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# An Integrated Framework for Modeling, Synthesis, Analysis, and Optimization of Coal Gasification-Based Energy and Chemical Processes

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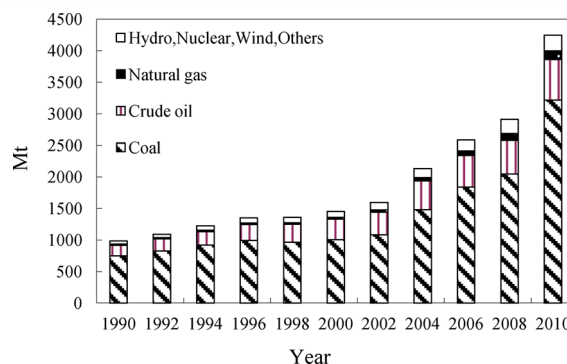
**ABSTRACT:** A framework is proposed for integration of unit modeling, process synthesis, analysis, optimization, and process design of coal gasification-based energy and chemical processes. The conceptual models of these processes are built by the modeling and synthesis blocks in the framework, which are the bases of analysis and optimization. A number of analysis techniques are employed in the framework to fully understand the characteristics of the processes and their performances from technical, economic, and environmental points of view. Life cycle assessment and sustainability analysis are also included in the framework. According to these multilayer analyses, optimization is included in this framework to explore the best process or the best operational parameter set. Because of the systematic integration of the above techniques, the proposed framework could provide a comprehensive study for coal gasification-based processes. Three coal gasification-based processes are selected as the study cases in this Article. They are an integrated gasification combined cycle process, a methanol production, and a coproduction process combining the first two processes. The exergy efficiencies and economic investments of the three processes are analyzed and compared. The key parameters for material distribution in the coproduction process are optimized from the exergy efficiency and the investment points of view.

## 1. INTRODUCTION

The heavy dependence on petroleum in the world gives rise to the rapid depletion of this energy resource. Exploitation of alternative energy resources is the prospective trend of the world's energy development. Coal has been broadly accepted as one of the important alternatives. It has been long used to produce varieties of chemicals, and to generate electricity and power. China has the unique energy structure featuring richness in coal but shortage in petroleum and natural gas. It has the largest proven coal reserve of 5.6 trillion tons, equivalent to 11.6% of the world total. However, the richness does not immunize China from the panic of increasing depletion of coal. Rapid industrialization brings the growing demand for energy, increasing the total energy consumption from 0.57 billion tons up to 3.06 billion tons standard coal.<sup>1</sup> Among this consumption, coal occupies the largest proportion, as shown in Figure 1.

The situation of the coal dominance in China is especially reflected on the electricity industry and chemical industry. 77% of coal is used to generate electricity, while 44% is used to produce chemicals.<sup>1</sup> A wide range of coal derivatives, including methanol, ammonia, dimethyl ether, ethylene propylene, and formaldehyde, are produced from coal. However, most production processes using traditional techniques suffer from low energy efficiency and high environmental pollution. These processes make the air unbreathable and hasten climate changes that are undermining China's economic sustainability. Developments of clean coal techniques are necessary for ensuring resource supply, energy security, environment improvement, and active response to global climate change in China.

Coal gasification is a major part of coal cleaning techniques. It converts useful elements in coal to a gaseous mix known as



**Figure 1.** Proportion profile of different energy consumptions in China.

syngas. This technique was first used in industrialized countries for generating electricity and producing value-added coal derivatives. With use of syngas, electricity, fuels, and different chemicals, such as ammonia, dimethyl ether, ethylene propylene, and formaldehyde, could be generated. A part of the coal gasification-based industry chain is shown in Figure 2. Since 2005, China had launched a new coal generation program, GreenGen, for establishing near zero-emission coal power plants. This program received support from the government and a group of enterprises led by China Huaneng Group. GreenGen established China's first Integrated Gas-

**Received:** June 14, 2012

**Revised:** October 11, 2012

**Accepted:** November 6, 2012

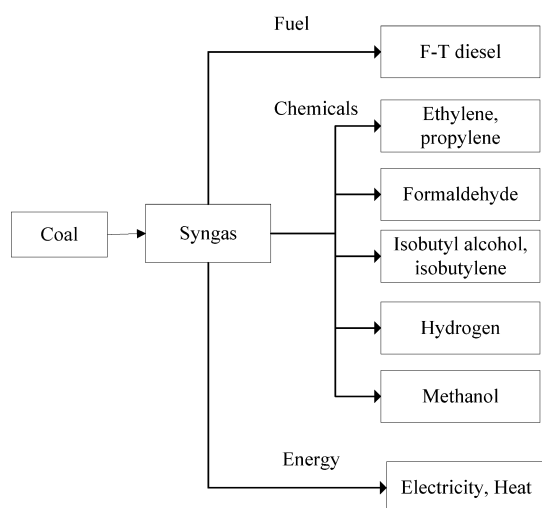


Figure 2. A part of the coal gasification-based industry chain.

research on technical, economic, and environmental performances based on economic metrics. They used this metric to quantify the environmental impacts, labors, and services, like the work of NETL.<sup>3</sup> Yet with this method, the analysis would change geographically and temporally. In addition, this research could not do systematic assessment on resource efficiency, energy efficiency, and environmental impact from the whole life cycle of products. Thus, the analysis based on economic metrics is incomprehensive and usually limited to the boundary of the production process. As understanding of process sustainability has increased, people believe that the comprehensive analysis of production processes and industry chains from their whole life cycle is necessary for avoiding inefficiency and then depression of new developed chemical and energy processes.<sup>11–13</sup> This Article aims to design a framework integrating different techniques of process system engineering for the comprehensive study of the coal gasification-based processes. A unit modeling block and a process synthesis block are used to build the conceptual models of the applied processes. To fully understand the essence of the processes and their impact on the surrounding environment, a large number of analysis techniques are employed. Sustainability analysis is a newly explored area from the ecological and social points of view. Depending on these analyses, more efficient and cleaner processes could be explored.

In section 2, the structure of the framework is proposed along with the introduction of the basic blocks and their functions. Basic units in coal gasification-based processes are modeled and simulated. Applied analysis techniques are described in the later part of this section, in the order of their four classes: thermodynamic analysis, techno-economic analysis, life cycle assessment, and sustainability analysis. Three case processes are studied in section 3. They are an electricity generation process, a chemical synthesis process, and finally a coproduction process. Exergy efficiencies and economic investment of the three cases are studied. Advantages of the coproduction process are explored through the comparison with the two single processes. The key parameters, with significant effects on material distribution, are optimized. Exergy saving ratio and investment saving ratio are used as the objectives of the optimization. Further development of the framework is discussed in the last section along with concluding remarks.

## 2. FRAMEWORK

The framework proposed in this Article integrates different techniques: unit modeling, process synthesis, analysis, optimization, and process design. Correspondingly, the framework consists of five building blocks, as shown in Figure 3.

The first block is the unit modeling block, in which the basic units of applied processes are identified. These units are then modeled and simulated. Generally, coal gasification-based processes are complicated but mainly consist of a few basic units: air separation unit (ASU), solid preparation and gasification (SPG), acid gas removal (AGR), sulfur recovery (SR), water gas shift (WGS), heat recovery steam generation (HRSG), combined cycle (CC), and chemical synthesis (CS). Other general units such as utilities, pump, and compressor could be easily found in commercial software. Some of the above units and their corresponding modeling methods will be described later in this Article.

The second block is the process synthesis block. The applied processes are synthesized by connecting those unit models

ification Combined Cycle (IGCC) demonstration power plant in Tianjing. On April 17th 2012, the GreenGen team successfully operated the gasifier at the Tianjin IGCC power plant. Until then, the gasification had successfully operated for 18 h, reaching 60% of the rated capacity.<sup>2</sup> There has been a large amount of research on IGCC, among which the research of the National Energy Technology Laboratory (NETL) of the U.S. Department of Energy is the representative. NETL established the baseline performance and cost estimates for fossil energy-based plants, based on models of different processes (including IGCC), by using data from published literatures and information from vendors and users. Such a baseline could be used to benchmark the progress of the fossil energy RD&D portfolio.<sup>3–5</sup> All of this previous research forms the basis of our work in this Article.

Because of the development of coal gasification techniques, electricity and chemicals could be generated at the same time from syngas. These coproduction processes have been studied in the literature.<sup>6,7</sup> They reported that coproduction could improve energy efficiency and reduce investment. However, the coproduction processes have to include an additional water gas shift process to adjust the  $H_2/CO$  of syngas for synthesizing different derivatives. The water gas shift process could be removed if the coal-based syngas mixed with the hydrogen-rich syngas generated from other energy resources, such as natural gas, shell gas, and biomass. These combinations could reduce the capital cost and the utility cost. The resulting processes are known as the cofeed and coproduction processes. In the early 21st century, a large amount of research on cofeed and coproduction processes sprung up. For example, Kreutz and his research team reported a series of research on the production of electricity and Fischer–Tropsch fuels from coal and biomass.<sup>8–10</sup> They found that the cofeed and coproduction process made the productivity of fuels increase twice and the  $CO_2$  emission reduce 10 times as compared to the single processes.

Although there have been a number of studies, it is commonly understood that the coal base processes are complicated while less investigated than petroleum-derived processes. In addition, current studies usually focus on assessing one of the performances, such as feasibility of designed processes, technical bottleneck, improvement of catalyst techniques, and equipment integration. There is also some

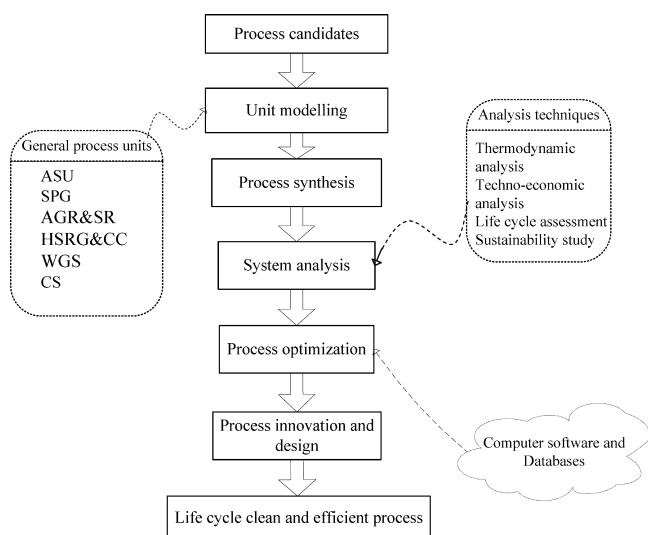


Figure 3. Outline of the proposed framework.

from the first block, with the use of mass flows and energy flows. The optimal structures of the processes are determined with use of different synthesis methods. According to the first and the second laws of thermodynamics, the synthesized processes are required to follow mass and energy balance and thermal energy flowing principles. In addition, the synthesized processes must be consistent with the basic chemical engineering requirements and other S.H.E. regulations.

The third block is the system analysis block. A number of analysis techniques are involved in the framework. In general, they could be classified into four categories, thermodynamic analysis, techno-economic analysis, life cycle assessment, and sustainability analysis, as shown in Figure 3. These techniques and their functions will be described later in detail.

The fourth block works for process optimization. Industrial processes are complex and generally modeled by nonlinear programming with integer variables determining the process structure. For example, an integer variable is used to determine whether there is an energy flow or a material flow between equipment "A" and equipment "B". Moreover, the search spaces of the optimizations are likely to be nonconvex and include one or more local optimal solutions. For the optimal process with the best performance on technical, economic, and environmental perspectives, the multiple objective optimization techniques are used for exploring the best trade-off.

The last block is the process design block. Missions for this block involve equipment selection, for example, selecting Lurgi, Texaco, or Shell gasifier for coal gasification; equipment related and operational parameter determination, for example, determining pressures and temperatures of the high pressure, the middle pressure, and the low pressure steams in IGCC; and mass and energy flow network design, for example, determining whether to recycle the unconverted syngas from the synthesizer or not, etc.

The above blocks comprise the backbone of the framework. Each of them uses a specific class of techniques. In the following sections, emphasis is on unit modeling, analysis of exergy efficiency and investment, and optimization.

**2.1. Modeling of Process Units.** Although coal gasification-based processes are complex and involve many different units, there are a few basic units, that is, ASU, SPG, AGR, SR, WGS, HRS, CC, and CS. To limit the length of this

Article, we explained the modeling of some units, that is, SPG, WGS, AGR, SR, and CS in this Article, while we ignored those of ASU, HRS, and CC. Methanol is selected as the coal derivative synthesized in the CS. This synthesis process is named as the methanol synthesis unit (MS). Yet it should be added that the concentrations of the outlet  $O_2$  and  $N_2$  from the ASU are 95% and 99.8%, respectively. The high pressure steam, the middle pressure steam, and the low pressure steam in the CC have their pressures and temperatures fixed to 12 MPa and 565 °C, 3.5 MPa and 563 °C, and 0.3 MPa and 243 °C.

The process modeling in this Article does not aim for new process design, but for process systems analysis, comparison, and decision making. Thus, several assumptions are made in our work. They are all Gibbs reactors are isothermal and isobaric; gaseous flowing activities in Langmuir–Hinshelwood and Hougen–Watson (LHHW) reactor do not include axial flow and backmixing flow; and the gasifier is modeled as a steady process obeying thermodynamic equilibrium. Products from the gasifier include  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ ,  $H_2O$ ,  $N_2$ ,  $H_2S$ ,  $NH_3$ ,  $COS$ ,  $SO_2$ , and solid C and S; ash content in raw materials does not participate in any chemical reaction. Following these assumptions, modeling and simulation of the unit processes will be described in following sections.

**2.1.1. SPG.** Solid preparation and gasification (SPG) unit, as shown in Figure 4, uses coal or coal char as the input materials

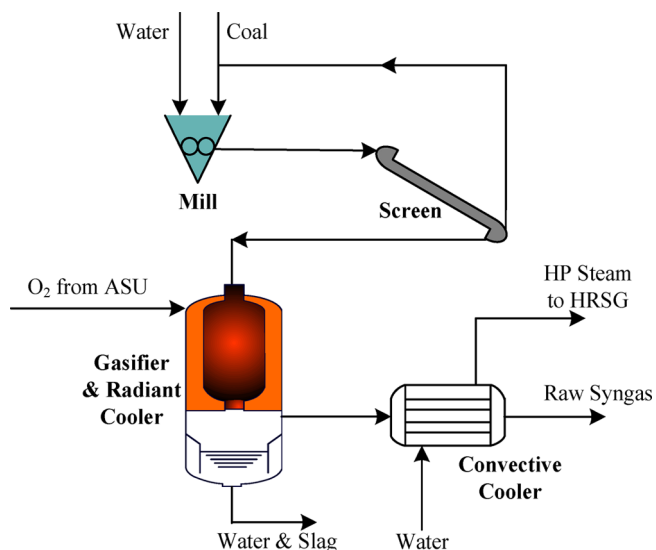
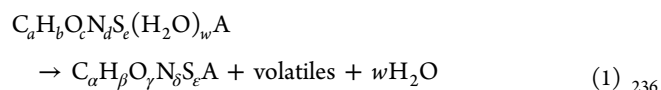


Figure 4. Process flowsheet of SPG.

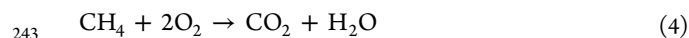
and  $O_2$ , steam, and  $H_2$  as the gasification media. For modeling, coal is formulated as  $C_aH_bO_cN_dS_e(H_2O)_wA$ . It is first converted into coke, volatiles, and water, with coke formulated as  $C_aH_bO_cN_dS_eA$ . This conversion is formulated as the formula:



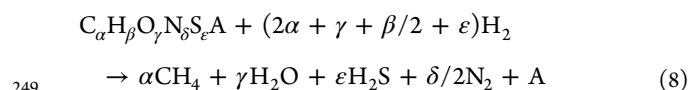
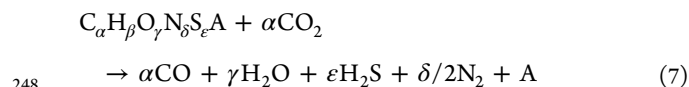
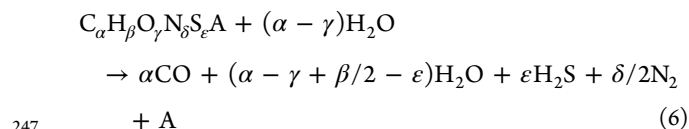
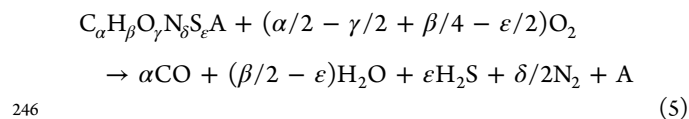
where A is ash. The composition of coke is determined by the mass balance in reaction 1. The volatiles are simply assumed as  $CH_4$ . It is combusted and converted to  $CO_2$  and  $H_2O$ . The reactions in the combustion are expressed by formulas 2–4.







244 With deficient  $\text{O}_2$ , coke is combusted and gasified. The main  
245 reactions are shown in formulas 5–8.



250 In our work, Texaco gasification, one of the major gasification  
251 techniques, was adopted. Coal and volatiles were defined as  
252 unconventional solids. Both the conventional solid and the  
253 unconventional solid in coal were modeled by the inherent  
254 class without particle size distribution, MIXCINC, in Aspen  
255 Plus. The reactions in the gasifier were modeled by the RStoic  
256 model. The process of coal gasification was modeled by the  
257 Gibbs free energy. The conversion rate of coal was fixed to  
258 0.95. The composition of the gaseous mix was determined  
259 according to the property of the input coal (i.e., analysis basis,  
260 dried basis, and sulfur content in coal). These gases mainly  
261 include CO and  $\text{H}_2$ , as well as a slight amount of  $\text{CO}_2$ , steam,  
262  $\text{N}_2$ ,  $\text{CH}_4$ ,  $\text{H}_2\text{S}$ , and traces of COS,  $\text{NH}_3$ , ash, and unconverted  
263 molten coal slag. The three-level heat exchanger was used and  
264 modeled by three HeatX models for recovery of the output  
265 syngas heat (NETL, 2010). The simulation was verified by  
266 comparing the composition of the output syngas with that of  
267 Zheng and Furinsky's work.<sup>14</sup> The comparison is shown in  
268 Table 1. It is seen that both compositions are similar with small  
269 relative errors  $REs \leq 1.5$ .

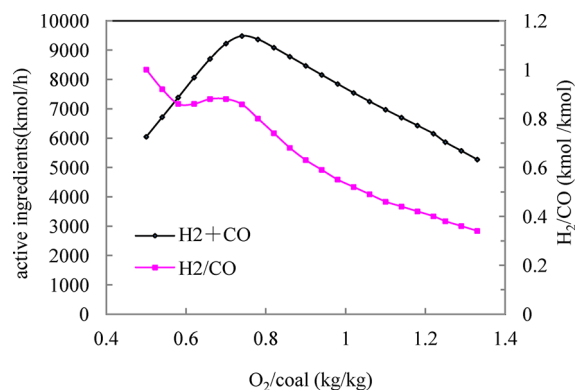
**Table 1. Comparison between Simulated SPG and Reference SPG<sup>a</sup>**

	our results	references	RE
CO (mol %)	39.5	38.4	0.78
$\text{H}_2$ (mol %)	27.7	28.6	0.63
$\text{CO}_2$ (mol %)	11.2	12.6	0.98
$\text{H}_2\text{O}$ (mol %)	19.3	17.4	1.34
$T$ ( $^\circ\text{C}$ )	1369	1371	1.41
$P$ (MPa)	4.2	4.3	1.06

<sup>a</sup>RE: relative error. Reference: Zheng and Furinsk, 2005.

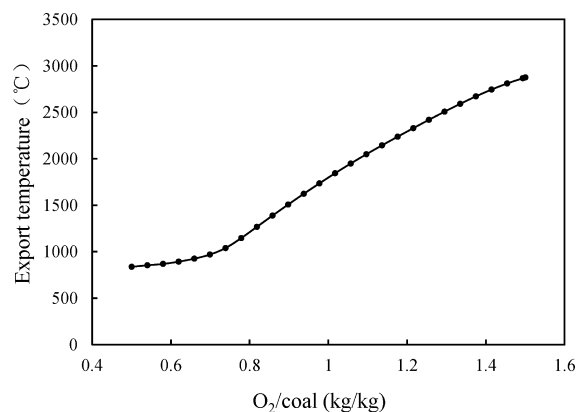
270 Because of the significance of gasification, it is necessary to  
271 analyze its key parameters. They are the ratio of the input  $\text{O}_2$   
272 and coal ( $\text{O}_2/\text{coal}$ ), the gasification temperature, the amount of  
273 active ingredients, and the sum amount of active ingredients  
274 ( $\text{H}_2$  and CO). For analysis of the three parameters,  $\text{O}_2/\text{coal}$  was  
275 taken as the designed parameter to study its relation with the  
276 other two parameters.

277 The relations between the  $\text{O}_2/\text{coal}$ , the  $\text{H}_2/\text{CO}$ , and the  
278 amount of active ingredients are shown in Figure 5, where the



**Figure 5.** Relation of  $\text{O}_2/\text{coal}$ ,  $\text{H}_2/\text{CO}$ , and moles of active ingredients.

$\text{O}_2/\text{coal}$  is designed to change from 0.5 to 1.3. It is seen that the  
 $\text{H}_2/\text{CO}$ , in general, declines as the  $\text{O}_2/\text{coal}$  increases. Yet the  
 $\text{H}_2/\text{CO}$  stays around 0.85 when the  $\text{O}_2/\text{coal}$  is between 0.6 and  
0.8. This is because, at first, the coal slurry in the gasifier is  
combusted to CO and  $\text{H}_2$  when  $\text{O}_2$  is insufficient. As  $\text{O}_2$   
increases, coal is oxidized into CO and  $\text{CO}_2$  so  $\text{H}_2/\text{CO}$   
decreases. Yet some CO is reacted with  $\text{H}_2\text{O}$  and converted to  
 $\text{CO}_2$  and  $\text{H}_2$ . This reaction increases the amount of  $\text{H}_2$  and  
meanwhile reduces that of CO. With these two reactions, the  
 $\text{H}_2/\text{CO}$  fluctuates in a small range. As  $\text{O}_2$  further increases, the  
oxidation of coal dominates in the gasification. The amount of  
CO increases so that the  $\text{H}_2/\text{CO}$  decreases again. When  $\text{O}_2$  is  
in excess,  $\text{H}_2$  and CO are oxidized into  $\text{H}_2\text{O}$  and  $\text{CO}_2$ . The  
oxidation of  $\text{H}_2$  has a higher priority due to its larger active  
energy so the  $\text{H}_2/\text{CO}$  keeps decreasing. The amount of active  
ingredients increases when the  $\text{O}_2/\text{coal}$  increases at the  
beginning and then quickly declines when the  $\text{O}_2/\text{coal}$  further  
increases over 0.8. This amount approaches the maximum  
when the  $\text{O}_2/\text{coal}$  lies between 0.7 and 0.8. Figure 6 shows the

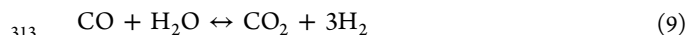


**Figure 6.** Relation between  $\text{O}_2/\text{coal}$  and export temperature.

relationship between the  $\text{O}_2/\text{coal}$  and the gasification temper-  
ature. In general, the temperature increases as the  $\text{O}_2/\text{coal}$   
increases because the oxidation of coal is an exothermic  
reaction. Depending on the above analysis, the  $\text{O}_2/\text{coal}$  is fixed  
at 0.8 in this Article for obtaining the maximal amount of active  
ingredients. At this time, the temperature is around 1386  $^\circ\text{C}$ ,  
which is in the temperature range that an industrial Texaco  
gasifier specifies, 1350–1400  $^\circ\text{C}$ .<sup>15</sup>

2.1.2. WGS. Water gas shift (WGS) converts CO to  $\text{H}_2$ .  
Thus, it is used to adjust  $\text{H}_2/\text{CO}$  in the syngas for the synthesis

of coal derivative chemicals. For modeling of the WGS, we used a middle-temperature shift reaction connected by a low-temperature shift reaction. Although these reactions are run at different temperatures, they could be formulated by the same reaction formula, as follows:



On the basis of LHHW,<sup>16,17</sup> the thermodynamics and kinetics of WGS could be formulated as eqs 10 and 11:

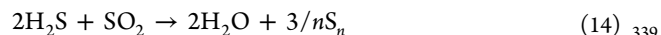
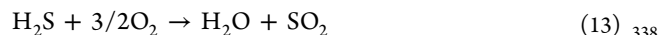
$$K_{\text{WGS}} = \frac{p_{\text{CO}_2} p_{\text{H}_2}}{p_{\text{CO}} p_{\text{H}_2\text{O}}} \quad (10)$$

$$r_{\text{WGS}} = 7.33e^{-7} \psi k_{\text{WGS}} \left( p_{\text{CO}} p_{\text{H}_2\text{O}} - \frac{p_{\text{CO}_2} p_{\text{H}_2}}{K_{\text{WGS}}} \right) \quad (11)$$

where  $i$  is the composite of the syngas ( $i = \text{CH}_4, \text{H}_2\text{O}, \text{CO}, \text{CO}_2$ );  $r$  (kmol/kg cat·s) is the reaction rate;  $\psi$  is the active factor affected by the used catalysts;<sup>18</sup>  $p_i$  is the partial pressure of composite  $i$ ;  $k_{\text{WGS}} = k_{0,\text{WGS}} \exp((-E_a)/(RT))$  is the reaction rate constant of reaction 12;  $K_{\text{WGS}}$  is the equilibrium constant;  $E_a$  (kJ/kmol) is the active energy; and  $R$  (kJ/kmol·K) is the gas constant.

**2.1.3. AGR and SR.** The crude syngas from the gasifier consists of impurities, ash, and acid gases ( $\text{SO}_x$ ,  $\text{NO}_x$ , and  $\text{CO}_x$ ). It is necessary to remove the acid gases before chemical synthesis. The gas clean process consists of an acid gas removal process and a sulfur recovery process. The flowsheet of this process is shown in Figure 7. In the first subprocess, the

Rectisol method was adopted to remove  $\text{CO}_2$ ,  $\text{H}_2\text{S}$ , organic sulfur compounds, cyanide, unsaturated hydrocarbon, etc. For sulfur recovery, a two-level CLAU conversion process was used. This two-level process includes a partial oxidation reaction of  $\text{H}_2\text{S}$ , formulas 12 and 13, and a CLAU catalytic reaction, formula 14.



The material flows were modeled as the conventional solids without particle size distributions, MIXCISLD, in Aspen Plus. Property method used IDEAL (ideal gas/Raoult's law/Henry's law). The integrated emission standard of air pollutants, GB16297-1996<sup>19</sup> provided that the sulfur recovery in coal gasification-based processes should be above 99.8%. For this emission standard, we added the low-temperature tail gas treatment process, SCOT in the SR.<sup>20</sup> The exhausted gas from CLAU is reheated for hydrogen reduction in the hydrogenation reactor. Next, hydrogenated gas is cooled by the downstream quench tower. The sulfur in the cold gas is recovered again in the absorber. The desulfurization solvent is recycled through the solvent regeneration tower. The desulfurized gas is combusted and then emitted to the environment. With this SCOT method, the sulfur recovery approached 99.87%, and the concentration of  $\text{SO}_2$  was less than 960 mg/m<sup>3</sup>.

**2.1.4. MS.** Methanol is one of the important platform chemicals. Its derivatives are widely used in industry. During the construction of the framework, we selected methanol synthesis for CS in this Article. Figure 8 shows the process of

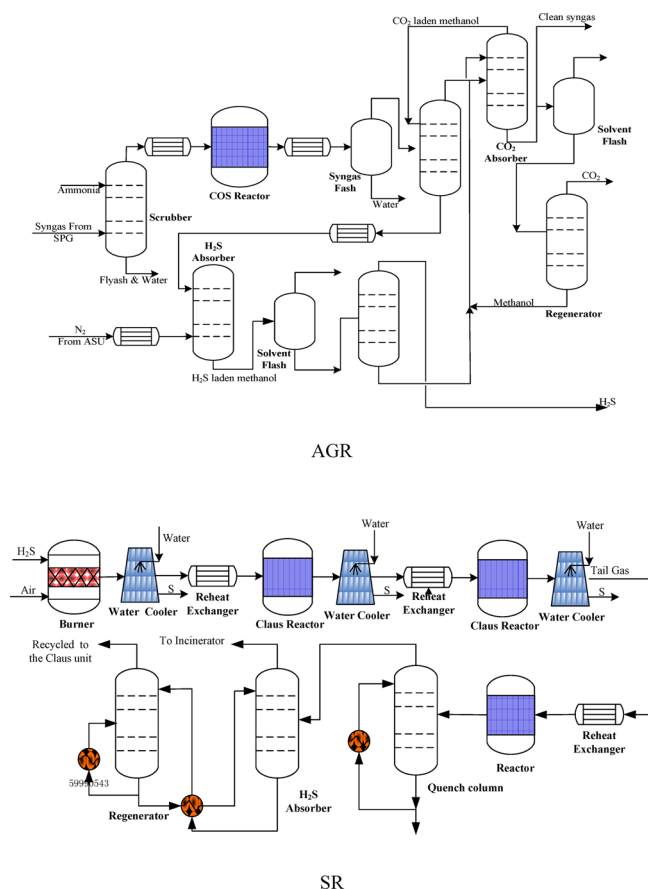


Figure 7. Process flowsheet of AGR and SR.

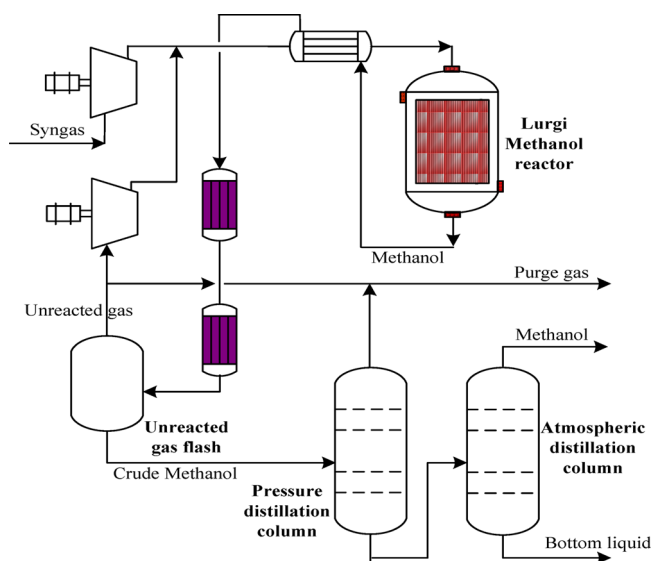
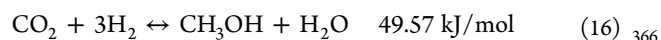
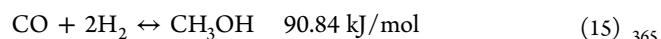
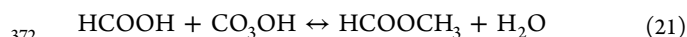
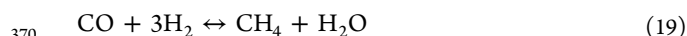
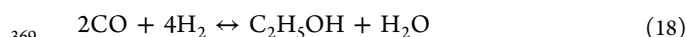
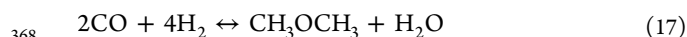


Figure 8. Process flowsheet of MS.

the methanol production. Different reversible reactions take place in the synthesis reactor because there are different substances,  $\text{CO}$ ,  $\text{CO}_2$ ,  $\text{H}_2$ ,  $\text{CH}_3\text{OH}$ ,  $\text{H}_2\text{O}$ , and  $\text{CH}_4$ . The main reactions are:



and the side reactions are:



Cu–Zn–Al catalyst was used in this reaction with its suitable temperature range from 200 to 300 °C and pressure range from 5 to 10 MPa. If ignoring the side reactions, the thermodynamics and kinetics model of reactions 15 and 16 are shown as follows:

$$K_{\text{MS},1} = \frac{p_{\text{CH}_3\text{OH}}}{p_{\text{CO}} p_{\text{H}_2}^2} = \frac{1}{p} \frac{y_{\text{CH}_3\text{OH}}}{y_{\text{CO}} y_{\text{H}_2}^2} \quad (22)$$

$$K_{\text{MS},2} = \frac{p_{\text{CH}_3\text{OH}} p_{\text{H}_2\text{O}}}{p_{\text{CO}_2} p_{\text{H}_2}^3} = \frac{1}{p^2} \frac{y_{\text{CH}_3\text{OH}} y_{\text{H}_2\text{O}}}{y_{\text{CO}_2} y_{\text{H}_2}^3} \quad (23)$$

$$\begin{aligned} -r_{\text{CO}} &= \frac{k_1 \left( p_{\text{CO}} p_{\text{H}_2}^2 - \frac{1}{K_{\text{p}_1}} p_{\text{CH}_3\text{OH}} \right)}{\left( 1 + K_{\text{CO}} p_{\text{CO}} + K_{\text{CO}_2} p_{\text{CO}_2} + K_{\text{H}_2} p_{\text{H}_2} \right)^3}, k_1 \\ &= 6.175 \times 10^{-16} \exp \left( \frac{5.043 \times 10^7}{RT} \right) \end{aligned} \quad (24)$$

$$\begin{aligned} -r_{\text{CO}_2} &= \frac{k_2 \left( p_{\text{CO}} p_{\text{H}_2}^3 - \frac{1}{K_{\text{p}_2}} p_{\text{CH}_3\text{OH}} p_{\text{H}_2\text{O}} \right)}{\left( 1 + K_{\text{CO}} p_{\text{CO}} + K_{\text{CO}_2} p_{\text{CO}_2} + K_{\text{H}_2} p_{\text{H}_2} \right)^4}, k_2 \\ &= 6.296 \times 10^{-20} \exp \left( \frac{6.997 \times 10^7}{RT} \right) \end{aligned} \quad (25)$$

where  $K_{\text{MS},1}$ ,  $K_{\text{MS},2}$  are the equilibrium constants of reactions 15 and 16;  $k_1$ ,  $k_2$  (kmol/m<sup>3</sup>·s·Pa) are the reaction rate constants;  $K_i$  is the absorption equilibrium constant; and  $y_i$  is the mole fraction of gaseous composite  $i$ .

Lurgi synthesis process was selected in this Article and modeled by using the Requil model in Aspen Plus. The key parameters for this synthesis are reaction temperature, reaction pressure, and recycle rate of the unconverted gas. The effects of these three parameters on methanol productivity were analyzed, respectively. For the reaction temperature, the temperature was designed to increase from 127 to 327 °C. The productivities are shown in Figure 9. Obviously, the productivity declines with an increasing rate. Referring to the book,<sup>21</sup> we fixed the temperature range for the synthesis between 200 and 300 °C. For the reaction pressure, we designed the pressure increasing from 7.05 to 9.05 MPa, and its effect is shown in Figure 10. Because the number of reactant molecules decreases as the reaction equilibrates, increasing reaction pressure is in favor of balancing the reaction and improving the efficiency of the catalyst. Certainly, the selection of the reaction pressure also depends on the activity of catalysts, the composition of syngas, and the heat balance of synthesizer, etc. For the recycle rate, we designed the rate increasing from 0.5 to 0.99. Its effect is shown in Figure 11. The productivity increases as the rate increases. However, increasing the rate blindly would incur the dramatic increasing of the compressors'

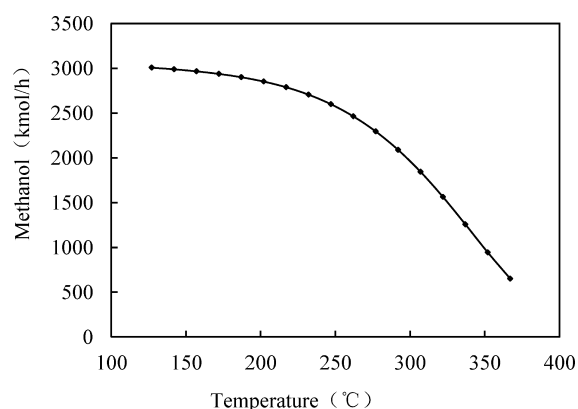


Figure 9. Relation between reaction temperature and methanol productivity.

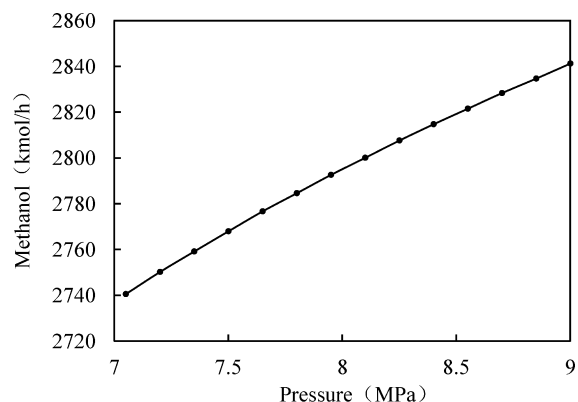


Figure 10. Relation between reaction pressure and methanol productivity.

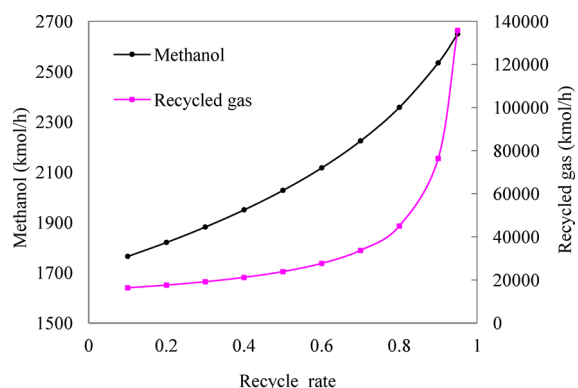


Figure 11. Relation of recycle rate, methanol productivity, and moles of recycled gas.

work and then electricity consumption. This work could be estimated roughly by the formula of shaft work:

$$W = p_1 V_1 \gamma / (\gamma - 1) [(p_2 / p_1)^{(\gamma-1)/\gamma} - 1] \quad (26)$$

where  $\gamma$  is the adiabatic index,  $p_1$  and  $p_2$  are the pressures of the unconverted gas before or after compression, and  $V_1$  is the volume of the unconverted gas before compression. It is seen that the work is proportional to the volume. The volume of gases is proportional to their moles at a constant pressure so the moles is proportional to the work. The relation between the recycle rate and the input gas moles is shown in Figure 11. The moles increases exponentially as the recycle rate increases

further over 0.8, as does the resulting work. This analysis gives us the important suggestion on the determination of the recycle rate.

**2.2. Theoretical Analyses.** There are a number of analysis techniques involved in the framework, which are classified into four categories: thermodynamic analysis, techno-economic analysis, life cycle assessment, and sustainability analysis. The following sections describe the analysis methods used in the four categories.

**2.2.1. Thermodynamic Analysis.** Van Gool<sup>22</sup> and Hammond<sup>23</sup> believed that the essence of energy conversion is explored not only by the first law of thermodynamics but also by the second law. The second law defines the quality of energy carriers, indicated by exergy. Chemical processes consist of material streams, heat streams, and work streams. The first and foremost step for exergy analysis is to calculate the exergy values of these streams, respectively. Exergy was defined as the maximum work that the stream could do from their initial state to dead state.<sup>24</sup> According to the classification,<sup>25</sup> exergy of a process stream consists of the physical exergy,  $Ex_{phys}$ , the chemical exergy,  $Ex_{chem}$ , and the change of mixing exergy,  $\Delta_{mix}Ex$ . The total exergy of a multicomponent material stream is calculated by eq 27.

$$Ex = Ex_{chem} + Ex_{phys} + \Delta_{mix}Ex \quad (27)$$

Exergy efficiency is used as one of the important indicators for energy conversion with the formulation as the ratio  $\eta$  of the output exergy  $Ex_{out}$  and the input exergy  $Ex_{in}$ :

$$\eta = \frac{Ex_{out}}{Ex_{in}} \quad (28)$$

Exergy losses of units, as shown in eq 29, are calculated to explore inefficient units.

$$Ex_{loss,j} = Ex_{in,j} - Ex_{out,j} \quad (29)$$

Exergy analysis of coal-based processes has been reported in much research. For example, it was used to explore the low-efficient units in power plants.<sup>26,27</sup> It was also used in analysis of IGCC to improve its energy efficiency.<sup>28</sup> With increased understanding of thermodynamic systems, exergy analysis was integrated with other techniques to reflect environmental impact, resource efficiency, and further sustainability.<sup>29–31</sup> The relationship with environmental impact exists because the exergy analysis of a system considers the difference between the states of the system and environment. As for the relationship with resource efficiency, some research regarded all natural resources as the carriers of exergy. In contrast, wastes were regarded as the invalid exergies without carriers so it would damage the environment. Combining the above ideas, exergy could be used for analysis of the environmental impact, the resource efficiency, and finally the sustainability of a production process.

As with the use of gasification techniques, energy processes and chemical processes are usually combined together for coproduction processes. Because the products include both energy and chemicals, it is inappropriate to simply add their exergies together in exergy analysis. Thus, we used the exergy saving ratio (ESR) to assess the exergy efficiency of the coproduction processes in this Article.<sup>32</sup> ESR is defined as the exergy saving of a coproduction process as compared to the equivalent single processes that produce the same amount of

products as those of the coproduction process, as in the following equations:

$$ESR = (Ex_{input}^{ref} - Ex_{input}^{co}) / Ex_{input}^{ref} \times 100\% \quad (30)$$

$$Ex_{input}^{ref} = \sum_{i=1}^N Ex_{input,i}^{sp} \quad (31)$$

$$ESR = \sum_{i=1}^M ESR_i \quad (32)$$

where  $Ex_{input}^{ref}$ ,  $Ex_{input}^{co}$ , and  $Ex_{input,i}^{sp}$  are the input exergies of the reference process, the coproduction process, and the single process with product  $i$ , respectively,  $ESR_j$  is the saving exergy of unit  $j$ , and  $N$  and  $M$  are the numbers of products and unit processes.

**2.2.2. Techno-economic Analysis.** Techno-economic analysis was also utilized in this framework. Different indicators, such as investment, reliability, and industrial safety, reflect the performances of applied processes.

Total investment  $I_{total}$  is an important index for assessing the economic performance. It consists of static investment  $I_{static}$  and production investment  $I_{product}$ .

$$I_{total} = I_{static} + I_{product} \quad (33)$$

$I_{static}$  consists of two subcosts: overnight cost  $I_{oc}$  and construction interest  $I_{ci}$ .  $I_{static}$  is calculated by the following equations:

$$I_{static} = I_{oc} + I_{ci} = I_{oc}(1 + \rho) \quad (34)$$

$$I_{oc} = \sum_j [\theta I_j^{ref} (C_j / C_j^{ref})^{sf}] \quad (35)$$

where  $\rho$ ,  $\theta$ , and  $sf$  represent the construction interest rate, the domestic made factor, and the scale factor, respectively;  $C_j$  and  $C_j^{ref}$  indicate the practical scale and the reference scale; while  $I_j^{ref}$  is the reference investment of unit  $j$ . Production investment  $I_{product}$  mainly consists of annual investment  $I_{annual}$ , material charge  $I_{material}$ , and maintenance charge  $I_{maintenance}$ . It is calculated by the following equations:

$$I_{product} = I_{annual} + I_{material} + I_{maintenance} \quad (36)$$

$$I_{annual} = I_{oc} \times CRF \quad (37)$$

$$CRF = r_{discount} / [1 - (1 + r_{discount})^{-n}] \quad (38)$$

where  $CFR$  is the capital recovery factor;  $r_{discount}$  is the discount rate; and  $n$  is the lifetime of the process.

Theoretically and practically, integration of different production processes leads to sharing of materials, utilities, equipment, and services. This will bring a large amount of saving in investment. Certainly, it is inadequate to use traditional investment calculation to assess coproduction processes. We used the investment saving ratio (ISR) to analyze the economic performance. Similar to the ESR, it is defined as the investment saving of a coproduction process as compared to the reference single processes. The formula of ISR is shown below.

$$ISR = (I^{ref} - I^{co}) / I^{ref} \times 100\% \quad (39)$$



$$I^{\text{ref}} = \sum_{i=1}^N I_i^{\text{sp}} \quad (40)$$

where  $I^{\text{ref}}$  is the investment of the reference process,  $I^{\text{co}}$  indicates the investment of the coproduction process, and  $I_i^{\text{sp}}$  is the investment of the single process that produces the product  $i$ .

**2.2.3. Life Cycle Assessment.** Life cycle assessment (LCA) analyzes production processes from the life cycle of their products. To date, there has been much research on life cycles assessment.<sup>33–35</sup> They used LCA as the decision-making tools for process design, selection, and optimization. However, application of LCA is limited due to the shortage of data at various life periods and means for evaluation of data quality. During the development of the framework, LCA was being conducted for evaluating the environmental impact of coal gasification-based processes. Several environmental indexes were used to measure different environmental impacts, global warming, acidification, photochemical oxidant formation, and human toxicity. The periods that had significant impact on environmental performance were explored.<sup>36</sup> However, in this Article, we focused on the modeling of the basic units and the synthesis of typical coal-based processes rather than the life cycle analysis and sustainability of the applied processes. The research on LCA will be reported in detail in coming papers.

**2.2.4. Sustainability Analysis.** Sustainability analysis is multiscale and multiobjective. It addresses broader perspective issues, which could be classified into a global one, regional one, industrial one, and technical one.<sup>37,38</sup> To date, there are numerous sustainability analysis tools developed. Ness et al.<sup>39</sup> proposed a generic framework, which involves three basic classes of tools, with the first using sustainability indicators; the second focusing on material flows, energy flows, and information flows of production processes; and the third focusing on the changes of policies and the feasibility of projects.

Combining exergy with life cycle assessment gives rise to exergy life cycle assessment (ELCA), which has been broadly accepted as the tool for sustainability analysis. Hau et al.<sup>40</sup> applied ecological exergy (the derivative of ELCA) on comparison between a coal-based electricity generation and a solar-based one. This analysis took ecological exergy of coal into consideration so that exergy efficiency of the coal-based generation is much lower than the solar-based one. Jorgensen<sup>41</sup> also suggested that ELCA unifies exergy measures of different raw materials, enabling for sustainability analysis. Beside ELCA, emergy is another tool for sustainability analysis. It unifies flows and storages of energy and materials with the same measurement: solar emergy. Lan et al.<sup>42</sup> suggested that emergy gives a way for quantified studies of ecological systems. Bakshi<sup>43</sup> proposed an emergy framework for both industrial processes and ecological processes. Ometto et al.<sup>44</sup> employed emergy into analysis of ecological agriculture processes. Zhou et al.<sup>45</sup> used emergy for analysis of biogas farming processes. During the construction of the framework, we mainly utilized the above two theories and the corresponding techniques for studying the sustainability of the coal-based processes. In fact, we are building the ELCA and the emergy models for the coal-based methanol production process. The data collections for the stages of raw material mining and production process have been finished. However, the study of the sustainability of coal-based processes will be reported in other papers.

**2.3. Process Optimization.** The last block in the framework is the optimization block. Optimizations of the applied processes are done according to the analyses from the analysis block. The coal gasification-based processes are usually complicated and involve a large number of discrete variables. These processes are normally modeled by MINLP programming. Until now, there has been much research on the optimizations of large-scale energy chemical processes based on different energy resources. For example, Floudas and his research team built the comprehensive superstructures for the different fuel and electricity production processes from different energy resources. They used the economic performances as the objectives for energy, power, and water integration of these processes.<sup>46–50</sup> On the study of optimization and corresponding techniques, our team has done a series of research on quantitative optimization method with time delay and parametric uncertainty,<sup>51</sup> multiobjective optimization for reactor network synthesis,<sup>52</sup> optimizing the initial conditions for batch processes,<sup>53</sup> and dynamic optimization of batch processes.<sup>54</sup> All of the above research gave the concrete base for establishing the optimization block in the framework. The optimization is multiobjective in the framework, because the technical performance, the economic performance, and the environmental impact of applied processes are analyzed in the analysis block. In fact, the analyses of these three aspects often conflict. The optimization is therefore to find the best trade-off of these three aspects. During the early stage of the framework construction, we did not do specialized analysis on the linearity and the convexity of the optimization problems. Optimization was implemented simply by combining Aspen Plus and Visual Basic in Microsoft Excel. The optimization code was written in the visual basic interface. More professional optimization will be done and reported in our future papers.

**2.4. Applied Software and Databases.** For construction of the framework, a number of engineering software tools and databases were used. Data for reaction thermodynamics, kinetics, and physical properties were obtained from DECHEMA and several other engineering databases. These databases were embodied in process modeling software tools. For process synthesis, several chemical process modeling software tools, Aspen Plus, gPROMS, and SuperPro, were used. For LCA, some databases are suitable for a specific series of products or to a specific geographical region (e.g., GREET, BUWAL). Some databases are general, for example, Ecoinvent and GaBi. The LCA software, SimaPro, was used in our framework for building the life cycle model. For sustainability analysis, SimaPro was integrated with ExerCom embedded in Aspen plus to build the ELCA models. For process optimization, a number of decision-making and engineering optimization tools, GAMS, OPTIMUS, and LINGO, were used in our work.

### 3. CASE STUDY

The framework was established for comprehensive study of coal-based chemical and energy processes. For case study, we selected three processes: an integrated gasification combined cycle process; a coal-based menthol production process; and a coproduction process combining the previous two. These three processes represent the three possible branches in the coal gasification-based product chain in Figure 2. The modeling and synthesis of the three cases are described in the following.

**3.1. Integrated Gasification Combined Cycle.** The flowsheet of the integrated gasification combined cycle

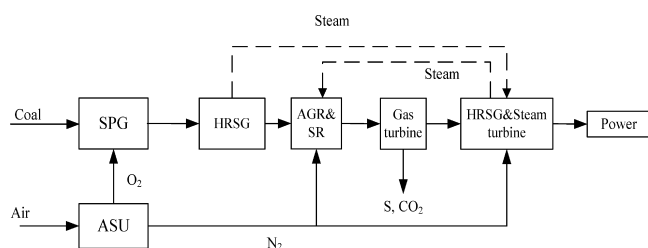


Figure 12. Schematic diagram of IGCC.

exergy loss are SPG, HSRG, and CC, with their percentages of the total losses as 18.4%, 13.6%, and 14.8% of the total loss, respectively. The exergy losses in the SPG are caused by the high irreversibility of gasification process, while the exergy losses in the gas turbine and the steam turbine are caused by the irreversibility of the combustions. The exergy losses in HSRG are caused by heat transferring. In contrast to the above units, exergy losses in the ASU and the SR are small. The exergy loss in the ASU is brought by different reasons, such as mechanical work done by the four-level cold compressors, the temperature difference for heat transferring at the top and the bottom of the distillation, and the heat transfer between liquid phase flows and gaseous phase flows. In the SR, the heat recovery boiler is the main source of exergy loss.

**3.2. Methanol Production Process.** In our work, the coal to methanol process (CTM) takes the same gasification process as that of IGCC. Its process flowsheet is shown in Figure 14. The crude syngas is separated into two parts in the CTM. One is directly fed to the gas clean process, while the other is fed to the WGS before the clean process. The two-part gases are cleaned and then fed into the methanol synthesis reactor together. The output stream is a mix of methanol, water, and unconverted syngas. The unconverted syngas is extracted by a liquid–gas separator, and most of it is fed back to the synthesis reactor while the rest is exhausted as the tail gas. Methanol solution is purified in the distillation tower. For the CTM synthesis, the molar ratio of the syngas need for water gas shift and the syngas need not was set as 47:53. Other operational parameters are shown in the column CTM of Table 2.

The material flow diagram of CTM is shown in Figure 15. There are several assumptions for drawing the material flow diagrams of the three case processes: (1) the water consumed in the production process was counted rather than that used for heat exchange and machine service; and (2) the recycled materials were not included in the diagrams but regarded as the inner material flows of the processes. According to the diagrams, the exergy analysis was done, and results were shown in Table 5. The main sources for exergy losses were found to be SPG, HSRG, and MS with their percentages as 16.9%, 12.1%, and 7.3%, respectively. The exergy loss in MS mainly consists of the work done for compression of syngas and the lost exergy in the tail gas. In addition, the heat exchanges of material flows at the inlet and the outlet of MS are irreversible, giving rise to the exergy loss. In CTM, WGS is for adjusting the  $H_2/CO$ , and its exergy loss is brought because of the irreversibility in the recovery of the steams. On the other side, the shift reaction is a relatively efficient process, with a small exergy loss, which is shown in the table.

**3.3. Coproduction Process.** The coproduction process (coal to methanol and electricity, CTME) is regarded as the integration of IGCC and CTM, with its process flow shown in Figure 16. For material sharing, the syngas from the gasifier is separated into two parts at point 1 ( $P_1$ ), with one for methanol production and the other for electricity generation. The unconverted syngas from the reactor is also separated into two parts, with one fed into the gas turbine and the other recycled back into the reactor at point 2. The molar ratio at  $P_1$  ( $r_{P1}$ ) and that at  $P_2$  ( $r_{P2}$ ) are two parameters, having significant effects on energy efficiency, input–output ratio, environmental emission, etc. In our work, we initially set that  $r_{P1} = 0.5$  and  $r_{P2} = 0.5$ , and detailed analysis and optimization of  $r_{P1}$  and  $r_{P2}$  will be studied later in this Article. Other operational parameters of CTME are listed in the column CTME of Table 2. Figure 17 is

Table 2. Reference Operational Parameters of IGCC, CME, and CTME

parameters	IGCC	CTM	CTME
O <sub>2</sub> supplied mole purity (%)	95	95	95
gasification pressure (MPa)	4.2	7.2	7.2
gasification temperature (°C)	1027	1027	1372
CO conversion ratio (%)		47	
H <sub>2</sub> /CO ratio in syngas		1.9	0.6
methanol synthesis temperature (°C)		272	272
methanol synthesis pressure (MPa)		8	8
unconverted recycling ratio		3.1	0
gas turbine outlet temperature (°C)	303		977
gas turbine expansion ratio	16		16
high pressure (MPa)	7.5		12
high temperature (°C)	497		565
intermediate pressure (MPa)	3.5		4
intermediate temperature (°C)	497		563
low pressure (MPa)	0.5		0.3
low temperature (°C)	243		243
pressure in captive power station (MPa)		3.5	
temperature in captive power station (°C)		497	

IGCC was analyzed with use of exergy theory. For exergy analysis, the material flowchart is shown in Figure 13. According to the material flowchart, we could do exergy analysis for IGCC. The chemical exergies were calculated from the standard chemical exergy of elements at the reference condition.<sup>55</sup> In this Article, we used the coal from Luzhou, China for the case study. The composition of this coal is shown in Table 3. According to Zhou,<sup>56</sup> the exergy value of this coal is 27 703.3 kJ/kg. Exergy analysis of IGCC was done, with the results listed in Table 4. It is found that the main sources for

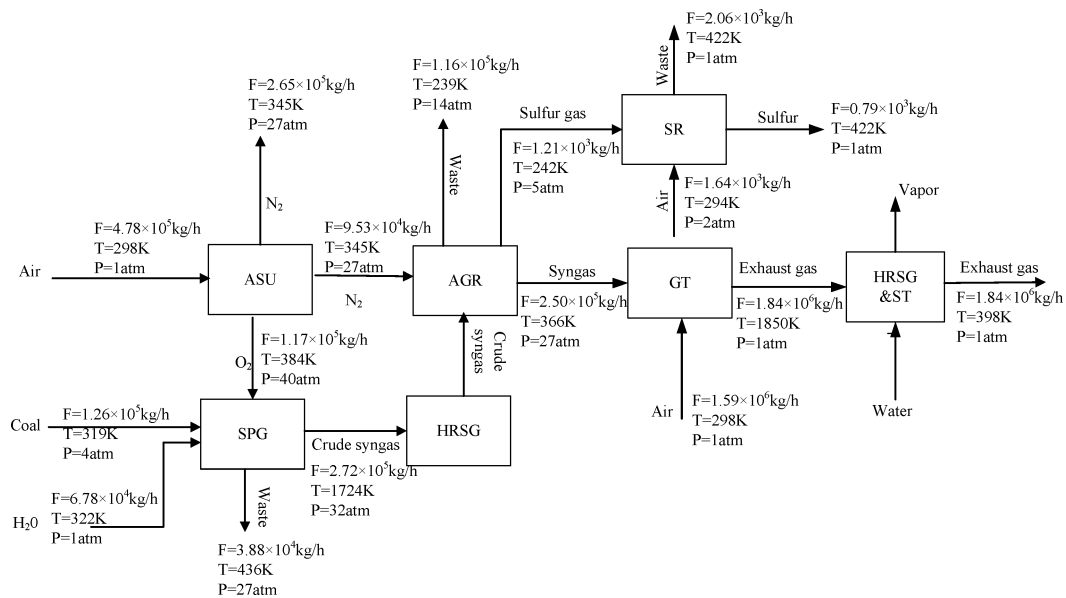


Figure 13. Schematic material flow diagram of IGCC.

Table 3. Composition of Coal at Luzhou, China

	coal					
	ash	C	H	N	S	O
mass fraction (%)	22	64	4.3	1.1	3.6	5

Table 4. Exergy Analysis of IGCC

	exergy (kJ/kg-coal)	ratio (%)
coal input	27 703	88.6
exergy input	3564	11.4
total input	31 267	100.0
ASU	2376	7.6
SPG	5753	18.4
AGR	500	1.6
SR	313	1.0
HRSG	4252	13.6
CC	4628	14.8
others	1407	4.5
total loss	19 230	61.5
output sulfur	94	0.3
electricity	11 944	38.2

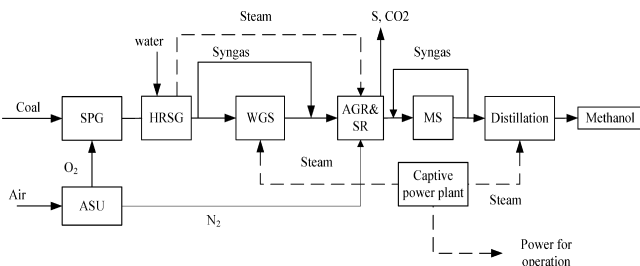


Figure 14. Schematic diagram of CTM.

the schematic material flow diagram of the coproduction. It is seen that  $8.28 \times 10^4$  kg/h unconverted syngas was fed into the CC unit for electricity generation. In addition to material sharing, there is heat shared between the two parts in the coproduction process. It could be found in the process of recovering waste heat from the gasifier and the steam turbine.

The recovered heat is used for electricity generation and methanol distillation. The exergy analysis of CTME is conducted and listed in Table 6.

**3.4. Comparison of the Three Processes.** We first compared the three processes with exergy analysis. The exergy efficiencies of the three cases and their unit processes were recorded and analyzed. It is seen that the exergy efficiency of CTME 46.9% is between those of IGCC and CTM. According to eqs 23 and 25, ESRs of units were calculated and shown in Figure 18. In general, the contributors of the reduction are the heat sharing and the material sharing. In MS, not all of the unconverted gas is recycled, but some is fed into the combined cycle for electricity generation. The excessive recycle would bring additional energy loss in the gas compression. Other heat sharing and material sharing is found to be the recovered heat used for the methanol distillation and the gas clean process. All of these heat sharings bring the exergy saving ratio to 1.52%, which is included in the "others" line in Figure 18. In addition, operational energy for equipments is internal-supplied in CTME, different from CTM in which the captive power plant is used to provide the energy. It is interesting that the exergy loss of CC does not decrease but increase by 0.94%. This is because the generated electricity in CTME is partially used for the syngas compression, the methanol distillation, and the water gas shift, etc. Thus, the output electricity is reduced.

Beside the comparison of energy efficiencies of the three processes, we also compared them from their investments. The investments were calculated on the basis of eqs 22–36. For investment comparison, we let the products of the coproduction process equal those of two single processes. The cost of the water consumed in the main processes was counted, while that in the auxiliary process, such as the heat exchange and the coal washing, was included in  $I_{\text{maintenance}}$ . Table 7 lists the reference investments of different units and the domestic made factors, referring to the works.<sup>56–58</sup> The nominal year for the investment estimation is 2008. Figure 19 shows the comparison of the capital investments of the three processes. It is seen that the coproduction saves cost on capital cost by 73 M\$. The product cost of the three processes was also calculated and is shown in Figure 20. For the production

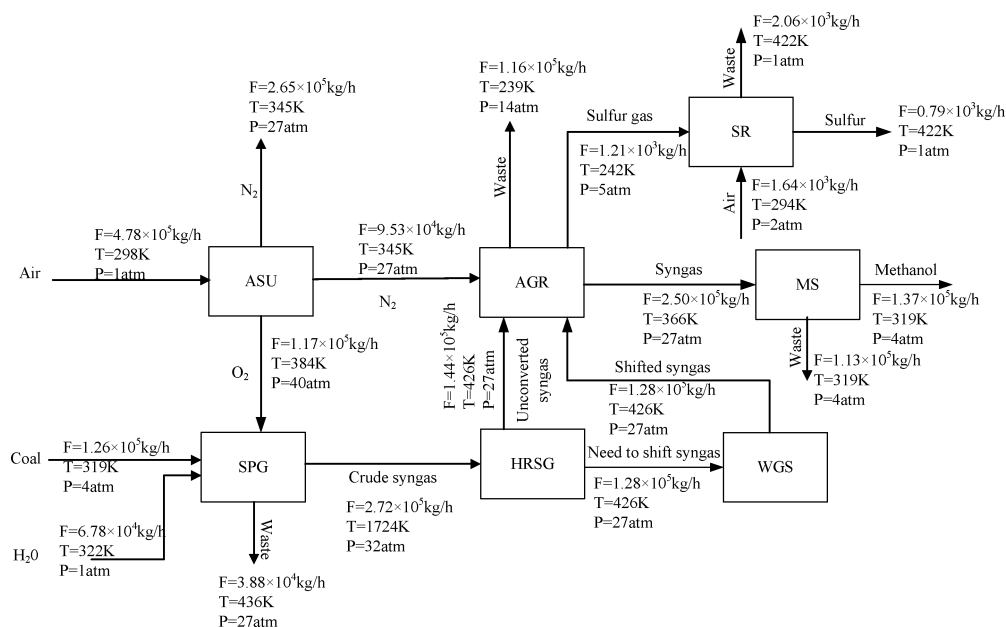


Figure 15. Schematic material flow diagram of CTM.

Table 5. Exergy Analysis of CTM

	exergy (kJ/kg-coal)	ratio (%)
coal input	27 703	81.8
exergy input	6164	18.2
total input	33 867	100.0
ASU	2303	6.8
SPG	5712	16.9
AGR	406	1.2
SR	135	0.4
HSRG	4102	12.1
WGS	610	1.8
MS	2472	7.3
others	1092	3.2
total loss	16 832	49.7
output sulfur	68	0.2
output methanol	16 967	50.1

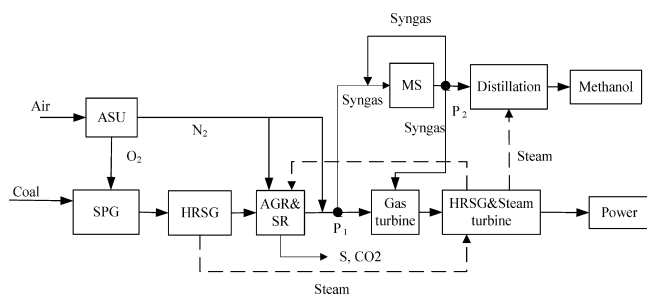


Figure 16. Schematic diagram of CTME.

**3.5. Process Optimization.** In the proposed framework, the last block is for process optimization in terms of technical, economic, and environmental points of views, respectively. In this Article, we optimized the two key parameters,  $r_{p1}$  and  $r_{p2}$  of CTME. These two parameters determine the distribution of raw materials between the two branches, the electricity generation and the methanol production. Before optimization, we assumed that the equipment operates at optimal states. In this Article, we mainly focused on the material flow parameters of the overall processes rather than the operational parameters, equipment parameters, and process configurations.

For optimization, we took *ESR* and *ISR* defined in section 2.2 as the objective functions. We summed the two objectives to simplify the multiobjective optimization into a single objective optimization. In our work, we equally assigned 50% to the weights of *ESR* and *ISR*, assuming that importance of these two objectives was the same. A new index, named as Energy and Investment Saving Ratio *EISR*, was proposed and formulated as:

$$EISR = (ISR + ESR)/2 \quad (41)$$

The classic Tabu search was used for optimization. Aspen Plus wrote the results of each iteration into Excel. The Visual Basic module in Excel was used to model the algorithm. In the algorithm, 10 local searches were done in an iteration. Tabu list was fixed to 10. The termination criterion was set as: The best state stays the same for 5; Total number of the iterations is larger than 50. For optimization using Tabu search, five runs from different initial  $r_{p1}$  and  $r_{p2}$  were done. Figure 21 shows a segment of average *EISRs* of the five runs as  $r_{p1}$  and  $r_{p2}$  took different values. The five runs are terminated at around 320 iterations. The changes of these two parameters lead to the changes in energy efficiency, investment, and product cost. It is concluded according to the figure that:

- (1) The surface of the figure is convex and has only one peak. This means that there is a set of  $r_{p1}$  and  $r_{p2}$  when *EISR* approaches the global optimal. As shown in Figure 21, the SPG, the AGR&SR, and the HRSG are shared for electricity generation and methanol production. This

investment, we set the discount rate to 0.1, the material cost to 600 RMB/t (\$95/t), and maintenance expense to 4% of the static investment. Similar to the capital cost, the product cost of the coproduction processes is lower than the sum of those of the two single processes (38 M\$/year). Thus, the coproduction process, which is the integration of electricity generation and methanol production processes, could increase energy efficiency and meanwhile save economic investment.



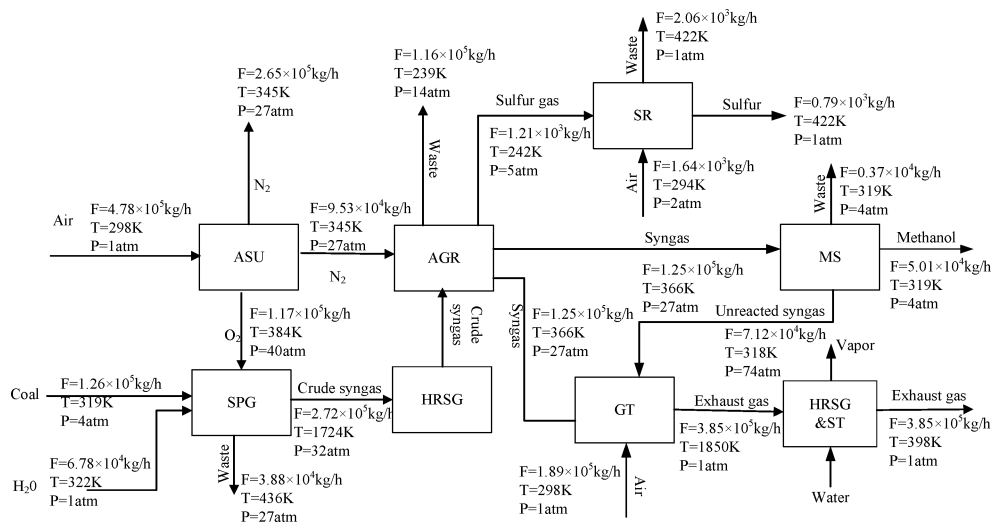


Figure 17. Schematic material flow diagram of CTME.

Table 6. Exergy Analysis of CTME

	exergy (kJ/kg-coal)	ratio (%)
coal input	27 703	88.5
exergy input	3585	11.5
total input	31 288	100.0
ASU	2264	7.2
SPG	5346	17.0
AGR	409	1.3
SR	220	0.7
HRSG	4056	12.9
CC	2830	9.0
WGS	0	0.0
MS	629	2.0
others	818	2.6
total loss	16 572	52.7
output sulfur	94	0.3
methanol	8490	27.5
electricity	6132	19.5

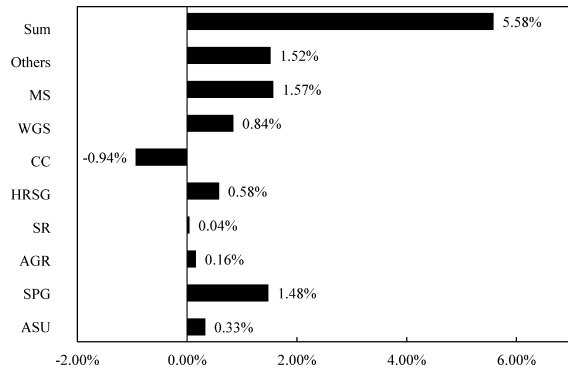


Figure 18. Exergy saving ratio of units of the coproduction process.

sharing makes  $I_{\text{static}}$  of the coproduction process less than the sum of  $I_{\text{static}}$  of the two single processes, so  $ISR > 0$ , while on the other side, the resulting material sharing and the energy sharing in the coproduction process makes  $ESR > 0$ .

(2) When  $r_{p1}$  is small, most of the syngas is used for electricity generation. The coproduction process is similar to IGCC, so  $EISR$  is small. As  $r_{p1}$  increases, the

syngas is increasingly separated for the methanol production. For energy sharing, electricity is self-consumed by the MS, the gas compression, and the methanol distillation. For material sharing, the unconverted gas is partially used for electricity generation. Thus,  $ESR$  and  $ISR$  increase. However, when  $r_{p1} > 0.3$ , most of the syngas is used for the methanol production. More and more energy is consumed for distillation and compression. The coproduction is similar to CTM, and thus  $EISR$  decreases.

(3) Similar as the effect of  $r_{p2}$ ,  $EISR$  first increases then decreases. The maximal  $EISR$  is approached when  $r_{p2}$  approaches to 0.7. With small  $r_{p2}$ , most of the unconverted syngas is used for electricity generation, leading to large generation capacity while small methanol productivity.  $ESR$  is low at this time, because the exergy efficiency of the methanol production process is in general higher than that of IGCC. As  $r_{p2}$  increases, the methanol productivity increases, so  $EISR$  increases. At this time, the recycled gas compressed into the reactor increases. This brings the increase of work done for gas compression, and as a result the increase of consumed electricity for the compressors. As discussed in section 2, the consumed electricity is increased exponentially as the recycle rate further increases over a certain value. Thus,  $EISR$  does not increase but drops rapidly.

(4) After optimization, we found that  $EISR$  approaches its maximum as  $r_{p1} = 0.3$  and  $r_{p2} = 0.7$ . This optimization provides the appropriate distribution of the syngas, both from the gasifier and from the reactor, between the electricity generation and the methanol production processes.

#### 4. CONCLUSIONS

This Article proposed a framework for integration of process modeling and simulation, process synthesis, analysis, optimization, and process design for coal-based energy and chemical processes. A number of analysis techniques were employed in the framework with four categories: techno-economic analysis, thermodynamic analysis, LCA, and sustainability analysis. For implementation, a series of engineering software tools and databases were employed. Three coal gasification-based

Table 7. Benchmark Case for Investments of the Study Cases

	benchmark	reference scale	scale factor	domestic made factor	reference investment (M\$)
ASU	oxygen supply	21.3 kg/s	0.50	0.50	45.70
coal preparation	daily coal input	27.4 kg/s	0.67	0.65	29.10
coal gasification <sup>a</sup>	daily coal input	39.2 kg/s	0.67	0.80	78.00
AGR&SR	sulfur output	29.3 mol/s	0.67	0.65	67.30
gas turbine <sup>b</sup>	electricity generation	276 MW	0.67	1.00	83.00
steam turbine, auxiliary	electricity generation	59.2 MW	0.67	0.65	94.70
WGS	material caloric value	1450 MW	0.67	0.65	28.00
synthesis and separation	syngas input	1081 mol/s	0.67	0.65	20.40

<sup>a</sup>Investment including that for waste heat boiler. <sup>b</sup>General Electric 109H gas turbine.

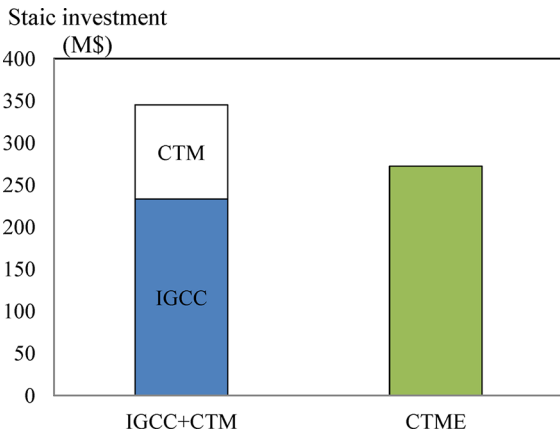


Figure 19. Static investment costs of IGCC, CTM, and CTME.

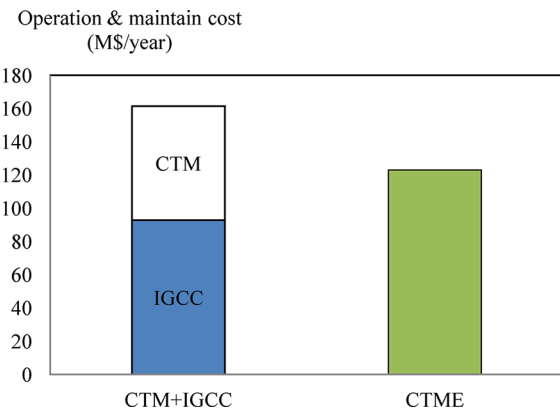


Figure 20. Product costs of IGCC, CTM, and CTME.

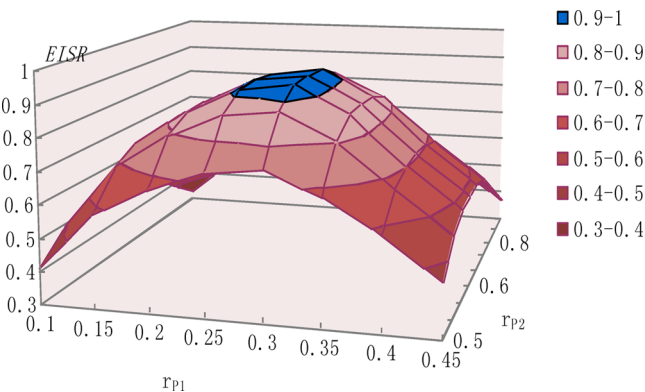


Figure 21. EISR as different  $r_{p1}$  and  $r_{p2}$ .

processes were taken as the study cases, IGCC, CTM, and CTME. Through unit modeling and process synthesis, the three cases were analyzed by using the exergy theory. For comparison of the three cases, the ESR and the ISR of CTME were calculated and analyzed. Results showed that the coproduction process has the higher exergy efficiency and the lower investment than the equivalent single processes. These improvements are mainly derived by the material and energy sharing and the equipment sharing in CTME. In addition, the self-supply electricity in CTME could eliminate the demand for the captive power plant. Some of the unconverted syngas is used for electricity generation instead of being recycled. This reduces the work of the compressors and their consumed electricity. In the end, we did optimization to explore the best material distribution between the electricity generation and the methanol processes, which was adjusted by  $r_{p1}$  and  $r_{p2}$ . This optimization used ESR and ISR as the two objectives.

In our future work, the framework will be expanded by including more analysis tools from technical, economic, and environmental points of views, respectively. The important tools are life cycle-related techniques, such as LCA, life cycle costing (LCC), ELCA, and so on. On the basis of these analyses, sustainability of processes will be concluded and then combined with optimization techniques to explore sustainable system processes. In addition, we will also model and simulate the coal-based processes with CCS. Techno-economic analysis and life cycle techniques will be used to explore the relation between the CCS and the overall processes. All of this work will be reported in a series of papers during the updating of the framework.

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### Notes

The authors declare no competing financial interest.

## ACKNOWLEDGMENTS

We would like to express our appreciation to the China NSF key project (no. 21136003), the National Basic Research Program (no. 2012CB720504), the Specialized Research Fund for the Doctoral Program of Higher Education (no. 20100172110016), and the China Postdoctoral Science Foundation (20100470922) for their great funding and support of this study.

## NOMENCLATURE

A = ash

AGR = acid gas removal

918 ASU = air separation unit  
 919 CC = combined cycle  
 920 CTM = coal to methanol  
 921 CTME = coal to methanol and electricity  
 922 HP = high pressure steam  
 923 HRSG = heat recovery steam generator  
 924 IGCC = integrated gasification combined cycle  
 925 LHHW = Langmuir–Hinshelwood and Hougen–Watson  
 926 LP = low pressure steam  
 927 MS = methanol synthesis  
 928 MP = middle pressure steam  
 929 SPG = solid preparation and gasification  
 930 WGS = water gas shift

### 931 Notations in Formulation

932  $a, b, c, d, e, w$  = mass fractions of elements C, H, O, N, S,  
 933  $\text{H}_2\text{O}$  in coal  
 934 CFR = capital recovery factor  
 935  $C_j$  = pactical scale of equipment  $j$   
 936  $C_j^{\text{ref}}$  = reference scale of equipment  $j$   
 937 EISR = energy and investment saving ratio of the  
 938 coproduction system, M\$  
 939  $E_a$  = activation energy, kJ/kmol  
 940 ESR = exergy saving ratio of the coproduction system, kJ  
 941  $\text{ESR}_j$  = exergy saving ratio of unit process  $j$ , kJ  
 942  $Ex_{\text{chem}}$  = chemical exergy, kJ  
 943  $Ex_{\text{phys}}$  = physical exergy, kJ  
 944  $Ex_{\text{in}}$  = input exergy, kJ  
 945  $Ex_{\text{out}}$  = output exergy, kJ  
 946  $Ex_{\text{loss}}$  = exergy loss, kJ  
 947  $Ex^{\text{sp}}$  = input exergy of single production system, kJ  
 948  $Ex^{\text{co}}$  = input exergy of coproduction system, kJ  
 949  $Ex_{\text{input}}^{\text{ref}}$  = reference input exergy, kJ  
 950  $Ex_{\text{input}}^{\text{co}}$  = input exergy of the coproduction system, kJ  
 951  $Ex_{\text{input}, j}^{\text{ref}}$  = input exergy of the single system with product  $i$ ,  
 952 kJ  
 953  $\Delta_{\text{mix}}Ex$  = exergy change of mixing, kJ  
 954 ISR = investment saving ratio of the coproduction system, M  
 955 \$  
 956  $i$  = composite of syngas ( $i = \text{CH}_4, \text{H}_2\text{O}, \text{CO}, \text{CO}_2$ )  
 957  $I_{\text{total}}$  = total investment, M\$  
 958  $I_{\text{product}}$  = investment for production, M\$  
 959  $I_{\text{static}}$  = static investment, M\$  
 960  $I_{\text{annual}}$  = annual investment, M\$  
 961  $I_{\text{material}}$  = material charge, M\$  
 962  $I_{\text{maintenance}}$  = maintenance investment, M\$  
 963  $I_{\text{oc}}$  = overnight cost, M\$  
 964  $I_{\text{ci}}$  = construction interest, M\$  
 965  $I_j^{\text{ref}}$  = reference investment of unit process  $j$ , M\$  
 966  $I^{\text{ref}}$  = reference investment, M\$  
 967  $I^{\text{co}}$  = investment for the coproduction system, M\$  
 968  $k_{\text{WGS}}$  = equilibrium constant  
 969  $k_1, k_2$  = reaction rate constants,  $\text{kmol}/\text{m}^3 \cdot \text{s} \cdot \text{pa}$   
 970  $K_i$  = absorption equilibrium constant  
 971  $K_{\text{MS}}, K_{\text{WGS}}$  = equilibrium constant of reactions in MS and  
 972 WGS  
 973  $n$  = lifetime of a system, years  
 974  $N$  = number of product in the coproduction system  
 975  $M$  = number of unit process in a system  
 976  $r$  = reaction rate,  $\text{kmol}/\text{kg} \cdot \text{cat} \cdot \text{s}$   
 977  $r_{\text{discount}}$  = discount rate  
 978  $r_{\text{point1}}$  = ratio of syngas at point 1  
 979  $r_{\text{point2}}$  = ratio of syngas at point 2  
 980  $R$  = gas constant,  $\text{kJ}/\text{kmol} \cdot \text{K}$

$p_i$  = partial pressure of gaseous composite  $i$  981  
 $sf$  = scale factor 982  
 $y_i$  = mole fraction of gaseous composite  $i$  983  
**Greek Letters** 984  
 $\alpha, \beta, \gamma, \delta, \varepsilon$  = mass fractions of elements C, H, O, N, S in 985  
 coke 986  
 $\eta$  = exergy efficiency 987  
 $P$  = construction interest rate 988  
 $\theta$  = domestic made factor 989  
 $\psi$  = active factor 990

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