1. Introduction

NETL’s Office of Research and Development (NETL-ORD) is conducting a research program in oxy-combustion and co-firing, the goal of which is to support the development and deployment of commercial oxyfuel power plants. NETL-ORD research projects are focusing on CFD modeling – including development of improved simulation tools, combustion testing, thermal cycle modeling, flue gas treatment and CO₂ capture, and materials performance. Results obtained from experimentation and operation of pilot-scale systems are coordinated with computational work to provide validation and verification of the simulation tools. The ultimate research goal is to provide useful information for the design and operation of oxyfuel “green-field” plants, as well as for re-powering or retrofitting existing coal-fired power plants to run on oxyfuel. An overview of NETL-ORD research, key results, and relationships to other NETL programs will be presented.
2. CFD Modeling and Combustion Research

NETL-ORD’s CFD modeling and simulations are performed using Fluent and OpenFOAM. These codes provide a selection of sub-models of varying fidelity (gas-solid reaction, radiation properties and turbulence) necessary for oxyfuel combustion simulation. Modifications are made to the built-in sub-models as necessary to improve predictive capability and/or computational efficiency.

This effort has several objectives. One is to provide computational support for experimental programs. Calculations are performed to supplement and interpret experimental measurements and to investigate operating regimes and furnace/burner designs. Predictions of the heat flux, temperature and composition at the wall provide the inputs for these models. Experiments measuring the radiative spectra of laminar syngas flames provide additional data for improving radiation sub-models. Literature data for coal reaction models are also supplemented by ongoing NETL research. Additional simulations are performed as part of an ongoing validation program. Validation targets include experiments from bench to industrial scale. CFD simulations are also used in connection with materials performance research projects.

A major goal of the development of high-fidelity computational models is to understand the impact of significantly higher concentrations of CO₂, water vapor, and possibly of oxygen associated with the oxy-combustion process. One NETL-ORD study is identifying key technical issues and data gaps in the literature for the reactivity of coals and blends of coal and biomass. Of particular interest are experimental results for char burnout and the adequacy of common char burnout models to accurately predict these data for oxy-combustion environments. These results will be used to recommend and guide future experimental and modeling work.

Experimental studies are in progress to measure radiative properties of oxyfuel flames. These studies use a mid-IR imaging spectrometer to measure the radiative properties of oxyfuel flames in a flat-flame laboratory burner facility. These measurements augment and extend the set of data pertinent to oxy-combustion applications and will be available for validation and improvement of simulation sub-models for oxy-combustion systems. In addition, coal reactivity studies in oxy-combustion environments may be initiated, if needed, as indicated by the results of the literature survey and analysis of models described above.

NETL-ORD also has an on-going effort to measure flame characteristics during pilot scale oxyfuel experiments and demonstrations within the United States. The aim of this effort is to quantify and profile the thermal radiation emissions from the flame, determine effective flame temperatures, and assess flame stability and spectroscopic properties. Thermal radiation profiles emitted from flames are measured using a series of total radiometers, passive thermal devices that are largely wavelength-independent in their response and which are optimized to accurately detect thermal radiation power changes through a specific calibration approach. Two monochromators and a CCD spectrometer are used to measure the spectral emissions of the flame as a function of wavelength from 280 nm to 5000 nm. Wien’s displacement law is utilized to determine the temperature of the hottest soot or ash emitters as proxies for estimating flame temperature at the location of the measurement. Flame data have been collected during pilot-scale air-firing and oxy-firing of different types of pulverized coal, as well as natural gas, and used to characterize changes in the flame as a function of process parameters, including switching from air to oxy-firing. Flame characteristics for various staging and oxygen injection strategies have also been documented to aid burner development. In general, oxygen adds a new variable that can be manipulated to optimize the flame characteristics in order to achieve efficient oxyfuel power generation. This is a critical element to the implementation of oxyfuel as part of an overall greenhouse gas emissions reduction strategy.

3. Thermal Cycle Modeling

Thermal cycle modeling with Gatecycle® software has been utilized to examine and improve existing pilot-scale oxy-combustion systems, and for evaluation of oxy-combustion retrofit and new design strategies for power plants. These models have identified multiple opportunities to minimize the impact of CO₂ capture on plant operation, efficiency and cost of electricity. Making improvements to oxygen production, avoiding FGD during flue gas recycle, implementing heat recovery, optimizing combustion, and minimizing air infiltration have the potential to greatly improve efficiencies and heat rates over baseline oxy-combustion systems.
4. Flue gas treatment

NETL-ORD’s Integrated Pollutant Removal (IPR®) incorporates the thermodynamic requirements for efficient multistage compression with energy recovery and CO₂ purification to improve the overall thermal efficiency of oxy-combustion. IPR uses condensing heat exchange after each compression stage to recover energy which is then used to pre-heat or reheat boiler combustibles and boiler feedwater. Current IPR research studies include computer modeling and verification, studies of trace element distributions, treatment of captured flue-gas water for re-use, and corrosion testing (see Materials Performance).

Power-plant thermal cycle models (Gatecycle®) and general chemical engineering models (ASPEN®) are used to examine the thermal integration of IPR® with the power plant and predict the effect of process variation on the distribution of flue-gas materials across the system. The models are built at multiple scales and are validated by data from a physical system processing a 100 lb/hr flue-gas slip-stream from an oxy-fired test boiler. Pressure, temperature, flow data, and gas and liquid compositions are collected throughout the process.

Early results from processing an oxy-fired flue gas from coal combustion show a 95% reduction in SO₂, a 78% reduction in water, and an 86% reduction in HCl in the first condensation operation of IPR®. Most of the remaining sulfur and chloride compounds, and some of the mercury, report to the water in higher-pressure condensing sections. Pressure may play a role in the oxidation state (and solubility) of gas components such as NOₓ and SOₓ, which in turn affect the capture rate of mercury. NOₓ compounds and CO become concentrated in the compressed CO₂ product.

Capturing CO₂ from fossil-fuel combustion generates a significant water product which can be tapped for use in the power plant and its peripherals. Water condensed in the IPR® process may contain fly ash particles, sodium (from pH control), and sulfur species, as well as heavy metals, cations and anions. NETL is developing a treatment approach for zero liquid discharge while maximizing available heat from IPR. Flocculation/coagulation, for removal of cations and fine particles, and reverse osmosis, for anion removal and scavenging of the remaining cations, are being studied as treatment steps. Fast, in-line treatment of water for re-use in IPR seems to be a practical step for minimizing water treatment requirements for CO₂ capture.

5. Materials performance

Corrosion in oxyfuel combustion is being examined fireside for superheater/reheater (SH/RH) conditions at 700 °C and water wall (WW) conditions at 450 °C, and also in the IPR® system. Most oxyfuel combustion retrofit designs use a flue gas recycle loop to match boiler heating rates found in air-fired systems. Each fireside corrosion test consists of alloy coupons (T22, T91, 347 and 617) exposed for 1000 hours at temperature, in a gas composition representative either of air-firing, oxy-combustion with recycle prior to FGD (SO₂ levels approximately 3 times higher than air-firing), or oxy-combustion with recycle after FGD (lower SO₂ levels and expected lower corrosion rates, but which results in lower efficiencies), and under a synthetic ash containing Na and K sulfates. For SH/RH tests, only oxidative conditions are considered. For WW tests both oxidative and reducing conditions are considered. The reducing conditions reflect wall regions near low NOₓ burners. The test results consist of metal recession rates (both from scaling and from internal oxidation/sulfidation) and metallographic examinations of the exposed coupons.

The IPR® system can be thought of as two linked systems, one low-pressure and the other high-pressure, with temperatures ranging from 15 °C to 170 °C across both systems. The environment in the low-pressure system is predominately an acidic aqueous environment laden with chlorides in certain sections, and the high-pressure system is predominately an acidic gaseous environment. Inline corrosion probes and electrochemistry-based corrosion measurement technology are being used to study the internal corrosion of the IPR system and the corrosion resistance of 316 SS and Inconel 625 in these environments. Alloy corrosion rates will be correlated with process and operational events over time. This will provide information on which alloy is more corrosion resistant in IPR’s environments and which process and operational events can be modified to reduce the corrosion. The data will be applied to selection of materials of construction and to inform process optimization.