The Integrated Pollutant Removal (IPR) Process for CO_2 Capture

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ABSTRACT:

A number of methods have been proposed to achieve near-zero emission power production using leading edge technologies.   The IPR method emphasizes the use of existing unit processes combined with available low-tech modifications to eliminate greenhouse gases from coal based power plants. The approach using a combined oxy-fuel/IPR process removes nitrogen from air for combustion, separates the solid combustion products from the gases and vapors, and can capture the entire vapor/gas stream for sequestration leaving near-zero emissions.

The IPR system can be applied to retrofit, repowering, upgrading, and greenfield plants. The actual application will depend on the context of the installation including fuel, location, local regulations, incentives, power plant design, power plant condition, rights-of-way, and transportation access.

BACKGROUND:

Governments are feeling pressure to find solutions to the problem of Global Warming. In spite of the uncertainty in the specific impact of Global Warming on local aspects of climate and the contribution of anthropogenic influences, the view of most scientists is that the Earth is heating up. The influence of green house gases will be debated for the near future, but it is important for the power generation sector to have available alternatives that reduce greenhouse gas output.

A typical modern sub-critical 400 MW coal burning power plant uses approximately 3,500mt/day of coal.   That coal produces approximately 40,000 mt/day of flue gas of which approximately 28,000 mt/day is hot N_2. The remaining combustion products include approximately 8,400 mt/day of CO_2, 2,000 mt/day of H_2O, 180 mt/day of SO_2, and 350 mt/day of ash (see Table 1).   A similar size oxy-fuel power plant burning the same coal produces the same amount of ash, CO_2, H_2O, and SO_2 with only approximately 44 mt/day of N_2. This reduces the flow of flue gas from an oxy-fuel combustion system to less than 12,000 mt/day due to the virtual elimination of N_2 flow.   Because of the reduction in total flow and the specific reduction in N_2, an oxy-fuel system is a good combustion system to feed into an IPR post-combustion system.

The IPR process first separates the solid combustion products from the gases and vapors. It then captures all, or most, of the gas/vapor stream for sequestration leaving near-zero emissions.   The IPR system captures combustion products, including CO_2, in a cascading series of related unit processes. The products of the IPR system are a neutralized acidic water stream containing particulates and particle-bound mercury, and a compressed

stream of CO_2 with some O_2, Ar, and N_2. High-purity CO_2 can be produced through an additional unit process if needed. If SO_2 can be tolerated in the compressed gas stream the neutralization unit process can be removed, lowering both capital and operating costs.

IPR takes a low-technology approach by using well proven standard industrial compression and scrubbing components and applying them in a new context for the capture of flue gas in power generation. IPR has used an oxy-fuel combustion system to promote combustion and concentrate the CO_2, H_2O and pollutants in the combustion product stream. To improve thermal efficiency, heat is recovered from the flue gas cooling and water removal process and compressor intercooling and recycled back to preheat the feed water or to provide other process heat.

Table 1: Comparison of a 400 MW computer model of an Oxy-fuel/IPR system with a computer model of a typical standard boiler exhaust. The yellow (first numeric) column contains information about the combustion products from a conventional boiler. The columns in blue contain information about combustion products from an oxy-fuel source going through the process of IPR.

	Conventional after economizer	Oxy-fuel/IPR Exhaust			
		After splitter	After 1st compression and condensation	After 2nd compression	After 3rd compression
Gas Flow (kg/hr)	1,716,000	449,400	377,000	367,900	367,900
Vol flow (m³/hr)	1,932,000	539,000	53,000	11,193	757
Inlet Pressure (kPa)	100.8	104.8	422	1,716	9,977
Inlet Temperature (°C)	132	260	28	24	29.6
Density (kg/m³)	0.8882	0.8334	7.09	32.87	486
H_2O (%)	8.32	32.43	0.90	0.17	0.04
Ar (%)	0.88	3.87	5.75	5.79	5.80
CO_2 (%)	13.68	59.84	88.93	89.59	89.70
N_2 (%)	73.42	0.51	0.75	0.76	0.76
O_2 (%)	3.50	2.46	3.66	3.69	3.70
SO_2 (%)	0.20	0.89	0.00	0.00	0.00

There are numerous proposed approaches to the recovery of CO_2 from flue gas[1]. The IPR system uses existing chemical or mechanical engineering components found in industries such as petrochemical, chemical, or process gas supply. Computer models[2] and experimental studies[3] of the application of these processes to the capture of multiple pollutants and CO_2 are refining the real-world design of the Integrated Pollutant Removal process. Recent research has shown that the CO_2 product can contain substantial impurities and still be suitable for sequestration schemes. In addition, since pipeline transport does not require liquid CO_2, the CO_2 can be delivered as an impure supercritical fluid. The relaxation of requirements for CO_2 purity and the removal of the need for liquefaction have greatly simplified the design and reduced both the complexity and cost of the system.

For engineering estimations, standard commercial software (GE GateCycle®)[a] has been used to provide proven algorithms for component simulations. This modeling work has focused on the distribution of energy throughout a 400 MWe power plant and the recovery of IPR process energy back into the power production cycle. The models show that the thermal efficiency found in current power plants can be approached in a plant with an IPR exhaust system.

Recent experimental work on the exhaust gases has focused on the removal of acidic gases in the beginning of the process and determination of the composition of the final CO_2–rich stream. Multiple thermodynamic simulation packages have been used to predict vapor behavior, however, experimental results better show the effects of contaminants on CO_2 properties near the critical point.

CAPTURE OF ALL POLLUTANTS:

As the IPR system has gone through its development stages the initial approach to capture 95% - 99% of CO_2 and to capture a significant portion of the associated pollutants[2] has changed to a goal of capturing all CO_2 and pollutants present in the exhaust of a power plant.   There will be some loss of CO_2 and pollutants during startup and shutdown, but during normal operation all of the combustion products will be captured in multiple product streams. Along with this 100% capture goal is the associated goal of reduction in both capital and operational costs.

The reason for the change from partial capture to total capture is the realization that CO_2 sequestration methods are, generally, more tolerant of tramp gases than was originally thought.   Due to the flexibility in the purity of the delivered CO_2, the energy associated with pollutant removal can be reduced and the cost of separation equipment can be, mostly, eliminated.   As an example, the original design of the IPR system produced 99.5% CO_2 as the pipeline product.   Tramp gases including N_2, O_2, and Ar were separated out in a unit process step that was capital intensive as well as energy intensive. That final separation step is now optional and the end user can determine if they require pure CO_2 or if they can deliver a compressed supercritical CO_2 with tramp O_2, Ar, N_2 to the sequestration site.   This decision will be sensitive to the site location and local regulations.   This final separation step is not an engineering consideration since the separation of these tramp gases from CO_2 has well known unit process solutions. This decision is now strictly determined by the economics of the specific installation and the tolerance of the sequestration method.

Capture of all tramp gases along with the CO_2 provides an incentive to keep the excess O_2 level as low as possible (as close to stochiometric combustion as possible) to minimize the amount of O_2 that is produced for combustion support and then is sent on to sequestration.   Since O_2 is being produced with an energy penalty to the process it is important to understand the combustion system to minimize O_2 use.   This is much more

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[a] The USDOE neither endorses nor recommends specific products. References to product names are for information purposes only and should not be interpreted as endorsement or recommendation.

of a concern with an oxy-fuel system than with an air fired system. If a final separation step is involved, the gases separated from the CO_2 product will be rich in O_2 and can serve as enriched feed for the air separation unit (ASU).

In some cases, the sequestration process can accept SO_2 in the mixture.   Initial tests involving geological sequestration where the CO_2 reacts with formations of magnesium rich deposits show that the addition of SO_2 in the reacting mixture will not interfere with the reaction of CO_2.   There are indications that the SO_2 might actually speed up the reaction under some conditions.   Sending a gas mixture containing SO_2 through a high pressure pipeline has a number of potential problems associated with it including toxicity, corrosion potential, and potential environmental issues.   However, if the location is such that SO_2 can be sequestered with the other gases it will result in a considerable cost savings by avoiding the costs associated with the removal of SO_2 from the exhaust. Consider the advantage of being able to squeeze all of the exhaust of a plant into a pipe and send it off for sequestration. The only pollutants left at the site for treatment and disposal will be ash, slag, and contaminated water that is condensed out of the flue gas stream.   The water will contain standard pollutants and will be treated using existing technologies to produce water meeting EPA standards.
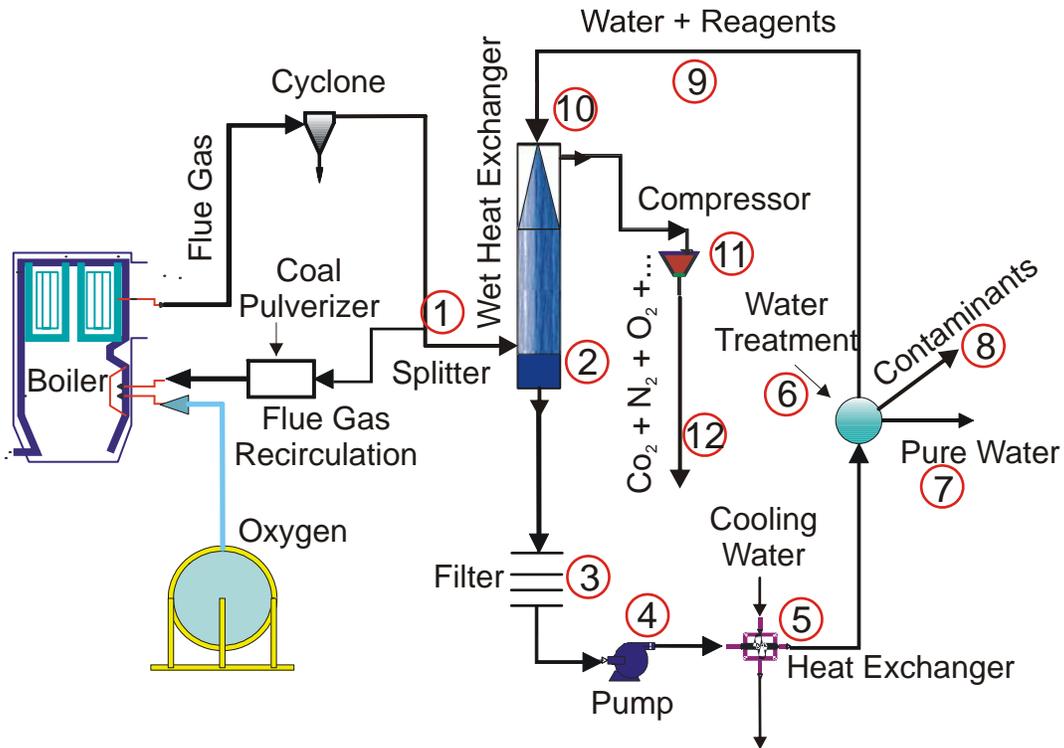

Figure 1: Overview of a simplified oxy-fuel/IPR system.

A LOW TECHNOLOGY APPROACH:

The IPR system is relatively simple, using only technologies that already exist in other contexts.   As can be seen in Figure 1, the IPR system consists of a flue gas cooling and

cleaning system (similar to existing scrubbing system) and then staged compression with energy recovery during intercooling.   The components of a system like this can be found in refineries, chemical plants, or petrochemical plants.   There is nothing unique about the components. It is the use in power generation and the emphasis on energy recovery that is different.

In Figure 1, the individual processes involved in the IPR approach can be seen (and are numbered) starting with the splitter:

1. The splitter divides the flue gas stream into a recycle stream which goes back toward the boiler and a treatment stream that goes on for processing.   Splitting of gas streams using ducts and dampers is a well known technology.   Measurement of stream volume (or mass) is not easy, but is done reliably in a number of process industries.   The splitter is used to ensure that the volume recycled into the boiler can maintain heat transfer in retrofit systems as well as keep flame temperatures within the design range.
2. The split portion of the stream which does not return to the boiler passes counterflow through a direct contact heat exchange where the hot fluid and the cooling fluid come into direct contact. If SOx is being removed, then the spray will contain a reagent to react with the SOx and form a salt for later removal, similar to the process in existing spray FGD systems. In the IPR system, the spray also serves to wash out particulates that are left in the stream.   Water vapor will initially condense at this point in the system.
3. The water portion of the stream then is passed through a filter to remove particulates.   This can use standard filters as found in common applications including power generation.
4. Pumps are used along the circuit to overcome friction and keep the pressure high enough to effectively spray fluid into the wet heat exchanger.   One pump is shown in this simplified diagram but there will be multiple pumps in the circuit.
5. An indirect heat exchanger (one where the hot fluid and the cooling fluid do not come into contact) is used to cool the recirculating spray water.   This exchanger is used to drop the temperature of the combined cooling water and water condensed from the exhaust gas to a temperature lower than the temperature achieved in the direct contact wet exchanger. Lowering the temperature more will allow more sensible cooling in the direct exchanger.   The materials used in this heat exchanger will be subject to an acid environment and will have to be designed to avoid corrosion problems.   The pH of the loop will be higher than that found in many chemical industry situations and the pressure will be relatively low.   There are indirect heat exchange systems available today that will be likely to meet the requirements for this application.
6. Water treatment takes place in the circulating loop. The main stream is treated to meet chemical requirements for recycling into the direct heat exchanger. A bleed stream is taken off the loop at this point, balancing the amount of water condensed in the direct contact heat exchanger.   Methods of water treatment that are used to polish discharge water from a number of process industries can be used in this environment.

7. The bleed stream is further treated to become a pure water stream meeting local discharge regulations. This water is treated using conventional methods such as reverse osmosis and/or ion exchange to remove contaminants.
8. Contaminants removed in step 7 are available for disposal.   This is similar to effluent streams from existing power plants, chemical plants, or other industrial sources.   Removal and disposal of these contaminants is common practice. There will normally be a fee associated with disposal of these contaminants.
9. Reagents are added to the water circulating towards the direct contact heat exchanger. If the direct contact heat exchanger is used to remove SOx then conventional reagents similar to those used in existing scrubbers will be used. If SOx will be sent on through the system for sequestration then the reagents added are to polish parameters such as pH.
10. Water is injected into the direct contact heat exchange system as a spray moving counterflow to the incoming exhaust gas. As is typical with spray absorbers, the water injection is designed to have a high surface contact area between the water and the exhaust gas.
11. The counter-flowing exhaust gas has lost moisture by the time it gets to the end of the direct contact heat exchanger and is then compressed in a series of stages. Only one compressor is shown here but an actual system will have multiple stages of compression.   It is important to note that a substantial portion of the compression work can be recovered through the use of cooler power plant working fluids (such as feedwater) to exchange heat with the compressed gases thereby adding heat to the working fluid at the same time we are improving the efficiency of the compressors.
12. The compressed gas can contain substantial tramp gases at this point. Indications are that this lower purity CO_2 (85% - 90%) stream is acceptable for many sequestration methods.   However, if a higher purity CO_2 (95% - 99%) stream is needed, an additional separation stage can be added at this point in the system.

As can be seen from Figure 1 and the narration above, there are no portions of the IPR system that require any new equipment or processes. This process uses existing technology and does not push the limits of any components or materials.   The IPR design is based on using plentiful US coal in standard PC boiler configurations to keep the cost of electricity competitive.

IPR can also be teamed with IGCC to produce a compressed CO_2 stream.   In this paper we are using the approach in which thermal efficiency is held approximately static at 34% in return for a dramatic reduction in emissions. The IPR approach can be available in the near term, can be maintained using existing technology, uses low maintenance components, and is designed to use the most available fuel in the US.

It is generally assumed that high-efficiency is synonymous with reduced CO_2 emissions. This is only true if the CO_2 is not captured. In like manner, if the generation method can use an abundant fuel such as PRB coal, the small increase in fuel consumption is not as important.

FUEL SAVINGS BY INCREMENTAL IMPROVEMENT IN THERMAL EFFICIENCY:

There is an interesting issue related to the incremental improvement in thermal efficiency. In fact, a one percent change in thermal efficiency at a lower thermal efficiency is more effective in saving fuel than the same one percent improvement in thermal efficiency at a higher base thermal efficiency. The reason for the difference can be seen in Figure 2. Instead of the relationship between heat rate (the number of BTUs required to produce one kWh) and thermal efficiency (the ratio of chemical fuel heating value to the power output) being a straight line it is curved with the curve flattening out at high heat rates (lower thermal efficiencies).
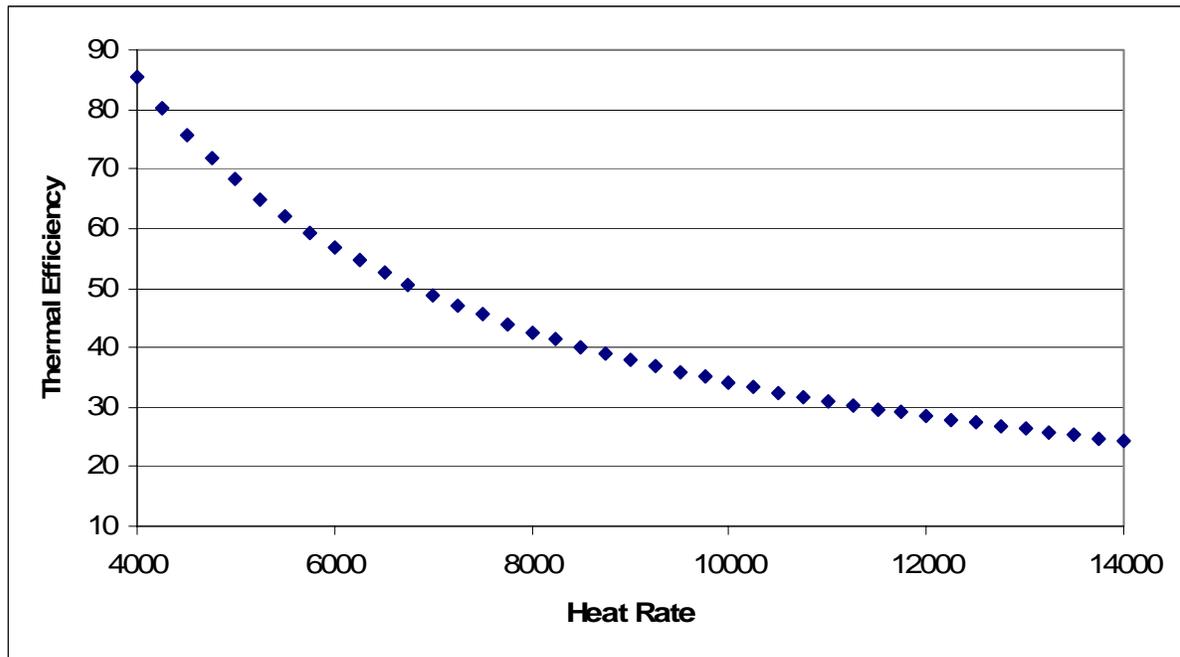

Figure 2: Relationship of heat rate (BTU/kWh) to thermal efficiency (%)

What Figure 2 shows us is that thermal efficiency changes slowly at high heat rates resulting in a larger change in fuel consumption per unit change in thermal efficiency.

This is illustrated in more detail in Figure 3 where it can be seen that the change in heat rate at low thermal efficiency (stepping from 18% thermal efficiency to 19% thermal efficiency saves approximately 998 BTU/kWh) is significantly higher than a 1% increase in thermal efficiency at a higher efficiency (improving from 55% thermal efficiency to 56% thermal efficiency saves approximately 111 BTU/kWh).

This is not to say that we should not be striving to improve thermal efficiency. It is to point out that there are two issues involved when improving thermal efficiency. The first is that it is more difficult to improve thermal efficiency as we get to higher numbers. At high efficiencies the way improvement is realized is by pushing the limits of materials

and processes.   That makes improvement at already high thermal efficiencies more costly. The second issue is that each equal increment of improvement in thermal efficiency at the higher efficiencies saves less fuel than an equal increment of improvement at a lower efficiency.

Consequently, we should be looking at improving the performance of the lower efficiency systems to get the most fuel savings per unit of improvement.   Admittedly, choosing 18% thermal efficiency versus 55% thermal efficiency is stretching the realm of credulity.   However, there are systems operating at the upper teens in the developing world.   There are also systems trying to move into the mid-fifties for thermal efficiency in advanced systems.   The bottom line is that improving that developing country system running at 18% by 1% will incrementally save approximately 9 times the fuel that would be saved by improving the advanced system running at 55% thermal efficiency by 1%. The observation for our project is that we should be working hard to improve the thermal efficiency of systems using IPR capture in the realm of 31% for a sub-critical system to 34% for a supercritical system as the baseline efficiency since it will have significant dividends in fuel savings.   The modern systems with a higher baseline thermal efficiency can tolerate slightly higher losses in thermal efficiency.
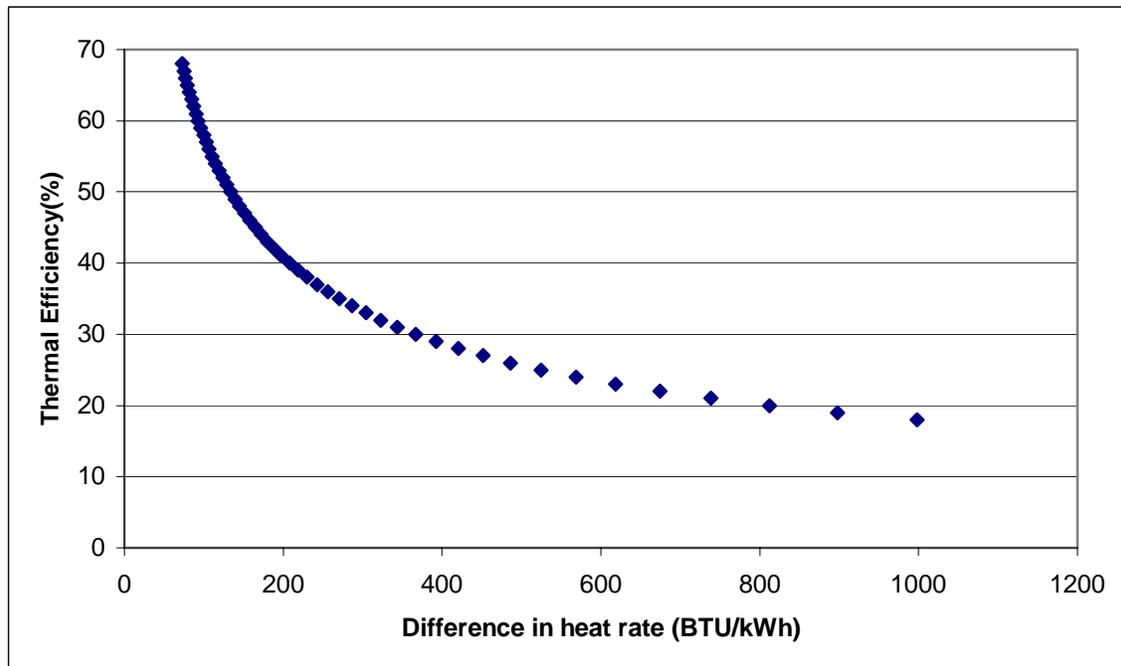

Figure 3: Change in heat rate(BTU/kWh) as a function of thermal efficiency (%).

ESSENTIAL CONCEPTS OF AN IPR SYSTEM:

- High CO_2 concentration in the source,
- Volume change with pressure,
- Moderate thermal efficiency, and,
- Recovery of energy into process fluids.

The IPR system relies on a concentrated CO_2 source.   This is different from the systems using solvents to remove the CO_2 from a less concentrated stream[4,5].   One of the simplest methods of producing a high CO_2 concentration in a combustion product stream is through the use of oxy-fuel (combustion of a hydrocarbon fuel with oxygen or oxygen enriched recycled combustion products).   The IPR system is a post-combustion processing system and can be used with alternative sources of CO_2 rich combustion products.   One aspect of the IPR approach is that the chemistry of the incoming stream is different from the chemistry we are used to dealing with in a standard PC boiler.   In the case of oxy-fuel, the chemistry and physics of the flame are notably different from that of a standard air-coal flame.   The heat transfer characteristics in the boiler can be changed significantly by having CO_2 and water vapor in the boiler at much higher concentrations than those found in a standard PC boiler.   Since CO_2 and H_2O are strong absorbers and emitters in the infrared, they change the radiant heat transfer from the flame to the boiler walls.   The difference in gas molecular weight also changes the temperature of the hot gases in the boiler, resulting in differences in both the radiant and convective zones when working with retrofit boiler systems.

As can be seen in Table 1, the combustion products from a typical 400 MW power plant are high in N_2 and contain approximately 14% CO_2.   In an oxy-fuel source corresponding to numbered position 1 in Figure 1, the N_2 content is seen to be less than 1% while the CO_2 content is near 60%.   Looking at the difference between the air-fuel combustion (yellow column in Table 1) and the oxy-fuel combustion (first blue column in Table 1) it can be seen that there are a number of benefits to the oxy-fuel exhaust including:

- a dramatic reduction in N_2 (which results in a corresponding reduction of NOx),
- a significant increase in H_2O (which can be condensed out),
- a high concentration of CO_2 (which we are capturing), and,
- a greatly reduced mass of exhaust gas (which reduces the size and cost of processing equipment).

One of the most remarkable effects of the IPR system is the reduction of volume of the combustion products.   Using Table 1, we can compare the volume of combustion products from a standard boiler with the volume of the combustion products at the exit from the IPR process.   The change from a volume of 1,932,000 m^3/hr to a volume of 757 m^3/hr is a ratio of approximately 2,500/1.   That translates to an ability to post-process this stream for pollutants such as elemental Hg with equipment that is 2,500 times smaller.

Another approach in the IPR system that differs from other pollutant removal systems is the recovery of energy from all sources in the system.   As an example, feedwater is used to cool (indirectly) the water used for spray in the wet heat exchanger.   As another example, the compressors can be intercooled using feedwater.   The cost of energy lost in compression will be important for all CO_2 capture systems and particularly those similar in nature to IPR.   Approximately 1/3 of the energy used for compression can be

recaptured and used to generate electricity.   This can make the difference between a process that is approximately 28% efficient and a process that is approximately 33% efficient.

Throughout the history of power generation, progress has been made by recovering small increments of energy.   Without taking advantage available energy through thermodynamic processes such as feedwater heating and steam reheating the thermal efficiency of power plants would be well below 30%.   However, the industry has continued to make progress a few tenths of a percent at a time.   We should carry on the tradition by saving energy where we can in the power cycle.   There have been studies of CO_2 capture that have not used any heat recovery from compression or condensation. Those studies show artificially low thermal efficiencies and demonstrate the need for heat recovery to lower the fuel consumption.
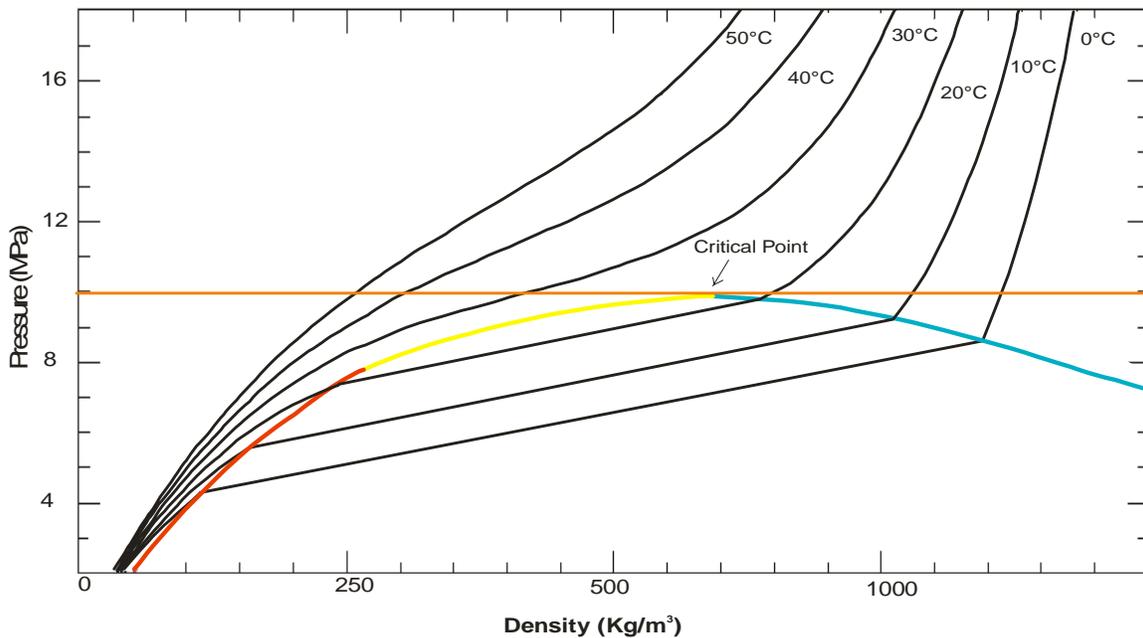

Figure 4: Liquid vapor relationship in compressed CO_2 (90%), Ar(5%), O_2 (4%)N_2(1%) system. Red curve represents saturated vapor line, blue line represents saturated liquid line, yellow curve represents interpolation due to instability of approximation methods.

THE ARGUMENT FOR THE DELIVERY OF IMPURE SUPERCRITICAL CO_2:

Earlier versions of the IPR system relied on high-pressure (2,500 psig – 5,000 psig) for CO_2 delivery.   The purpose of the high pressure was to produce as much liquid CO_2 as possible for a pure CO_2 sequestration product.   As research continued it became clear that we could use a less pure CO_2 stream under supercritical conditions instead of a liquid stream of pure CO_2 for some sequestration technologies.   For typical mixtures of CO_2, O_2, Ar, and N_2 found in IPR flue gas, the pseudo-supercritical pressures are in the range of 1,500 psig to 2,200 psig.   Approximations for mixed fluid behavior can be calculated using the NIST REFPROP[6] software.   An example of the behavior of a typical oxy-fuel IPR gas mixture can be seen in (Figure 4).   The lower the pseudo-supercritical pressure

of the mixture, the less energy can be used in compression.   In order to prevent 2 phase flow in the mixture, we want to stay above the pseudo-critical pressure.   In the case of the mixture shown in Figure 4, the pseudo-critical pressure is less than 10 MPa (1,450 psi).   The actual pseudo-critical point for a mixture like this is hard to calculate due to the differences in the critical points of the individual components.   The approximation of ternary mixtures of CO_2, N_2, and O_2, has been shown to be close to the approximation of a binary mixture of CO_2 and any one of the other constituents (O_2, N_2).

As the concentration of the tramp gases increases the pressure needed to stay above the pseudo-critical point (to avoid two phase flow) increases. However, it is not expected that the fluid will be composed of more than approximately 20% of tramp gases (and probably considerably less as seen in the last column of Table 1. At 20% we can keep the pressure below 2,200 psig and be able to avoid two phase flow.   At a composition similar to that shown in Table 1, the pressure can stay as low as 1,500 psig. If we can deliver impure CO_2 and can include SO_2 as well as NOx in the mixture, much of the energy penalty for compression goes away since we need to compress for pipeline delivery anyway.   The only penalty is the energy cost for O_2.   Another advantage to delivering impure CO_2 is that the process of gas separation can be avoided.   This reduces the capital investment and reduces operating costs.

SUMMARY:

The IPR approach to capture CO_2 uses existing technology in a new context.   Through the principles of high CO_2 concentration in the feed, combustion product volume reduction, and energy recovery, the IPR system can be retrofitted to existing plants or serve as the back end on next generation combustion systems.   The product of the IPR system can be a raw stream containing a supercritical mixture of predominantly CO_2 with associated Ar, O_2, and N_2, or a purified stream of CO_2 (depending on the tolerance of the sequestration method).   This approach is a viable method for application if incentives for CO_2 sequestration become available.

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