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Operating Experience and Performance Characteristics of a Gas-Oxy Combustion Technology at Total's Carbon Capture and Storage Demonstration Plant

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B&W babcock & wilcox power generation group

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Abstract

In January 2010, TOTAL began operating Europe's first fully integrated carbon capture and storage (CCS) demonstration facility, in Lacq, near Pau in southwestern France. The pilot plant uses 99% pure oxygen, produced by an air separation unit (ASU), which is substituted for air to combust natural gas in a 30MW$_{th}$ industrial boiler. The original air combustion boiler had been refurbished and modified suitable for oxy combustion. Flue gas leaving the boiler consisting primarily of water vapor and carbon dioxide (CO_2) gas is further conditioned in a compression and purification unit (CPU). Dried carbon dioxide gas travels through a 30 kilometer pipeline to be injected into a depleted natural gas reservoir, 4500 meters underground in a porous sedimentary rock formation extending over two square kilometers.

In December 2011, in collaboration with Babcock & Wilcox Canada Ltd. (B&WC) and Air Liquide (AL) of France, Total embarked on a comprehensive testing campaign at Lacq to gain in-depth knowledge of oxy-combustion technology. The following observations were made comparing oxy combustion to air combustion:

- The boiler operated with much higher efficiencies, as well as lower furnace exit gas temperatures (FEGT) in oxy mode
- Incident heat fluxes increased in oxy mode as measured at various locations in the furnace for the same load cases
- Percentages of heat absorption between the furnace and convection pass including superheater, steam generating banks, and economizer were similar for both air- and oxy-combustion modes
- Nitrogen oxides (NO_X) emissions trended lower as the load increased in oxy-combustion mode

Introduction

Total's Lacq facility in southwestern France is Europe's first fully integrated carbon capture and storage (CCS) pilot project that includes a natural gas extraction and processing plant; natural gas-fired power plant; CO_2 compression and purification unit. The project is a unique combination of oxy combustion and onshore storage demonstration which was launched to demonstrate Total's commitment to the well-being of local communities to find new industry for Lacq, to fight climate change, and to develop CCS technology.

Planning for the project began in 2005 followed by a public announcement in 2007. Boiler refurbishment and modification, as well as construction of the ASU and CPU, took place in 2008 and 2009. The 1950s vintage two-drum low pressure superheater boiler underwent major refurbishment and modification to extend its service life as well as making it suitable for oxy-gas combustion. Air Liquide provided an air separation unit based on the cryogenic process and four oxy-gas burners. Total installed a 3-stage CO_2 compression and purification unit to remove moisture in the CO_2 gas to less than 50 ppm as well as to increase the pressure to over 27 bar (392 psi). The dried CO_2 travels through a 30-kilometer pipeline to enter a second compression station located at the Rouse reservoir. CO_2 is then injected into a depleted natural gas reservoir, 4500 meters underground. Figure 1 depicts an overall view of the Lacq facility as well as a CCS process.

In January 2010, the Lacq facility was commenced into full CCS operational mode. The pilot plant uses 99% pure oxygen which is substituted for air to combust natural gas. In December 2011, in collaboration with B&WC and AL of France, Total embarked on a comprehensive testing campaign at Lacq to gain in-depth knowledge of oxy-combustion technology.

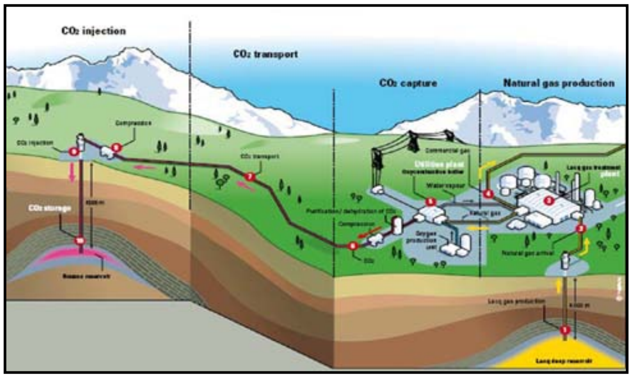

Fig. 1 Lacq facility and CCS process.

Process Description, Process Flow Diagram, and Component Description

Figure 2 presents a simplified process flow diagram representing the overall configuration of the CCS process. The ASU separates atmospheric air into its primary components: oxygen and nitrogen as well as some argon and other rare inert gases. Oxygen greater than 95% purity is piped directly to the boiler area to feed the oxy burners. Nitrogen gas is piped to the CO_2 dehydration unit. Spent nitrogen from the dehydration unit is exhausted to the atmosphere thru its own stack. Natural gas fuel from a separate Lacq processing unit supplies fuel to the oxy burners. Moisture in flue gas is condensed in the gas conditioning unit in a washing process when contacted with water. Cooled dehumidified flue gas, primarily CO_2, is then processed via three stages of compression and dehydration in the CPU where CO_2 is further dried to less than 50ppm moisture and compressed to 27 bar (392 psi) pressure.

Dried CO_2 is finally transported to the Rouse reservoir for permanent storage. In air mode, mostly used for starting up the boiler or when either ASU or CPU is shut down for maintenance, the flue gas leaving the boiler is exhausted to the flue gas stack. Description of individual systems and processes mentioned above are provided in the following sections.

Air separation unit

The air separation unit was supplied by Air Liquide. The technology is based on a cryogenic distillation process which requires very tight integration of heat exchangers and separation columns to obtain a good efficiency. The process consists of several steps: 1) it begins with a particulate free pre-filtered air; 2) the air is then compressed. During compression, condensed water is removed in inter-stage coolers. The compressed air then passes thru a molecular sieves bed which removes remaining water vapor as well as carbon dioxide; 3) the processed air then passes through an integrated counter flow heat exchanger where it is cooled by the product cryogenic streams. Part of the air is liquefied to form liquid oxygen. The remaining nitrogen rich gas is distilled to almost pure nitrogen in a high pressure distillation column.

Cryogenic distillation is a mature technology which has been used in the production gas industry to separate air into nitrogen and oxygen gases for several decades. The process efficiency, thus energy consumption in the air separation process, is a function of the desired oxygen purity. In oxy combustion, an oxygen purity of 95% or greater is desirable. The higher level of oxygen purity minimizes the nitrogen

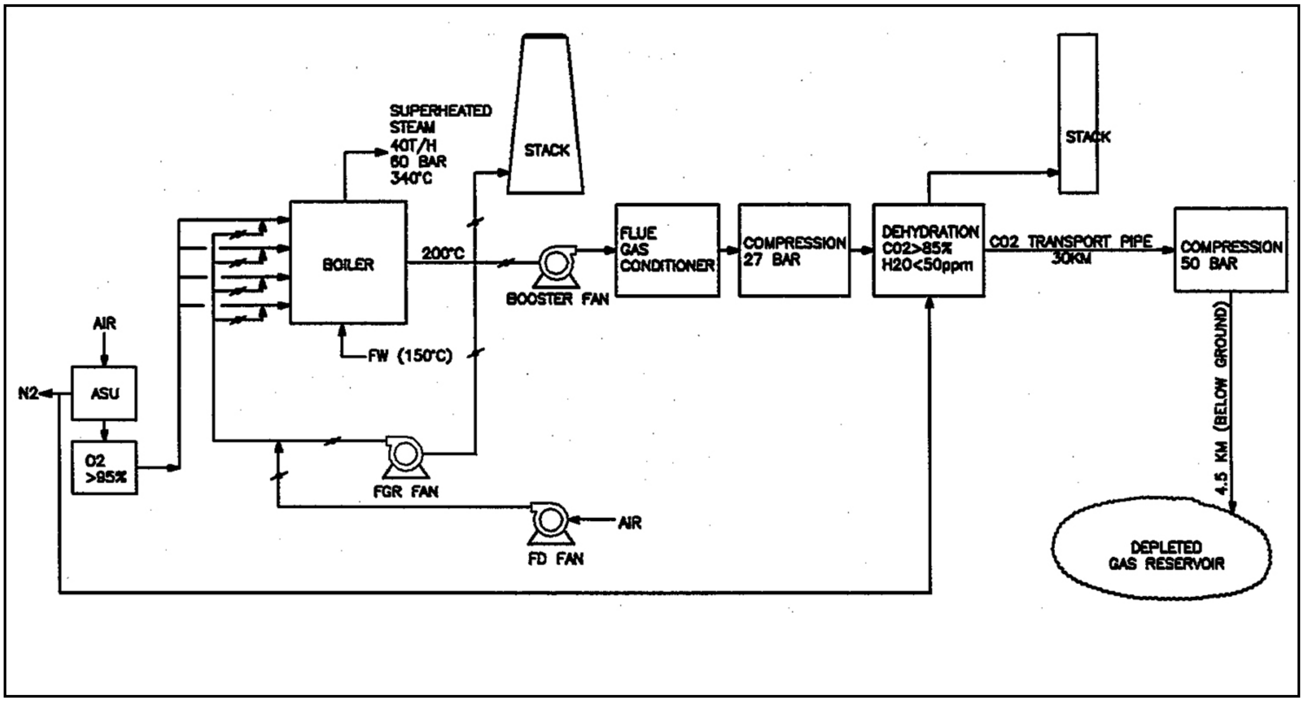

Fig. 2 Simplified process flow diagram for the CCS plant at Lacq.

content in the oxygen stream. This in turn will limit the formation of NO_X during the combustion process. NO_X in the flue gas stream will eventually convert into nitric acid which is detrimental to the downstream flue gas conditioning equipment, especially CO_2 gas compressors and CO_2 gas transport pipelines.

Gas-oxy burners

Gas-oxy burners were supplied by Air Liquide. They are located on the front wall of the boiler, arranged 2 high by 2 wide. As shown in Figure 3, the AL oxy-gas burner is very unique in design. It is designed to burn natural gas with pure oxygen. Oxygen and the flue gas recycle (FGR) stream are individually piped, controlled and metered for each burner. A windbox is not required nor included in the configuration. O_2 supply is split into two streams. Primary core O_2 feeds about 15% of the total required O_2 to the center core of the burners. The secondary O_2 stream feeds the remaining O_2 to the outer zone. All four burners are designed to swirl in the same direction, clockwise when looking from the boiler front wall toward the steam drum.

Air Liquide's involvement in oxy combustion has spanned a couple of decades in the metal smelting industry. AL's engagement in oxy combustion at Lacq is its first large-scale application in the power industry. The AL gas-oxy burners at Lacq have been in operation for over two years demonstrating successful operation and low maintenance. The fixed vane burners require no moving parts, thus minimizing maintenance.

Two-drum industrial boiler

The 1950s vintage industrial boiler (Figure 4) was originally supplied by a local boiler company. A major life extension and modification program was executed in 2008 to accommodate oxy combustion. The boiler is a low pressure, two drum superheated unit. Major components include the furnace, single stage superheater, boiler bank (saturated surface), and external finned tube economizer.

The furnace is 4.5 m (14.76 ft) wide and 7.2 m (23.62 ft) high measured between the centerlines of the two drums.

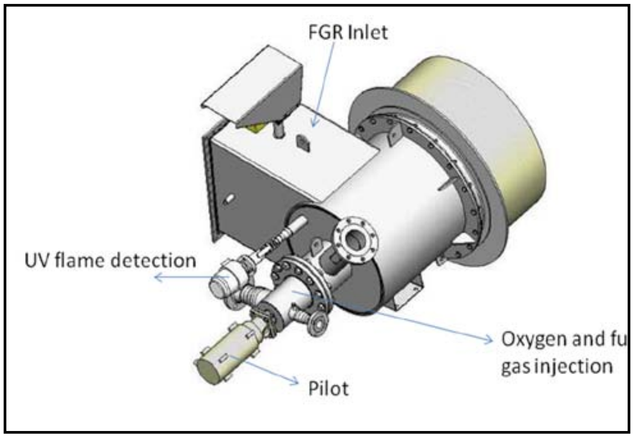

Fig. 3 Gas-oxy burner.

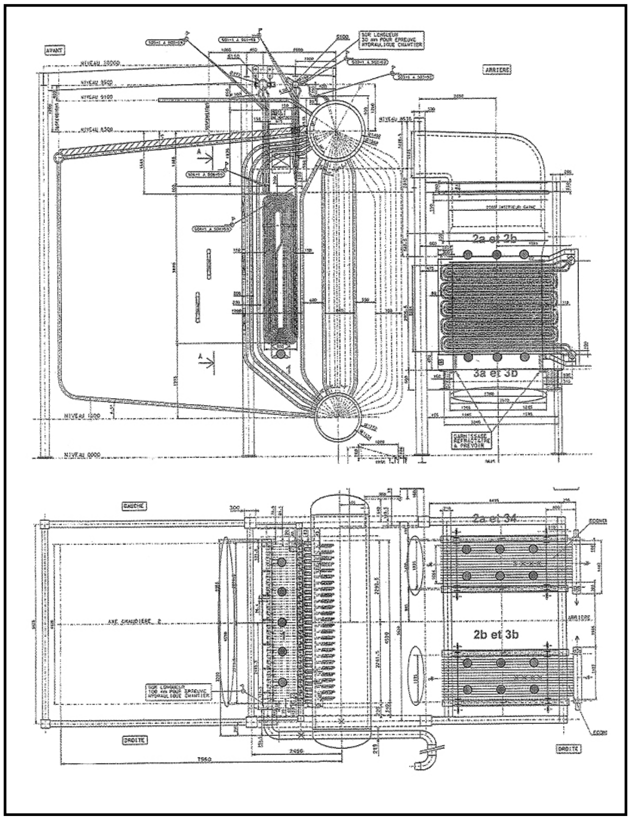

Fig. 4 Original boiler showing thermocouple grids for flue gas temperature measurements.

The non-membrane construction furnace walls are made up of 83 mm (3.27 in.) outside diameter tubes which are held together by interlocking brick forming a solid wall behind the tubes. Refractory extrudes through the gap between the tubes to the tube centerline leaving half the tube crown exposed to hot flue gas.

The unique coil designed superheater is comprised of 38 mm (1.49 in.) outside diameter tubes. The elements are equally spaced across the width of the boiler. From a heat transfer perspective, the coil design is characterized with a combination of alternating parallel flow and counter flow with the flue gas flow parallel to the tube length (long flow).

The boiler banks include three passes consisting of a combination of cross flow and long flow design. The majority of heat transfer takes place in the long flow section where flue gas flows along the tube length. The remainder of heat absorption is conducted with a cross flow at the entrance and exit of the boiler bank cavity. The flue gas exits the boiler bank at the top part of the third pass to a flue connecting to the economizer banks.

The split design finned tube economizer consists of two identical tube banks located at the back end, outside of the boiler setting. Each economizer bank receives one-half of the flue gas flow leaving the boiler.

In the past two years, the boiler has been operated mostly in oxy mode. The boiler was in air mode only when it was in start-up or when either the ASU or CPU was shut down for maintenance. Noticeable differences were observed during oxy combustion including a 5% improvement in boiler efficiency and reduction in final steam temperature as compared to air operation for the same boiler operating conditions.

Flue gas conditioner

The flue gas leaving the boiler consists primarily of CO_2 and moisture, by volume basis up to 33% and 63%, respectively. The moisture must be removed prior to entering a compression process. This is accomplished by passing the flue gas through a two-stage flue gas conditioner consisting of two cylindrical washing vessels as shown in Figure 5. Flue gas temperature is reduced and sub-cooled from 200°C (392°F) to about 25°C (77°F) thus removing most of the moisture.

The flue gas conditioning unit was initially designed for conditioning the flue gas stream from both gas and heavy fuel oil operations. In addition to the task of removing moisture, the first stage of the conditioner also removes particulates from operation with heavy oil fuel firing instead of gas. Operating experience to date indicates that for gas operation, a single-stage conditioner would be sufficient to remove all the moisture in the flue gas stream.

CO_2 compression unit

Flue gas leaving the flue gas conditioner contains mostly CO_2 gas at near ambient temperature. It then undergoes a three-stage compression process to boost the pressure to 27 bar (392 psi), the level required to transport it through a 30 km pipeline to its final storage site. As shown in Figure 6, the compression process features three stages of reciprocating engines driven by an electric motor.

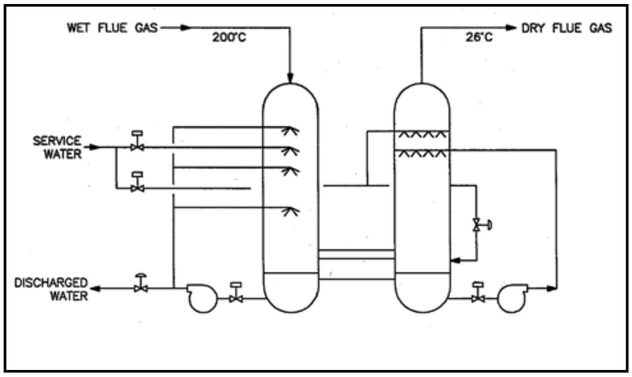

Fig. 5 Flue gas conditioning unit.

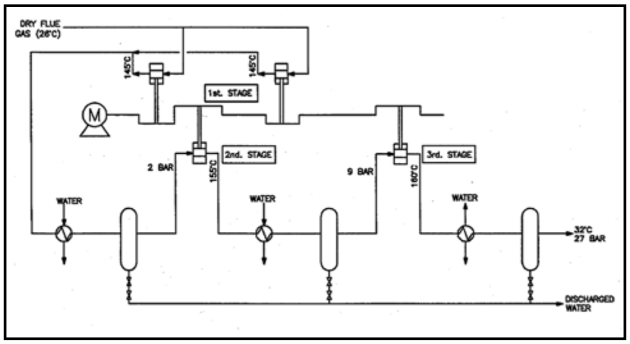

Fig. 6 Compression and purification unit.

As the CO_2 is compressed, the temperature is raised through heat of compression. Interstage coolers reduce the CO_2 temperature passing it through a water-cooled tube and shell heat exchangers. During the compression and cooling process, moisture in the CO_2 gas is condensed and removed in water columns located downstream of each interstage cooler.

The third stage compression unit has been experiencing abnormal wear requiring frequent replacement of its cylinder liners. The acceleration of the wear rate was due largely to the presence of moisture and nitric acid in the compressed flue gas coupled with the fact that the liners were made of carbon steel material. For the next generation compressor, stainless steel liners would be preferred to reduce compressor maintenance.

CO_2 dehydration unit

To avoid potential pipeline corrosion problem dues to hydrates formation at low temperature, a dehydration process (Figure 7) is used to remove moisture in the CO_2 gas stream prior to transporting it to the final injecting well. Wet CO_2 gas is brought into a production absorber, where water vapor is absorbed by dried nitrogen, which is used as a dehydration agent. Wet nitrogen in the production absorber is purged to the stack when it is switched to the regeneration cycle.

Test Preparations and Measurement Procedures

Water, steam and flue gas operating data were collected during performance tests. Other test data included heat fluxes, furnace exit gas temperatures and gas temperatures at various locations in the convection pass. Test ports and thermocouple grids were installed for these measurements as described in the following section.

Fuel analysis: Natural gas fuel is supplied from Lacq's production well. Ultimate fuel analysis is provided in Table 1.

Thermocouple grids: Five thermocouple grids were installed in the superheater outlet as well as economizer

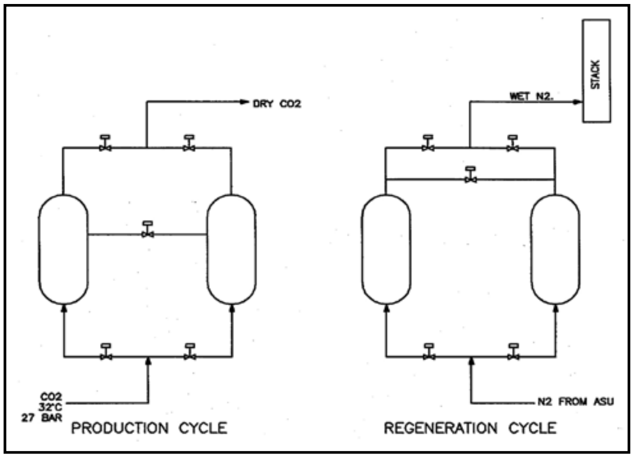

Fig. 7 CO_2 dehydration unit.

Table 1
Fuel Ultimate Analysis

Composition	% by volume
Methane CH_4	92.564
Ethane C_2H_6	4.284
Propane C_3H_8	1.472
Butane C_4H_{10}	0.935
Pentene C_5H_{10}	0.348
Benzene C_6H_6	0.103
Nitrogen N_2	0.250
others	0.044
HHV (Btu/ft^3)	1096
HHV (KJ/m^3)	40,836

inlet and outlet flues. Figure 4 illustrates locations of five thermocouple grids to measure flue gas temperature.

FEGT measurements: Furnace exit gas temperatures were measured via traverse during every performance test. Figure 8 illustrates the high velocity thermo probe used during the tests. See Figure 9 for locations of the test ports to measure FEGT.

Heat flux measurements: Incident heat flux measurements on the boiler side walls, as well as boiler front wall, were performed using ellipsoidal radiometer probes (Figures 10, 11, and 12) at boiler loads ranging from 30% to 88%.

The ellipsoidal radiometer developed by the International Flame Research Foundation (IFRF) measures the total radiative flux incoming from the flue gas facing the tip of the radiometer. The instrument consists of a water-cooled ellipsoidal cavity with a highly reflective gold plated surface. The aperture opening is located at one focus and the sensing element is located at the other focus. The ellipsoidal cavity focuses all the radiation entering the orifice onto the surface of the thermopile. The thermopile is a heat flow plug of stainless steel with two thermocouple junctions at each end. Total radiative incident heat flux is determined based on the following principle:

$$\Phi_T = \alpha_S \Phi_R - \varepsilon_S \sigma T_S^4 + h_C(T_G - T_S)$$

Where:

Φ_T = total heat flux absorbed by the surface area
α_S = surface absorption = 0.85
Φ_R = incident radiative heat flux
ε_S = surface emissivity
σ = Stephan-Boltzman constant
T_S = surface temperature
h_C = convective heat transfer coefficient
T_G = flue gas temperature
Φ_C = incident convective heat flux = $h_C(T_G - T_S)$

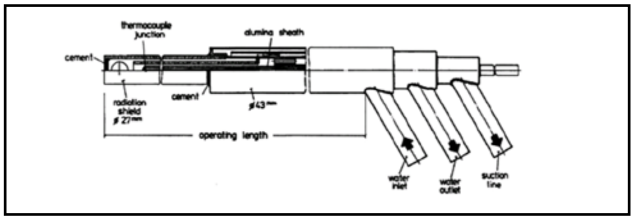

Fig. 8 High velocity thermo probe for FEGT measurements.

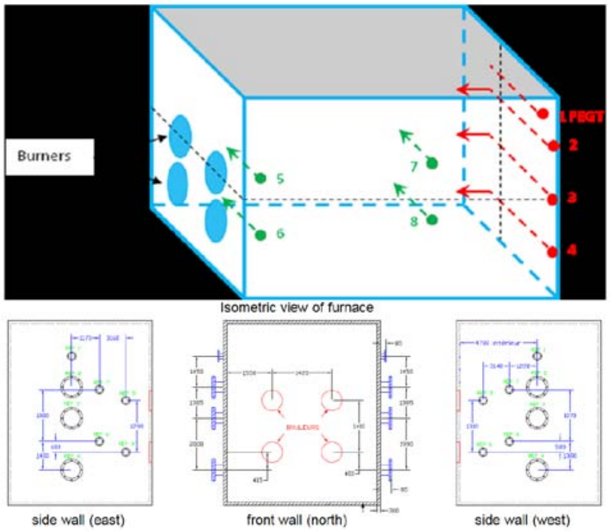

Fig. 9 Location of test ports for heat flux and FEGT measurements.

The ellipsoidal radiometer was calibrated in a vacuum facility in Total's laboratory for both radiation and convection sensors prior to and after tests for the specified Lacq furnace configuration.

A straight probe (Figure 11) was used for measurement of incident heat flux imposing on the furnace side walls. The probe is inserted to penetrate into test ports to align the probe tip with the plane of the furnace side wall tubes (see Figure 13).

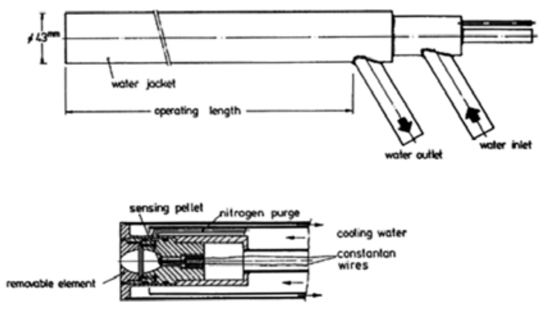

Fig. 10 Ellipsoidal radiometer probe.

Fig. 11 Straight probe.

Fig. 12 Curved probe.

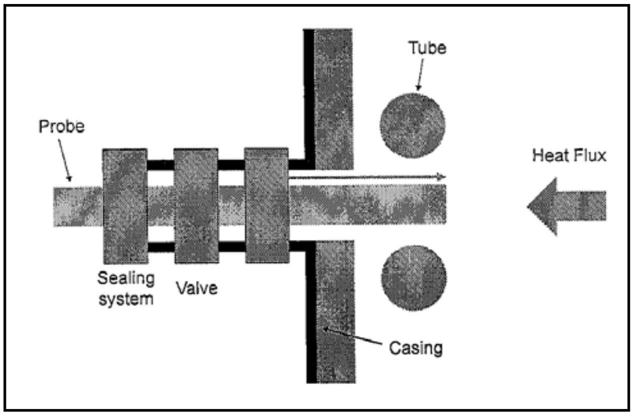

Fig. 13 Probe location for side wall heat flux measurement.

A curved probe (see Figure 12) was used for front wall heat flux measurements. The probe was inserted into ports 2, 3 and 4. Traversed measurements from the furnace sidewall to the furnace center were conducted through each port for both the east and west sides.

Both air- and oxy combustion tests were conducted with the same excess O_2 leaving the economizer for the same operating load tests. However, in oxy-combustion mode, a large quantity of flue gas leaving the economizer was recycled back to the boiler via the burners to boost the convective heat transfer in the convection pass. The flue gas recycle (FGR) was set between a ratio of 2.5 and 3.0 which is the ratio of the volumetric flow of flue gas recycled to the burners over the volumetric flow of flue gas leaving the boiler (entering the CPU).

Test Results and Discussion

Air and oxy combustion

Combustion air contains approximately 21% oxygen and 78% nitrogen. In oxy combustion, fuel is burned with nearly pure oxygen. The absence of a large percentage of nitrogen will reduce the flue gas mass flow by more than 75% as compared to air combustion. Boiler performance is affected both positively and negatively with a substantial reduction in flue gas mass flow through the boiler. The majority of heat absorbed in the superheater, saturated surface (boiler bank) and economizer sections is by convective heat transfer which is primarily a function of flue gas mass flow. An original air boiler that is converted into oxy combustion, like the boiler at Lacq, would suffer substantial reduction in final steam temperature due to a large reduction in flue gas mass flow. Therefore, the flue gas mass flow must be increased to a level comparable to that with air combustion. Closed loop flue gas recirculation with a ratio of 2.75 would theoretically be required to make up for the shortfall in flue gas. A large increase in boiler efficiency is the positive benefit of reduced net flue gas exiting the boiler to the CPU in oxy-combustion mode. Overall heat loss in the dry exit gas is directly proportional to exhausted flue gas mass flow and its exit temperature. For the Lacq boiler, as shown in Table 2, the boiler efficiency improvement is on the order of 5%.

The combustion byproduct flue gas, from an air boiler that fires with natural gas fuel, contains about 12 to 13% of

Table 2
Boiler performance comparison for 35t/hr (77,157lb/hr) load

	Air combustion	Oxy combustion
Excess O_2 (% volume – wet basis)	2.60	2.60
Excess air equivalent (%)	3.98	3.98
Flue gas from combustion byproduct [(Kg/hr)/(lb/hr)]	37,650 / 83,000	9,957 / 21,950
Flue gas recycle [(Kg/hr)/(lb/hr)]	Nil	27,693 / 61,050
Flue gas recycle ratio	-	2.78
FEGT (°C°/F)	1,132 / 2070	1,053 / 1927
Flue gas temperature leaving economizer (°C°/F)	194 / 382	204 / 399
Boiler efficiency (HHV basis - %)	82.62	87.37
Furnace absorption (%)	47.90	47.31
SH absorption (%)	16.06	16.16
Economizer absorption (%)	5.30	5.90
Furnace screen + boiler bank absorption (%)	30.74	30.63

Note: absorption is percentage of total boiler absorption moisture by volume. This level of moisture increases to over 60% in oxy combustion with full rate of flue gas recirculation. For the same flue gas mass flow through the convection pass, for two comparable cases of air and oxy combustion operating with the same boiler load, there is a slight increase in the convection pass overall heat absorption in the case of oxy combustion compared to air combustion. However, in terms of the heat absorption split between the furnace and individual sections in the convection pass, the proportion remains almost unchanged between the two combustion modes as shown in Table 2.

Heat flux

It is always desirable to know the tube temperature in the water-cooled furnace walls, especially those tubes that are in close proximity of the burner zone subjected to the highest flame temperature. Heat flux is usually calculated based on outside surface thermocouple temperature, mid wall thermocouple temperature, thermal conductivity and equivalent tube thickness (surface to depth). A chordal thermocouple imbedded in the tube is the most reliable and commonly used temperature measurement method for furnace wall tubes. Unfortunately, due to lack of access, it was not feasible to install chordal thermocouples. Therefore, an alternative heat flux measurement using an ellipsoidal radiometer probe was employed. Although its measurement results do not provide a definitive conclusion for design purposes, the relative value comparison of readings between air and oxy combustion for the same boiler load can provide some insights regarding heat fluxes.

By comparing heat flux measurement values for the same measuring point for air and oxy combustion for the same boiler load, readings were approximately 50% higher for oxy combustion (Figure 14). This may suggest that water tubes are subjected to higher metal temperature in oxy combustion than in air combustion. The Lacq boiler has been operating in oxy combustion mode for over two years, and water wall tubes do not show any sign of distress. Water-wall tube design temperature is usually based on saturation temperature at boiler design pressure plus a temperature margin which is based on fuel type and firing characteristics. This design margin is adequate to cover heat flux variation associated with oxy combustion.

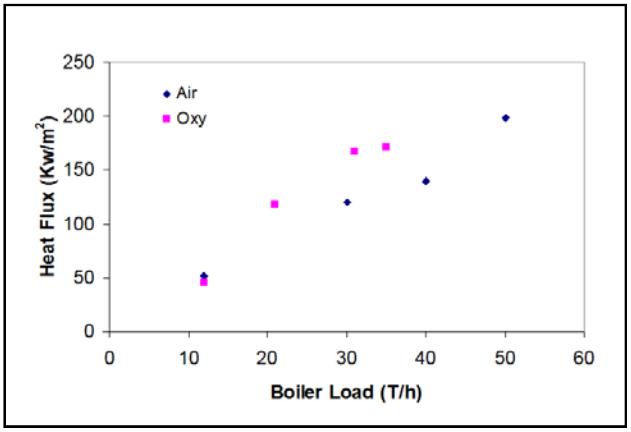

Fig. 14 Heat flux comparison on boiler west side.

Furnace exit gas temperature (FEGT)

Furnace exit gas temperature is a key design parameter in new boiler furnace sizing as well as for design of the convective heating surface. FEGT is a function of net fuel heat input, which is heat available (HA) for furnace absorption divided by the total furnace effective projected radiant surface (EPRS). Measured FEGT values for both air and oxy combustion across boiler load ranges, when plotted against the HA/EPRS parameter, exhibit the same trend (Figure 15). FEGT values with air combustion is consistently above the FEGT values for oxy combustion within a band of 100 to 150°F.

An oxy-combustion boiler, whether newly designed specifically for oxy combustion or original air design boiler retrofitted for oxy combustion, would require a large quantity of recycle flue gas to enhance convective heat transfer. A series of FEGT tests were conducted with varied FGR ratios from 2 to 3 to establish behavior characteristics of FEGT versus FGR. As the FGR starts to increase from 2.0, FEGT begins to climb higher and peak at 2.5 then it starts to trend downward. For the Lacq boiler, FGR of 2.5 is deemed an optimum point, as far as FEGT and corresponding final steam temperature are concerned.

NO_X emissions

NO_X is an undesirable pollutant produced from the reaction of nitrogen and oxygen gases at high temperature during combustion. In an air boiler, NO_X emissions contribute to smog and acid rain, while in an oxy boiler, it potentially causes corrosion in compressors and CO_2 gas transport pipelines. There are three NO_X formation mechanisms: fuel NO_X, thermal NO_X, and prompt NO_X. The dominant mechanism depends on the fuel burned whether coal, oil or natural gas.

Fuel NO_X is formed from the oxidation of fuel-bound nitrogen which is strongly dependent on fuel/air stoichiometry (oxygen availability). However, it is independent of variation in combustion zone temperature. Fuel NO_X can contribute as much as 80% of total NO_X emissions when combusting

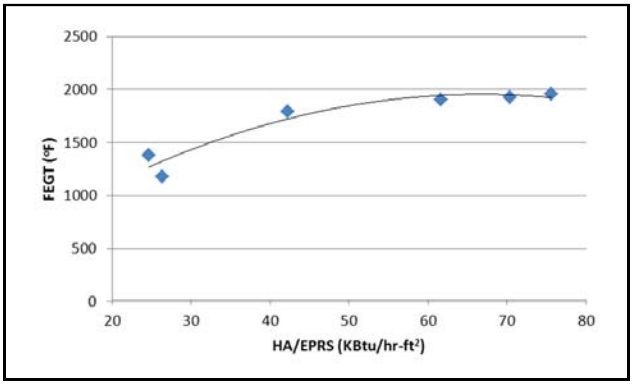

Fig. 15 FEGT curve for oxy combustion.

coal and as much as 50% when combusting oil. However, it is not important in natural gas combustion, especially at Lacq where fuel-bound nitrogen content is only 0.25% (by volume basis) of total fuel.

Thermal NO_X refers to NO_X formed through high temperature dissociation/oxidation of nitrogen in combustion air. Its formation is a function of the three Ts: time, temperature and turbulence. For oil and natural gas firing, thermal NO_X contributes up to 50% of total NO_X emissions. In oxy-combustion with 99% pure oxygen, the remaining nitrogen content in the oxygen stream is very small. However, there are a number of possible sources of air leakage into the combustion and flue gas streams. These include burner instrument air and atmospheric air. An oxy-combustion boiler operates with a large quantity of recycle flue gas. Therefore, atmospheric air infiltration into the flue gas stream through observation ports, flue isolation dampers and FGR fans would increase the contribution rate of thermal NO_X. At Lacq, both the furnace and convection pass were pressurized above atmospheric pressure to eliminate air infiltration into the flue gas stream except for a small amount via instrument air to burners. Thus, thermal NO_X formation was kept to a minimum.

The third NO_X formation mechanism is prompt NO_X from the reaction of molecular nitrogen and hydrocarbon species. It is formed in the earliest stages of combustion in the lower temperature fuel rich flame zone. It is not important in coal and oil combustion. However, it is a predominant contributor of total NO_X emissions in gas-oxy combustion.

In an air boiler firing natural gas, the overall NO_X versus HA/EPRS curve exhibits a gradual concave parabolic characteristic which tends to decline from full load to 50% load. After that point it increases as boiler load is reduced toward minimum load. In gas-oxy combustion, as shown in Figure 16, NO_X emissions trended lower as boiler load increased. Furthermore, NO_X emissions from oxy combustion at Lacq are very low as compared to a gas-air fired boiler with low NO_X combustion system for the same heat input. In addition, its NO_X production rate was not sensitive to excess O_2 or quantity of FGR. However, it was quite sensitive to the way in which oxygen was introduced to the burner via primary and secondary O_2 zones.

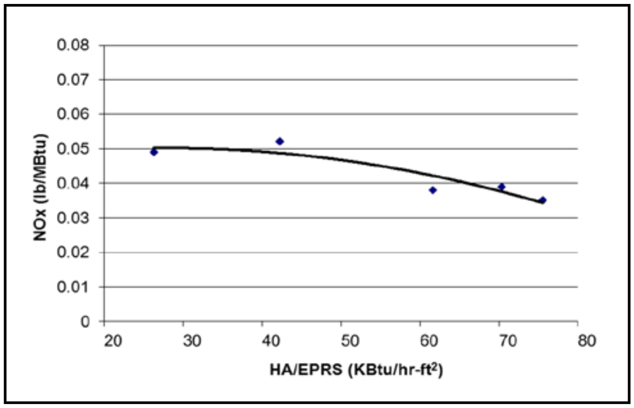

Fig. 16 NO_X curve for oxy combustion.

Conclusion

Operating experiences to date at Total's carbon capture and storage demonstration plant have shown that an original air combustion boiler can be retrofitted with minimal modifications, which are limited to oxy burners and flue gas recirculation system, to successfully operate in oxy combustion. Depending on the level of flue gas exit temperature, a boiler efficiency improvement on the order of 2 to 5% is possible due to a large reduction in combustion byproduct flue gas. A flue gas recirculation ratio of between 2.5 and 3.0 would be required in an air boiler retrofit to maintain satisfactory operation of the boiler. Care should be taken to avoid air leakage into the furnace and flue gas stream to minimize formation of thermal NO_X. Operating with positive pressure above atmospheric has proved successful in minimizing air leakage into the boiler. NO_X emissions are low in oxy combustion, however its presence in the flue gas stream causes deterioration of compressor liners. Water wall tubes in the furnace appeared to be subjected to higher heat fluxes in oxy combustion than in air combustion, but the variation is deemed within design tolerance of an air-combustion furnace. This is evident as the furnace tubes do not show any signs of distress after more than two years of operation in oxy-combustion mode.

The Lacq testing campaign has provided an opportunity to acquire real time operational data, and to enrich our understanding with respect to operation and performances of gas-oxy combustion. The data collected will enable us to calibrate and refine design and simulation models which will be beneficial to the next generation gas-oxy combustion plant for carbon capture and storage.

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