Proof of Concept for Integrating Oxy-Fuel Combustion and the Removal of All Pollutants from a Coal Fired Flame

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The USDOE/Albany Research Center and Jupiter Oxygen Corporation, working together under a Cooperative Research and Development Agreement, have demonstrated proof-of-concept for the integration of Jupiter's oxy-fuel combustion and an integrated system for the removal of all stack pollutants, including CO_2 , from a coal-fired flame. The components were developed using existing process technology with the addition of a new oxy-coal combustion nozzle. The results of the test showed that the system can capture SOx, NOx, particulates, and even mercury as a part of the process of producing liquefied CO_2 for sequestration. This is part of an ongoing research project to explore alternative methods for CO_2 capture that will be applicable to both retrofit and new plant construction.

BACKGROUND

The use of fossil fuel for the production of electrical power has traditionally been viewed as being at odds with environmental requirements. There is no fundamental reason why electric power generation must cause pollution. The underlying reason for pollution has always been economic. Given enough resources, any competent mechanical or chemical engineer can demonstrate a completely pollution-free device for generating electrical power. However, it is not clear that all of the devices in such a system would generate a net power surplus, let alone be capable of generating power in an economically competitive environment. The production of electric power with near-zero emission of pollutants and greenhouse gases is the subject of intense research throughout the world.

The USDOE/ARC (Albany Research Center) approach to a viable method for the net-positive, economical generation of electric power using fossil fuels includes a capture technology that requires an oxy-fuel fired combustion system. Jupiter Oxygen Corporation has developed a proprietary patented oxy-fueled combustion method applied to power generation boilers. The oxy-fuel combustion gas streams are much easier to treat with the USDOE/ARC technology than are conventional boiler flue gases. The two organizations are now cooperating on combining the two approaches into an integrated fossil fueled net-positive power generating system with near-

zero emission of pollutants and with thermal efficiencies near 33% - the equivalent of baseline conditions.

Jupiter's oxy-fuel combustion process employs a relatively simple concept in which substantially pure oxygen for the combustion process (in the absence of air) is used to oxidize the fuel. There are several important advantages of using an oxygen gas mixture other than air:

- Lack of nitrogen in the gas mixture reduces NOx
- Increased O₂ content reduces the mass of combustion products
- Recirculation of flue gas offers the potential to recover substantial amounts of heat otherwise lost up the stack
- O₂ content can be adjusted to change flame temperature and control radiant heat transfer

The IPR (Integrated Pollutant Removal) system as developed by ARC is also a relatively simple concept. The basis of the process is the separation of condensable vapors (such as water and CO_2) from the non-condensable gases (such as O_2 and N_2) using compression and cooling. In the process, pollutants such as SOx, NOx, and particulates are removed with the condensate streams.

Cooperation between USDOE/ARC and Jupiter Oxygen Corporation has been on-going for approximately two years under a CRADA (Cooperative Research and Development Agreement). These groups have worked together to develop a framework for integrating the two concepts into a single system. Computer modeling has shown that the two component sections of the system should work together as a system with near-zero emissions. However, previous experimental work was limited to the design and testing of small subsets of the total system.

Previous computer modeling work indicated that the system could be constructed using existing technology applied in novel ways, and that no breakthrough technologies would be required. Additionally, none of the components would require extensive re-design work. The system could be assembled using standard chemical engineering and mechanical engineering processes adapted in unique ways. Following the system modeling process, there appeared to be no fundamental obstacles to building a functioning system, and it was determined that a demonstration apparatus could, in fact, be assembled using 'off the shelf' components and standard engineering practices. However, as is the case with many theoretical systems and computer models, the reality usually presents unforeseen challenges and surprises.

In August of 2004, Jupiter Oxygen Corporation issued a challenge to USDOE/ARC to help design and build a functioning Oxy-Fuel/IPR system within three months using technology which could be readily acquired. ARC agreed and the two organizations put together an approach in which both teams used ingenuity and standard components to rapidly put together the complex of systems. The resultant flurry of activity produced a successful demonstration of the combined technologies on November 3rd, 2004.

TECHNICAL APPROACH

The ARC has been modeling IPR systems since 2000 using the commercially available power plant modeling software GateCycle¹. While other organizations have built custom models using a more fundamental methodology, the ARC approach has been to model system components using industry standard software. When less well-developed processes are involved they can be modeled either off-line or through the use of macros within GateCycle. The advantage of using the GateCycle modeling tool is that we can be sure of reliable responses from the bulk of the power plant common equipment such as steam turbines, feedwater heaters, economizers, and other conventional equipment.

For components that are not found in the typical power plant we went to the petrochemical industry, the petroleum refining industry, the air liquefaction industry, and the chemical industry. There we found a number of processes that can be applied directly to the IPR and oxy-fuel systems.

Jupiter Oxygen Corporation has developed their oxy-fuel burner systems for over 8 years. The experience they have gained in oxy-fuel systems has been coupled directly with the ARC experience in boiler design, operation, and maintenance to enable the design of both retrofit applications and new boiler designs. The two organizations have been working together for two years to develop a better understanding of this new application and the most effective way to apply it in the power generation industry. Between the working knowledge of these two organizations and the expertise available throughout the other process industries it required little engineering design time to be able to put together a bench-scale (250,000 BTU/hr thermal) proof-of-concept system. At these low flow rates, it was easy to find off-the-shelf component equipment capable of sustained operation. When the exact equipment specifications were not available, the designs were modified to accommodate existing equipment. This unique approach allowed the rapid design and construct a working bench-scale system.

DESIGN

Design work consisted of the following six phases, which are discussed in more detail below:

- 1. Initial design was based strictly on flow estimates from a scaled-down power plant computer model.
- 2. Critical portions of the system were identified and simplified without compromising the fundamental functionality in order to make it a valid test bed.
- 3. Commercially available equipment was identified that approximated the calculated flows of the initial system.
- 4. Complex portions of the system were simplified to make it easier to construct without impacting the validity of the demonstration.
- 5. Instrumentation and sampling systems were identified to ensure the appropriate and accurate measurement of experimental parameters.
- 6. Connections, valves, piping, safety equipment, auxiliary equipment, and layout were designed based on the components and instrumentation.

¹ Commercial products mentioned in this paper are neither endorsed nor recommended by the USDOE. The USDOE does not endorse or recommend commercial equipment, processes, or software.

Initial designs for the system came from a GateCycle computer model of a 400 MW power plant (Figure 1). The model included the steam generator as well as the power production turbines and their auxiliary equipment. It was clear that it was not necessary to produce power with the proof-of-concept system since the goal was to show that IPR could be successfully integrated with oxy-fuel systems. After closer examination it was determined that it was not even necessary to generate steam. Our concentration was on capture of the combustion products after they had given up most of their energy through heat transfer to the power plant working fluid. The mechanism for the transfer of heat to the working fluid was not critical for this particular experiment. Based on the decision to eliminate the steam cycle the design became more focused.

The second estimation of the system flows came from the GateCycle computer models (Figure 2) of a 250,000 BTU/hr coal combustion system with an attached IPR system. This system lacks any parts of the steam cycle. Instead the steam and water flowing in the system are being used to cool the combustion products (as happens in a steam generator). The GateCycle software is good at modeling the initial combustion products; however, there are no commercial software systems that are good at estimating the behavior of those combustion products under high pressure IPR conditions. Estimations of liquid and vapor phase compositions for compressed CO_2 are difficult due to the lack of data and the lack of reliability of standard equations of state near the critical point of vapors. In the case of CO_2 liquefaction we are working close to the critical point due to the temperature of cooling water at power plants. This makes the estimation of compression work and the required temperature of cooling water problematic to calculate using standard software.



Figure 1: Simple computer model of a 400 MW power plant

In a commercial system we would look closely at energy recovery as well as methods for conserving water. In the demonstration system we were interested only in proving the concept of CO_2 liquefaction and pollutant removal using an oxy-fuel combustion system to generate feed gas. This allowed further simplification of the system to make the cooling water a once-though process instead of tying it to the working fluids in a steam cycle, and the change is reflected in the model shown in Figure 2.

Figure 2: GateCycle model of Jupiter/ARC experimental system (see Figure 3 for a key to the components)



Figure 3: Key for Figures 1 and 2



In the computer model (Figure 2) the exit vapor stream was calculated to consist predominantly of liquid CO_2 with both N_2 and O_2 either dissolved in the liquid CO_2 or carried on as gas. It was considered that there existed the possibility that there would be significant solubility of tramp gases (primarily N_2 and O_2) in the CO_2 and there was not expected to be any gas released to the atmosphere (all gases would be "squeezed" into the liquid CO_2). In order to predict the actual partitioning of the non-condensable gases between a vapor phase and liquid phase, thermodynamic software was applied using complex equations of state including Redlich-Kwong, Peng Robinson, and the van der Waals equations. Unfortunately, they gave inconsistent results and proved inaccurate when checked against the minimal binary and ternary experimental data that are available. The ARC is currently conducting research to better define the behavior of the ternary $N_2/O_2/CO_2$ systems and expects to be able to use those data for full scale designs by 2006.



Figure 4: Overview of experimental system as implemented. Numbers are explained in the text.

For this demonstration project, there was no need for a high degree of accuracy in the partitioning of the gases. At this scale, the components that were specified were done so in order for the system design to be robust enough to accommodate for the uncertainty that existed for the many variables, including those that dealt with solubility. The system was therefore somewhat over-designed to avoid any problems of optimization. The calculated pressures and flows for the experimental system are shown in Figure 4. The main components of the system are numbered and consist of:

- 1. Oxy-fuel combustor
- 2. Filter for large particulates
- 3. Near-ambient pressure wet heat exchanger
- 4. First stage compressor
- 5. Intermediate pressure wet heat exchanger
- 6. Second stage compressor
- 7. Third stage compressor
- 8. Fourth stage compressor
- 9. Shell_and_tube heat exchanger
- 10. Mercury trap
- 11. Gas accumulator
- 12. Exhaust

Based on the fuel heating value of Illinos #6 coal, a fuel flow of approximately 27 lb/hr was calculated. Approximately 40 lb/hr of CO₂ was used to support the flow of the coal, with approximately 52 lbs/hr of pure O₂ to maintain the combustion with flame stability. The calculated total flow was approximately 119 lb/hr. The combustion system was designed to burn coal at the calculated rates and to cool the exhaust to less than 300°F before it reached the baghouse/filter (#2 in Figure 4). In order to achieve the cooling of the gas in a small container, copper coils were used to line the interior of the combustion chamber was designed to ensure that water would not boil in the coils (opposite of what would be done in a steam generator). It was decided to have cold walls instead of insulated walls to ensure sufficient, prompt radiant transfer from the hot flame to the tube walls simulating the combustion conditions in a power plant (the temperature is lower in this system but closer than it would have been using an insulated system).

<u>A</u> full_scale system would require some method for removing the particulates from a recycled flue gas stream. Even though no recirculation was used here, a rough particulate filter was installed between the first wet heat exchanger and the combustion chamber. This also served to ensure the temperature of the combustion gas was below 300° F. About one lb/hr of ash was collected in this filter leaving about 118 lb/hr of combustion gases.

After the rough particulate filter, the hot exhaust gases flowed into a spray of water in the first of two counter-flow direct contact heat exchangers (#3 in Figure 4). The water cooled the gas to the point where hot water vapor condensed, removing soluble pollutants and entrained particulate mater. Approximately 13 lb/hr of the water vapor condensed out of the combustion products at this step. The gases and vapors remaining totaled_approximately 105_lb/hr.

Once through the first direct contact (wet) heat exchanger the combustion products were compressed to approximately 175 psig in a low pressure compressor, where the gas was also heated by the compression process. Remaining vapors then went to the second direct contact heat exchanger (#5 in figure 4) where they were cooled again and approximately 4_lb/hr more water vapor was condensed out, leaving the compressed vapor stream with approximately 101_lb/hr of combustion gases.

Following the second wet heat exchanger, the cool, saturated gas_was again compressed to approximately 250_psig. During compression the temperature of the gas rose again. The original design (and the built system) included a third wet heat exchanger to cool this gas. However, it was determined that the counter-flowing water was not necessary and the mass of the exchanger and the associated tubing were enough to intercool the gas between compressors.

A third compressor boosted gas pressure from approximately 250 psig to approximately 680 psig. Again, the gas heated but the tubing connecting this compressor to the next compressor was sufficient to intercool the gas. The final compressor was specified to boost the 680 psig vapor to pressures as high as 2,000 psig. However, these vapor mixtures were not compressed to

 $^{^{2}}$ The USDOE does not endorse or recommend commercial equipment, processes, or software.

this maximum pressure. The extra pressure was present to give flexibility in combustion gas mixtures.

After the final compression, the hot compressed vapor was passed through a shell-and-tube condenser. At this point the vapors were dropped to below their dew point and condensation began. The condensate was directed to a converted high-pressure gas storage bottle which acted in this system as the CO_2 tank to hold the high-pressure liquid.

The high-pressure non-condensable gases, diluted with CO_2 vapor, were then passed through a mercury filter to trap the mercury remaining in the system. After the mercury trap the high pressure gas was bled into an accumulator at a lower pressure, which was put into place in order to give the system a measure of flexibility with regard to the flow rates in the system. From the accumulator the exhaust gas was expanded to the atmosphere. This bleed stream, however was put into place strictly for the purpose of collecting and analyzing the gas composition. Were it not for the experimental requirements, the system could have easily been completely closed.

CONSTRUCTION

The primary components for the combustion system consisted of a modified Venturi Volumetric feeder tied to a custom designed burner, using needle valves and standard flow meters to control the gas flows. The boiler simulator consisted of a sealed drum with cooling water circulated through copper loops to lower the exhaust gas temperatures to those expected in a power generating boiler. A typical bag from a baghouse was used for ash removal, and a small chamber was constructed to simulate actual performance

The post combustion systems consisted of compressors, wet heat exchangers, and an accumulator. The wet heat exchangers and accumulator were manufactured in-house while the compressors, both single and two stage, were purchased directly from equipment suppliers.

The physical layout of the system was broken out into three sections, or 'benches'. The first consisted of the combustion side and baghouse, the second held the first two wet heat exchangers and first and second compression stages. The last 'bench' consisted of the third wet heat exchanger and the final, high pressure compression stage with gas collection and sampling.

OPERATION

Operation of the system took place in six stages:

- 1. Low pressure shake-down and leak testing
- 2. High pressure integrity testing
- 3. Wet heat exchange testing
- 4. Bench 2 testing
- 5. Bench 2 and 3 testing
- 6. Full system testing

CONCLUSIONS / RESULTS

Standard approaches to the elimination of pollutants in flue gas rely on numerous separate processes, each of which uses a distinct method for removal. The USDOE/Albany Research Center and Jupiter Oxygen Corporation. have developed and demonstrated an approach that rather than reducing the emissions of a single pollutant, combines their technologies in a control "package" which effectively removes virtually all of the pollutants simultaneously from the flue gas. The technique involves the redesign of the boiler system to re-circulate the flue gas (simulated in the experiment with the use of CO₂ as the carrier gas for the coal), the use of pure oxygen to support the combustion and the condensation of the vapors in the flue gas to remove effectively all of the pollutants, including particulates, CO₂, SO_X and NO_X, as well as mercury and fine particulate matter. This practical removal of carbon dioxide adds an entirely new dimension to the science of flue gas treatment and handling.

During the demonstration, combustion gas tests showed NO_x levels at 0.088 pounds per million British thermal units (lb/10⁶ Btu), well below the target of 0.1 lb/10⁶ Btu. With the addition of the Integrated Pollutant Removal system, more than 80% of the carbon dioxide, and 99% of the SO_x and particulates were captured. Particle-bound mercury was efficiently removed from the flue gas, and mercury vapor was concentrated at the end of the process, where proven technologies could be used to collect it. Based upon the results of these tests, it is anticipated that future optimization of the combined systems could capture more than 95% of the CO₂ and over 90% of the mercury while maintaining ultra-low NO_x.

The projected fuel savings and other increased efficiencies are such that the cost of this combined process could be competitive with current combustion technologies. Additionally, the prospect of new regulatory requirements are causing power plant designers to revisit the conventional approaches used to remove pollutants which would only serve to improve the economics behind this approach. Cost estimates for the combined ARC/Jupiter Oxygen system are on the order of 5.1 cents/kWh with amortized capital expenses including a production cost of 1.7 cents/kWh (both figures are lower than traditional air-fired power plants). However, further testing and demonstration on a commercial scale plant would be needed to prove out the system.

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