

Synthesizing Optimal Flowsheets: Applications to IGCC System Environmental Control

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A new process synthesis capability implemented in the public version of the ASPEN chemical process simulator is demonstrated via an illustrative case study of a complex flowsheet. The objective of the case study is to minimize the cost of an advanced integrated gasification combined-cycle (IGCC) plant design featuring hot-gas cleanup, subject to environmental constraints. The problem is formulated as a mixed integer nonlinear programming (MINLP) optimization problem, involving the selection of both an optimal process configuration and optimal design parameters for that configuration. Performance and cost models of the IGCC system developed for the ASPEN simulator, along with the newly developed process synthesis capability, are used. As a first step, alternative in situ and external desulfurization are considered as process alternatives.

1. Introduction

There is a significant interest today in the ability of integrated coal gasification combined-cycle (IGCC) systems to provide electricity reliably and at lower cost relative to conventional fossil fuel alternatives. The ability of IGCC systems to meet stringent environmental emission standards is another attractive feature of this technology. Environmental control systems, however, account for a significant part of the cost and complexity of IGCC systems. Current systems require cooling of the gas stream prior to cleanup, thus generating a significant wastewater stream which must be treated in addition to the air pollutant and solid waste streams normally associated with coal-based electric power generation. Hot-gas cleanup systems offer the potential for significantly simplifying and reducing the cost of environmental control for many IGCC systems. This is currently the subject of intensive research and development.

In addition to the technical aspect of IGCC technology, there is also a strong need for "systems" research to

identity the best ways of configuring IGCC systems and of incorporating advanced cleanup and other technology to produce electricity at minimum cost. For example, the most common design for sulfur removal using hot-gas cleanup is through the use of solid sorbents. Sulfur capture occurs either through the addition of a solid reactant in the gasifier (i.e., in-bed desulfurization), or by external desulfurization of the flue gas (e.g., zinc ferrite process) or by a combination of these two methods.

A number of computer models have been developed by the U.S. Department of Energy's Morgantown Energy Technology Center (DOE/METC) to allow analysis of different configurations of IGCC systems using the ASPEN process simulator (e.g., Stone, 1985). Selection of the optimal flowsheet configuration and the optimal design parameters, however, requires additional capabilities in the modeling environment.

A new process synthesis capability that has been implemented in ASPEN (Diwekar et al., 1992) provides a tool to obtain the optimal flowsheet configuration and design

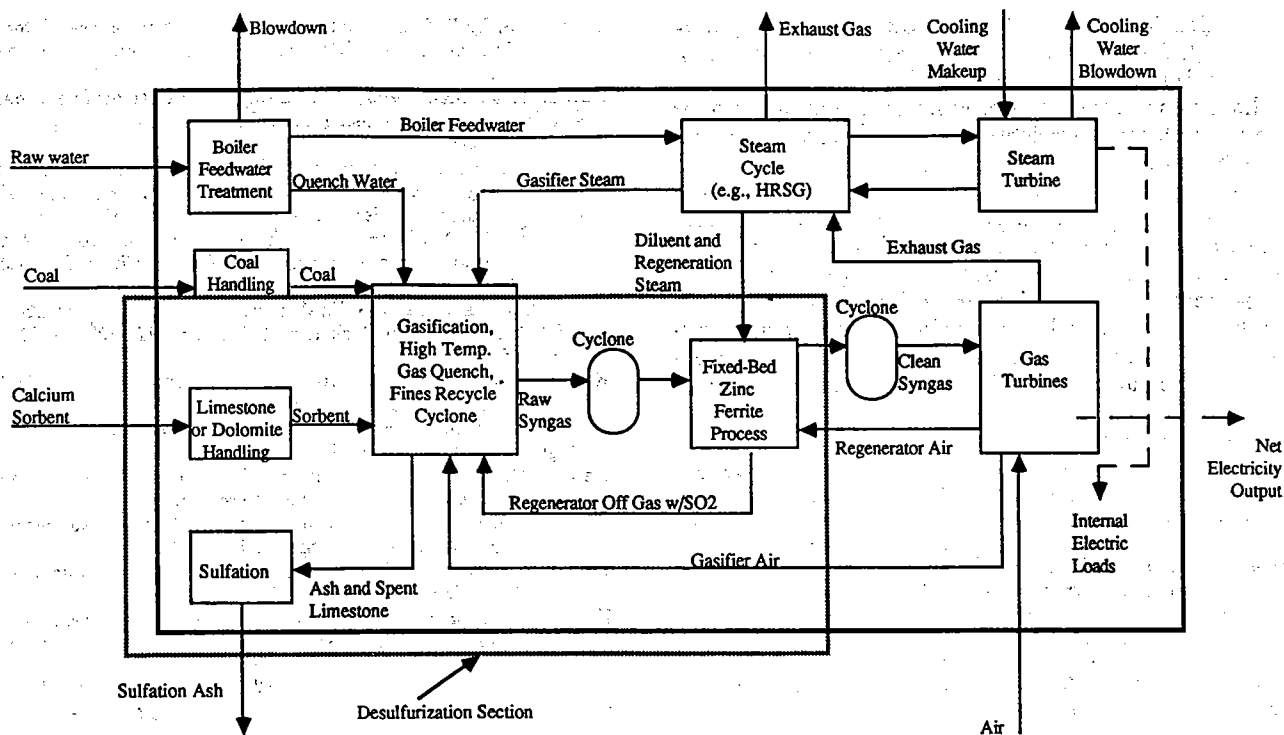


Figure 1. Simplified schematic of air-blown fluidized bed gasifier IGCC system with hot gas cleanup.

optimization. This process synthesis capability is based on a mathematical programming approach which requires solution of a mixed integer nonlinear programming (MINLP) problem.

The major steps involved in the MINLP process synthesis are as follows:

1. Formulation of a flowsheet superstructure with embedded alternative flowsheet structures, representing alternative process configurations.

2. Modeling the superstructure as an MINLP problem of the form

$$Z = \min_{x,y} C(x,y) \quad (1)$$

subject to

$$h(y,x) = 0 \quad (2)$$

$$By + g(x) \leq 0 \quad (3)$$

$$y \in Y; \quad x \in X$$

where

$$Y = \{y | Ay \leq a, y \in \{0,1\}^m\}; \quad X = \{x | x^L \leq x \leq x^U, x \in R^n\}$$

The continuous variables, x , represent flows, operating conditions, and design variables. The binary variables y denote the existence or nonexistence of process units and, hence, the process configuration. $C(y,x)$ represents the objective function. Equation 2 is the process equation representing the equality constraints, which may be nonlinear. Equation 3 represents the design specification and logical constraints, in the form of linear inequalities.

3. Decomposition of the superstructure into an initial flowsheet and subsystems of nonexisting units which are contained in alternate flowsheet configurations (for additional details please refer to Kravanja and Grossmann (1989)).

4. Extraction of the optimal design from the superstructure by solving the corresponding MINLP problem.

In this paper the MINLP process synthesis capability added to the public version of ASPEN (Diwekar et al., 1992) is used to select the optimal plant configuration and optimal design parameters for an IGCC system using hot-gas cleanup. The optimal synthesis is based on a superstructure containing three alternative configurations for sulfur removal, i.e., in-bed desulfurization, gas stream desulfurization, and a hybrid system combining both methods.

2. Advanced IGCC Systems and Technological Alternatives for Sulfur Removal

A number of variations in IGCC design exist, based primarily on gasifier design and fuel gas cleanup strategy. Conventional IGCC designs, including those of existing demonstration plants, feature oxygen-blown gasification with "cold" fuel gas cleanup. These designs consist of serial applications of a number of low-temperature gas cleaning processes, each of which removes one or more contaminant species from the fuel gas. Such systems require cooling of the fuel gas.

Significant performance and cost advantages can be gained if the need for fuel gas cooling can be eliminated, thus improving plant thermal efficiency and eliminating the need for heat exchangers and process condensate treatment. Advanced designs using hot-gas cleanup systems employ cyclone or barrier filtration for particulate control and solid sorbents for removal of sulfur species either "in situ" during gasification or "externally" from the fuel gas.

One of the most promising hot-gas cleanup configurations is an air-blown Kellogg-Rust-Westinghouse (KRW) IGCC system. A schematic of this technology is shown in Figure 1. The hot-gas cleanup system features in-bed desulfurization in the fluidized bed gasifier with limestone or dolomite, subsequent sulfur removal from the fuel gas with a zinc ferrite sorbent, and high-efficiency cyclones and ceramic filters for particulate removal. The off-gas from the zinc ferrite reactor, which contains sulfur compounds, is recycled to the gasifier. The advantages of such a system

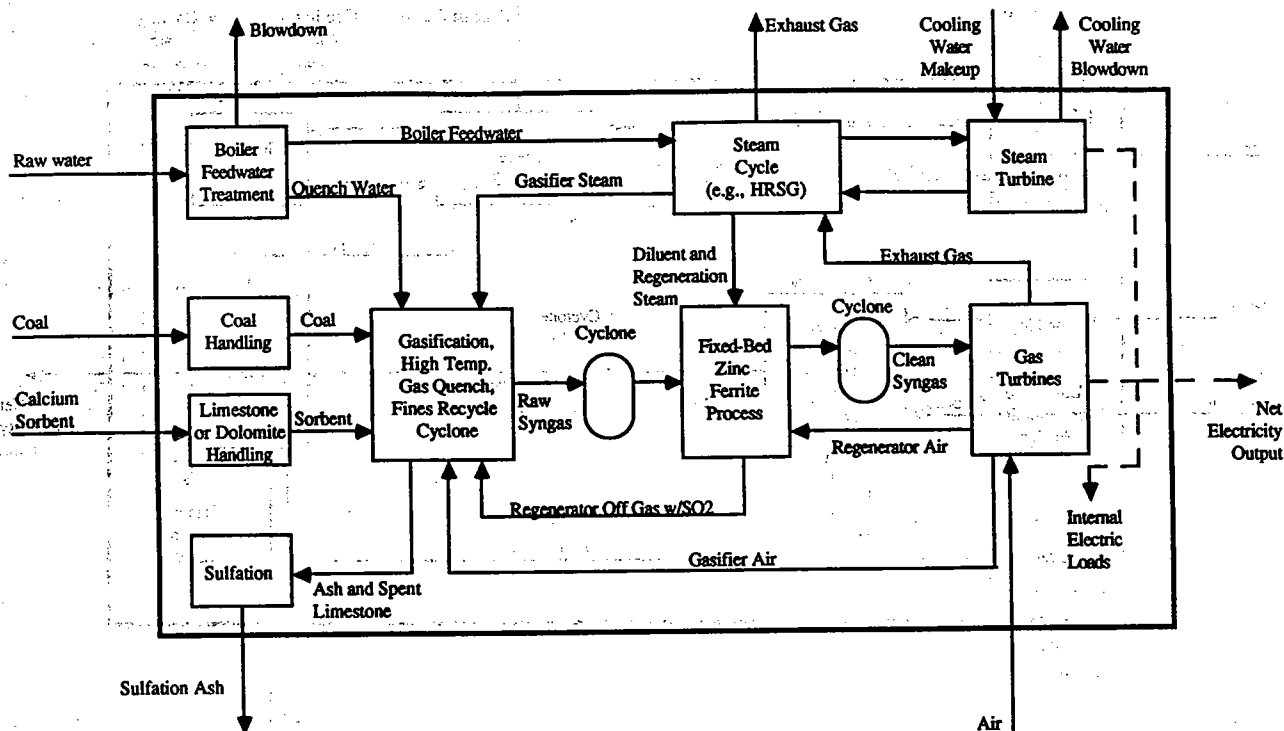


Figure 2. Schematic of air-blown fluidized bed gasifier IGCC system with hot gas cleanup (combined in-bed and gas stream desulfurization).

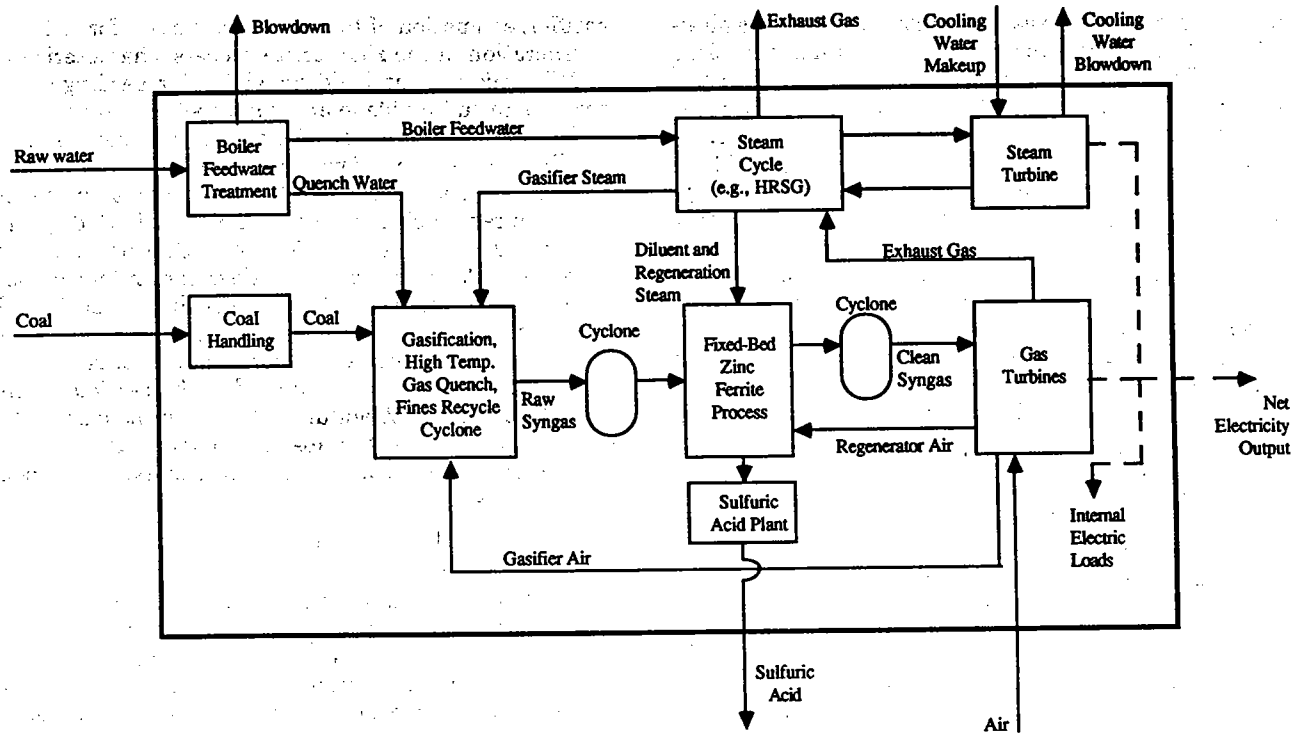


Figure 3. Schematic of air-blown fluidized bed gasifier IGCC system with hot gas cleanup (gas stream desulfurization only).

compared to a base case oxygen-blown system with cold-gas cleanup are that (1) it does not require an expensive and energy-consuming oxygen plant; (2) it eliminates the capital costs associated with sulfur recovery (i.e., all sulfur is disposed with the spent limestone or dolomite); and (3) it reduces the amount of fuel gas cooling required prior to combustion in the gas turbine, thereby improving the plant thermal efficiency. Testing of an air-blown KRW-based system with hot-gas cleanup at the process development unit (PDU) level has been conducted (Haldipur et al., 1988, 1989) and studies of several KRW IGCC hot-gas cleanup system concepts have been performed by

Southern Company Services (1990) and Fluor Daniel, Inc. (Earley and Smelser, 1988; Smith and Smelser, 1987). Additional details regarding the process technology are discussed by Frey (1991).

Alternative process configurations may have cost advantages over the system with both in-bed and external desulfurization. In addition, the split of the sulfur removal burden between the gasifier and external desulfurizer affects process performance and cost. To study these interactions and to identify the optimal process design and optimal values of key design parameters, three alternative flowsheet topologies (Figures 2-4) are evaluated using the

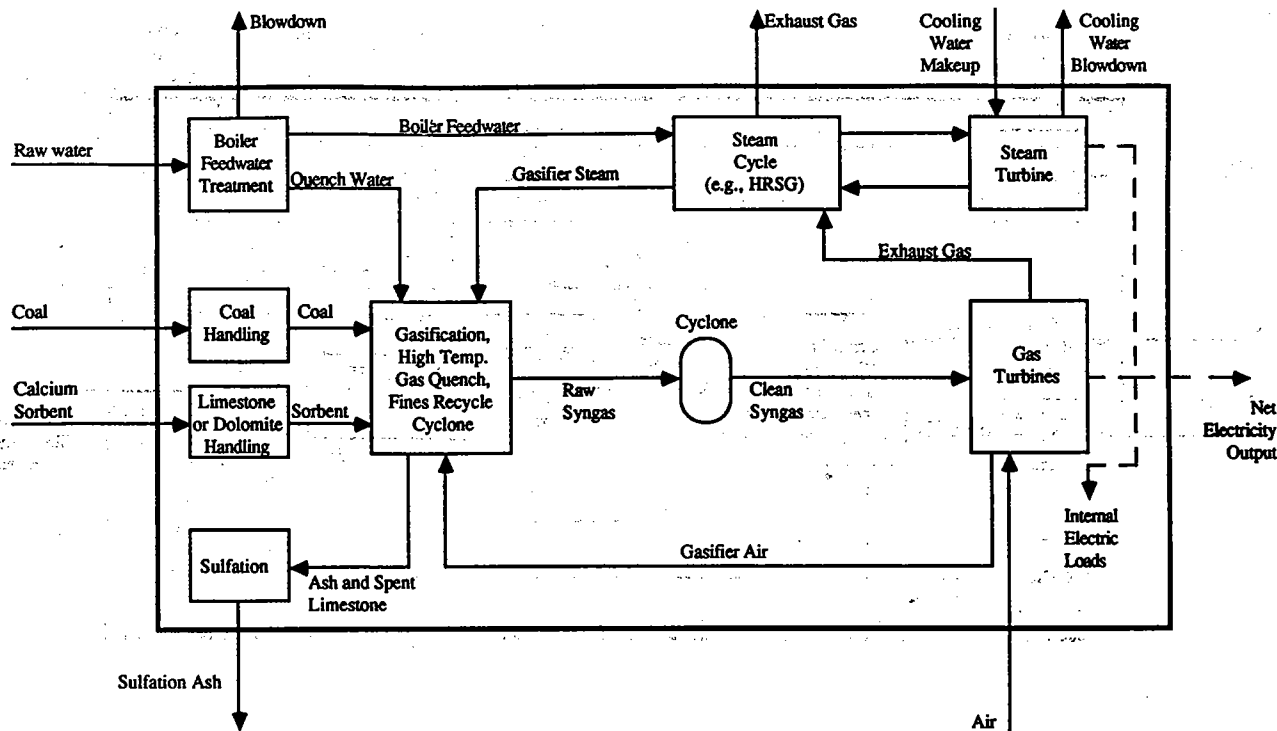


Figure 4. Schematic of air-blown fluidized bed gasifier IGCC system with hot gas cleanup (in-bed desulfurization only).

MINLP process synthesis capability. These three alternatives differ in the approach used for sulfur emissions control. They employ different combinations of in-bed gasifier desulfurization and external zinc ferrite desulfurization: (1) in-bed desulfurization with external "polishing" zinc ferrite desulfurization (Figure 2); (2) external "bulk" zinc ferrite desulfurization only with by-product recovery (Figure 3); (3) in-bed desulfurization only (Figure 4). The third case may not be a realistic design because it produces a relatively high sulfur loading in the gas turbine fuel. It is included in this analysis for illustrative purposes. In the two cases where in-bed desulfurization is used, the spent calcium-based sorbent from the gasifier, which contains calcium sulfide (CaS), must be sulfated prior to landfilling in order to comply with RCRA standards for nonhazardous wastes. Thus, a circulating fluidized bed boiler is used as a sulfator. In the case where only external zinc ferrite desulfurization is employed, the sulfator is eliminated, along with the limestone handling requirement, but there is an addition of a sulfuric acid plant for byproduct recovery.

2.1. Performance Models. Performance models for all three flowsheet topologies have been devised based on a public domain ASPEN input file developed by DOE/METC. The METC model of an air-blown KRW system with hot-gas cleanup employs in-bed gasifier desulfurization using limestone, fixed-bed zinc ferrite external desulfurization, an advanced gas turbine design (e.g., the General Electric MS7001F), and a reheat steam cycle (Craig, 1988). This model includes 73 unit operation blocks in four major plant sections, and about a dozen major design specifications and FORTRAN blocks. Similar models are documented by Stone (1985).

Modifications to the METC model have been made by Frey (1991, pp 64-72, 263-270). Through the addition of unit operation blocks, FORTRAN blocks, design specifications, and chemical species components (as appropriate), other process areas were added or represented in more detail. These modifications include the addition of the sulfation process area, estimation of ammonia yield in the

gasifier, estimation of fuel gas consumption for reductive regeneration in the zinc ferrite process, characterization of NO_x emissions, and additional detail regarding the gas turbine, including blade and vane cooling requirements.

To evaluate the effect of alternative assumptions regarding in-bed desulfurization efficiency, two simple models have been considered for the relationship between the required calcium-to-sulfur molar ratio and gasifier per-pass desulfurization efficiency. Unfortunately, few data are available to support the development of a detailed model. Haldipur et al. (1989) report that 10-20% of the sulfur in the coal is not released when gasification occurs without sorbent. They also report that a calcium-to-sulfur ratio of 2.6 is likely to be required for high-calcium limestone to achieve 90% sulfur capture. On the basis of this information, and in the absence of more detailed (and proprietary) parametric information, a simple linear model was assumed:

$$R_{\text{Ca/S}} = a_1(\eta_s - b_1) \quad (4)$$

The parameters of this model are $a_1 = 3.47$ and $b_1 = 0.15$. This model is likely to be conservative in overpredicting the Ca/S ratio for a given removal efficiency below 90% efficiency, since the relationship is more likely to be nonlinear with exponentially increasing Ca/S for higher removal efficiencies. Therefore, an alternative exponential model was also considered:

$$R_{\text{Ca/S}} = a_2 \left(\frac{\exp(\eta_s - b_2) - 1}{1 - \eta_s} \right) \quad (5)$$

The parameters of this model are $a_2 = 0.233$ and $b_2 = 0.15$. This functional form was selected to return a Ca/S ratio of 2.6 for a removal efficiency of 90% and an unbounded value for a 100% removal efficiency. The two models are compared in Figure 5. Generally, the linear model estimates higher Ca/S ratios than the exponential model for removal efficiencies less than 90%. In implementation, both models are also bounded by the minimum stoichio-

Table I. Major Process Areas for Each Flowsheet Topology

area no.	flowsheet topology (key features)		
	in-bed desulfurization with zinc ferrite	zinc ferrite only with byproduct recovery	in-bed desulfurization only
10	air boost compression	air boost compression	air boost compression
20	coal handling	coal handling	coal handling
25	limestone handling		limestone handling
30	gasification, high-temperature gas cooling, ash and particulate removal, coal pressurization	gasification, high-temperature gas cooling, ash and particulate removal, coal pressurization	gasification, high-temperature gas cooling, ash and particulate removal, coal pressurization
35	sulfation		sulfation
50	zinc ferrite desulfurization	zinc ferrite desulfurization	
60		sulfuric acid plant	
80	steam, condensate, boiler feed water	steam, condensate, boiler feed water	steam, condensate, boiler feed water
91	gas turbine	gas turbine	gas turbine
92	HRSG ^a	HRSG	HRSG
93	steam turbine	steam turbine	steam turbine
100	general facilities	general facilities	general facilities

^aHRSG = heat recovery steam generator.

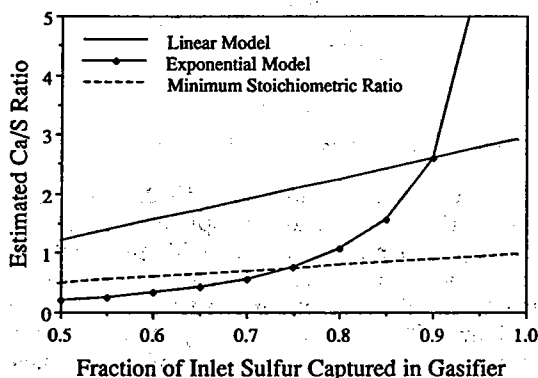


Figure 5. Comparison of alternative models for Ca/S molar ratio as a function of gasifier in-bed per-pass sulfur capture rate.

metric ratio required to achieve sulfur capture by the calcium sorbent.

To represent alternative flowsheet topologies in a single simulation, splitters (ASPEN unit operation blocks for dividing inlet streams into two or more outlet streams) and mixers were employed, plus binary control variables to control mass flow rates (please see section 3).

2.2. Cost Models. A key limitation of the IGCC performance model developed by METC was the lack of a directly coupled cost model. To overcome this limitation, cost models of selected IGCC systems, including the air-blown KRW system with hot-gas cleanup, were developed by Frey and Rubin (1990). These cost models were based on a review of approximately 30 comprehensive conceptual design studies prepared for DOE, the Electric Power Research Institute (EPRI), and the Gas Research Institute (GRI), as well as other studies which focused on specific process components. The models provide "preliminary" estimates of process capital, annual, and levelized costs based on the standard costing method developed by EPRI (1986).

In order to link process flowsheet parameters with economic cost models, the method employed was to model all costs at the level of major plant sections for each IGCC technology. Table I shows a list of the sections used for the three alternatives flowsheet topologies analyzed in this paper. Altogether, there are a dozen major process areas.

As noted in Table I, there are a few major differences between the three flowsheet topologies. For the cases in which in-bed gasifier desulfurization is present, limestone (sorbent) handling and spent sorbent sulfation is required. In the cases where the zinc ferrite process is used, the cost for this process area must be estimated. Furthermore, where sulfuric acid byproduct is recovered, the cost of a

sulfuric acid plant is also estimated.

The direct capital cost of each process section was estimated on the basis of analytic relationships between direct cost and key performance parameters (Frey and Rubin, 1990). After summing the individual section costs, the total direct cost of each IGCC system is sensitive to approximately two dozen performance parameters, in addition to process design parameters such as the number of operating and spare trains of equipment.

For each IGCC system, the total capital cost is then obtained as the sum of (1) the total direct cost for all major process areas; (2) indirect costs, such as engineering and home office fees; (3) contingency costs; (4) allowance for funds during construction; (5) preproduction (startup) costs; and (6) initial catalysts and chemicals inventory costs. In addition to capital cost models, models for annual costs also have been developed (Frey and Rubin, 1990). These include fixed (e.g., operating labor and maintenance) and variable (e.g., consumables) costs. The consumption rates of chemicals and catalysts were modeled on the basis of data from published design studies. Examples of key consumables include limestone sorbent, zinc ferrite sorbent, and sulfuric acid plant catalyst. Another key variable cost is ash disposal. The annual cost models also include byproduct credit for sulfuric acid recovery where appropriate.

The total revenue requirement for an electric power plant is often the key parameter of interest, and is typically expressed as the cost of electricity. The cost of electricity model for each system includes economic and financial parameters, plus models for the power consumption of major plant sections (Frey and Rubin, 1990). A generalized cost model for all three flowsheet topologies was developed from the existing cost models, with a modification for calculating gasifier direct cost (presented below) for the case studies presented here. Binary control variables are used to describe the flowsheet topologies given in Table I (please see section 3), and process area capital and annual costs are included or excluded as appropriate for each design option. The generalized cost model has been coded in FORTRAN and implemented as a set of subroutines along with the ASPEN performance model.

The gasifier process area direct cost model was revised to account for differences in coal throughput for the different gasification modes considered in this case study. The direct cost model consists of two parts. One is a method for estimating the required number of gasifier trains based on the coal processing capacity, type of oxidant, and whether in-bed desulfurization is employed. The second is a model for estimating direct capital cost as a

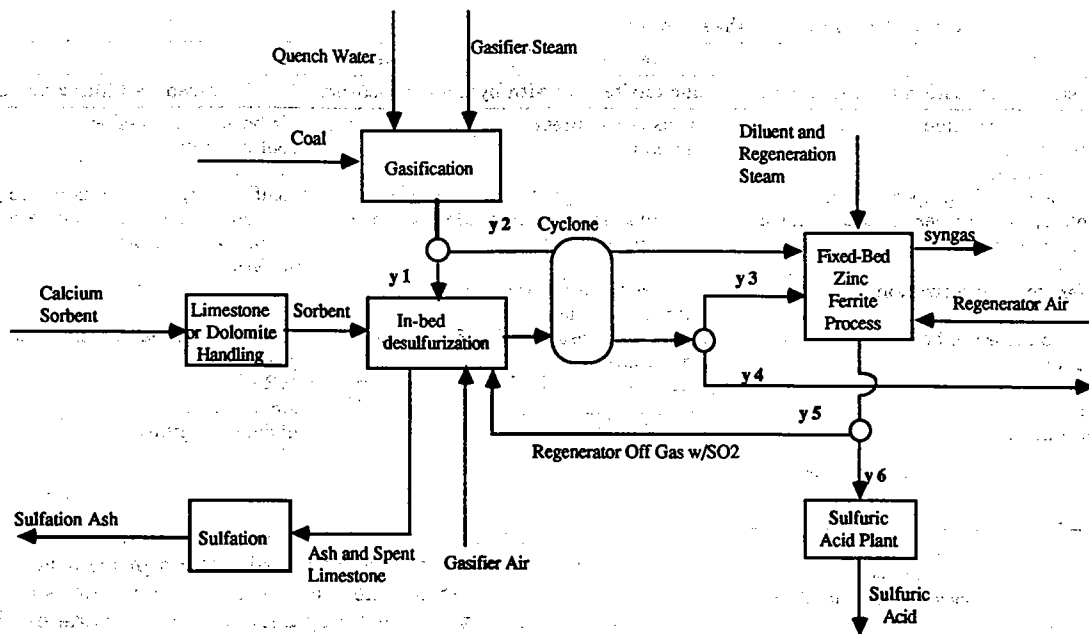


Figure 6. Superstructure for the desulfurization section in the KRW IGCC system with hot gas cleanup.

Table II. Gasifier Oxidant and Sorbent Operating Modes and Relative Number of Gasifier Trains

operating mode	rel no. of trains, f_t
oxygen-blown, without sorbent	1
oxygen-blown, with sorbent	0.8
air-blown, without sorbent	1.9
air-blown, with sorbent	1.3

function of the number of gasifiers employed.

A model for the required number of gasifiers for an oxygen-blown system without in-bed desulfurization has been developed previously (Frey and Rubin, 1990) and is given here:

$$N_{O,G} = \text{INT}(0.557 + 9.10 \times 10^{-4} m_{cm,G,i}) \quad (R^2 = 0.936; n = 15) \quad (6)$$

where $\text{INT}(x)$ returns the integer value closest to the real value x . The number of operating gasifiers is estimated on the basis of the coal feed rate, on a dry and ash-free basis (tons/day). This model assumes that gasifier vessels are of approximately the same size and design (e.g., operating pressure of 465 psia, operating temperatures of 1850–1900 °F or less). The relative number of operating trains required for different modes of operation (e.g., oxygen- versus air-blown, with and without in-bed desulfurization) are estimated on the basis of multiplicative factors developed previously by Frey (1991, p 494), as given in Table II.

The revised equation is thus

$$N_{O,G} = \text{INT}\{f_t(0.557 + 9.10 \times 10^{-4} m_{cm,G,i})\} \quad (7)$$

The value of f_t depends on the gasification mode and is given by Table II.

The total number of trains is estimated based on the notion of availability of the gasifier. The general equation for the total number of trains is

$$N_T = \text{INT}(N_{O,G}/A_v) \quad (8)$$

where A_v is the availability. The default for A_v is assumed to be 0.85.

On the basis of an analysis of two studies by Fluor (Smith and Smelser, 1987; Earley and Smelser, 1988), a study by Southern Company Services (1990), and a pre-

vious cost model (Frey and Rubin, 1990), the cost of each standard KRW gasifier train is approximately \$20 million for typical operating conditions of 465 psia and 1850 °F. From the Fluor studies, the cost per train for an oxygen-blown system with heat exchangers for gas cooling is about \$22 million. This number was adjusted to account for air-blown operation and for the use of water quench instead of heat exchange for fuel gas cooling (see Frey and Rubin (1990), pp 97–99). The Southern Company Services study does not use the same process area battery limits as the Fluor studies, but a comparison of the two suggests agreement with the Fluor studies. Thus, the total direct cost of the gasification process area, including recycle gas compression, ash handling, high temperature gas quench, and fines collection, is estimated to be (1989 dollars)

$$DC_G = 16500N_T(I_c/351.5) \quad (9)$$

3. Solution Strategy

The first step in the solution of the process synthesis problem is to develop the superstructure containing all alternative designs to be considered for the optimal solution. The objective is minimize the leveled cost of electricity (C_{elec}), which can be stated as

$$\min C \quad (10)$$

$$C - C_{elec} = 0 \quad (11)$$

The superstructure for the three alternative desulfurization configurations for the advanced IGCC system is shown in Figure 6. There are a total of three additional splitters, one additional mixer, and six binary decision variables, i.e., two decision variables (branches) per node. Each binary variable represents the presence ($y = 1$, where $y \in Y$) or absence ($y = 0$) of the branch associated with that variable. At the first node, the two decisions involved are in-bed desulfurization ($y_1 = 1$) or only external desulfurization by the zinc ferrite process ($y_2 = 1$). Since these two decisions are mutually exclusive, the following constraint is added in the optimization problem

$$y_1 + y_2 = 1 \quad (12)$$

If in-bed desulfurization is selected, then at the second node there is a choice of the zinc ferrite process after in-bed desulfurization ($y_3 = 1$) or the cyclone separator marking

the end of the desulfurization section ($y_4 = 1$). These two decisions also are mutually exclusive and apply only in the presence of in-bed desulfurization ($y_1 = 1$). This leads to

$$y_3 + y_4 = y_1 \quad (13)$$

The last node in the desulfurization process exists on the zinc ferrite branch. If the zinc ferrite process is selected, the decision about whether the SO_2 should be recycled ($y_5 = 1$) or passed to the sulfuric acid plant ($y_6 = 1$) is decided at this node. Again, these events are mutually exclusive and are considered only when the zinc ferrite process is selected ($y_4 = 0$). The recycle alternative is applicable only for combined in-bed and gas stream desulfurization ($y_1 = 1, y_3 = 1$). Coupled with the zinc ferrite only option is the presence of the sulfuric acid plant ($y_6 = 1$). These conditions are represented by the following logical constraints:

$$y_5 + y_6 = 1 - y_4 \quad (14)$$

$$y_6 = y_2 \quad (15)$$

The six binary decision variables along with the above four constraints (eq 12–15) result in three feasible alternative technologies:

1. In-bed desulfurization plus external "polishing" by zinc ferrite desulfurization

$$Y_1 = (1, 0, 1, 0, 1, 0) \quad (16)$$

2. Gas stream desulfurization only, via zinc ferrite with byproduct recovery

$$Y_2 = (0, 1, 0, 0, 0, 1) \quad (17)$$

3. In-bed desulfurization only, via limestone or dolomite injection

$$Y_3 = (1, 0, 0, 1, 0, 0) \quad (18)$$

As an example of the implementation of the binary variables, consider the binary variable y_1 . We introduce this variable into the appropriate performance and cost model equations to represent the presence of unit operations associated with sorbent handling. Thus, the direct capital cost of the limestone handling section is given by

$$DC_L = y_1(1160 + 0.026m_L)(I_c/351.5) \quad (19)$$

The gasifier coal throughput is affected by the use of limestone sorbent. Therefore, the "train factor" described previously is given by (for air-blown mode only)

$$f_t = 1.8 - 0.5y_1 \quad (20)$$

The gasifier direct cost is then calculated using the model presented in eqs 7–9. The direct cost of the sulfation process area is given by

$$DC_S = y_1\{13.0(m_{\text{lash}})^{0.639}\}(I_c/351.5) \quad (21)$$

The binary variable y_1 is applied similarly to the equations used to estimate annual costs for the affected process areas.

The purpose of this exercise is to obtain a flowsheet configuration and design variables which minimize the leveled cost of electricity, given an environmental constraint on total sulfur emissions:

$$E_{\text{SO}_2} \leq \frac{0.015 \text{ lb of SO}_2}{10^6 \text{ Btu}} \Rightarrow E_{\text{SO}_2} \leq \frac{6.45 \text{ g of SO}_2}{\text{GJ}} \quad (22)$$

This constraint is far more stringent than current emission regulations in the U.S., but is representative of the capability of advanced IGCC technology.

The continuous decision variables selected for this preliminary study are the in-bed desulfurization efficiency (η_s), the zinc ferrite absorption cycle time (t_a), and the

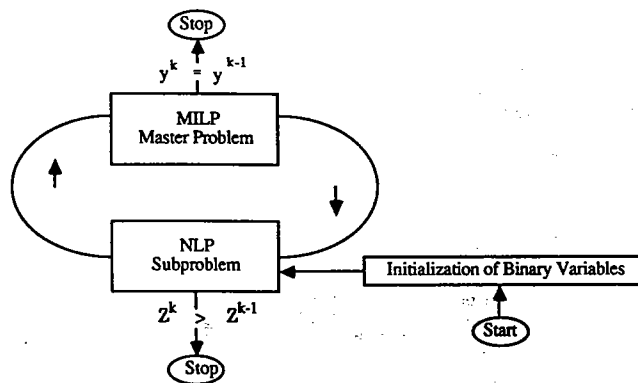


Figure 7. Main steps in the GBD/OA/ER/AP algorithm.

maximum vessel height to diameter ratio (L/D) for the zinc ferrite absorbers. The in-bed desulfurization efficiency determines the limestone sorbent requirement and removal of residual sulfur evolved in the zinc ferrite process area. This variable is allowed to vary up to 90% per pass gasifier sulfur removal. In the absence of sorbent about 15% of the sulfur is removed in the gasifier bottom ash; this gives the lower limit for the efficiency. The zinc ferrite absorption cycle time is allowed to vary from 30 to 172 h and the zinc ferrite vessel height to diameter ratio ranges from 2 to 4.

The MINLP problem at this stage consists of three continuous and six binary decision variables, the above four equality constraints for the binary logical variables (eqs 12–15), the environmental constraint on total sulfur emissions (eq 22), and three inequality constraints (eq 23–25) for the three continuous decision variables, which are related to the binary variables via upper and lower bounds.

$$0.15 \leq \eta_s \leq (0.75y_1 + 0.15) \quad (23)$$

$$30(y_2 + y_3) \leq t_a \leq 172(y_2 + y_3) \quad (24)$$

$$2(y_2 + y_3) \leq L/D \leq 4(y_2 + y_3) \quad (25)$$

Although this completes the mathematical formulation of the MINLP problem, the implicit nature of the sequential modular simulator, in which performance relationships are not written explicitly as analytical equations, calls for additional decision variables and an equal number of additional implicit equality constraints. This is because the MINLP environment is based on the generalized Benders decomposition/outer approximation method/equality relaxation/augmented penalty function (GBD/OA/ER/AP) algorithm (Diwekar et al. 1992). This algorithm involves two levels of optimization as shown in Figure 7. These are (1) an upper-level mixed integer linear programming (MILP) master problem and (2) a lower-level nonlinear programming (NLP) problem. The MILP master problem has the role of predicting new binary variables, while the NLP problem provides new continuous variables. The MILP master problem represents the linearized NLP problem with non-fixed binary variables, since, at each stage, the MILP master problem obtains the linearization information from the NLP optimizer.

The relationship between input design variables and the output variables used to calculate the objective function is represented as a nonlinear equality constraint given by eq 11. Equation 11 is an implicit constraint in the ASPEN simulator. This constraint and similar implicit constraints must be made explicit for the purpose of providing the information needed by the MILP master problem for linearization of the black-box relations. For this purpose, new decision variables are created, and these are equated

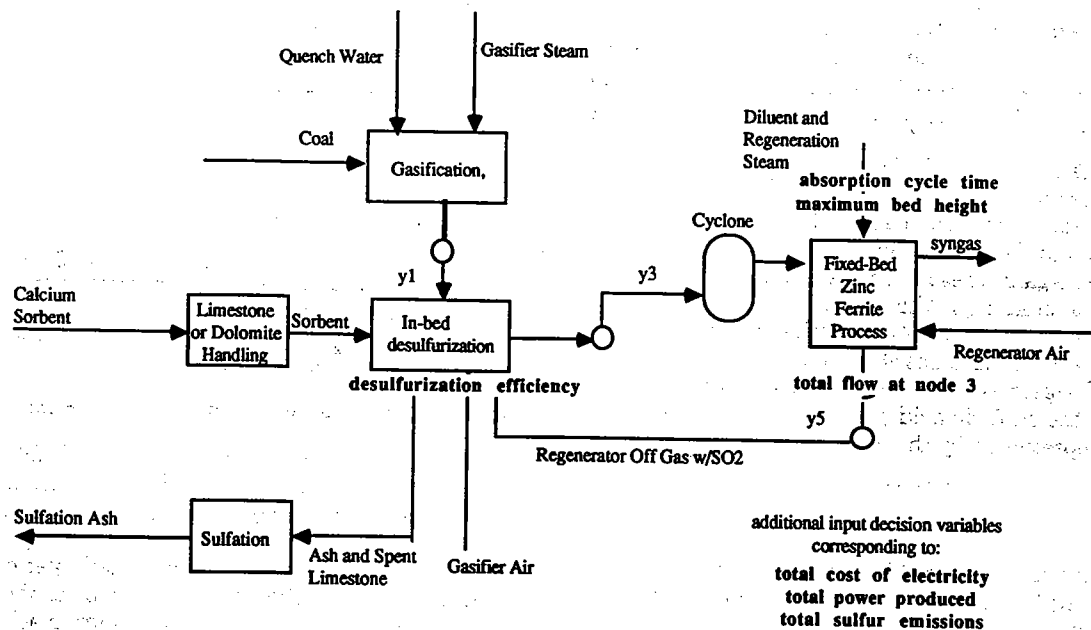


Figure 8. Initial flowsheet and the associated continuous decision variables (boldfaced) for the decomposition strategy.

to the output variables from the flowsheet configurations. This procedure ensures that the original MINLP problem remains the same, while at each stage the MILP master problem receives increasing information from the NLP optimizer. This procedure leads to a proper optimal solution in fewer iterations.

For the evaluation of the advanced IGCC sulfur removal alternatives, the additional output variables for which constraints were made explicit (eqs 26-30) are sulfur

$$x_4 - C_{T-H_2SO_4} \quad (26)$$

$$x_5 - C_{H_2SO_4} \quad (27)$$

$$x_6 - P_{T-H_2SO_4} \quad (28)$$

$$x_7 - P_{H_2SO_4} \quad (29)$$

$$x_8 - E_{SO_2} \quad (30)$$

emissions, in pounds of SO_2 per million Btu (or grams per gigajoule) of coal throughput; annualized cost of electricity without the sulfuric acid plant, in millions of dollars; annualized cost of the sulfuric acid plant, in millions of dollars; total power generated without power consumption in sulfuric acid plant, in MW-h; and power consumed for the sulfuric acid plant, in MW-h.

These additional variables are needed for the proper estimation of the linear approximations of the objective function and the emission constraint in the MILP master problem. The implicit constraint for the levelized cost of electricity, in mills (one-thousandth of a dollar) per kW-h of electricity generation (eq 11), can then be replaced by

$$C - \left(\frac{C_{T-H_2SO_4} + C_{H_2SO_4}}{P_{T-H_2SO_4} - P_{H_2SO_4}} \right) = C - \frac{x_4 + x_5}{x_6 - x_7} = 0 \quad (31)$$

The sulfuric acid plant annualized cost and power consumption are related to the binary variables via upper bounds (and) given by

$$P_{H_2SO_4} \leq y_6 UP_{H_2SO_4} \quad (32)$$

$$C_{H_2SO_4} \leq y_6 UC_{H_2SO_4} \quad (33)$$

The main difficulty which is encountered in the process synthesis calculations is having to optimize "dry units" with

zero flows, which are temporarily turned off in the superstructure. This problem of zero flows in the nonexistent units calls for the decomposition strategy proposed by Kocis and Grossmann (1989). The decomposition strategy establishes the required linear approximations of the constraints related to the nonexistent units for the master problem. It involves proper selection of initial flowsheet and subsystems.

The initial flowsheet selected for this problem (Figure 8) is the hybrid system containing the in-bed desulfurization combined with the zinc ferrite process option (Y_1). It can be seen that the initial flowsheet selected is the only configuration which provides information about flow rates at all the interconnection nodes (y_1/y_2 , y_3/y_4 , y_5/y_6) and has all the units included in it except the sulfuric acid plant. The modular nature of the cost model allowed capture of the information about the variables associated with the sulfuric acid plant (eq 27 and 29), by considering the subsystem containing the sulfuric acid plant only. The continuous decision variables associated with the initial flowsheet are in-bed desulfurization efficiency, zinc ferrite absorption cycle time, maximum height-to-diameter ratio of the vessel for zinc ferrite process, total annualized cost of electricity, total power, levelized cost of electricity, sulfur emissions, and the entering flow rate for the splitter at the y_5/y_6 node. The subsystem variables include the annualized cost of and power consumed in a sulfuric acid plant.

4. Results and Discussion

The problem of selecting an optimal flowsheet configuration and optimal design variables is solved using the newly developed process synthesis environment. The ASPEN input file for this problem contains the MILP master unit operation block, the NLP optimizer for initial flowsheet and subsystem, and the entire flowsheet superstructure. The resulting optimal flowsheet structure was found to be the hybrid system shown in Figure 8. This result was obtained assuming either a linear or nonlinear models to estimate the calcium-to-sulfur ratio for in-bed desulfurization (eqs 4 and 5).

The values of the optimal design variables are shown in Table III. The levelized cost of electricity of this optimal flowsheet is found to be 53.76 mills/kW-h (or 53.76 dollars per megawatt-hour) of electricity assuming a linear model

Table III. NLP Optimization Results for the Different Sulfur Removal Alternatives

technological alternatives	model for $R_{Ca/S}$	desulfurization efficiency	absorption cycle time	maximum length/diam	levelized cost of electricity
in-bed and gas stream desulfurization	linear	0.89	150.5 h	2.29	53.76 mills/kW-h
	nonlinear	0.81	84.45 h	4.00	52.09 mills/kW-h
gas stream desulfurization only	linear	0.15	30 h	2.00	62.30 mills/kW-h
	nonlinear	0.15	30 h	2.00	62.30 mills/kW-h
in-bed desulfurization only	linear	infeasible	n/a	n/a	infeasible
	nonlinear	infeasible	n/a	n/a	infeasible

and 52.09 mills/kW-h assuming a nonlinear model. Given the environmental constraint of total plant SO_2 emissions less than or equal to 0.015 lbs per MMBtu of coal throughput (eq 22), the in-bed desulfurization system alone was found to be an infeasible option in both cases. The optimal cost of electricity from the zinc ferrite gas stream desulfurization option alone was determined to be 62.30 mills/kW-h for both cases, which exceeds the optimal cost of combined in-bed desulfurization and zinc ferrite option by over 15%.

The optimal cost for the system with the nonlinear model is approximately 3% less expensive than the solution for the system with the more conservative linear model. However, more notable differences are in the optimal in-bed desulfurization efficiency, absorption cycle time, and the maximum length-to-diameter ratio for the zinc ferrite reactor vessels. In the case of the linear model, a higher desulfurization efficiency is predicted than for the nonlinear model. From Figure 5 it is clear that the Ca/S ratio for 81% removal with the nonlinear model is substantially lower than the Ca/S ratio required for 89% removal as estimated using the linear model. Thus, the limestone sorbent requirement is lower, but the sulfur loading into the zinc ferrite system is significantly increased.

In the case of the linear Ca/S ratio model, space velocity constraints in the zinc ferrite model have a pronounced effect on the initial sorbent charge requirement, leading to a relatively long cycle time of 150 h. For the nonlinear case, the increased sulfur burden to the zinc ferrite system results in the need for a higher sorbent charge, and larger process vessels to accommodate the charge. Hence, the length-to-diameter ratio for this case is higher to accommodate the larger volume of sorbent, while the sorbent is consumed more rapidly, resulting in a lower cycle time.

Alternative KRW-based IGCC systems with hot-gas cleanup have been evaluated previously by Southern Company Services (1990). In that study, the hybrid in-bed and external desulfurization system was identified as the optimal configuration. The in-bed desulfurization efficiency was the only decision variable used, and its optimal value was reported to be 88%. The study assumed a zinc ferrite absorption cycle time of 172 h and vessel height-to-diameter ratio of 2.8:1, which were held fixed. The levelized costs of the external desulfurization only case were reported to exceed those of the hybrid case by approximately 20%. Thus, there is generally good agreement between the results reported here and those of the earlier study.

However, the current work differs from the Southern Company Services (1990) study in that the approach here allowed not just one, but three (and potentially more) design parameters to vary simultaneously in search of the optimal solution. Furthermore, the possible alternative flowsheet configurations were considered simultaneously as part of an integrated computer modeling approach in a single computer simulation, whereas separate estimates were developed by Southern Company Services for each flowsheet and then compared.

While the Southern Company Services result for the optimal in-bed removal efficiency is bounded by the alternative cases considered in this study, the case study results suggest that the optimal solution and, hence, the proper design basis for the hybrid IGCC system is sensitive to the assumptions regarding the in-bed desulfurization performance model. Thus, more detailed characterization of the in-bed desulfurization system is merited.

With the new MINLP process synthesis capability, it was possible to solve this problem in one MINLP iteration, which includes one MILP and one NLP (along with one NLP subsystem) iteration. The individual NLP optimization of each alternative would have taken three evaluations to get to the optimal configuration. Furthermore, it was not necessary to specify that in-bed alone cannot meet the SO_2 constraint; the infeasibility of the in-bed only option was detected automatically by the MINLP synthesizer.

Thus, the computationally efficient process synthesis capability described here provides advantages for the identification of optimal design solutions when several alternative flowsheets are possible and many design parameters may vary simultaneously.

5. Conclusion

This paper has addressed the problem of selecting an optimal configuration and design for an advanced IGCC plant featuring hot-gas cleanup systems constrained by environmental regulations. This problem has been formulated as an MINLP optimization problem. Necessary implicit constraints have been made explicit, and a proper decomposition strategy has been identified. A new process synthesis capability developed for the ASPEN simulator has been used to solve the MINLP problem. This marks the first large-scale application of the new process synthesis capability implemented in the ASPEN sequential modular simulator. The case study illustrated the use of the MINLP capability for selection of both optimal flowsheet configuration and optimal design parameters. While the case study confirmed the results of the work of other researchers, it demonstrated the computationally efficient and robust mathematical programming approach afforded by the new process synthesis capability.

In the future, other configurations for advanced IGCC systems will be evaluated. The effect of uncertainty on optimal configurations and designs also will be part of our future research.

Nomenclature

- A_v = availability of the gasifier (fraction)
- C_{elec} = levelized cost of electricity (mills/kW-h)
- $C_{H_2SO_4}$ = annualized cost of sulfuric acid plant (106 \$/year)
- $C_{T-H_2SO_4}$ = annualized cost of electricity without sulfuric acid plant (106 \$/year)
- D = diameter of the zinc ferrite absorber (ft)
- DC_G = direct cost of the gasification process area (\$1000)
- DC_L = direct cost of the limestone handling process area (\$1000)

DC_S = direct cost of the sulfation process area
 E_{SO_2} = total sulfur emissions (lb of SO_2 /MMBtu)
 I_c = Chemical Engineering plant cost index
 L = height of the zinc ferrite absorber (ft)
 L/D = length-to-diameter ratio
 $m_{cm,G,i}$ = mass flow of dry, moisture-free coal into the gasifier process area (tons/day)
 m_L = mass flow of limestone into the limestone handling process area (lb/h)
 m_{lah} = mass flow of spent limestone sorbent and ash from the gasification process area to the sulfation process area (lb/h)
 $N_{O,G}$ = number of operating gasifier trains
 N_T = number of total gasifier trains (includes operating plus spares)
 $P_{H_2SO_4}$ = power consumption in sulfuric acid plant (MW-h)
 $P_{T-H_2SO_4}$ = total power generated without the power consumption in sulfuric acid plant (MW-h)
 $R_{Ca/S}$ = calcium-to-sulfur ratio
 t_a = zinc ferrite cycle time (h)
 $UC_{H_2SO_4}$ = upper bound for $C_{H_2SO_4}$
 $UP_{H_2SO_4}$ = upper bound for $P_{T-H_2SO_4}$
 x = continuous variables, represent flows, operating parameters, etc.
 y = binary variables, denote the existence or nonexistence of process units
 Z = objective function

Greek Symbol

η_s = desulfurization efficiency

Literature Cited

- Craig, K. R. *ASPEN input file: KRW fluidized bed IGCC power plant (hot-gas cleanup)*. U.S. Department of Energy Morgantown Energy Technology Center: Morgantown, WV, June 22, 1988.
- Diwekar, U. M.; Grossmann, I. E.; Rubin, E. S. An MINLP Process Synthesizer For a Sequential Modular Simulator. *Ind. Eng. Chem. Res.* 1992, 31, 313-322.
- Earley, P. I.; Smelser, S. C. "Design and Economics of a Plant to Convert Eastern Bituminous Coal to Pipeline Gas or Power Using KRW Gasifiers with In-Bed Desulfurization". Report No. GRI-86/0166; Gas Research Institute: Chicago, IL, September 1988.
- EPRI. "TAG(tm)—Technical Assessment Guide, Volume 1: Electricity Supply—1986". Report No. P-4463-SR; Electric Power Research Institute: Palo Alto, CA, December 1986.
- Frey, H. C. Probabilistic Modeling of Innovative Clean Coal Technologies: Implications for Technology Evaluation and Research Planning. Ph.D. Dissertation, Carnegie Mellon University: Pittsburgh, PA, May 1991.
- Frey, H. C.; Rubin, E. S. "Stochastic Modeling of Coal Gasification Combined Cycle Systems: Cost Models of Selected Integrated Gasification Combined Cycle (IGCC) Systems". Report No. DOE/MC/24248-2901 (DE90015345); U.S. Department of Energy Morgantown Energy Technology Center: Morgantown, WV, June 1990.
- Haldipur, G. B.; Smith, K. J.; Datta, S.; Cherish, P. Fluidized Bed Coal Gasification With Hot Gas Cleanup. In *Proceedings of the Eighth Annual Gasification and Gas Stream Cleanup Systems Contractor's Review Meeting*; Kothari, V. P., Longanbach, J. R., Eds.; DOE/METC-88/6092 (DE88010253); U.S. Department of Energy Morgantown Energy Technology Center: Morgantown, WV, 1988; Vol. 1, pp 32-44.
- Haldipur, G. B.; Schmidt, D. K.; Smith, K. J. "A 50-Month Gasifier Mechanistic Study and Downstream Unit Process Development Program for the Pressurized Ash-Agglomerating Fluidized-Bed Gasification System, Volumes 1 and 2, Final Report". Report No. DOE/MC/21063-2740; U.S. Department of Energy Morgantown Energy Technology Center: Morgantown, WV, March 1989.
- Kocis, G. R.; Grossmann, I. E. A Modelling/Decomposition Strategy for MINLP Optimization of Process Flowsheets. *Comput. Chem. Eng.* 1989, 13, 797-819.
- Kravanja, Z.; Grossmann, I. E. PROSYN—An MINLP synthesizer. Presented at the Annual AIChE Meeting, San Francisco, 1989.
- Smith, J. T.; Smelser, S. C. "Designs and Economics of Plants to Convert Eastern Bituminous Coal to Methane Using KRW Gasifiers With and Without In-Bed Desulfurization, Topical Report". Report No. GRI-87/0160; Gas Research Institute: Chicago, IL, January 1987.
- Southern Company Services. "Assessment of Coal Gasification/Hot Gas Cleanup Based Advanced Gas Turbine Systems, Final Report". Contract No. DE-FC-21-89MC26019; U.S. Department of Energy Morgantown Energy Technology Center: Morgantown, WV, December 1990.
- Stone, K. R. "ASPEN Simulations of Fluidized-Bed and Entrained Flow Integrated Gasification Combined-Cycle Power Plants". Report No. DOE/METC-85/4027 (DE85013620); U.S. Department of Energy Morgantown Energy Technology Center: Morgantown, WV, July 1985.

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