On the Thermal and Kinetic Performance of a Coal-CO_2 Slurry-fed Gasifier:
Optimization of CO_2 and H_2O flow using CO_2 skimming and steam injection

Cristina Botero*, Randall P. Field, Howard J. Herzog, Ahmed F. Ghoniem

Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA 02139

Abstract

Coal-CO_2 slurry feed provides significant thermal advantages when compared to conventional coal-water slurry for feeding pressurized entrained-flow gasifiers, assuming complete carbon conversion. However, substituting H_2O by CO_2 in the feeding system affects the heterogeneous gasification kinetics and could reduce carbon conversion. This work examines CO_2 slurry skimming, or flashing, in combination with steam injection, as a way to increase conversion by controlling and optimizing the flow of CO_2 and H_2O entering the reactor. We use multiscale computational tools developed at MIT to examine the thermal and kinetic performance of a gasifier with CO_2 and H_2O injection and quantify how it affects the overall plant economics. The gasifier is part of a plant producing clean syngas with a H_2:CO ratio of 2.0 after water-gas shift. The slurry feeding system uses the Phase Inversion-based Coal-CO_2 Slurry (PHICCOS) method to prepare coal-CO_2 slurry from bituminous coal and lignite. The results show that the minimum syngas production cost is \$132.9/kNm^3 for bituminous coal and \$128.6/kNm^3 for lignite when the gasifier operates with a fixed outlet temperature of 1,400℃ and 1,300℃, respectively. This economic optimum is achieved when CO_2 skimming to dry feed conditions is combined with the injection of 0.23 kg of steam per kg of coal (dry basis). Multivariable optimization is currently being conducted to include the effect of reactor temperature, among other important operating and design variables, into the optimization process. The effect of uncertain parameters on the process technoeconomics is also being studied.

Keywords: Coal, Gasification, Slurry, CO_2, Technoeconomics

1. Introduction

Coal-water slurry (CWS) is currently the least capital-intensive technology for feeding pulverized coal into a pressurized entrained-flow gasifier (EFG) [1]. Unlike lock-hopper-based dry feeding systems, CWS is attractive because of its simplicity and the high pressures that it can achieve. Nonetheless, heating up liquid water to the high temperature of >1300℃ at which EFGs typically operate requires large amounts of thermal energy. Slurry-fed EFGs are thus inefficient, when compared to dry feeding systems, which is especially problematic for low-rank coal. Besides increasing fuel consumption, this leads to high capital and operating costs in the air separation unit (ASU) required to supply O_2 for autothermal reactor operation.

Coal-CO_2 slurry is being studied as a more efficient alternative to CWS feed in plants with carbon capture [2–6]. Liquid CO_2 -or supercritical CO_2 with liquid-like density- is available in such plants and is especially appealing as a slurrying medium as a result of its lower enthalpy of vaporization, heat capacity, and viscosity, among others [4].

The thermodynamic benefits of CO_2 slurry feed are significant: a gasifier with this feeding system and complete carbon conversion is predicted to have an 11%-points (%-pt.) higher cold gas efficiency (CGE) than one with CWS feed, for lignite, and a 7%-pt. advantage for bituminous coal [4]. The CGE for the latter is 82%, on a higher heating value basis, and is hence in the same range as the 78-83% typical for commercial lock-hopper based dry-fed reactors [7].

*Corresponding author

Email address: cbotero@mit.edu (Cristina Botero)

Slurry preparation has proved to be a challenging step in a coal-CO_2 slurry-fed system. The Phase Inversion-based Coal-CO_2 Slurry (PHICCOS) preparation and feeding system has been proposed as a way to address this challenge [5, 8]. Unlike other slurry preparation methods being studied [2], the PHICCOS system operates at ambient temperature and without the use of lock hoppers.

Despite the thermodynamic appeal of liquid CO_2 in the feeding system, recent work has shown that injecting CO_2 instead of H_2O with the feed leads to the production of high concentrations of CO in the gasifier. This slows down the heterogeneous gasification kinetics directly, through inhibition of the steam and CO_2 gasification reactions, as well as indirectly, through a slower diffusion of reactants and products in the pores of the pulverized coal particles. Once this is accounted for, a 7%-pt. reduction in the carbon conversion is predicted in a gasifier operating with CO_2 slurry, relative to a reactor with CWS feed and the same volume and outlet temperature [6].

The conversion reduction caused by the presence of CO_2 has a significant impact on the gasifier performance. For example, for bituminous coal, once the conversion reduction is accounted for, the CGE of a gasifier with CO_2 slurry feed is the same as that of a CWS-fed gasifier, i.e. no performance advantage is predicted. The gasifier outlet temperature must be raised by over 100K to make up for the conversion reduction. This increases oxygen consumption by 10-20%, further contributing to the already high capital and operating costs incurred in the ASU. The overall economic attractiveness of increasing the gasifier temperature as a means to raise conversion in a coal-CO_2 slurry-fed reactor is currently being studied.

As an alternative to, or in addition to, increasing the reactor temperature, the carbon conversion in a CO_2 slurry-fed gasifier could be increased by addressing the actual source of the slower gasification kinetics: the fraction of CO_2 in the gasification agent. This fraction can be reduced by either injecting less CO_2 or more H_2O into the reactor, both of which can be practically implemented.

The flow of CO_2 entering the gasifier can be reduced by flashing -or evaporating- CO_2 from the pressurized coal-CO_2 slurry. This process, which has been suggested in the past and is also known as CO_2 skimming [9], requires only a small amount of thermal energy, if any, due to the proximity of CO_2 in the slurry to its saturation line. Furthermore, the flow of H_2O in the feed can be increased by injecting steam to the gasifier in a way similar to how dry-fed gasifiers operate.

This work uses multiscale analysis to evaluate the technoeconomics of CO_2 skimming and steam injection as a way to control the fraction of CO_2 and H_2O in the feed of a PHICCOS-fed gasifier. The thermal benefits of CO_2 are combined with the kinetic benefits of H_2O in order to optimize plant economics. The final product is clean syngas with a H_2:CO ratio of 2.0 after water-gas shift, which can be used for the production of synthetic liquid fuels. The only application-specific component of this analysis is the H_2:CO ratio of the syngas, which can be adjusted without major hurdles in the shift reactor. Hence, the results of this study are applicable to any syngas application, including the production of synthetic fuels and chemicals, as well as power generation in an Integrated Gasification Combined Cycle (IGCC) power plant with carbon capture.

This paper begins by discussing the motivation for studying gasification in an environment of mixed CO_2 and steam. A PHICCOS-fed gasifier with CO_2 skimming and steam injection is introduced as an attractive platform to do this. The modeling methodology and tools used for the analysis are then presented. This is followed by a discussion of the main findings, which show that an optimum flow of CO_2 and H_2O exists, which is a tradeoff between kinetics, thermodynamics, and costs and leads to the most favorable plant economics. The work concludes with an insight into the main challenges associated with the proposed feeding system and an overview of ongoing and future work.

2. Gasification in a Mixed Environment of H_2O and CO_2

The motivation behind combining CO_2 and steam injection in an EFG can be better understood by considering the chemical and thermal processes taking place inside an autothermal reactor and how these affect its performance.

2.1. Thermochemistry in an autothermal entrained-flow gasifier

The main gasification reactions occurring in an EFG are the steam-gasification and CO_2-gasification (Boudouard) reactions. These yield a mixture of H_2 and CO known as synthesis gas, or *syngas*, and are given by

$$C_{(s)} + H_2O \rightleftharpoons CO + H_2 \qquad +131 \text{ MJ/kmol} \tag{1}$$
$$C_{(s)} + CO_2 \rightleftharpoons 2CO \qquad +172 \text{ MJ/kmol} \tag{2}$$

respectively [10]. The heats of reaction are given above and the positive sign indicates that the reactions are endothermic.

Oxygen is required in the gasifier for oxidation reactions such as [10]

$$C_{(s)} + \tfrac{1}{2}O_2 \rightarrow CO \qquad -111 \text{ MJ/kmol} \tag{3}$$
$$CO + \tfrac{1}{2}O_2 \rightarrow CO_2 \qquad -283 \text{ MJ/kmol} \tag{4}$$
$$H_2 + \tfrac{1}{2}O_2 \rightarrow H_2O \qquad -242 \text{ MJ/kmol.} \tag{5}$$

These exothermic reactions enable the reactor to operate at autothermal conditions by providing the energy necessary a) for the endothermic pyrolysis and gasification reactions, b) to heat up the reactants, and c) to make up for any heat losses to the environment. This is schematically illustratred in Figure 1.

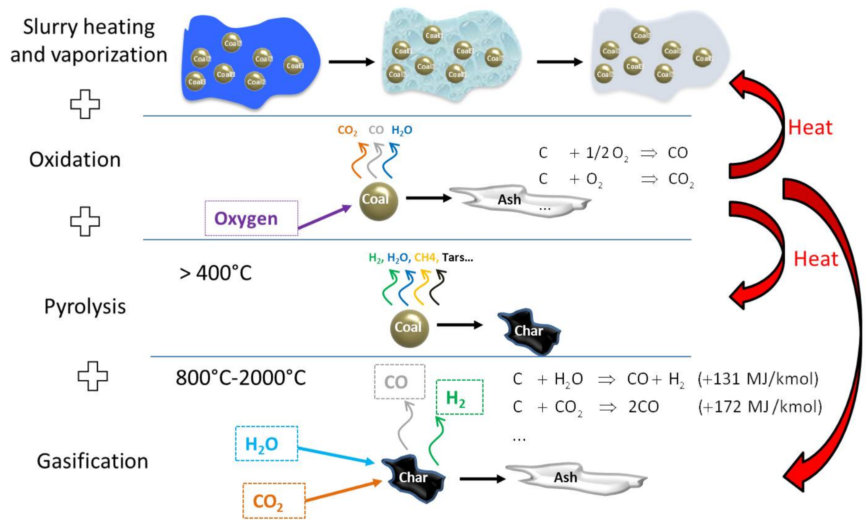

Figure 1: Schematic of autothermal operation of an oxygen-blown, entrained flow coal gasifier.

2.2. Quantifying gasifier performance

The cold gas efficiency is the most commonly used measure of gasifier performance. It is defined as the fraction of the feedstock's chemical energy that is recovered in the cooled gaseous product. It is calculated from the heating value and mass flow ($\dot{m}$) of the gasifier feed and product gas streams, according to:

$$CGE = \frac{(\dot{m}_{\text{gas}})\,(HHV_{\text{gas}})}{(\dot{m}_{\text{feed}})\,(HHV_{\text{feed}})}, \tag{6}$$

where a higher heating value (HHV) basis has been used.

The CGE has a thermal and a kinetic component. The thermal component is an indication of how energy-intensive the reactor is, i.e. how much feedstock must be oxidized -rather than gasified- in order to maintain autothermal operation. In eq. (6), the thermal performance is contained in the heating value of the gas, since oxidation products have a negligible heating value.

Feedstock heating is a major loss of thermal performance in an EFG. This is especially problematic for gasifiers with CWS feed due to the high heat capacity and vaporization enthalpy of H_2O. The potential for better thermal performance in a reactor with coal-CO_2 slurry feed has been the main motivation for conducting research in this field [4]. Beyond making the gasifier more efficient, a good thermal performance implies that less oxygen is consumed for reactions (3)-(5), reducing the high capital and operating costs of the air separation unit.

The kinetic gasifier performance is a measure of how fast the chemical reactions are. It can be quantified through the fraction of carbon that was converted to gas, also known as carbon conversion, and is contained in $\dot{m}_{\text{gas}}$ in eq. (6).

Note that just like the HHV of the produced gas is not a direct indication of carbon conversion, the latter says nothing about the characteristics of the product: a carbon conversion of 100% could mean that the entire feedstock has been oxidized to CO_2, producing a gas with no heating value. Hence, neither the thermal performance alone nor the kinetic performance alone is sufficient to characterize gasifier operation. The CGE includes both components and is hence a much more attractive performance measure than carbon conversion or syngas heating value alone.

2.3. Optimizing the fraction of H_2O and CO_2 in the feed

In view of the thermal advantages of CO_2 as slurrying medium and of the kinetic advantages of H_2O as a gasification agent it is desirable to combine these two in order to optimize gasifier performance and economics. This is schematically illustrated in Figure 2, which shows qualitative gasifier performance trends based on preliminary calculations. For a given total flow of gasification agent (CO_2+H_2O), extreme cases of gasification only with steam and only with CO_2 are shown, as well as intermediate cases in which both steam and CO_2 are injected.

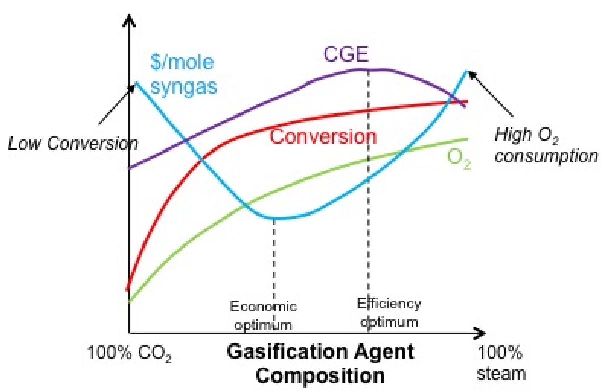

Figure 2: Qualitative performance and cost trends as a function of the gasification agent composition

The figure shows that a gasifier with CO_2 as the sole gasification agent has low O_2 consumption but also low carbon conversion. On the other hand, a steam-injected gasifier with no CO_2 injection converts more of the coal's carbon content to gas but at the cost of a higher oxygen demand. Overall, the gasifier performance can be optimized by combining steam injection and CO_2 injection to achieve the best tradeoff between oxygen consumption and conversion, i.e. a maximum gasifier cold gas efficiency. This efficiency optimum may or may not coincide with the economic optimum. Given the high cost of producing pure O_2, the latter is likely to lie to the left of the performance optimum, as the figure shows, where the CGE is lower but so is the oxygen consumption.

2.4. PHICCOS feed with CO_2 skimming and steam injection

The injection of CO_2 and steam into a pressurized EFG can be implemented regardless of the characteristics of the reactor and feeding system. Nonetheless, CO_2 slurry-fed reactors are an especially appealing platform: CO_2 is inherently contained in the feed and can be combined with steam injection to operate the gasifier at the optimum conditions in Figure 2.

The PHICCOS feeding system has been proposed as a way to feed coal to a high-pressure entrained flow gasifier by using liquid CO_2 instead of water. Unlike other proposed coal-CO_2 slurry feeding systems, PHICCOS has the unique advantage that the slurry is prepared at ambient temperature and without the use of lock hoppers. This can be achieved by using CWS as an intermediate and by taking advantage of the surface properties of coal, as it has been described in more detail elsewhere [5, 8].

The concept behind a PHICCOS-based EFG with CO_2 skimming and steam injection is depicted in Figure 3 for a gasifier operating at 55 bar.

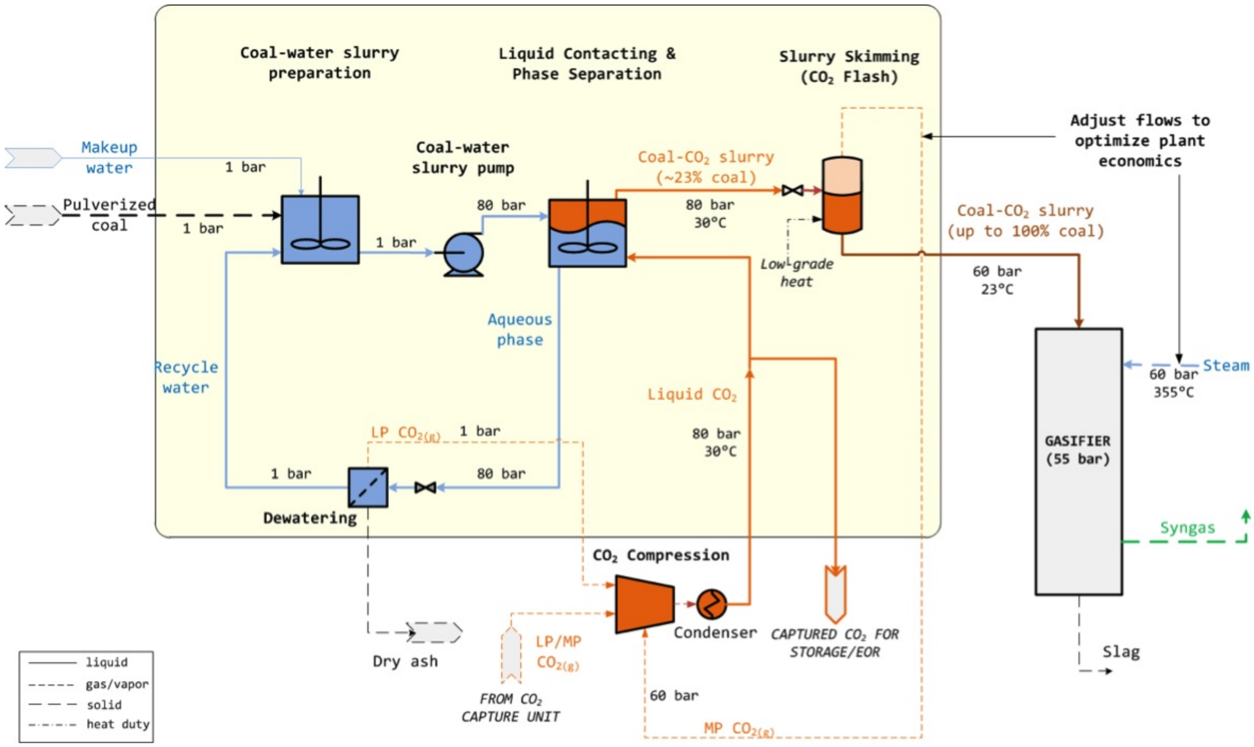

Figure 3: PHICCOS-fed gasifier with CO_2 slurry skimming and steam injection.

CWS is first prepared at ambient pressure and pumped to a pressure of 80 bar. It is subsequently mixed with liquid CO_2 at the same pressure and near ambient temperature. Due to its mostly hydrophobic surface, coal particles accumulate in the lighter CO_2 phase while most of the coal moisture and hydrophylic matter remains in the aqueous phase. This phenomenon, known as phase inversion, produces two distinct phases: a coal-rich CO_2 slurry phase and a mineral-rich aqueous phase [11–13]. The feedstock in the CO_2 phase has been experimentally observed to have a low moisture and ash content [11]. This *upgrading* effect is thought to be among the most attractive features of the PHICCOS feeding system, especially for low-rank coal [5].

The coal-CO_2 slurry produced through the PHICCOS process has a low coal loading of close to 20% [11]. Nonetheless, the amount of CO_2 injected into the gasifier can be reduced as much as required by skimming, or flashing, the CO_2 out of the pressurized slurry before it is injected to the reactor, see Figure 3.

For a gasifier operating at 55 bar, like the one shown in the figure, this is done by reducing the pressure of the dilute coal-CO_2 slurry from 80 bar to 60 bar in a flash unit, where it enters the liquid-vapor two-phase region. Due to its proximity to the critical point, where the saturated liquid and saturated vapor line meet, the amount of heat required to evaporate CO_2 in the flash unit is very low at 120 J/kg CO_2. Furthermore, low-grade heat is sufficient, since the saturation temperature of CO_2 at 60 bar is only 23°C.

The gasifier operation can be optimized by combining CO_2 skimming with steam injection. The latter is common practice in dry-fed gasifiers like Shell's, which operate with a steam to dry coal ratio of about 0.11, by weight (wt.). [1]. CWS-fed reactors, on the other hand, do not have the possibility of optimizing the gasification agent flow, since the amount of H_2O in the slurry is dictated by the slurry rheology. Evaporating excess water from the slurry would be prohibitively expensive, given the thermophysical properties of water.

3. Methodology

Multiscale analysis is used in this work to study the technoeconomics of a syngas production plant based on entrained-flow gasification of bituminous coal and lignite, see composition in Table A.4. The reactor considered operates with PHICCOS feed, CO_2 skimming, and steam injection. The degree of CO_2 skimming and steam injection are optimized to yield the minimum syngas production costs.

The scope of the plant considered is schematically illustrated in Figure 4. The plant produces clean syngas, made primarily of H_2 and CO, and includes the coal handling and feeding system, the gasifier, and the main syngas processing units.

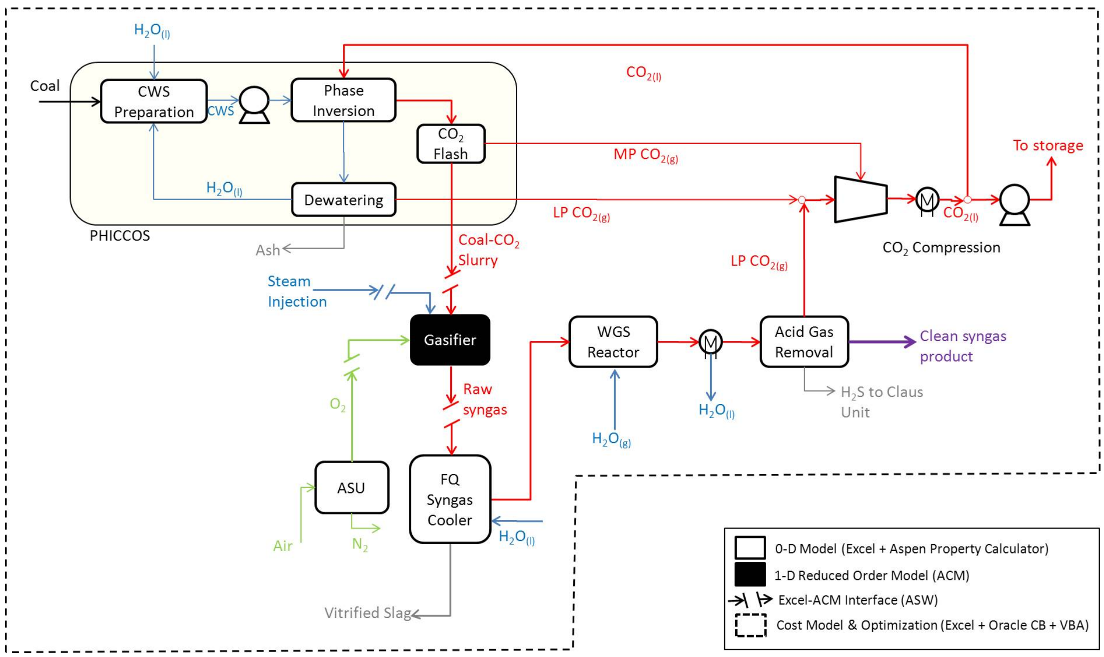

Figure 4: Scope of syngas production plant considered and summary of tools used for the analysis. Coal preparation and handling, ash handling, and Claus unit were not modeled but are included in the cost model.

Coal-CO_2 slurry is prepared at 80 bar with the PHICCOS system and concentrated in a flash unit before being introduced into a steam-injected gasifier operating at 55 bar. The raw syngas produced in the gasifier is saturated with water in a full-quench (FQ) cooler, leaving at a temperature of 225°C. Its H_2:CO ratio is then adjusted in a water-gas shift (WGS) reactor. The target H_2:CO ratio for the shifted syngas depends on the application. A ratio of 2.0 was assumed here, which is close to that required for the production of synthetic liquid fuels.

The shifted syngas is brought to 40°C and hereby freed of the majority of its H_2O content, which leaves the cooler as condensate. The cool syngas enters an acid gas removal (AGR) unit, where the majority of its CO_2 and H_2S content is separated. The clean, pressurized syngas leaving the AGR is the final product. It has a pressure of about 50 bar, consists of mainly CO and H_2 and, with minor modification in the WGS reactor operation, can be used for any application requiring syngas, e.g. a Fischer-Tropsh process for synthetic liquid fuel production, an IGCC plant, methanol synthesis, etc.

The modeling approach and tools used are also indicated in Figure 4. Except for the gasifier, all process units are modeled as 0-D components using Microsoft Excel [14] and Aspen Properties Excel Calculator [15]. For the gasifier, a detailed, 1-D reduced order model is used, which includes a high-pressure chemical kinetics submodel and is capable of predicting carbon conversion. The gasifier ROM is implemented in Aspen Custom Modeler (ACM) [16] and is linked to Excel through Aspen Simulation Workbook (ASW) [17].

The economics of the plant are assessed with a cost model, also in Excel, that uses the performance and equipment size from the simulation results as an input. The economic figure of merit is the syngas production cost per mole of exported syngas.

The cost model is used to find the optimum flow and composition of the gasification agent fed to the reactor, i.e. that leading to a minimum syngas production cost. The optimizer and tools in the Excel-based software Crystal Ball (CB) [18] are used for this purpose. The latter is linked to ASW through Visual Basic for Applications (VBA) [19].

3.1. System-level model of plant

The steady-state, system-level plant model is a collection of Excel-based mass and energy balances for the feeding system and for all the main syngas processing units (gas cooling, WGS, AGR), except for the gasifier. These 0-D models are used to estimate stream conditions and equipment size from within Excel.

The 0-D models in Excel are surrogates to a more detailed Aspen Plus representation of the corresponding process units described elsewhere [4, 20]. The main plant characteristics and modeling assumptions used are listed in Table 1. The simplicity of the surrogate models used here and their ability to run from within Excel make these more appropriate when a large number of simulations is required. This is the case for the present work, where sensitivity studies and optimization are conducted.

Table 1: Key assumptions for system-level model

Feedstock	
Coal type	Illinois # 6 Bituminous North Dakota Lignite
Coal composition	as-received (see Table A.4)
Feeding system	PHICCOS [4] with CO_2 flash
Gasifier	
Thermal input	1,700 MJ/s (2 x 850 MJ/s)
Oxygen supply	ASU: 1,370 kJ_e/kg [4, 20]
Syngas cooling	Full-quench to 224°C
Gas Conditioning	
Water-gas shift reactor	
Temperature	224°C
Steam addition	30% excess
Shifted syngas composition	H_2:CO = 2:1 (molar)
Acid gas removal	2-stage Selexol: 167 kJ_e/kg CO_2 [1]
Carbon capture	98% (local)
Auxiliary power consumption [1]	
CO_2 compression	
CO_2 captured in AGR	896 kJ_e/kg CO_2
MP CO_2 from PHICCOS	173 kJ_e/kg CO_2
LP CO_2 from PHICCOS	1,517 kJ_e/kg CO_2
Coal handling, BOP, and others	144 kJ_e/kg coal (ar)

The main data used for modeling the performance of the PHICCOS feeding system is listed Table A.5 in the Appendix. It is based on experimental observations of phase inversion of bituminous coal with CO_2 [11]. Due to the upgrading effect of the PHICCOS feeding system, the gasifier feedstock is expected to have a low ash and moisture content for both bituminous coal and lignite [5]. For both coals, an ash content of 10 wt.-% (dry basis) and a moisture content of 10 wt.-% were assumed. The coal lost to the aqueous phase during phase inversion and phase separation is assumed to account for 5% of the as-received coal enthalpy.

The solid and liquid phases of the PHICCOS feeding system are treated separately in the system-level model. The solid phase is modeled by separating ash, moisture, and coal from the feedstock to comply with the performance characteristics in Table A.5. The liquid phase flows are estimated by accounting for the required slurry loading and the solubility of CO_2 in H_2O at the conditions of interest.

The syngas is assumed to be composed only of H_2, CO, CO_2, and H_2O and its flow and composition are imported from the gasifier ROM. Syngas full-quench is represented as an adiabatic saturation process and the WGS reactor modeled by conducting equilibrium calculations at a constant temperature. The acid gas removal model is a black box separating 98% of the CO_2 content from the sour syngas.

3.2. Reduced order model of the gasifier

A detailed 1-D, steady-state reduced order model (ROM) of a high-pressure EFG was used to simulate the gasifier operation and export the syngas conditions and oxygen requirement to the 0-D system model. The model was developed by Monaghan and Ghoniem and uses a network of idealized reactors to represent fluid mixing and recirculation inside the gasifier. The heterogeneous gasification kinetics are modeled using a Langmuir-Hinshelwood expression based on experimental measurements at high pressure [21]. A relative reactivity factor (ψ) is used to account for reactivity differences between coals of different ranks. Internal mass transfer limitations at high temperatures are accounted for through the effectiveness factor approach. The ROM [22–25] and its high-pressure kinetic submodel [6] have been described in detail elsewhere.

The main characteristics of the gasifier studied are summarized in Table 2. It resembles General Electric (GE) design and its dimensions [26] are similar to that of the commercial-scale (1,800 ft^3) GE gasifier operating at the Tampa Polk Power Station [27]. Unlike a GE gasifier, however, the reactor is steam-injected and is not fed with CWS but with coal-CO_2 slurry via the PHICCOS feeding system. Furthermore, the gasifier can be operated in dry feed mode if the entire CO_2 content of the slurry is flashed out before injection.

Table 2: Key data for gasifier ROM

Type	Single-stage, entrained flow
Feeding System	PHICCOS with CO_2 skimming
Volume	2 x 51 m^3
Diameter	3.0 m
Gasification agent	Steam and/or CO_2
Pressure	55 bar
Relative reactivity factor (ψ)	8.3 (Bituminous coal)
	9.8 (Lignite)
Outlet temperature	1,400℃ (Bituminous coal)
	1,300℃ (Lignite)

The relative reactivity factor of bituminous coal in Table 2 was estimated from conversion data from the GE gasifier used in the Cool Water IGCC Demonstration Project for a similar coal [6, 28]. The outlet temperature of 1,400℃ is that required to achieve a carbon conversion of 98% in the gasifier, when fed with conventional CWS. This is the conversion reported for a CWS-fed GE gasifier of the same size as that considered here and processing Illinois # 6 coal at 55 bar [1].

The reactivity factor reported for lignite was estimated from its fixed carbon content based on a correlation by Botero et al. [6]. The gasifier outlet temperature of 1,300℃ is that at which a GE-type gasifier with lignite-water slurry feed would operate in order to achieve full conversion. This is the minimum temperature, below which the CH_4 content of the syngas becomes excessively high for most applications [10].

3.3. Economic model

The figure of merit used as here to quantify plant performance is the cost of producing one unit of clean syngas. The syngas consists of H_2 and CO at a molar ratio of 2:1.

An economic model was developed in Excel to estimate the syngas production cost. The model is based on the standard methodology used by National Energy Technology Laboratory (NETL) for the comparative assessment of power plant performance [29], which was modified for syngas production applications, where appropriate.

The total overnight capital cost (TOC) of the plant is first estimated and then annualized by using a capital charge factor. The latter depends on the applicable finance structure of the plant and on the capital expenditure period and is reported in Table A.6 in the Appendix, together with other economic assumptions. A plant capacity factor of 90% was assumed, which is greater than the 80% typical of IGCC plants [1], but less than the 95% of a syngas production plant based on a fluidized-bed gasifier [30].

The bare erected costs (BEC) and process contingencies (PRC) for the main, state-of-the-art (SOA) equipment in the syngas production plant are based on NETL's estimates for similar equipment of comparable size in an IGCC plant [1, 31]. Equipment within the PHICCOS feeding system was costed based on Westinghouse's economic assessment of a commercial-scale coal-CO_2 phase inversion unit [5, 11]. A high process contingency of 40% was used for the PHICCOS units given the early stage of development of this technology.

Balance of plant (BOP) equipment, cooling water system, accessory electric plant, instrumentation and control, improvements to sites, and buildings and structures were not taken from NETL's estimates for IGCC plant data but were estimated as a percentage of the total Bare Erected Cost (BEC). The percentages were taken from a similar syngas production plant in Case 1A of NETL's comprehensive study of syngas production technologies from fluidized-bed gasifiers [30].

The latter source was also used for estimating operating and maintenance (O&M) costs in the plant, excluding electricity as well as oxygen and nitrogen purchase costs. These gases are produced in the plant's ASU.

Steam and electricity are assumed to be imported rather than produced locally. Process integration is not considered. The fuel value of steam was used to calculate the cost of steam in Table A.6, assuming a boiler efficiency of 90% [32].

Finally, the Chemical Engineering Plant Cost Index (CEPCI) [33] was used to convert all equipment costs to 2011 U.S. dollars, where necessary. Furthermore, the *six-tenths factor rule* [34] was used to scale all reference BECs for the individual pieces of equipment to the capacity predicted by the simulations.

3.4. Cases studied

The effectiveness of steam injection and CO_2 skimming are assessed here as a means to overcome the low conversion in a slurry-fed gasifier with PHICCOS feed, when compared to one with CWS feed. Both bituminous coal and lignite were studied in order to cover a broad range of coal ranks.

A gasifier with CWS feed was used as a reference to set the gasifier temperature at a fixed value. The latter was left unchanged during the analysis in order to isolate the effects of CO_2 and steam injection. Hence, the gasifier operates at a constant outlet temperature of 1,400℃ for bituminous coal feed and 1,300℃ for lignite. The effect of temperature on conversion in a gasifier with coal-CO_2 slurry feed has been reported elsewhere [6] and is currently being studied when in conjunction with CO_2 skimming and steam injection.

The moisture and ash content of the feedstock delivered by the PHICCOS feeding system to the gasifier is expected to be the same for both coals, see Table A.5. The main difference between the two feedstocks is thus in their fixed carbon and oxygen content, as well as in the temperature at which they are gasified.

4. Results and Discussion

This section presents and discusses the effect of CO_2 slurry skimming (i.e. of different slurry loadings) and of H_2O injection on the performance and economics of a syngas production plant with a PHICCOS-based gasifier operating on bituminous coal or lignite. The gasifier outlet temperature is constant and corresponds to that of a CWS-fed gasifier with the same feedstock.

4.1. Gasifier efficiency optimum

The specific oxygen consumption and carbon conversion in the gasifier are presented in Figure 5 as a function of the coal-CO_2 slurry loading and steam injection ratio. The asymptotic behavior of the conversion surface in the figure reflects the Langmuir-Hinshelwood kinetics underlying the gasifier model. These account for the effect of product inhibition at high conversions [6].

The results in the figure show that the conversion and oxygen consumption in a reactor with bituminous coal and with lignite follow similar trends. Due to its higher reactivity, a gasifier operating on lignite and an outlet temperature of 1,300℃ has a comparable -only slightly lower- carbon conversion than a bituminous coal-fed reactor operating with an outlet temperature of 1,400℃. For the conditions studied here, carbon conversion is incomplete for both feedstocks, and is in the range of 82%-96% for bituminous coal and 84%-93% for lignite.

Even though its conversion is comparable, the specific oxygen consumption in a gasifier operating on lignite is about 15% lower than for one with bituminous coal. Despite its higher moisture content, gasification of as-received (ar) low-rank coal is known to consume only slightly more oxygen than gasification of high-rank coal [35]. This is a result of the high oxygen content of this feedstock, see Table A.4. For the same moisture and ash content, hence, the specific oxygen consumption of lignite is expected to be lower. This is the case in a PHICCOS-fed gasifier, where both coals are likely to have a comparable content of ash and moisture due to the upgrading effect of the feeding system [5].

Figure 5 shows that steam injection, which increases the reactant partial pressure in the gasifier, is an effective means of increasing carbon conversion. This is particularly true at low conversions, where the reaction rate is limited by the low reactant partial pressure. At higher conversions, the role of product inhibition gains increasing importance and the kinetic benefits of steam injection are modest, as shown by the curvature of the conversion surfaces in the figure.

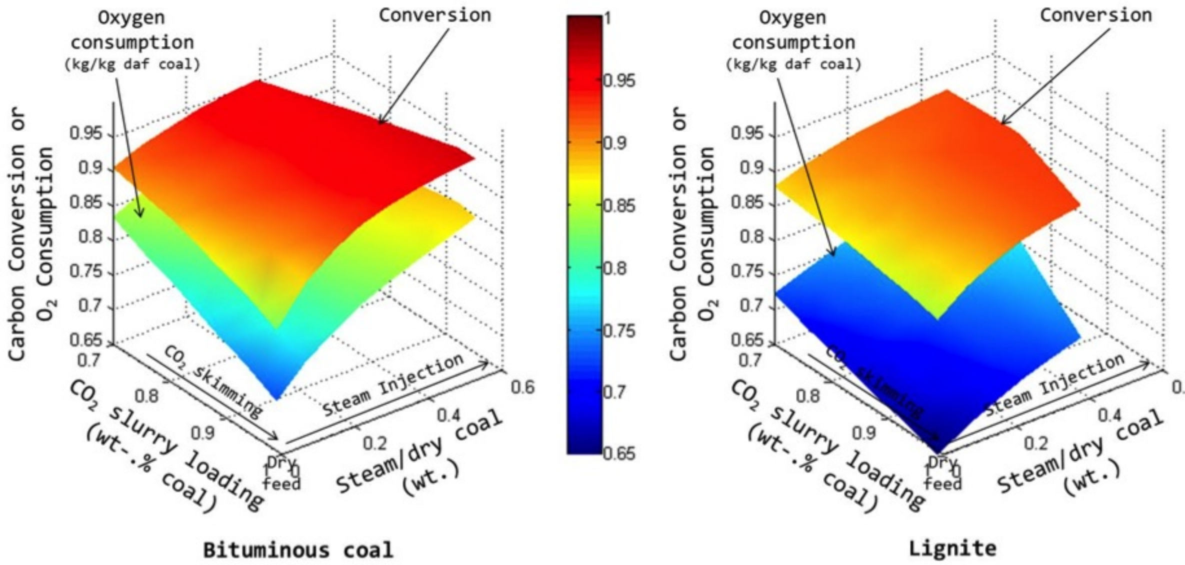

Figure 5: Specific oxygen consumption (bottom surface) and carbon conversion (top surface) for a PHICCOS-fed gasifier operating with bituminous coal (left, 1,400℃) or lignite (right, 1,300℃).

While beneficial for the gasifier kinetic performance, steam injection is detrimental to its thermal performance. Oxygen consumption increases when steam is injected, as seen in the bottom surfaces in Figure 5: additional oxygen is required not only to sustain the endothermic gasification of additional carbon, but also to heat up the injected steam.

Skimming the CO_2 from the slurry to achieve higher loadings is always beneficial for the thermal performance of the gasifier, as the bottom surfaces in Figure 5 shows: O_2 consumption drops since CO_2 skimming reduces the thermal load of the slurrying medium in the reactor. At low steam injection ratios, however, increasing the slurry loading also reduces conversion: once again, under these conditions the conversion is limited by the low reactant partial pressure.

The overall tradeoff between the kinetic and thermal gasifier performance at different levels of steam injection and CO_2 skimming can be observed in Figure 6, which shows the gasifier cold gas efficiency.

The results show that CO_2 skimming to loadings above an average of 80% is detrimental to the gasifier efficiency when no steam is injected into the reactor. Under these conditions, the reactant partial pressure is very low so the oxygen savings arising from CO_2 skimming are over-weighed by the conversion reduction it causes, leading to a CGE decrease. As more steam is injected into the gasifier, however, the reactant partial pressure becomes high enough that the the thermal load of the injected CO_2 dominates. The latter is significantly less than that of steam, but must still be accounted for. As the figure shows, starting at a steam injection ratio of about 0.1, CO_2 skimming up to dry feed conditions (100% loading) always yields the highest cold gas efficiency for a given steam/coal ratio.

The curvature of the surfaces in Figure 6 shows that for any given slurry loading, an optimum steam injection ratio exists, i.e. that leading to a maximum CGE. It represents the best tradeoff between the kinetic benefits of steam and the thermal load it represents. The optimum steam/coal ratio increases with slurry loading: steam injection is especially beneficial when little CO_2 enters with the feed, in which case the gasification kinetics are limited by the low partial pressure of the gasification agents.

The maximum CGE of a PHICCOS-fed gasifier operating with bituminous coal and an outlet temperature of 1,400℃ is 78.5%. For lignite, the maximum CGE at 1,300℃ is 75.9%. For both feedstocks, the efficiency optimum is achieved when CO_2 is entirely skimmed out of the slurry (i.e. for 100% coal loading, dry feed conditions) and when steam is injected at a ratio of 0.4 kg/kg daf coal.

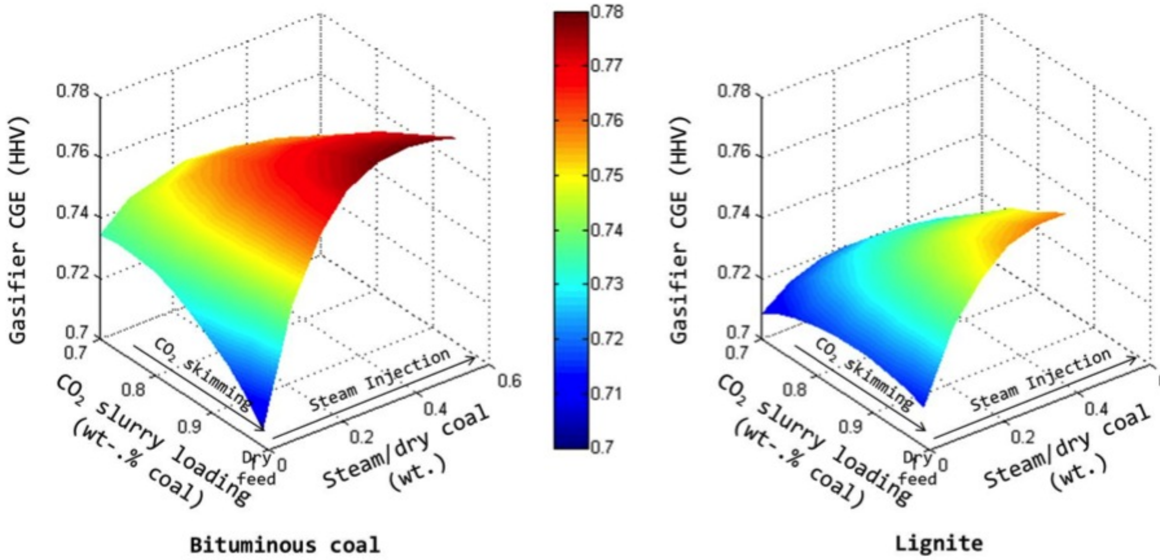

Figure 6: Gasifier cold gas efficiency as a function of coal-CO_2 slurry loading and steam/coal ratio for a gasifier operating with bituminous coal (left, 1,400℃) and lignite (right, 1,300℃).

4.2. Plant economic optimum

The total cost of producing 1,000 normal cubic meters (kNm^3) of clean syngas with a H_2:CO ratio of 2:1 is presented in Figure 7 as a function of the coal-CO_2 slurry loading and of the steam injection ratio.

The minimum syngas production cost in a PHICCOS-fed gasifier operating with bituminous coal and an outlet temperature of 1,400℃ is \$132.9/kNm^3. For a gasifier operating with lignite and an outlet temperature of 1,300℃, the minimum cost is \$128.6/kNm^3. For both feedstocks, the plant economic optimum is achieved when CO_2 is completely flashed out of the slurry, in combination with steam injection at a ratio of 0.23 kg/kg ar coal.

The economic optimum is located at a lower steam injection ratio than the performance optimum. This is predominantly due to the cost of producing additional oxygen when steam is added. Oxygen production consumes a large amount of electricity and, most importantly, is associated with very high capital costs. The latter is the dominating factor in the economics of a syngas production plant [36].

The performance and economics of the PHICCOS-fed syngas production plant at the economic optimum are summarized in Table 3. The calculated syngas production cost of \$12-13/GJ is lower than the \$15-19/GJ (2010 cost basis) reported in the literature for a plant based on a fluidized bed gasifier [36]. This comparison should be treated with caution, however, since the estimates not only contain uncertain parameters, whose effect is being studied, but they also originate from different cost sources. An estimate of the syngas production cost with commercial technologies is currently being carried out with the same cost model as that used for a PHICCOS-fed plant.

Furthermore, only the CO_2 slurry loading and steam injection ratio were optimized here. In order to study the effect of these two variables in isolation, the gasifier outlet temperature was maintained at a fixed value for each feedstock. The influence of the gasification temperature alone has also been studied before and is known to have a significant effect on plant performance and economics [6]. Operating the lignite-fed gasifier at 1,400℃ instead of 1,300℃, for example, reduces the minimum syngas production cost in the PHICCOS-fed plant to \$124.5/kNm^3. Multivariable optimization -including temperature- is the subject of current work.

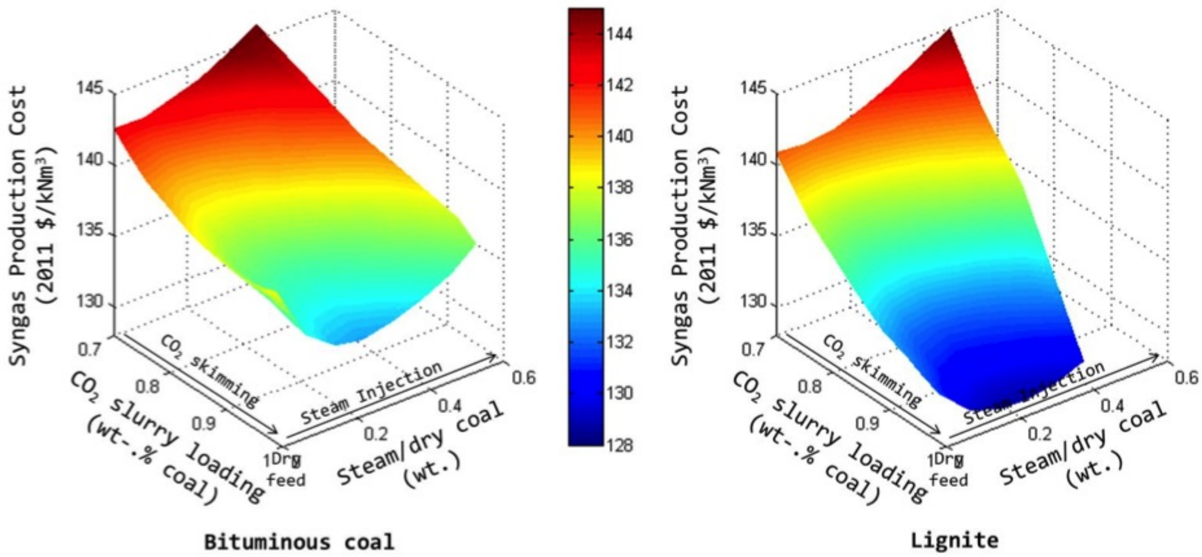

Figure 7: Syngas production cost as a function of coal-CO_2 slurry loading and steam/coal ratio for a gasifier operating with bituminous coal (left, 1,400℃) and lignite (right, 1,300℃).

Table 3: Plant performance and economics at economic optimum for a syngas production plant with a gasifier operating at 55 bar and an outlet temperature of 1,400℃ for bituminous coal and 1,300℃ for lignite. The product is clean syngas with a molar H_2:CO ratio of 2.0. The gasifier temperature has not been optimized.

		Bituminous	Lignite
Feed			
Coal feed (ar)	kg/h	238,783	402,705
CO_2 slurry loading	wt.% coal	100%	100%
Steam injection	kg/kg dry coal	0.23	0.23
O_2 consumption	kg/kg daf coal	0.81	0.71
Gasifier Performance			
Carbon conversion		93%	91%
Cold gas efficiency	(HHV)	77.4%	75.5%
Plant Performance			
Syngas flow	kN^3/h	377,516	367,466
Net thermal efficiency	(HHV)	62.5%	59.1%
Syngas production cost	**\$/kN^3**	**\$ 132.9**	**\$128.6**
	(\$/GJ)	(\$ 12.4)	(\$ 12.0)

5. Conclusions and Outlook

The Phase Inversion-based Coal-CO_2 Slurry (PHICCOS) feeding system has been suggested as a more efficient alternative to conventional coal-water slurry feed in gasification-based plants with carbon capture. While coal-CO_2 slurry has significant thermal advantages, a lower carbon conversion is expected in an environment with a high ratio of CO_2:H_2O in the feed.

Multiscale analysis was used to study CO_2 skimming and steam injection as a way to control the flow of CO_2 and H_2O injected into a PHICCOS-fed gasifier and improve carbon conversion. Both bituminous coal and lignite feedstocks were considered within the scope of a gasification-based plant producing clean syngas with a H_2:CO ratio of 2.0.

The optimum gasifier efficiency was found to be a tradeoff between the gasification kinetics and the thermal load associated with the injection of the gasification agents H_2O and CO_2. A maximum gasifier cold gas efficiency is achieved when the CO_2 content of the slurry is flashed completely to yield a dry feed, in combination with steam injection at a ratio of 0.4 kg per kg of coal (dry basis).

For a fixed gasifier temperature of 1,400℃ for bituminous coal and 1,300℃ for lignite, the minimum syngas production cost in the plant is \$132.9/kNm^3 and \$128.6/kNm^3, respectively. The plant economic optimum is achieved at a steam injection ratio of 0.23 kg per kg of dry coal.

The optima unveiled in this study do not consider the influence of gasification temperature, which has been studied in isolation before and is known to have a significant effect on plant technoeconomics. Future work will conduct multivariable optimization in order to include this in the analysis, together with the effect of other important operating and design variables and of their uncertainty. Furthermore, a comparison with commerical syngas production technologies will be carried out and the potential practical challenges associated with the transport of a nearly dry solid from the CO_2 skimming vessel to the gasifier of a PHICCOS-fed system with full skimming will be addressed.

Acknowledgments

The authors would like to acknowledge BP for the financial support and Aspen Technology, Inc. for the simulation software.

References

[1] National Energy Technology Laboratory , Cost and performance baseline for fossil energy plants, revision 2, Technical Report DOE/NETL-2010/1397 (2010).

[2] Dooher J., Marasigan J., Goldstein H. N., Liquid CO_2 slurry (LCO2) for feeding low rank coal (LRC) to gasifiers, in: 37th International Technical Conference on Clean Coal and Fuel Systems, Clearwater, FL (USA), 2012.

[3] Dooher J. P., Castaldi M. J., Rubin D., Phillips J. N., Schoff R., Evaluation of low rank coal/liquid CO_2 slurries for generic, single-stage, slurry-fed gasifiers, in: 35th International Technical Conference of Coal Utilization and Fuel Systems, Clearwater, Florida (USA), 2010.

[4] Botero C., Field R. P., Herzog H., Ghoniem A. F., Performance of an IGCC plant with carbon capture and coal-CO_2-slurry feed: Impact of coal rank, slurry loading, and syngas cooling technology, Industrial & Engineering Chemistry Research 51 (36) (2012) 11778–11790.

[5] Botero C., Field R., Herzog H., Ghoniem A., Coal-CO_2 slurry feed for pressurized gasifiers: Slurry preparation system characterization and economics, in: Greenhouse Gas Technologies Conference (GHGT-11), Kyoto, Japan, 2012.

[6] Botero C., Field R. P., Herzog H. J., Ghoniem A. F., Impact of finite-rate kinetics on carbon conversion in a high-pressure, single-stage entrained flow gasifier with coal-CO_2 slurry feed, Applied Energy 104 (0) (2013) 408–417.

[7] Black & Veatch , Holcomb generation expansion project: Coal technology selection study, Technical Report 144102 (2006).

[8] Botero C., Field R., Herzog H., Ghoniem A., Method for preparing a slurry of pulverized solid material in liquid or supercritical carbon dioxide, U.S. Patent Application No.: 61/712954 (2012).

[9] McNamee G. P., White G. A., Use of lignite in texaco gasification-based-combined-cycle power plants, Technical Report AP-4509, Prepared by Energy Conversion Systems, Inc. for Electric Power Research Institute (1986).

[10] Higman C., van der Burgt M., Gasification, 2nd Edition, Elsevier, 2008.

[11] Westinghouse Electric Corporation , Development of the LICADO coal cleaning process, Technical Report DOE/PC/79873-T1; (1990).

[12] Cooper M., Muenchow H. O., Chiang S. H., Klinzing G. E., Morsi S., Venkatadri R., The licado coal cleaning process: A strategy for reducing SO_2 emissions from fossil-fueled power plants, in: Energy Conversion Engineering Conference, 1990. IECEC-90. Proceedings of the 25th Intersociety, Vol. 4, 1990, pp. 137–142.

[13] Chi S. M., Interfacial properties and coal cleaning in the licado process, Ph.D. thesis, University of Pittsburgh (1986).

[14] Microsoft Corporation , Microsoft Excel 2010 (2010).

[15] Aspen Technology, Inc. , Aspen Properties Excel Calculator, Version 7.3 (2011).

[16] Aspen Technology, Inc. , Aspen Custom Modeler, Version 7.3 (2011).

[17] Aspen Technology, Inc. , Aspen Simulation Workbook, Version 7.3 (2011).

[18] Oracle , Oracle Crystal Ball Clasroom Edition, Release 11.1.2.2.000 (2012).

[19] Microsoft Corporation , Microsoft Visual Basic for Applications, Version 7.0 (2010).

[20] Field R. P., Brasington R., Baseline flowsheet model for IGCC with carbon capture, Industrial & Engineering Chemistry Research 50 (19) (2011) 11306–11312.

[21] Muehlen H. J., van Heek K. H., Juentgen H., Kinetic studies of steam gasification of char in the presence of H_2, CO_2 and CO, Fuel 64 (7) (1985) 944–949.

[22] Monaghan R. F. D., Ghoniem A. F., A dynamic reduced order model for simulating entrained flow gasifiers: Part I: Model development and description, Fuel 91 (1) (2012) 61–80.

[23] Monaghan R. F. D., Ghoniem A. F., A dynamic reduced order model for simulating entrained flow gasifiers. part ii: Model validation and sensitivity analysis, Fuel 94 (0) (2012) 280–297.

[24] Monaghan R. F. D., Ghoniem A. F., Simulation of a commercial-scale entrained flow gasifier using a dynamic reduced order model, Energy & Fuels 26 (2) (2011) 1089–1106.

[25] Monaghan R. F. D., Dynamic reduced order modeling of entrained flow gasifiers, Phd thesis, Massachusetts Institute of Technology (2010).

[26] Chinese Academy of Sciences (CAS) and U.S. Department of Energy (DOE) , The United States of America and the People's Republic of China experts report on integrated gasification combined cycle technology, Technical Report DOE/FE-0357 (1996).

[27] Mc.Daniel J. E., Tampa Electric Polk Power Station Integrated Gasification Combined Cycle Project, Technical report, Prepared by Tampa Electric Company for The U.S. Department of Energy (2002).

[28] Electric Power Research Institute , Cool Water coal gasification program: Final report, Technical Report GS-6805 (1990).

[29] U.S. Department of Energy, National Energy Technology Laboratory (NETL) , Quality guidelines for energy system studies: Cost estimation methodology for NETL assessments of power plant performance, Technical Report DOE/NETL-2011/1455, NETL (2011).

[30] U.S. Department of Energy, National Energy Technology Laboratory (NETL) , Industrial size gasification for syngas, substitute natural gas and power production, Technical Report DOE/NETL-401/040607, U.S. Department of Energy, National Energy Technology Laboratory (2007).

[31] U.S. Department of Energy, National Energy Technology Laboratory (NETL) , Cost and performance baseline for fossil energy plants, volume 3a: Low rank coal to electricity: Igcc cases, Technical report, U.S. Department of Energy, National Energy Technology Laboratory (NETL).

[32] U.S. Department of Energy , Energy Efficiency and Renewable Energy, Advanced Manufacturing Office: benchmark the fuel cost of steam generation (2012).

[33] Lozowski D., Economic indicators, Chemical Engineering 120 (1) (2013) 63–64.

[34] Peters M., Timmerhaus K., West R., Plant Design and economics for chemical engineers, 5th Edition, McGraw-Hill, 2003.

[35] Klosek J., Smith A. R., Solomon P. R., The role of oxygen in coal gasification, in: Proceedings of the Eighth Annual Industrial Energy Technology Conference, Houston, TX (USA), 1986.

[36] International Energy Agency , Energy Technology Systems Analysis Program, Technology Brief S01: Syngas Production from Coal (2010).

[37] U.S. Energy Information Administration , Annual coal report 2011 (2012).

[38] U.S. Energy Information Administration , Electric power monthly (January 2013).

AppendixA. Appendix

Table A.4: Coal composition (dry basis)

Rank	**Bituminous**[1]	**Lignite** [31]
Seam	Illinois # 6	North Dakota
	Proximate Analyses (weight %)	
Moisture (ar)	11.12	36.08
Ash	10.91	15.43
Volatile Matter	39.37	41.49
Fixed Carbon	49.72	43.09
HHV, kJ/kg	30,506	24,254
	Ultimate Analyses (weight %)	
Carbon	71.72	61.88
Hydrogen	5.06	4.29
Nitrogen	1.41	0.98
Chlorine	0.33	0.00
Sulfur	2.82	0.98
Ash	10.91	15.43
Oxygen	7.75	16.44

Table A.5: Key data for PHICCOS model [5, 11]

CWS loading	20%-wt. ar coal
Flow of CO_2	CO_2:CWS = 0.5 (wt.)
Phase inversion	
Pressure	80 bar
Temperature	30 ℃
Residence time	5 min.
CO_2 flash	
Pressure	60 bar
Temperature	23 ℃
Heat duty	< 0.12 kJ/kg CO_2
Overall performance	
Enthalpy recovery	95%
Coal product ash	10%-wt. (dry)
Coal product moisture	10%-wt.

Table A.6: Key data for economic model

Cost basis	2011	U.S. Dollars ($)	
Cost index	CEPCI		[33]
Plant capacity factor	0.9		
Levelization factor	1.268		[29]
Capital			
Bare erected costs	*see Reference*		[1, 30, 31]
Capacity scaling exponent	0.6		[34]
EPC costs	9.4%	of BEC	[1, 31]
Process contingency			
SOA equipment	0-20%	of BEC	[1, 31]
PHICCOS	40%	of BEC	[29]
Project contingency	16%	of BEC+EPC+PRC	[1, 31]
Owner's costs	23%	of TPC	[1, 31]
Capital charge factor	0.1243		[29]
Operation & Maintenance			
Fixed O&M costs (1st year)	2.6%	of Total Plant Cost	[30]
Variable O&M costs (1st year)	0.450	$/GJ syngas (HHV)	[30]
Fuel			
Bituminous coal price	50.80	$/tonne	[37]
Lignite price	15.72	$/tonne	[37]
Utilities			
Electricity	6.46	c/kWh	[38]
Steam (60 bar)	8.3	$/1000 kg (bituminous coal)	
	4.6	$/1000 kg (lignite)	