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ABSTRACT

The U.S. Department of Energy (DOE) is investigating CO_2 recovery from fossil-fuel cycles as a greenhouse gas mitigation strategy. Recognizing this, we compared two integrated gasification combined-cycle (IGCC) plant designs based on the Shell entrained-flow gasifier. One option, called the "co-product case," uses high-sulfur Illinois #6 coal to produce electricity and hydrogen (H_2) as energy carriers. At the same time, 90% of the carbon dioxide (CO_2) is recovered for disposal in geological storage or for use, such as in enhanced-oil recovery (EOR). The second option, called the "base case," is a conventional IGCC power plant releasing CO_2 by combustion of the synthesis gas in a gas turbine. Process design has been aided by the use of the ASPEN-Plus© simulation for critical design areas. Special attention is paid to the transport issues for the CO_2 product, because transportation technology is a determinant of product specifications, which affect plant design. Separating and purifying the H_2 for fuel cell use should yield an impressive gain in overall process efficiency, offsetting the losses in efficiency from recovery and compression of CO_2 to supercritical conditions.

OBJECTIVE: LOW-GREENHOUSE-IMPACT GASIFICATION CYCLES

Plant Design Basis

The Shell (entrained-flow) coal gasification system has been selected as the basis for the co-product plant. The energy and environmental performances of the co-product plant are compared with those of a base-case plant that also uses the Shell gasification technology but produces only electricity as a salable product. The base-case integrated gasification combined-cycle (IGCC) plant and the co-product plant are substantially different in design. The most significant common elements are the use of the Shell gasifier and the consumption of the same amount and type of coal. Principal features and differences are summarized in Table 1.

Shell Gasification-Based Combined Cycle with Hydrogen, Electricity, and CO_2

Figure 1 presents an overview of some of the critical process areas of the co-product plant, clarifying the differences noted in Table 1. The plant is conceptually divided into five main plant areas. Each area consists of a set of related processes. The processes, in turn, consist of equipment or unit operations, and process streams connect these components. A two-digit taxonomy has been adapted for consistency in referring to these plant elements. The first digit designates the plant area, while the second designates the process. Table 2 presents a summary and comparison of the plant performance for a base-case IGCC plant, which is the proposed plant.

Coal Mining, Coal Washing, Transportation, and Preparation

Coal characteristics and the impacts of the coal-preparation circuit appear in Table 3. The mining, coal-sizing, and washing circuits are considered integral to the design of the gasification system. An underground mine near Seeser, Illinois, supplies Illinois #6 coal by using long-wall continuous mining that feeds 4,502 tonnes/day of raw coal to a washing circuit employing a jig, two crushers, three screens, a centrifuge, and a thickener. This set-up provides a more uniform product in the 5 × 1.5-in. (13 × 4-cm) size range with considerable reduction of the ash and modest reductions of pyritic sulfur. Employing this washing circuit considerably reduces the tonnage of coal shipped by rail to the plant because the mining operation brings in roof and floor material. Calculations show that 81% of the energy from the raw coal reaches the product. At the same time, only 65% of the original tonnage of coal needs to be transported and handled. We have assumed that cleaning-plant refuse is returned to the mine. The water use is 38.8 L/tonne of raw coal, and electricity use is 6.4 kWh/tonne of raw coal. As a consequence of shaking and

abrasion, coal losses of 0.05%/100 mi (0.031%/100 km) of rail transport are included. The cleaning delivers a coal (Table 3) with a lower heating value of 26.235 J/kg.

Raw Materials Preparation

A material balance for the major process streams appears in Tables 1–3. The front end of the plant is nearly unchanged through Area 20. Hence, the gasification; heat recovery, particulate removal, and COS hydrolysis follow the base-case performance as originally modeled by EG&G.

11-Coal Preparation

After delivery by unit train, a pulverizing circuit prepares the coal for transport into the gasifier by using hot inert nitrogen from the 12-Air Separation Unit. In pulverizing and transporting the coal, further drying takes place so that a net 2,700 tonnes/day of coal is feed to the gasifier. The coal is combined with steam in transport, but it does not mix with oxygen until the gasifier.

12-Air Separation Unit (ASU)

A cryogenic unit provides 2,320 tonnes/day of oxygen feed to the gasifier at 95% purity. Nitrogen at 2.1% and argon at 2.9% are inert diluents that carry through the rest of the cycle.

13-Water Treatment

Conditioning of raw water for feed to the boiler and gasifier is essential so that steam service maintains a high efficiency. The process consumes 79 tonnes/day of steam as a chemical reagent in the gasification, while a further 145 tonnes/day is consumed in the 31-Shift block. Sour water and blow-down streams also are treated in the plant.

Gasification

21-Gasification

The Shell gasifier receives the dry coal feed into an oxygen-blown, entrained-flow slagging unit that operates at 25 bar. The gasifier exit conditions are controlled by a feedback system on the oxygen so that the exit temperature before quench is 1,371^{o}C. One critical design decision is to employ a gas recycle stream from the 24-COS Hydrolysis block rather than quench the hot raw gas with a water spray. Using the gas recycle stream significantly reduces the water treatment from this system, as contrasted with other commercial oxygen-blown, entrained-flow gasifiers.

22-Heat Recovery and 23-Particulate Removal

The raw gas product has considerable enthalpy that is converted to steam and employed for power generation. Because of the dust loading coming off the gasifier, the design of these sections presents some particularly challenging issues related to materials of construction, fabrication, and heat-transfer. A dust-free raw product gas at 232°C with a minor pressure drop is delivered for 24-COS Hydrolysis treatment.

24-COS Hydrolysis

This section converts the COS produced in gasification to H_2S. It is included in the 20-gasification process block because nearly 30% of the product stream is recycled to the raw gas exiting the gasifier to serve as a quench. Any HCl (and nearly all the ammonia) entering with the raw gas stream is captured in this section and reports to the sour water.

30-Gas Conversion

31-Shift Reaction

The shift reaction uses 145 tonnes/day of steam to convert CO in the gasifier product stream to CO_2 and hydrogen. The reaction takes place in two beds of sulfur-tolerant shift catalyst. The first bed of lower-activity catalyst yields a 76% conversion. The temperature of the shift product from the first stage must be returned to 233°C so that 98% conversion in the second bed is feasible. Because these reactions are exothermic, cooling of the shift product from the two stages provides an additional 4.9 MW of power in the 32-Heat Recovery process block.

40-Gas Separation and Purification

41-H_2S Recovery

Glycol-based absorber-stripper processes for H_2S and CO_2 are commonly employed for gas cleanup. Commercial systems generally employ an optimized mixture of five or more glycols; however, the vendors of these systems have warned that the physical properties data for their mixtures are not well simulated when data in the open literature are used. The current ASPEN 10.2 simulation solely employs tetra-ethylene glycol di-methyl ether ($C_{10}H_{22}O5$) as a surrogate for the commercial mixture. Using this physical solvent and a 25 molar % water mix, more than 98% of the H_2S is captured in this section. This H_2S is recovered for treatment in the 44-Claus process block that will yield a sulfur product. The next stage of glycol-based scrubbing recovers a very high fraction of residual H_2S so that a product specification of 10 ppm H_2S in the

turbine fuel is met. While the glycols are more selective for H_2S than for CO_2, nearly 60% of the CO_2 is captured here.

42-CO_2 Recovery

A second glycol-based absorber-stripper system is employed for polishing so that a total of 90% of the CO_2 is captured for recovery and pipeline transport. After drying, 6,000 tonnes/day of CO_2 is compressed to 143 bar and transported from the plant by using a super-critical pipeline. Commercial experience shows that other species (such as H_2S) are permissible in co-mixtures with CO_2 for injection into underground reservoirs.

43-Pressure Swing Adsorption

This approach is commonly used in the purification of hydrogen. It is a semi-continuous process, which yields 324 tonnes/day of a very high purity hydrogen product, with some minor argon dilution. The blow-down product from this system has a significant heating value and is employed as a turbine fuel for power generation.

50-Power Generation

51-Combustion Turbine; 52-Heat Recovery Steam Generator; 53-Steam Cycle

These process areas are configured so that after the gas turbine (61.95 MW), the heat recovery steam generator employs three steam pressures. Additional output from steam cycle with incorporation of raw gas cooling is 86.63 MW, and low-pressure turbine output from shift system heat is 4.9 MW for a total power generation of 153.48 MW. The plant's internal power requirements are based on this power: -82.4 MW delivering a net of 71.1 MW to the busbar. By examining the power balance over the entire plant, it is clear that most of the power is being exported over the fence as hydrogen.

COMPARATIVE COSTS OF POWER CYCLES

Table 5 includes the results of this assessment for the costs of a Shell IGCC system with CO_2 capture and H_2 generation, including the costs of transportation and CO_2 sequestration in a table showing the comparative costs of several fossil-based and non-fossil-based energy cycles. In 1996, the California Energy Commission (CEC) undertook a broad survey of pricing for various power-generating technologies [CEC, 1996] that was combined with CO_2 inventory data for the same power-generating technologies from the U.S. DOE Energy Information Agency [DOE, 2000]. Consistent with these numbers, a recent EPRI study has compared all the cost estimates

for 90% CO_2 capture systems that also appear in Table 5 [Holt, 2000]. Transportation costs for the CO_2 assume a fully developed infrastructure cost of \$7.82/tonne CO_2 [Doctor, 2001], as compared against first system costs of \$25/tonne CO_2 [Doctor, 1994]. No adjustments were made for the 1996 costs of natural gas because the CEC did not structure its report so that fuel costs could be manipulated separately, but with the necessary adjustments, the costs of turbine combined-cycle systems would be comparable with or higher than those if CO_2 sequestration is included. The costs of sequestration in the field are based on the observation that during the 1999-2000 time period, breakeven for CO_2-flooding EOR required crude prices higher than \$12/bbl oil. If the typical utilization of 5,600 standard cubic feet/bbl of oil is employed, this equates to \$2.14/1,000 standard cubic feet of CO_2 or \$34/tonne CO_2.

CONCLUSION

This process design employs a Shell IGCC cycle in a "Vision 21" multiproduct plant with low greenhouse impact. Hydrogen can be cogenerated with electricity and delivered to consumers at very high purities. The selection of a very high purity hydrogen product stream benefits the high-efficiency performance of fuel cells and yet still meets the internal power needs of the IGCC and yields a revenue stream from electricity sales. The introduction of "shift" to increase the hydrogen content of the gasifier product also benefits the CO_2 recovery, which has inherent cost advantages if it is largely removed before the combustion turbines.

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Table 1. Comparison of Design Basis for Three Power Cycles

Process	Base Case – Electricity	H2 – Electricity Co-Product
Gasification	Shell gasification with cold gas cleanup. Raw gas is produced at 1,006°C and 24 bar.	
Ash removal	This is a slagging gasifier with slag quench.	
Air separation	Cryogenic air separation with partial integration where N_2 used as diluent for combustion turbine	
High-temperature gas cooling/particulate removal	Used to raise high-pressure, superheated steam	Also used for combustion turbine fuel gas preheat
COS hydrolysis	Single stage to form H_2S and CO_2	
Shift reaction	Not applicable	Two-stage shift to convert raw gas to high H_2 and CO_2 content
H_2S recovery	MDEA	Glycol used for improved selectivity (H_2S vs. CO_2)
Acid gas treatment	Claus-SCOT using filtered raw gas as SCOT reagent	Claus-SCOT using H_2 product as reagent
CO_2 removal	Not applicable	Glycol
H_2 purification	Not applicable	Pressure Swing Adsorption
Combustion turbine fuel	Synthesis gas cleaned of sulfur and particulates	Residual gas rejected by PSA
Steam cycle heat source	Gas turbine exhaust	Gas turbine exhaust and heat recovery from shift reaction

Table 2. Comparison of Plant Performance for Three Power Cycles

Item	Base Case – Electricity	H_2 – Electricity Co-Product Case
Coal consumption, tonnes/day	2,877 Coal LHV = 820.1 MW	2,877 Coal LHV = 820.1 MW
Gas turbine power, MW	272.3	62.0
Steam cycle power, MW	188.8	91.5
Internal power consumption, MW	- 48.3	-82.4
Net electricity, MW	412.8	71.1
H_2 production (equivalent MW)	0	423.2 – 100% fuel cell efficiency 275.1 – 65% fuel cell efficiency 194.7 – 46% fuel cell efficiency
CO_2 product, tonnes/day	0	6,000
CO_2 emissions, tonnes/day	6,724	724

Table 3. Coal Mining and Cleaning, Rail Transport and Delivery to IGCC

11-Coal Preparation	As-Received Basis, wt %			Raw	Coal	Coal		Coal
	Raw Coal	Clean Coal	Dry Coal	Coal	to train	to pulverizer		to gasifier
	wt%	wt%	wt%	tonne/d	tonne/d	tonne/d	tonne/d	kg/h
Ultimate Analysis				4,506	2,884	2,877	2,701	112,530.5
Moisture	10.10	11.12	5.33		321	320	144	5,997.9
Ash	26.19	9.70	10.33		280	279	279	11,626.5
Sulfur	3.68	2.51	2.67		72	72	72	3,008.5
MAF Values								
83.15% Carbon	48.984	63.751	67.904		1,839	1,834	1,834	76,412.8
5.87% Hydrogen	3.458	4.501	4.794		130	129	129	5,394.4
1.63% Nitrogen	0.960	1.250	1.331		36	36	36	1,497.9
0.38% Chlorine	0.224	0.291	0.310		8	8	8	349.2
8.97% Oxygen	5.284	6.877	7.325		198	198	198	8,243.2
100%	100.000	100.000	100.000		2,211	2,877	2,701	112,530.5

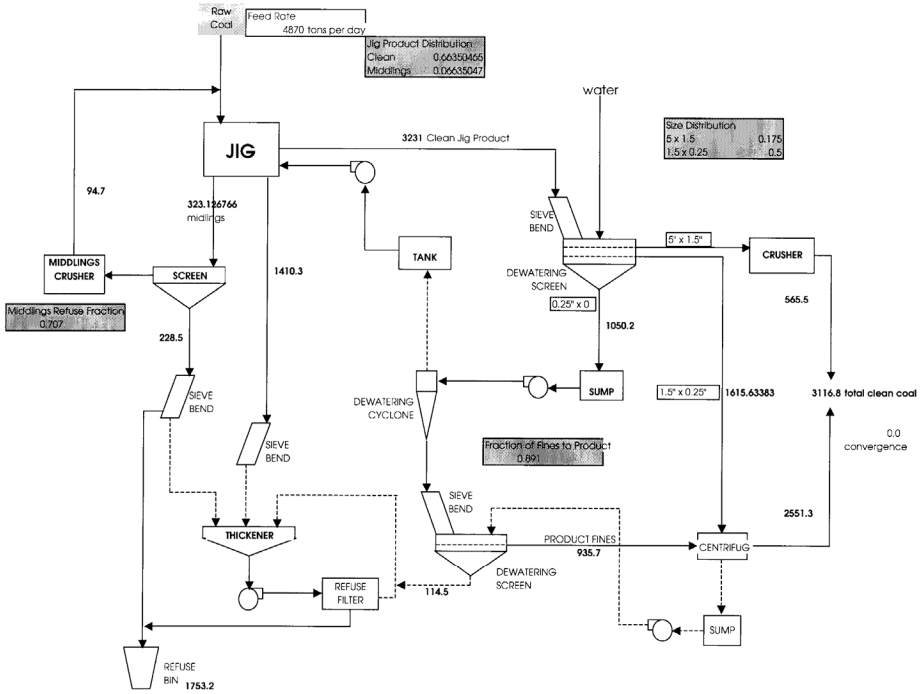

Table 4. IGCC Major Process Streams

21 - Gasification	1	2	3	4	5		6	7	8	
22 - Heat Recovery	Coal feed	Dust from	Nitrogen for	Steam	Oxygen	TOTALin	Slag	Dust	Raw gas	TOTALout
23 - Particulates	to gasifier	recycle	coal transport					to gasifier	product	
Mass Flow kg/hr	(pulverized)	Str #8						Str #2	(dust-free)	
O2	8,243.21	0.00	68.62	0.00	91,446.47	99,758.30	0.00	0.00	0.00	0.00
N2	1,497.93	0.00	8,488.63	0.00	1,752.32	11,738.88	0.00	0.00	11,738.36	11,738.36
AR	0.00	0.00	47.73	0.00	3,509.80	3,557.53	0.00	0.00	3,557.53	3,557.53
H2	5,394.39	0.00	0.00	0.00	0.00	5,394.39	0.00	0.00	5,805.80	5,805.80
CO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	170,278.14	170,278.14
CO2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	9,986.25	9,986.25
H2O	5,997.87	0.00	0.00	3,272.37	0.00	9,270.25	0.00	0.00	3,771.25	3,771.25
CH4	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	63.74	63.74
H2S	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	2,920.32	2,920.32
CL2	349.21	0.00	0.00	0.00	0.00	349.21	0.00	0.00	0.00	0.00
HCL	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	361.57	361.57
NH3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	5.80	5.80
COS	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	481.41	481.41
CARBON	76,412.81	27.55	0.00	0.00	0.00	76,440.37	523.52	27.55	0.00	551.07
SULFUR	3,008.52	0.00	0.00	0.00	0.00	3,008.52	0.00	0.00	0.00	0.00
ASH	11,626.53	611.74	0.00	0.00	0.00	12,238.27	11,623.06	611.74	0.00	12,234.80
Total Flow kg/hr	112,530.47	639.29	8,604.98	3,272.37	96,708.56	221,755.68	12,146.58	639.29	208,970.27	221,756.04
Total Flow cubic M/hr			287.4	263.0	2,813.0				54,236.3	
Temperature C	15.56	337.85	40.00	367.78	95.94		1,371.11	337.85	1,371.11	
Pressure bars	1.0	25.17	27.21	34.01	32.11		25.00	25.17	25.00	

Table 4. IGCC Major Process Streams (Continued)

24 - COS Hydrolysis **31 - Shift** **41 - H2S Glycol** **42 - CO2 Glycol**	8 Raw gas to Hydrolysis	9 Hydrolysis Product	10 Shift Feed	11 Shift Product	12 H2S Glycol Feed	13 H2S Glycol Product	14 CO2 Glycol Clean-Gas	15 CO2 to Sequestration
Mass Flow kg/hr								
O2	1.31E-08	1.31E-08	1.31E-08	1.31E-08	1.31E-08	0	0	
N2	11,738.36	25,878.66	11,738.30	11,738.30	11,738.30	11,021.74	10,224.02	
AR	3,557.53	7,842.30	3,557.19	3,557.19	3,557.19	3,028.15	2,483.73	
H2	5,805.80	12,799.46	5,805.71	18,002.98	18,002.98	18,002.98	18,002.98	
CO	170,278.14	375,399.00	170,277.23	819.51	819.51	751.95	677.64	
CO2	9,986.25	22,772.42	10,329.41	276,588.75	276,588.75	109,285.91	26,661.30	249,927.45
H2O	3,771.25	1,143.78	518.80	564.74	564.74	14.14	6.80	trace
CH4	63.74	140.51	63.74	60.59	60.59	42.31	28.22	
H2S	2,920.32	6,945.58	3,150.45	3,150.44	3,150.44	40.76	0.07	<1% volume
CL2	0.00	0	0	0	0	0		
HCL	361.57	0.25	0.25	0.25	0.25	6.71E-02	7.71E-03	
NH3	5.80	0.02	0.02	0.02	0.02	0	0	
COS	481.41	6.77	6.77	6.77	6.77	7.71E-03	0	
Glycol-C10H22O5	0	0	0	0	0	5.20E-02	1.30E-02	
Total Flow kg/hr	208,970.17	452,928.75	205,447.87	314,489.54	314,489.54	142,188.07	58,084.78	
Total Flow cubic M/hr	54,236.30	11,205.41	13,280.55	13,344.94	11,151.71	9,131.15	7,600.40	
Temperature C	1371.1	37.8	236.3	37.8	-9.4	1.7	-7.2	37.8
Pressure bar	25.0	22.3	31.2	29.9	29.8	29.5	29.1	142.9

Table 4. IGCC Major Process Streams (Continued)

43 - PSA	14	16	17	18	19
51 - Power	PSA Feed	Hydrogen	Turbine Feed	Air	Flue gas to HRSG
Mass Flow kg/hr					
O2	0	0	0	111,379.28	75,158.42
N2	10,224.02	1.02	10,222.99	364,548.01	374,770.83
AR	2,483.73	0.55	2,483.48	6,386.87	8,870.34
H2	18,002.98	29,767.52	4,500.75	0	0
CO	677.64	0.15	677.57	0	0
CO2	26,661.30	5.88	26,658.61	0	27,800.73
H2O	6.80	1.00E-03	6.80	0	40,291.32
CH4	28.22	6.00E-03	28.21	0	0
H2S	6.53E-02	0	6.53E-02	0	0
CL2	0	0	0	0	0
HCL	7.71E-03	0	7.71E-03	0	7.71E-03
SO2	0	0	0	0	1.16E-01
NOx	0	0	0		10ppm
Glycol-C10H22O5	5.90E-03	0	0	0	0
Total Flow kg/hr	58,084.77	29,775.12	44,578.49	482,314.16	526,891.77
Total Flow cubic M/hr	7,600.40	5,129.59	2,491.94		
Temperature C	-7	38	38	21	656
Pressure bar	29.1	34.0	34.0	1.0	1.0

Table 5. Comparative Electric Power Generating Costs (mills/kWh)

	Cost - Base	Cost - CO_2	Transport	Reservoir	TOTAL
Natural Gas with co-gen [1]	41.5	59.5	2.6	11.4	73.5
Hydroelectric	82.0	0.0	0.0	0.0	82.0
IGCC Coal	52.4	65.7	5.7	25.1	96.5
IGCC Coal + CO_2 + H_2 [2]	--	75.9	5.7	25.1	106.7
PC Coal with co-gen	50.5	82.5	6.8	29.8	119.1
Nuclear	125.5	0.0	0.0	0.0	125.5

1. Natural Gas pricing from 1996 assumed
2. Fuel cells @ 65% efficiency; H_2 = $9.00/1000 standard cubic feet to cover transport and

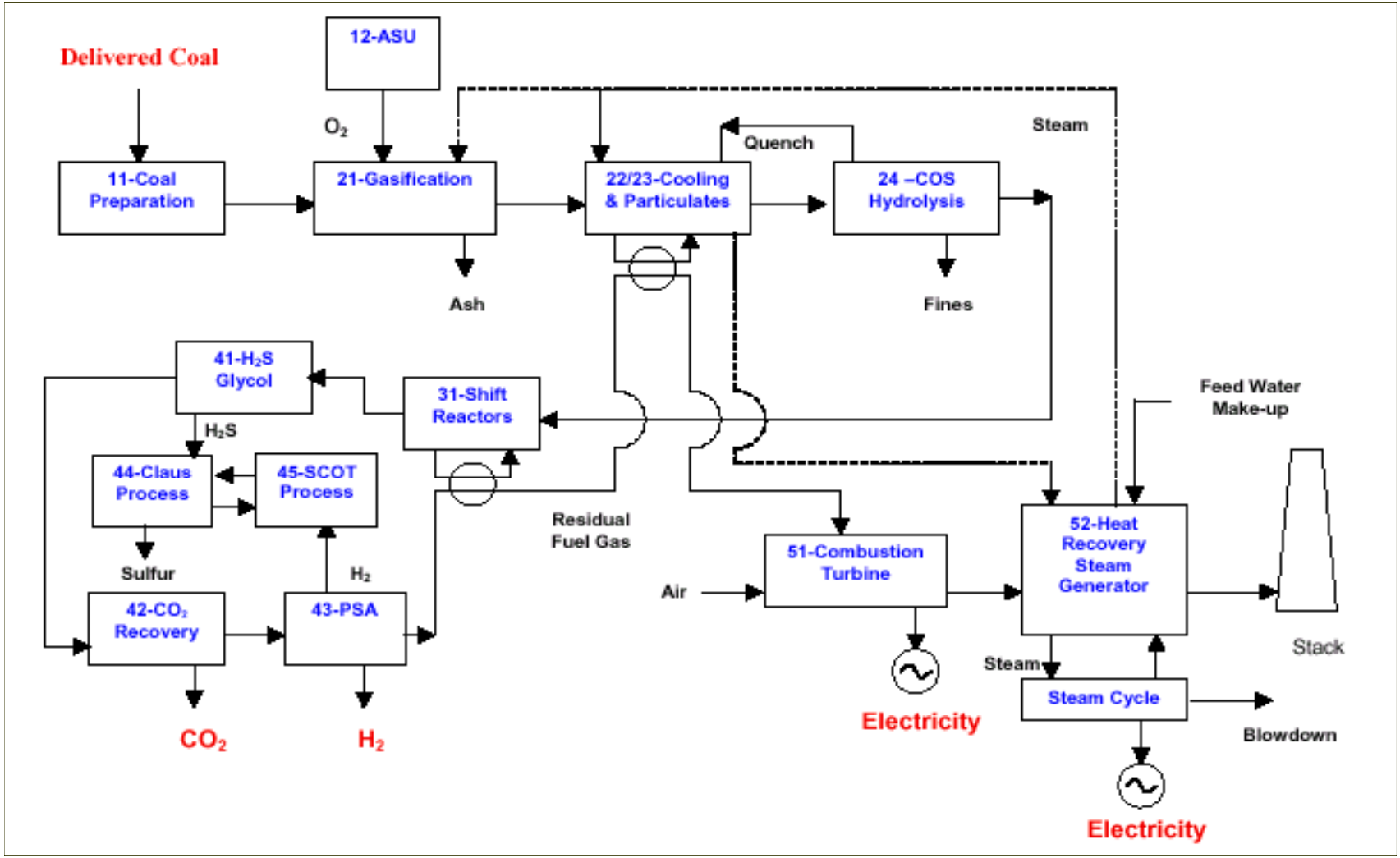

Figure 1. Shell IGCC with Hydrogen and Electricity Production