feedstock ratio.

**Table 3**  
Energy consumption of AGR unit.

Items	Specification	Metric	Consumption
Circulating cooling water	25 °C	t/h	3170
Electricity	\	kW	5400
LP steam	0.5 MPa	t/h	41
MP steam	2.5 MPa	t/h	20
LP nitrogen	0.5 MPa	m <sup>3</sup> /h	3240
Cryogenic energy	−40 °C	kW	28,800

Crude syngas from the gasifier is sent into an exhaust boiler to generate high temperature steam with 0.5 MPa. Next the crude syngas is sent into a water scrubber to remove most of the particulate matter and gaseous impurity which is easily dissolved in water, such as phenol and ammonia. Then the produced syngas is sent into AGR unit for further cleaning.

As shown in the right part of Fig. 3, AGR unit contains an absorber (uses chilled methanol to absorb H<sub>2</sub>S, COS and CO<sub>2</sub>), a CO<sub>2</sub> desorption column (desorbs CO<sub>2</sub> from the CO<sub>2</sub>-rich methanol), an H<sub>2</sub>S concentrator (collects and concentrates H<sub>2</sub>S from the H<sub>2</sub>S-rich methanol) and a methanol regenerator. Modeling and simulation of the water scrubber and AGR unit has been presented in the previous works on improved Rectisol methanol washing technology from parts of the authors [13]. Table 3 lists the energy consumption of AGR unit in CGtSNG.



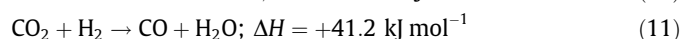
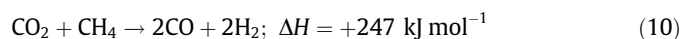
**Fig. 3.** Subsystem flowsheet of coal gasification and syngas clean.

## 2.2. COG separation and dry methane reforming

In most of the coal based chemical process, especially the process using Texaco or Shell gasification technologies, the source of CO<sub>2</sub> emission mainly comes from the WGS unit. The existent coal based co-feed processes usually achieve the goal of CO<sub>2</sub> reduction by reducing the shift degree of WGS unit. However, due to the resource allocation limitation, the present CtsNG processes in China adopt lignite as the feedstock and use Lurgi or similar gasification technology. The advantage of Lurgi gasification is that the H/C ratio of the gasification syngas is as high as 2.1–2.4. That means the shift degree could be so low that the energy consumption could be decreased in the WGS unit. The disadvantage of Lurgi gasification is the high CO<sub>2</sub> generation. Syngas from Lurgi gasification contains 35% CO<sub>2</sub>, which accounts for more than 80% of the CO<sub>2</sub> emission in CtsNG. In co-feed process for producing SNG, only use hydrogen-rich resource to reduce the shift degree is not an effective solution for reducing CO<sub>2</sub> emission and energy consumption. The proposed co-feed process in this paper adds a DMR unit to recycle the CO<sub>2</sub> from Lurgi gasification unit.

DMR offers valuable environmental benefits. It is an effective way to recycle CO<sub>2</sub> from the coal gasification process [14]. In DMR reaction, excessive H<sub>2</sub> would inhibit reaction equilibrium of DMR. And there may occur the side reaction, a reverse WGS reaction. This reaction would take away the useful H<sub>2</sub> in the syngas, and it is a strongly endothermic process and consumes considerable amount of energy [14]. COG has to be separated into H<sub>2</sub> and CH<sub>4</sub> before DMR. A pressure swing adsorption (PSA) unit is used for COG separation. The separated CH<sub>4</sub> from COG will be sent into DMR unit and H<sub>2</sub> will be mixed with the syngas for adjusting the H/C ratio in the later step. The subsystem flowsheet of COG separation and dry methane reforming is shown in Fig. 5.

Main and side reaction in DMR is shown as Eqs. (10) and (11). Ni–Co based catalyst is used in DMR unit with the reaction temperature of 700 °C. In such condition, the conversion rate of CH<sub>4</sub> and CO<sub>2</sub> could be achieved up to 95% [15]. Peng–Robinson state equation is used as the equilibrium properties method in Aspen Plus. And RGibbs reactor model is used in simulations [16].



RGibbs reactor model predicts a thermodynamically limited maximum methane conversion of 95% at 700 °C and 0.4 MPa. Referred to the work of Luyben [16], the expressions used for the forward and reverse reaction rates of Eq. (10) are given in the following equations.

$$r_F = 5 \times 10^{-9} e^{-50,000/RT} P_{\text{CH}_4} P_{\text{CO}_2} \quad (12)$$

$$r_R = 1.094 \times 10^{-22} e^{-195,900/RT} P_{\text{CO}} P_{\text{H}_2} \quad (13)$$

For the reverse water gas shift reaction of Eq. (11), a simple kinetic expression is assumed with the following equation referred to the work of Luyben [16].

$$r_{\text{RWGS}} = 10^{-11} e^{-50,000/RT} P_{\text{CO}_2} P_{\text{H}_2} \quad (14)$$

By mass balance calculation, the quantity of CH<sub>4</sub> sent into DMR accounts for 1.5% of the SNG production while it reduced 60% CO<sub>2</sub> emission.

Syngas for methanation is obtained from the mixture of coal gasification syngas, DMR syngas, and H<sub>2</sub> from COG.

## 2.3. Methanation

Main reaction in methanation is shown in Eq. (15). Eq. (16) only occurs when CO concentration is very low [17]. Referred to the CtsNG project of Datang Corporation, subsystem of methanation selects Davy methanation technology as the simulation model. The flowsheet of this subsystem is shown in Fig. 6. Four sequential adiabatic reactors are used to convert syngas into SNG. The first two reactors are bulk methanation reactors, in which most of the CO, CO<sub>2</sub>, and H<sub>2</sub> will convert into CH<sub>4</sub>. The last two reactors are trim methanation reactors. Unreacted gas from bulk methanation reactors is sent into trim methanation reactors for converting the rest of the CO, CO<sub>2</sub>, and H<sub>2</sub>. Methanation reactions are strongly exothermic. The heat released from methanation is recovered by the waste-heat boiler for generating high temperature and pressure stream. Peng–Robinson state equation is used as the equilibrium properties method in Aspen Plus and RPlug reactor model is used in simulations [17].

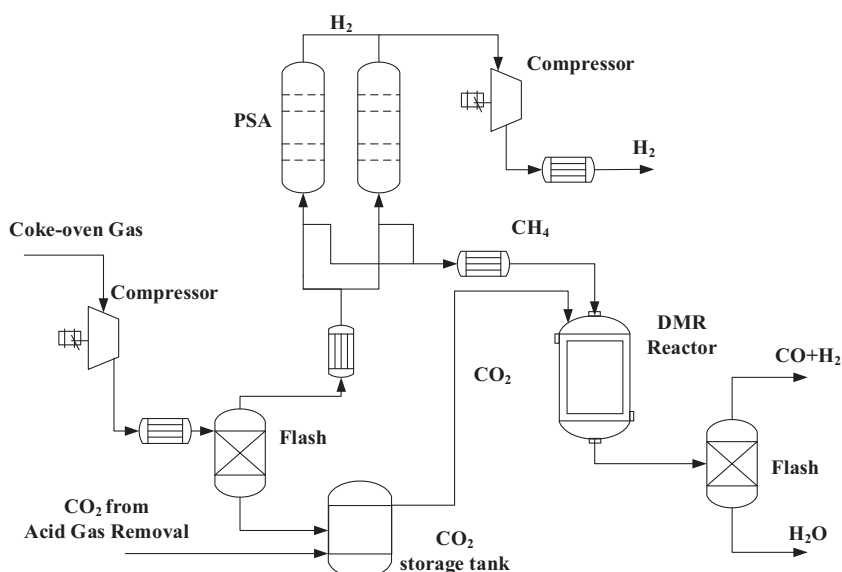
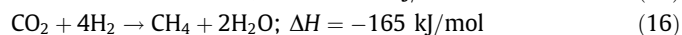
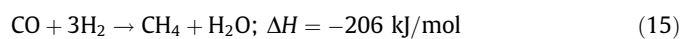


Fig. 5. Subsystem flowsheet of COG separation and DMR.

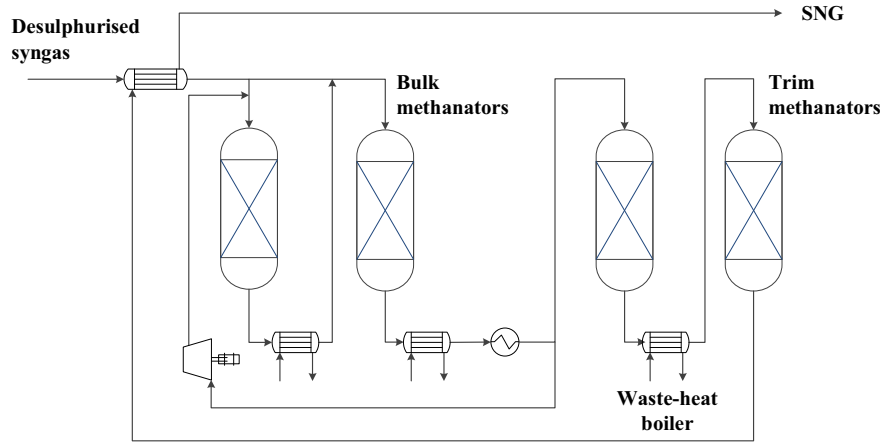


Fig. 6. Subsystem flowsheet of methanation.

Yadav and Rinker [18] developed the empirical correlations for the methanation of CO [Eq. (15)] as part of a laboratory data program. The correlation is as follows:

$$r = \frac{A}{C^2} \frac{P_{CO}}{P_{H_2}^{0.5}} \quad (17)$$

where  $r$  is the reaction rate in  $\text{mol (g s)}^{-1}$ ,  $P$  refers to partial pressure in kPa, and  $A$  and  $C$  are empirical constants determined at each temperature.

The data reported by Yadav and Rinker [18] at 503, 513 and 529 K were fitted to a simple Arrhenius equation as follows:

$$r = 0.314e^{1300(\frac{1}{T} - \frac{1}{513})} \frac{P_{CO}}{P_{H_2}^{0.5}} \quad (18)$$

Proper modeling of the methanation should include the effect of the backward reaction as equilibrium is approached. The complete version of Eq. (15) is as follows:

$$r = A_C 0.314e^{1300(\frac{1}{T} - \frac{1}{513})} \frac{P}{P_{H_2}^{0.5}} \left( y_{CO} - \frac{y_{CH_4} y_{H_2O}}{y_{H_2}^3 P^2 K_{CO}} \right) \quad (19)$$

$$K_{CO} = e^{(-38.4523 + 26.270/T)} \quad (20)$$

where  $P$  is the pressure in kPa,  $y$  is the component mole fraction,  $K_{CO}$  is the equilibrium constant for Eq. (15) and  $A_C$  is the catalyst activity factor.

For the methanation of  $CO_2$ , the same kinetic expression for  $CO_2$  is used as for  $CO$ , recognizing that the concentration of  $CO_2$  in the process is very small. The rate of Eq. (16) is as follows:

$$r = A_C 0.314e^{1300(\frac{1}{T} - \frac{1}{513})} \frac{P}{P_{H_2}^{0.5}} \left( y_{CO_2} - \frac{y_{CH_4} y_{H_2O}^2}{y_{H_2}^4 P^2 K_{CO_2}} \right) \quad (21)$$

$$K_{CO_2} = e^{(-33.923 + 21.621/T)} \quad (22)$$

#### 2.4. Process simulation of CGtSNG

The main operation conditions and specifications are shown in Table 4. By simulation, data for each main stream is obtained and shown in Table 5, which includes compositions, temperatures, and pressures. The stream number in Table 5 is referred to Fig. 1. Fig. 7 shows the steam balance of CGtSNG based on the simulation data of Aspen Plus.

Table 4

Main operation conditions of CGtSNG process.

Unit	Operation conditions	Value
Coal gasifier (Lurgi)	Gasification pressure, MPa	4
	Gasification temperature, °C	>800
	Carbon conversion, %	99.9
DMR reactor	Reaction pressure, MPa	0.4
	Reaction temperature, K	~700
	Carbon conversion, %	95
Pressure swing adsorption	Hydrogen yield, %	>90
	Number of absorber	2
Acid gas removal	Solvent	Rectisol®
	Overall $H_2S$ removal yield, %	99.5
Methanation	Residual content of CO and $CO_2$	<10 ppm

Table 5

Simulation results of CGtSNG process.

Stream No.	1	2	3	4	5
Mole fraction, %					
CO	14.5	21.2	5.8	17.9	Trace
$CO_2$	31.9	1.8	2.8	1.5	0.7
$H_2$	39.00	57.2	59.5	60.8	1.7
$O_2$	0.3	0.4	1.6	0.4	0
$CH_4$	12.8	18.8	24.4	18.9	96.8
$C_2^+$	0.6	0.2	1.4	0.2	0
$H_2S$	0.7	0	0	0	0
$N_2$	0.2	0.4	4.5	0.3	0.8
Mole flow, kmol/h	26982.9	18707.2	45200.9	66732.1	22321.4
Temperature, °C	180	40	25	40	40
Pressure, MPa	4	4	0.1	3.3	3.3

#### 2.5. Key operational variables analysis

In CGtSNG process, syngas for methanation is obtained by the mixture of coal gasification syngas, DMR syngas, and  $H_2$  from COG. Insufficient COG feedstock rate would result in low H/C ratio of syngas, thereby reduce the yield of SNG production. And insufficient  $CH_4$  from COG could not react with whole content of  $CO_2$ . As a result, the reduction of carbon emission would be little. On the other hand, excessive COG feedstock rate would bring about high H/C ratio of syngas. Redundant  $H_2$  not only waste the hydrogen resource, but also bring problems in SNG refinement. Suitable range of feedstock rate for COG needs to be found.

H/C ratio of the syngas is a very important indicator for methanation reaction. The optimal H/C ratio for methanation is in the range 3.05–3.2. The change of H/C ratio with different feedstock ratio of COG to coal is analyzed in this paper.

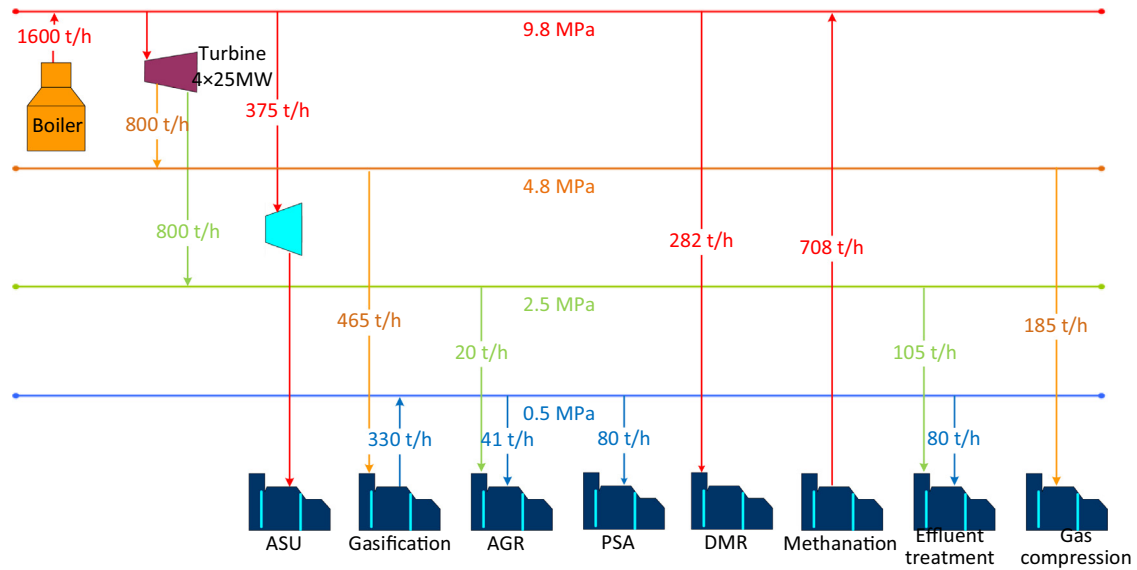


Fig. 7. Steam balance of CGtSNG.

Consumed feedstock of coal affects resource utilization, energy efficiency, and economic benefit. It also embodies the contribution of the system to CO<sub>2</sub> emission. Efficiency of carbon use (treated as an element) is taken as an investigation object when analyzing the technical performance of the process. Elementary carbon efficiency is defined as the ratio of effective carbon compounds of the outlets to effective carbon compounds of the inlets streams [19]. For CGtSNG process, coal (carbon content of lignite is valued as 75 wt.%) and CH<sub>4</sub> in COG are inlet effective compounds, SNG is outlet effective compound. Elementary carbon efficiency of CGtSNG is defined as following Eq. (23).

$$f = \frac{F_{\text{SNG}}^{\text{out}}}{F_{\text{Coal}}^{\text{in}} + F_{\text{COG}}^{\text{in}}} \times 100\% \quad (23)$$

where  $f$  is the elementary carbon efficiency,  $F_{\text{Coal}}^{\text{in}}$  is the mole flow rate of carbon content in coal,  $F_{\text{COG}}^{\text{in}}$  is mole flow rate of CH<sub>4</sub> in COG, and  $F_{\text{SNG}}^{\text{out}}$  is mole flow rate of SNG product.

The impact of COG/coal feedstock ratio on H/C ratio is plotted in Fig. 8. With increase in the feedstock ratio, the H/C ratio rises due to the CO content of methanation syngas gradually decrease. When the ratio of COG/coal feedstock reaches 0.8–1.0, and H/C ratio is in the range 3.05–3.2.

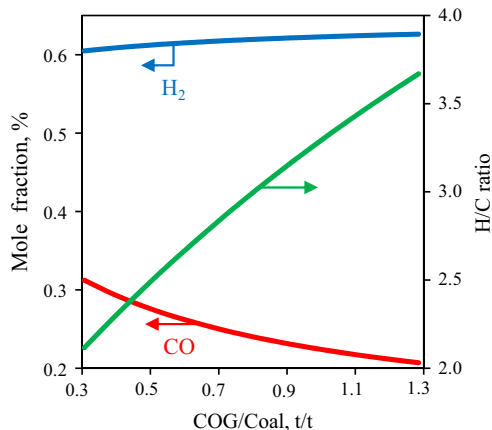


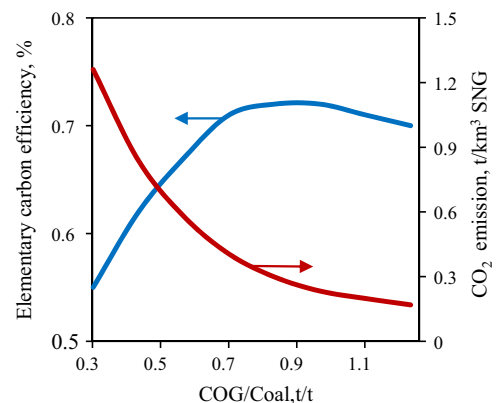
Fig. 8. Impact of COG/coal feedstock ratio on H/C ratio.

The impact of COG/coal feedstock ratio on elementary carbon efficiency and CO<sub>2</sub> emission is plotted in Fig. 9. The efficiency of elementary carbon increases with the increase in the feedstock ratio. When the ratio of COG/coal feedstock reaches around 0.9, almost all CO<sub>2</sub> from AGR unit is reacted, the increasing of elementary carbon efficiency becomes smooth. Continuing raise the feedstock would lead to the waste of the raw material and makes a decrease in elementary carbon efficiency.

In consequence, COG/coal feedstock ratio for CGtSNG process should be around 0.8–0.9. In such conditions, the elementary carbon efficiency reaches up to 70% and the value of H/C ratio is adequate for the methanation process.

### 3. Coal and coke-oven gas to SNG process without DMR

A co-feed process of coal and COG to produce SNG without DMR unit (CGtSNG w/o DMR) is designed in this paper, shown in Fig. 10. In this process, desulfurated coke-oven gas is directly mixed with coal gasification syngas. After mixture, the syngas with suitable H/C ratio is sent into methanation unit. Compared with CGtSNG, the PSA and DMR units are cancelled in this process. It avoids the energy consumption of matter conversion. However, CO<sub>2</sub> in this process cannot be recycled without DMR reaction.

Fig. 9. Impact of COG/coal feedstock ratio on elementary carbon efficiency and CO<sub>2</sub> emission.



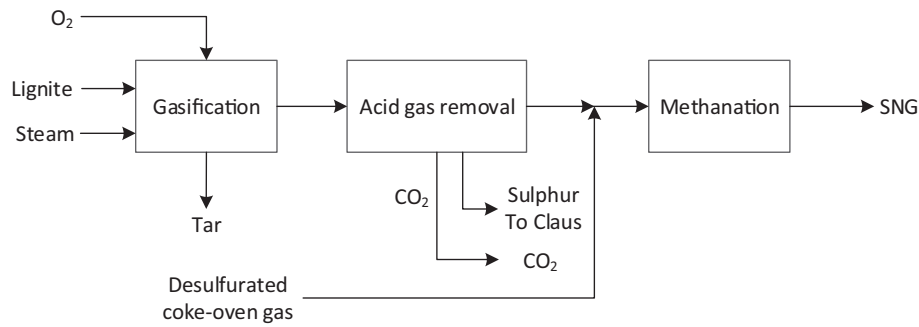


Fig. 10. Schematic diagram of CGtSNG w/o DMR process.

#### 4. Techno-economic analysis of CGtSNG process

##### 4.1. Energy efficiency

Table 6 shows the consumption, product output, and energy efficiency of CtSNG, CGtSNG, and CGtSNG w/o DMR processes. Utilities consumption is calculated by the lignite coal consumption for fuel, including steam and power generation. High elementary carbon efficiency makes CGtSNG process need much smaller amount of coal, as raw material, to produce the same amount of SNG. That means the scale of coal gasification, AGR, and ASU units of CGtSNG are also smaller than CtSNG. The total energy input of CGtSNG is significantly reduced. As a result, energy efficiency of CGtSNG is 4.1% higher than CtSNG process. CGtSNG w/o DMR process do not make use of CO<sub>2</sub>. However, WGS unit is canceled in CGtSNG w/o DMR process and the waste of elementary carbon convention is avoided. The energy efficiency of CGtSNG w/o DMR is 7.6% higher than CtSNG process.

##### 4.2. CO<sub>2</sub> emission and effluent discharge

CO<sub>2</sub> emission and effluent discharge of CtSNG, CGtSNG, and CGtSNG w/o DMR with 4.0 billion m<sup>3</sup>/y SNG production capacity is shown in Fig. 11. The total CO<sub>2</sub> emission of CtSNG  $7.85 \times 10^6$  t/y, and the mass flow of waste water is 1670 m<sup>3</sup>/h. In CtSNG process, the waste water from gasification unit accounts for 98% of the total effluent discharge [7]. Hence, the calculation of effluent in this paper only considers the waste water from coal gasification unit. The waste water is generated by crude coal gas washing. The amount of waste water is depended on the amount of crude coal gas. The total amount of waste water of CGtSNG and CGtSNG w/o DMR could be obtained by calculating the amount of the crude coal gas.

The increase in elementary carbon efficiency of CGtSNG makes the process emit less CO<sub>2</sub> and the application of DMR unit takes use

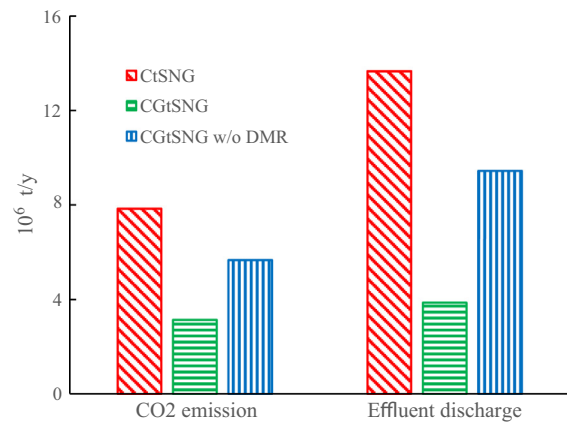


Fig. 11. CO<sub>2</sub> emission and effluent discharge.

of the CO<sub>2</sub> from coal gasification. As a result, CO<sub>2</sub> emission of CGtSNG decreases by 60% in comparison of CtSNG. The consumption for lignite in CGtSNG process is around 29% of that in CtSNG process. That means the amount of crude syngas from coal gasification of CGtSNG is much smaller than CtSNG. Water usage for crude syngas scrubbing would have a corresponding decrease. The result shows the effluent discharge of CGtSNG is reduced by 72%. This will reduce the water consumption for the production process and relieve the pressure on effluent treatment.

For CGtSNG w/o DMR process, CO<sub>2</sub> is not reused. However, the canceled WGS unit makes this process generate less CO<sub>2</sub>. And the introduction of COG as the raw material also saves some coal consumption. As a result, CO<sub>2</sub> emission of this process decreases by 27.8% and the effluent discharge decreases by 30.9%.

##### 4.3. Economic performance

Economic performance for CGtSNG process with the production capacity of 4.0 billion m<sup>3</sup>/y SNG is analyzed and made a comparison to the CtSNG of Datang Corporation.

##### 4.3.1. Total capital investment

Estimation method of total capital investment is followed with the authors' previous works [20].

The total capital investment includes fixed investment and variable cost. Fixed investment is estimated by Eq. (24), according to the benchmark case shown in Table 7.

$$I_2 = \theta \cdot I_1 \cdot \left( \frac{Q_2}{Q_1} \right)^n \quad (24)$$

where  $I_1$  and  $Q_1$  are the fixed capital investment and the production capacity of the current project;  $I_2$  and  $Q_2$  are the fixed capital

Table 6  
Consumption, product output, and energy efficiency.

Item	CtSNG [7]	CGtSNG	CGtSNG w/o DMR	Lower heating value
Consumption				
Lignite, t	1.55E7	4.47E6	1.07E7	14.4 MJ/kg
Coke-oven gas, m <sup>3</sup>	–	8.1E9	2.73E9	17.4 MJ/m <sup>3</sup>
Lignite for fuel	3.75E6	3.61E6	2.78E6	14.4 MJ/kg
Water, t	1.83E7	6.42E6	1.03E7	–
Total energy input, GJ	2.77E8	2.57E8	2.42E8	–
Products output				
SNG, m <sup>3</sup>	4.0E9	4.0E9	4.0E9	35.99 MJ/m <sup>3</sup>
CO <sub>2</sub> emission, t	7.85E6	3.14E6	5.67E6	–
Total energy of SNG, GJ	1.44E8	1.44E8	1.44E8	–
Energy efficiency, %	51.9	56.0	59.5	–

**Table 7**

Summary of investment data for main equipment components.

Unit	Benchmark	$Q_i$	$\theta$	$I_1$ ( $10^8$ CNY)
Air separation [11]	Oxygen supply	21.3 kg/s	0.50	2.83
Coal gasification [21]	Daily coal input	39.2 kg/s	0.80	4.84
Pressure swing adsorption [11]	Hydrogen supply	21.3 kg/s	0.50	2.83
Dry methane reforming [11]	Material caloric value	716 MW	0.65	3.84
Water gas shift [22]	Coal caloric value	1377 MW	0.67	2.47
Acid gas removal [22]	CO <sub>2</sub> absorption	2064.4 mol/s	0.65	2.03
Methanation [23]	Syngas input	10,810 mol/s	0.65	1.26

**Table 8**

Ratio factors for capital investment.

Component	Ratio factor (RF, %) [24]
(1) Direct investment	
(1.1) Equipment	100
(1.2) Installation	48
(1.3) Instruments and controls	24
(1.4) Piping	57
(1.5) Electrical	29
(1.6) Buildings(including services)	71
(1.7) Land	5
(2) Indirect investment	
(2.1) Engineering and supervision	48
(2.2) Construction expenses	43
(2.3) Contractor's fee	19
(2.4) Contingency	33
(3) Fixed capital investment	477
(4) Working capital	80
(5) Total capital investment	557

investment and the production capacity of the planned project;  $\theta$  is the domestic-made factor;  $n$  is scale exponent,  $n = 0.6$  is a common value for chemical processes.

The components of the total capital investment could be determined according to their ratios to the equipment investment. The ratios are shown in Table 8 and the calculation follows Eq. (25)

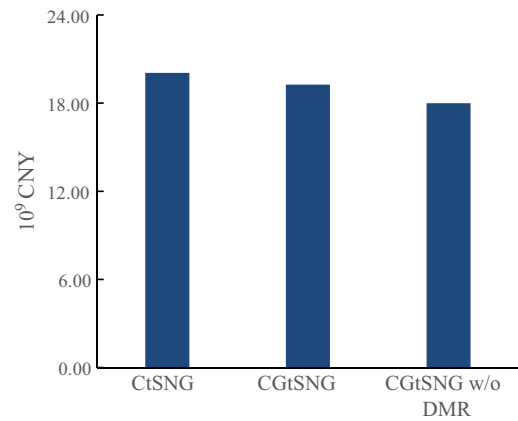
$$TCI = I \cdot \left( 1 + \sum_i RF_i \right) \quad (25)$$

where TCI the total capital investment,  $I$  is the fixed capital investment, and  $RF_i$  is the ratio factor of capital investment of component  $i$ .

Total capital investment of CtSNG, CGtSNG, and CGtSNG w/o DMR processes is shown in Fig. 12. Total capital investment of the CtSNG of Datang Corporation is around  $2.006 \times 10^{10}$  CNY [7]. Based on the benchmark of CtSNG, total capital investment of CGtSNG and CGtSNG w/o DMR processes with same production capacity could be estimated to be around  $1.926 \times 10^{10}$  CNY and  $17.99 \times 10^{10}$  CNY. CGtSNG adds a COG separation and DMR units. Correspondingly, the scale of acid gas removal unit becomes smaller. At the same time, number of the gasifiers reduces to a quarter of the CtSNG process because of the improvement of the coal utilization efficiency. CGtSNG w/o DMR process canceled the WGS unit in CtSNG and PSA and DMR units in CGtSNG. As a result, the total capital investment for CGtSNG and CGtSNG w/o DMR processes are reduced by 4% and 10%.

#### 4.3.2. Production cost

Estimation method of production cost is based on the work of the authors' previous works [24]. Unit price of raw material and utilities are listed in Table 9 based on the average price in China in 2015. Since the waste COG from coking plant is not a kind of

**Fig. 12.** Total capital investment.**Table 9**

Estimation of production cost.

Component	Economic assumptions
(1) Raw material	Lignite 120 CNY/t, Coke-oven gas 0.43 CNY/m <sup>3</sup>
(2) Utilities	Water 2 CNY/t, electricity 0.7 CNY/kW h, steam 42 CNY/GJ
(3) Operating & maintenance	
(3.1) Operating labor	300 labors 10,000 CNY/labor/year
(3.2) Direct supervisory and clerical labor	20% of operating labor
(3.3) Maintenance and repairs	2% of fixed capital investment
(3.4) Operating supplies	0.7% of fixed capital investment
(3.5) Laboratory charge	15% of operating labor
(4) Depreciation	Life period 20 years, salvage value 4%
(5) Plant overhead costs	60% of (3.1 + 3.2 + 3.3)
(6) Administrative costs	2% of product cost
(7) Distribution and selling costs	2% of product cost
(8) Product cost	(1) + (2) + (3) + (4) + (5) + (6) + (7)

commodity, it is hard to find the price of COG. Therefore, the price of COG is estimated by comparing the heating value of COG and electricity based COG combustion power generation, as shown in Eq. (26). The price of electricity is referred to the generation price of Inner Mongolia in 2015 [25].

$$\frac{C_{COG}}{C_{Ele}} = \frac{Q_{COG}}{Q_{Ele}} \cdot \lambda \quad (26)$$

where  $C_{COG}$  is the price of COG;  $Q_{COG}$  is the heating value of COG;  $Q_{Ele}$  is the heating value of electricity;  $\lambda$  is the efficiency of COG combustion power generation, adopted as 30%;  $C_{Ele}$  is the generation price of Inner Mongolia in 2015, adopted as 0.3 CNY/kW h.

Comparison of the production cost for CtSNG, CGtSNG and CGtSNG w/o DMR is shown in Fig. 13. As discussed before, the total capital investment of CGtSNG is lower than that of CtSNG. And the



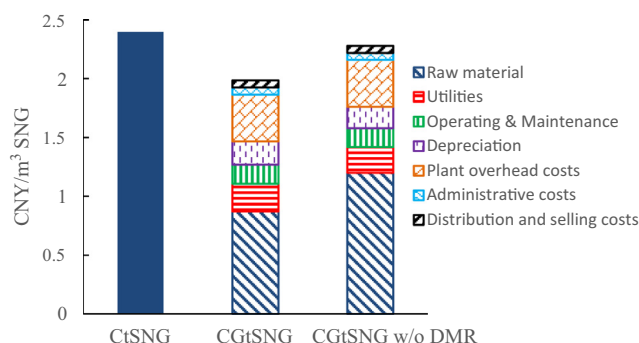


Fig. 13. Production cost.

feedstock of the raw material reduces a lot due to the high carbon utilization efficiency. Therefore, the production cost of CGtSNG is approximately 2 CNY/m<sup>3</sup> SNG. Production cost of CGtSNG is 16.7% lower than that, 2.4 CNY/m<sup>3</sup>, of CtSNG. Production cost of CGtSNG w/o DMR is 2.28 CNY/m<sup>3</sup> SNG, which decreased 5% due to the lower total capital investment and less raw material consumption.

## 5. Conclusions

This paper proposes a novel coal and COG to SNG process. In CGtSNG process, methane in COG is used for DMR reaction for reducing CO<sub>2</sub> emission. And H<sub>2</sub> in COG is used for adjusting the H/C ratio of the methanation syngas. Key operational variables are analyzed. The result shows the elementary carbon efficiency reaches up to 70% and the H/C ratio of the syngas is appropriate for the methanation when COG/coal feedstock ratio is around 0.8–0.9. A process of coal and COG to SNG without DMR unit is also proposed. Although CGtSNG w/o DMR process do not make use of CO<sub>2</sub>, the CO<sub>2</sub> emission and energy consumption are also decreased by system integration.

The major advantages of the proposed CGtSNG process are: (1) CGtSNG enables to take advantage of the wasted COG in China's coke industry, what allows to optimize the allocation of resources and enhances the economic value of COG. (2) CGtSNG has high energy efficiency and low emission. The energy efficiency of CGtSNG and CGtSNG w/o DMR is 4.1% and 7.6% higher than that of CtSNG. CO<sub>2</sub> emission of CGtSNG and CGtSNG w/o DMR decrease by 60% and 27.8% in comparison to the CtSNG process, with the effluent discharge decrease by 72% and 30.9%. (3) CGtSNG has advantaged in economic performance. Production cost of CGtSNG and CGtSNG w/o DMR is 16.7% and 5% lower than that of CtSNG.

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