PRESSURIZED OXY-FUEL COMBUSTION FOR MULTI-POLLUTANT CAPTURE

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SUMMARY

The novel[i] pressurized oxy-fuel approach known at the ThermoEnergy Integrated Power System (TIPS) is a concept designed to produce energy (electricity, steam) and liquid CO_2 from coal with near zero air emission of priority and toxic pollutants. The increased system pressure enables use of gas-to-liquid steam-hydroscrubbing to collect and remove pollutants and recover latent heat from water entrained or produced in the combustion process. The pressurized oxy-fuel approach also enables CO_2 to be recovered as a pressurized liquid through direct condensation. Emerging improvements to oxygen separation technology will significantly improve the economics of oxy-fuel processes.

Pressurized oxy-fuel technology addresses two major issues affecting the future use of the country's coal resource. These are (1) economic capture of criteria and toxic pollutants (such as mercury) from the diverse power and steam generators needed, and (2) economic capture of CO_2 from the larger power and steam generators used by utilities and large industrial facilities. Decades of research and billions of dollars have been invested in developing coal gasification as a means to address these issues via the Integrated Gasification Combined Cycle (IGCC) technology and this approach is taken as the base for comparison of the proposed oxy-fuel alternative, TIPS. In contrast to IGCC, TIPS requires fewer unit operations and can use proven oxygen sensor instrumentation for process control. It is intrinsically more reliable technology on which to base the nation's coal-to-steam-and-power future.

Economics for IGCC and TIPS are similar for new large utility plants using high-rank coals as a fuel. TIPS has superior projected economics for all other cases we have examined including: sub-bituminous and lignite fuel, retrofit of existing coal plants, smaller power plants (5 to 100 MW), and industrial steam plants. TIPS provides the captured CO_2 product as compressed liquid or solid (dry ice) ready for beneficial use or sequestration.

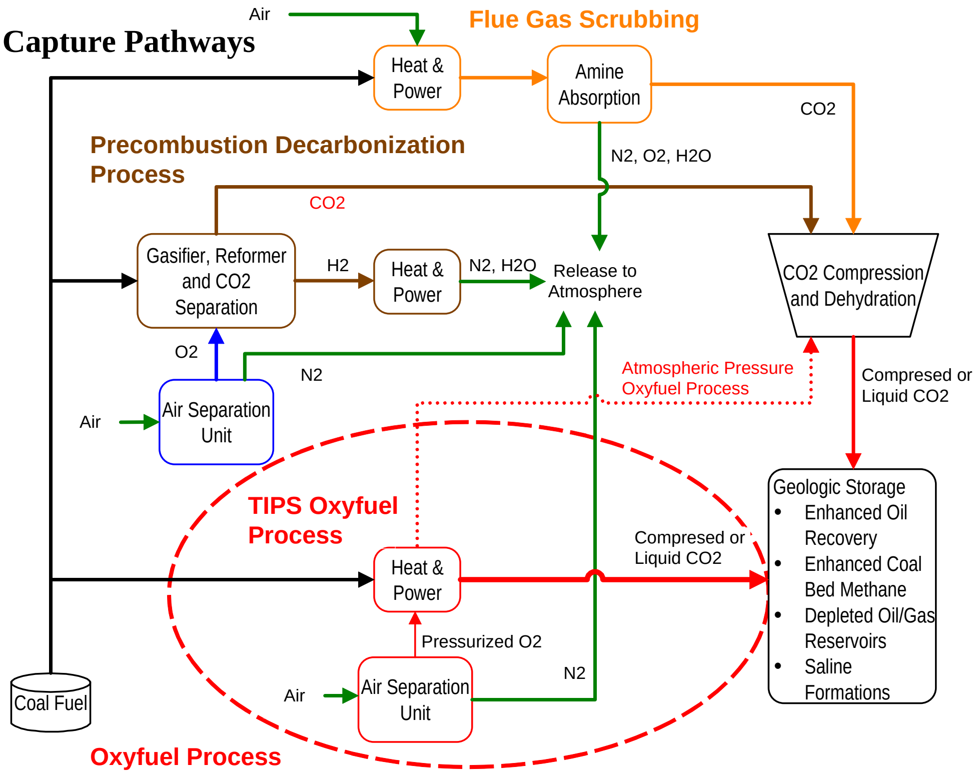
INTRODUCTION

The three basic methods for capturing CO_2 are shown in the diagram from the CO_2 Capture Project, revised for coal based power systems in Figure 1.[ii] Pressurized oxy-fuel combustion takes a direct path to capture CO_2 without the formation of intermediate chemicals or solvent solutions. In comparison, conventional post-combustion flue gas scrubbing uses a solvent solution, and pre-combustion decarbonization creates and separates hydrogen prior to combustion. Neither post combustion scrubbing nor pre-combustion decarbonization recover the latent heat of vaporization of the entrained or produced water in the exaust. The latent heat losses are particularly significant for low-rank, high-moisture coal.

TIPS PROCESS DESCRIPTION

TIPS process combustion takes place at pressures between 4.83 and 8.96 MPa. (700 and 1300 psia) Increasing the pressure of combustion shifts the temperature at which water, CO_2 mercury and acid gases condense. The elevated pressure and condensation temperature process conditions enable TIPS to utilize heat transfer, mass transfer and liquid vapor equilibrium regimes well suited to capture of

Figure 1. CO_2 Capture Pathways



pollutants and CO_2. Elevating the pressure enables TIPS to use the phenomenon of nucleate condensation at temperatures in the range of 262 to 303°C (503 to 577°F) in a heat exchanger that simultaneously recovers heat and condenses and captures pollutants. Two components of pressurized oxy-fuel technology central to achieving the twin goals of efficiency and pollution control are the air separation plant to provide oxygen under pressure, and the condensing heat exchanger to capture both pollutants and heat from the combustion gases.

Energy Efficiency Of The High-Pressure Oxygen Supply

Pressurized oxy-fuel firing of coal eliminates energy lost in the exhaust nitrogen by eliminating the nitrogen and recovers the latent heat of vaporization of both the produced and entrained water. The energy cost of separating air and pressurizing the oxygen is roughly 20% of the total energy contained in the coal.[iii] Therefore, improvements to air separation technology are especially significant to oxy-fuel processes and fortunately new improvements are now entering commercial development. In comparing the components of TIPS with a modern pulverized coal power plant, the capital cost of a conventional air separation plant used by TIPS is slightly less than the capital cost of the conventional desulfurizing unit for the pulverized coal plant.[iv]

Condensing Heat Exchanger Recovers Heat And Captures Pollutants

The TIPS configuration enables one simple device, the condensing heat exchanger, to collect particulates, acid gases and mercury into a condensed phase that is roughly 2,500 to 3,500 times smaller than the volume of gas treated by conventional atmospheric pressure flue gas clean-up systems. The nucleate condensation phenomenon is so rapid and so efficient that, once developed, the TIPS process may be less costly than current atmospheric pressure systems requiring particulate collection, desulfurizers, de-NOx and mercury abatement equipment.

Figure 2 shows a flow schematic of a coal-fired TIPS process. The large colored numbers 1 through 4 on Figure 2 correspond to the large colored numbers and the pressure and temperature conditions shown in Figure 3. The curves in Figure 3 correspond to the pure component liquid vapor equilibrium lines for mercury, water, sulfur dioxide and carbon dioxide. The curves show the effect of increasing the pressure on condensation temperature of the main exhaust gases. The higher pressure corresponds to a higher liquid-vapor equilibrium temperature for the gaseous components of the exhaust. TIPS makes use of this pressure-induced temperature shift to enable the recovery of heat, condensation of pollutants and removal of particulates from the exhaust gases. Position 1 shows the point at which a dirty exhaust gas would exit the radiative section of the pressurized combustion and heat transfer unit. Liquid water sprayed into this gas rapidly evaporates and cools the exhaust gas to position 2. The large surface area of the droplets in the liquid spray provides rapid heat transfer. Although the mass of the gas stream, exhaust gas plus evaporated water, is increased, the volumetric flow decreases due to the cooling of the gas. The water spray cools the exhaust gas and rapidly moves the temperature and pressure of the combined stream to position 2. Position 2 is at the liquid-vapor equilibrium line for water at the system pressure. The heat energy that was in the hot exhaust gas was not lost during the cooling process. It was transformed into the latent heat of vaporization of the evaporated water. This moisture laden exhaust stream in the current example then goes into a condensing heat exchanger at a temperature of 302°C (575°F) and a pressure of 8.8 MPa (1,276 psia). This heat exchanger transfers the heat from the moisture-laden exhaust to the boiler feed water. The boiler feed water is substantially cooler, typically ~27°C (80°F) than the temperature at which the moisture in the exhaust gas will condense. The large temperature difference between the boiler feed water and the moisture in the exhaust gas drives rapid heat transfer to condense the water in the exhaust gas and rapidly and efficiently to heat the boiler feed water. The temperature of the moisture laden exhaust gas stream, 302°C (575°F) does not change significantly as it transfers heat to the boiler feed water until the bulk of the water vapor has been condensed to liquid. This enables the heat exchanger in the current example to maintain a temperature difference suitable for efficient heat transfer. The exhaust gas is cooled below the liquid vapor equilibrium temperature to position 3. At position 3, the bulk of the moisture and energy has been removed from the exhaust gas and the remaining gas, primarilly CO_2, is further cooled to position 4. At position 4, carbon dioxide in the exhaust gas has condensed into a liquid.

When steam or carbon dioxide are cooled and condensed into a liquid, surface forces cause the condensing gas to preferentially condense on solid or liquid surfaces. When the liquid water and carbon dioxide gas condense, the liquid material preferentially goes onto the surfaces of particles to make big droplets. Big droplets with little seed particles in them are easily removed. This phenomenon of nucleate condensation or steam hydroscrubbing exhibits very high particle capture efficiencies.[v] [vi]

Use of Commercial Process Control and Unit Operations

The pressurized oxy-fuel system benefits from two key combustion control elements. For a given flow of oxygen and coal, the CO_2 recycle and water concentration of the feed coal-water slurry can be varied to adjust the flame temperature independent of the coal and oxygen flow ratio. Increased pressure and decreased flame temperature favor CO_2 production and inhibit the back reaction of CO to C. This offers the possibility for minimizing soot formation.[vii] In an oxidizing environment, the high-pressure and low-temperature condition in the condensing heat exchanger favors SO_3 and NO_2 formation.[viii] These oxidized compounds react with water to form sulfuric and nitric acid and are readily scrubbed from the CO_2 gas. The second key element is that robust high-temperature oxygen sensors are a proven technology that will allow combustion process oxygen concentrations to be monitored and controlled on a real time basis. Control of comubustion conditions assures a fully oxidized exhaust gas and allows downstream equipment and refractories to operate at constant conditions.

Most of the unit operations required to implement TIPS are proven in existing industrial applications or under active development. Cryogenic air separation technology is known and research is underway to improve its efficiency as well as develop advanced ion-transport membrane oxygen separation technology. The combination of known techniques, equipment and unit operation with the novel TIPS configuration make it an attractive and simple alternative to complex Integrated Gasification Combined Cycle (IGCC) schemes.

COMPARISON OF PRESSURIZED OXY-FUEL AND IGCC WITH CARBON CAPTURE

IGCC with carbon capture and TIPS share elements of similarity along with striking differences. Figure 4 shows a schematic of an IGCC configuration designed for carbon capture. A description and mass and energy balance of this IGCC configuration is in a comparative report by Argonne National Laboratory.[ix] Figure 4 shows the lowest cost, most efficient IGCC configuration presented in this report. It was oxygen fired and used glycol for both hydrogen sulfide and carbon dioxide recovery and it is used as the reference case for comparison with TIPS. Comparing Figures 2 and 4 show the relative simplicity of TIPS and the relative complexity of IGCC. Table 1 provides a comparative list of major operations used by IGCC with and without carbon capture and TIPS.

The conclusion we draw from these figures and tables is that TIPS is a combustion system that burns fuels to make steam and power in a way that allows capture of pollutants and CO_2. IGCC with carbon capture is a chemical plant that converts coal into intermediate chemicals, processes and refines those chemicals and then burns the chemicals in a combined cycle power plant. The facts on the ground and recent energy and chemical price changes support the view that the refined chemical produced by IGCC is often more valuable to subsequent chemical processes than the electricity that can be produced by its combustion.

Table 2 compares TIPS with IGCC with and without carbon capture under conditions favorable to IGCC – high-rank coal and a large utility power plant. The TIPS case was assumed to use an existing state-of-the-art high-temperature multiple reheat steam turbine. The IGCC case reported in the Argonne study was developed before the recent revisions to gas turbine firing temperatures by General Electric. According to a recent report by the Electric Power Research Insititute, (EPRI) examining several studies on IGCC with carbon capture:[x]

> “In order to maintain blade temperatures similar to those in NGCC service (where GE offers Long Term Service Agreements (LTSAs)) they have reduced the firing temperature. This adversely affects both gas turbine and steam turbine performance so that the heat rates are typically 200-400 Btu/kWh higher than those reported in prior years.”

Table 1. Operations List of IGCC With and Without Carbon Capture with Pressurized Oxy-fuel Combustion (TIPS)

	Case>	IGCC No Carbon Capture	IGCC With Carbon Capture	Pressurized Oxy-fuel
	Units	**Base**	**Case 1**	**TIPS**
Base Unit Operations				
Gasifier Oxidant		Oxygen	Oxygen	Oxygen
Sulfur Recovery		Glycol	Glycol	H2SO4 condensation
CO2 Recovery		none	Glycol	Direct condensation
Topping Cycle		Turbine	Turbine	None
Bottoming Cycle		Steam	Steam	Steam
Air Separation Plant	tons/d	2,347	2,347	8,294
Oxygen Compressor	tons/d	2,347	2,347	8,294
Air Compressor	tons/d	none	none	none
High Temp Heat Exchangers		yes	yes	yes
Condensing Heat Exchanger		none	none	yes
Water/Gas Shift Reactor		none	yes	none
Steam for WGSR	tons/d	none	2,875	none
Recycle Gas Fan		none	none	yes
CO2 Compressor - 10 bar		none	yes	none
CO2 Expander	MW	none	none	**10.9**
Claus Plant		yes	yes	none
Additional Unit Operations				
SCR NOx Treatment Plant		needed	needed	none
Sulfided Carbon Gas Filter		needed	needed	none
Liquid Treatment		none	none	needed
Waste Disposal				
Ash Disposition		landfill	landfill	Cement
Sulfur Disposition		Elemental S	Elemental S	SA or Gypsum
Spent Carbon Filter/Hg		landfill	landfill	none
Liquid Sludge		none	none	landfill

The following adjustments were made to the IGCC cost and performance estimates in Table 2:

- A selective catalytic reactor (SCR) was added to remove NOx generated by the IGCC system to make its envrionmental performance closer to that projected for TIPS.
- A CO_2 expander was added to TIPS to expand the produced CO_2 to the same pressure as the CO_2 exiting the IGCC case.
- The 1996 cost data were updated to 2005 using the Engineering News Record construction index.
- The estimate heat rates reported by Argonne were corrected for lower gas turbine operating temperatures by adding 300 Btu/kWh to the previously estimated heat rate.

Table 2. Comparison of IGCC With and Without Carbon Capture with Pressurized Oxy-fuel Combustion (TIPS)

	Case Units	IGCC No Carbon Capture Base	IGCC With Carbon Capture Case 1	Pressurized Oxy-fuel TIPS
Component*				
Base Plant Capex	$/kW	$ 1,332	$ 1,485	$1,496
CO2 Capex	$/kW	$-	$202	
NOX Capex		$110	$110	
Total Capex	**$/kW**	**$ 1,442**	**$ 1,797**	**$1,496**
Cost Component*				
Base Plant Capex	$/kW	$ 1,730	$ 1,928	$1,943
CO2 Capex	$/kW	$-	$262	$-
NOX Capex		$143	$143	$-
Total Capex	$/kW	$ 1,873	$ 2,334	**$1,943**
Total Capex - GE LTSA Spec.	**$/kW**	**$ 1,940**	**$ 2,416**	
Coal Energy Input	MMBtu/hr	3,839	3,839	3,839
Gross Power Input	MW	458	446	458
Gross Power Input - GE LTSA	MW	444	434	
In Plant Power Use	MW	44.7	68.9	102
Net Plant Output w/o SCR	MW	413.5	377.5	n/a
Net Heat Rate w/o SCR	Btu/kWhr	9,284	10,170	n/a
Therm Eff. HHV w/o SCR	%	36.8%	33.6%	n/a
SCR parasitic loss est.	MW	4.00	4.00	n/a
Net Plant Output w SCR	MW	410	374	**366**
Net Heat Rate w SCR/equivalent	Btu/kWhr	9,375	10,278	10,479
Therm Eff. HHV w SCR	%	36.4%	33.2%	32.6%
Heat rate addition - GE LTSA	Btu/kWhr	300	300	
Actual heat rate estimate	Btu/kWhr	9,675	10,578	
Actual Net Plant Output w SCR	**MW**	**395**	**361**	**366**
Actual Overall Efficiency	**%**	**35.3%**	**32.3%**	**32.6%**

* ENR Ratio 1996 to 2005 = 1.29858

The single largest parasitic loss for both IGCC and TIPS is the air separation unit. The data in Table 2 are based on the use of a standard cryogenic air separation plant that has a specific performance of 279 kWh/tonne of 93-95% oxygen. Since TIPS uses substantially more oxygen than IGCC, the parasitic load to generate this oygyen in comensurately higher and this is reflected in the in-plant energy use shown in Table 2. Further, TIPS has only a single thermodynamic cycle and IGCC has two. However, IGCC loses energy in the unburned carbon remaining in the ash, in the unrecovered heat from water added to the synthesis gas before it passes through the water gas shift reactor, from the water added as a diluent to the hydrogen fuel fed to the gas turbine and the latent heat of vaporization of the water formed during the combustion of hydrogen and finally in the sensible heat of the nitrogen and excess oxygen leaving the Heat Recovery Steam Generator. In addition, there is significant pumping energy used in the circulation and refrigeration of glycol for hydrogen sulfide and CO_2 recovery. Another small chemical loss is the hydrogen lost with the hydrogen sulfide and ammonia. Due to the intrinsic energy losses in the water-gas shift process and the fuel dilution for the gas turbine, refrigeration for the glycol system, the maximum energy potential the combined cycle has to work with is actually less than the lower heating value of the fuel. The losses at each step are small but there are many steps and the losses add up. The lowering of the temperature rating of the gas turbine in the combined cycle by GE also reduces the overall efficiency.

In contrast to IGCC, TIPS recovers the latent heat of vaporization of the produced, entrained and added water in the condensing heat exchanger and has no diluent nitrogen in the final exhaust. This allows the TIPS thermodynamic cycle to utilize the higher heating value of the fuel with minimal thermal losses. As a result, the net efficiency of IGCC with carbon capture is essentially the same as TIPS with intrinsic carbon capture, even under conditions favorable to IGCC. The advantage for TIPS lies in its simplicity and lower capital and operating cost. TIPS does not require costly catalysts and does not need sulfided activated carbon sorption for mercury capture. The ash and/or slag from TIPS will be fully combusted and retain little or no carbon and will be suitable for use in cement manufacture.

EFFECT OF TECNOLOGY ADVANCES IN AIR SEPARATION

Emerging Ion-Transport Membrane (ITM) technology is poised to revolutionize oxygen separation. Recent research results show that the ITM technology is steadily proving out its potential and the Department of Energy and major air separation companies estimate that capital and energy costs for oxygen separation plants will be reduced by roughly 35% compared to the traditional cryogenic process.[xi] Since TIPS uses substantially more oxygen than IGCC, the relative effect of ITM technology on the economics and performance of TIPS plants will be significant. Table 3 incorporates the capital and operating savings projected for the ITM technology into the basic cost and performance economics for the same example shown in Tables 1 and 2. The cost and efficiengy of both technologies improve but the relatively larger effect of ITM on TIPS is apparent.

Table 3. Effect of Air Separation Technology Capital Cost and Energy Use Projections on IGCC with Carbon Capture and TIPS

	Case Units	**IGCC With Carbon Capture Case 1**	**Pressurized Oxy-fuel TIPS**
Total Capex	$/kW		**$1,631**
Total Capex - GE LTSA Spec.	**$/kW**	**$ 2,136**	
Actual heat rate estimate	Btu/kWhr	10,345	9,942
Actual Net Plant Output w SCR	**MW**	**369**	**386**
Actual Overall Efficiency	**%**	**33.0%**	**34.3%**

COMPLEXITY, SCALE AND RELIABILITY

Despite recent improvements, one of the significant perceptions regarding IGCC is that that it is complex and unreliable. The DOE Policy Office lists reliability as the number 1 factor in why IGCC (without CO_2 capture) has failed to gain significant inroads into the power sector.[xii] As indicated in Figure 4, IGCC with CO_2 capture is significantly more complex than IGCC without CO_2 capture and additional complexity is likely to exacerbate industry concerns regarding reliability. While CO_2 capture adds complexity to all power plant configurations, it adds the least complexity to the pressurized oxy-fuel or TIPS approach. TIPS has significantly fewer and simpler unit operations, less complexity and greater intrinsic reliability. This is specifically the case when comparing TIPS with IGCC with carbon capture because unlike other gasification applications such as Fischer-Tropsch or Haber-Bosch chemical production, IGCC with carbon capture for power production has no storage capability for intermediate products. It is not realistically feasible to store meaningful quantities of either synthesis gas or hydrogen. This means every unit operation of the entire IGCC with carbon capture process train must be on-line for the system to operate and any shutdown of any un-spared component can shutdown the entire system. This highly integrated series of chemical processes is where complexity detracts from reliability.

Economically successful petrochemical facilities are typically very large. There are few economically viable small refineries or basic chemical plants. The same economic driving forces that apply to these plants apply to IGCC with carbon capture facilities due to their similar nature. In contrast, the fewer and simpler unit operations required by pressurized oxy-fuel offer more favorable scale economics. Air separation plants are manufactured in a variety of sizes and the new ITM systems are, by their nature as ceramic membranes, amenable to smaller scale. Interest in TIPS for combined heat and power located in non-attainment areas is growing as the price of natural gas increases.

FUEL FLEXIBILITY

The high moisture and relatively high hydrogen content of low-rank coal produced by a number of large western coal mines make it a good candidate for pressurized oxy-fuel firing. In high-moisture, low-rank western coal, latent heat and exhaust gas losses amount to 18 - 20% of the total energy contained in the coal.[xiii] Pressurized oxy-fuel firing of coal eliminates nitrogen and recovers the latent heat of vaporization of both the produced and entrained water and is well suited to use the abundant supply of western coal supplies. A recent study from EPRI compared several IGCC technologies and several coal sources and concluded:[x]

> "IGCC for Low Rank Coals – Need for Gasification Improvements
>
> With these estimates the current E Gas IGCC does not appear to compete with PC plants for PRB coals and lignites. Most IGCC studies have been based on using bituminous coals. The entrained flow gasifiers of Texaco, Shell and E Gas all perform better with the lower ash lower moisture bituminous coals. Given the abundance and low cost of US resources of low rank coals such as Powder River Basin (PRB) and the Texas and North Dakota Lignites there is a great need to improve the performance of IGCC with these coals."

CONCLUSIONS:

While detailed studies are not yet available for the performance of TIPS pressurized oxy-fuel technology under a wide range of applications, preliminary estimates presented here indicate that in a carbon capture scenario:

- TIPS performance comparable to IGCC for high-rank coal and superior for low-rank coal,
- TIPS simplicity enhances process reliability and lessens dependence on scale,
- Proven oxygen monitoring instrumentation offers the oxy-fuel approach convenient, real-time process control of system chemistry that, at present, has not been developed for IGCC,
- Implementation of advanced air separation technolgies, while improving both IGCC and oxy-fuel technologies, have a greater positive effect on the oxy-fuel approach.

Future utilization of coal resources for heat and power will require a technology capable of capturing CO_2 as well the priority and toxic pollutants associated with coal utilization. To be effective, this technology must be cost competitive with existing technologies and able to use low rank as well as high rank coals. The TIPS pressurized oxy-fuel technology described here is a strong candidate to meet these requirements that merits closer examination.

Figure 2. Pressurized Oxy-Fuel Power Plant Schematic
ThermoEnergy Integrated Power System (TIPS)

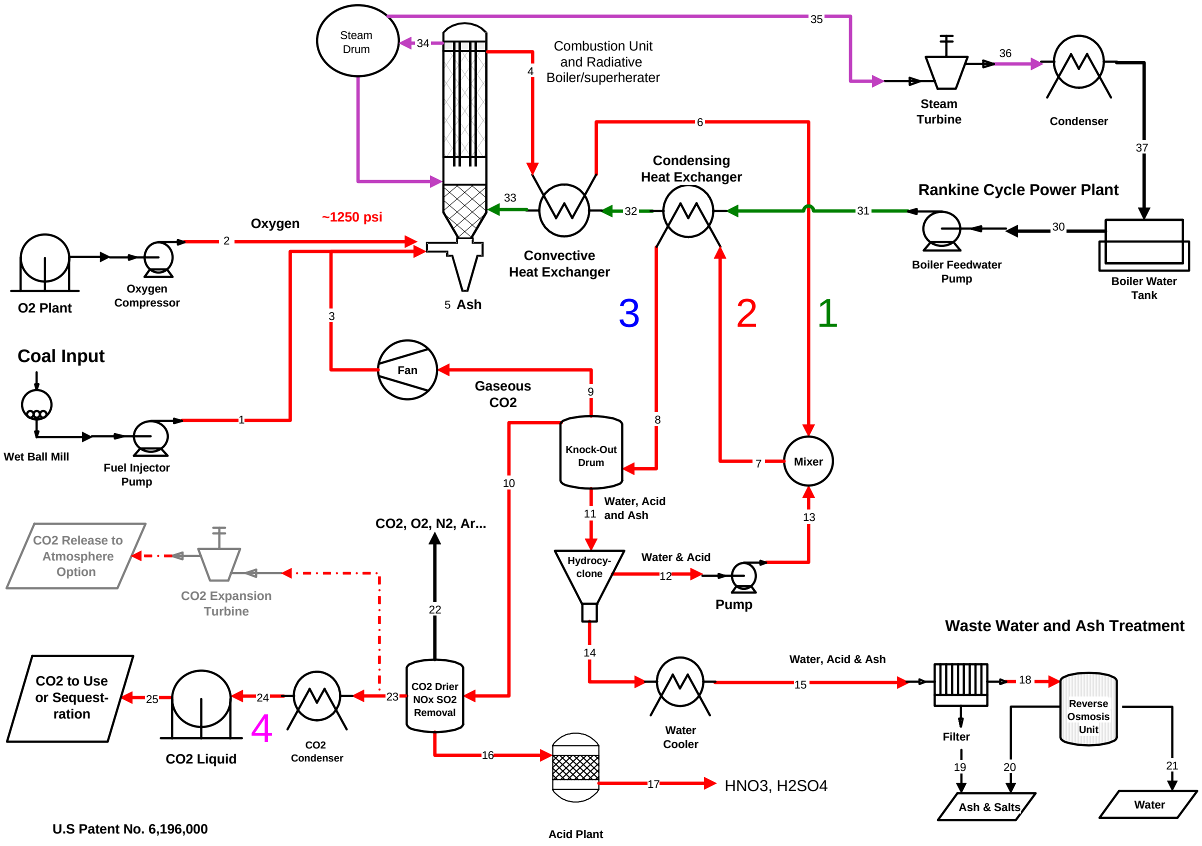


Figure 3. Pure Component Liquid-Vapor Equilibrium

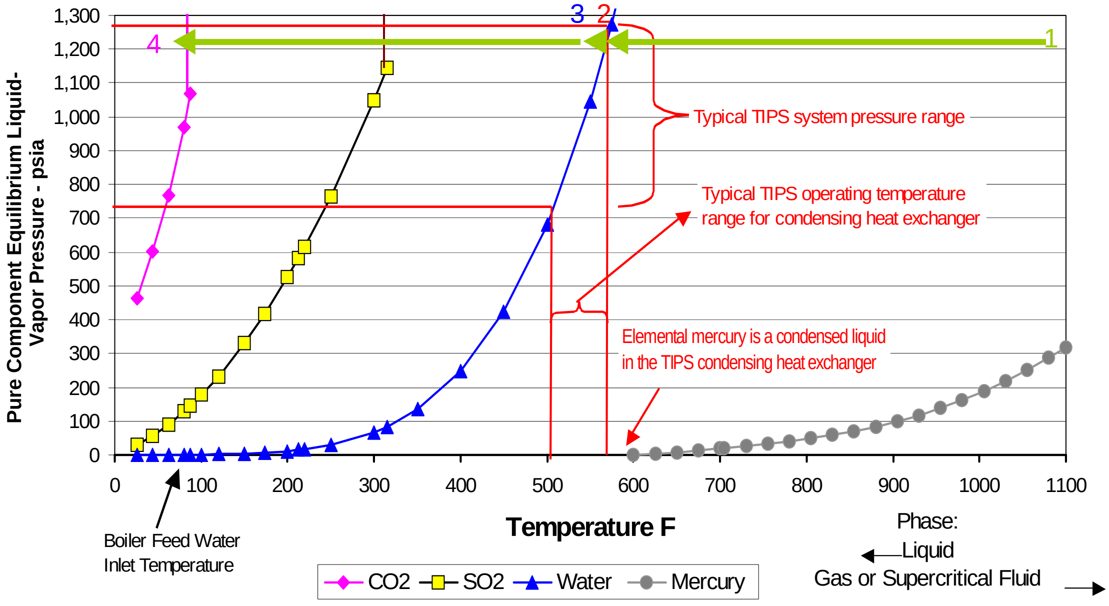
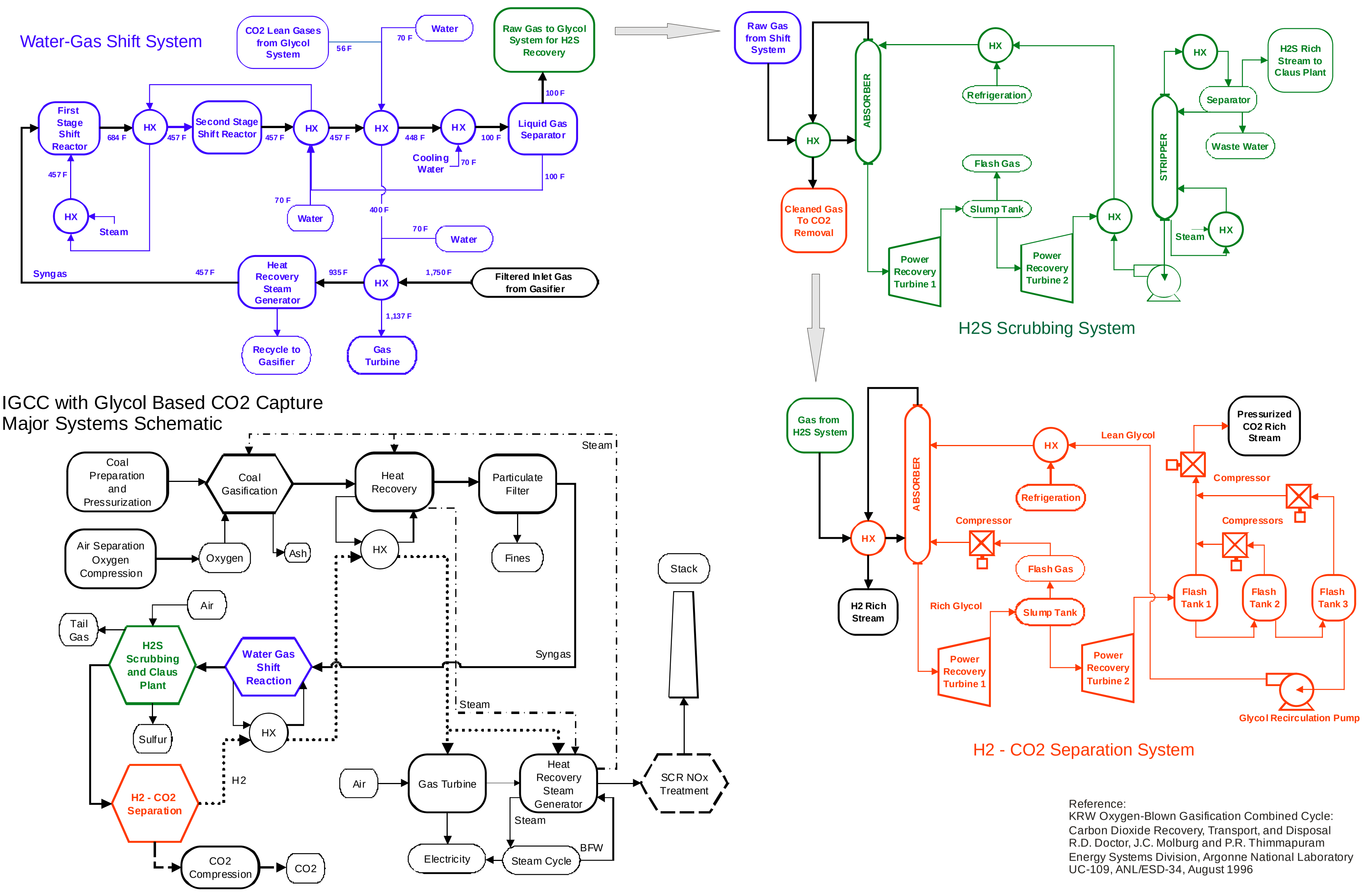


Figure 4. IGCC With Glycol Based CO2 Capture – Major Systems Schematic



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[viii] Ibid. pp 206 and 212.

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