PHASE 3 – CLEAN AND SECURE ENERGY FROM COAL

A. OBJECTIVES (Phase 3)

The University of Utah (the Recipient), via their Institute for Clean and Secure Energy (ICSE), shall pursue research to utilize the vast energy stored in our domestic coal resources and to do so in a manner that will capture CO$_2$ from combustion from stationary power generation. The research is organized around the theme of validation and uncertainty quantification (V/UQ) through tightly coupled simulation and experimental designs and through the integration of legal, environment, economics and policy issues. The results of the research will be embodied in the computer simulation tools which predict performance with quantified uncertainty; thus transferring the results of the research to practitioners to predict the effect of energy alternatives using these technologies for their specific future application. Overarching project objectives are focused in three research areas and include:

1. Clean Coal Utilization for Power Generation ‘Retrofit’ through
   - Oxy-Coal Combustion – To ultimately produce predictive capability with quantified uncertainty bounds for pilot-scale, single-burner, oxy-coal operation. This research brings together multi-scale experimental measurements, advanced diagnostics and computer simulations. The efforts shall focus on ignition and coal-flame stability under oxy-coal conditions. This predictive tool developed under this effort will form the basis for application to full-scale, industrial burner operations.
   - High-Pressure, Entrained-Flow Coal Gasification – To ultimately provide a simulation tool for industrial entrained-flow integrated gasification combined cycle (IGCC) gasifier with quantified uncertainty. This project’s target is to develop a prototype simulation tool, perform preliminary uncertainty quantification on a pilot-scale gasifier, and to begin to predict heat transfer by radiation and convection, coal conversion, soot formation, and synthesis gas composition with quantified uncertainty.
   - Chemical Looping Combustion (CLC) – To develop a new carbon-capture technology for coal through CLC and to transfer this technology to industry through a numerical simulation tool with quantified uncertainty bounds. The specific research target for this project is to quantitatively identify reaction mechanisms and rates, explore operating options with a laboratory-scale bubbling bed reactor, develop process models and economics and demonstrate and validate simulation tools for a pilot-scale fluidized bed. This task will focus primarily on CuO/Cu$_2$O.

2. Secure Fuel Production by in-situ substitute natural gas (SNG) production from deep coal seams – The primary objective of this research is to explore the potential for creating new in-situ technologies for production of SNG from deep coal deposits and to demonstrate this in a new laboratory-scale reactor. The systems concept for the SNG is to use this premium fuel produced from coal in natural gas, combined cycle (NGCC) power generation or compressed and used as a transportation fuel (CNG). This underground coal pyrolysis (UCP) technology leaves large portions of the carbon from the coal in the ground. The research will focus on the development of simulation tools, the collection of process thermo-chemical parameters, and the development of CO$_2$ absorption isotherms from the laboratory test facility.
3. **Environmental, Legal, and Policy Issues** - Given that that carbon capture and storage for coal utilization has yet to receive public acceptance, numerous environmental, legal and policy issues need to be addressed if these technologies are to be applied. The Recipient shall address the legal and policy issues associated with carbon management strategies in order to assess the appropriate role of these technologies in our evolving national energy portfolio.

B. **SCOPE OF PROJECT (Phase 3)**

The Recipient shall perform a range of academic research tasks addressing issues common to clean-coal utilization for power generation by oxy-coal combustion, IGCC, and chemical looping; and for SNG production by thermal processing from deep coal seams. The Recipient shall:

- Conduct a set of tasks that builds from the strengths of the ICSE to provide experimental results and simulation tools to aid the development, production and utilization of coal for power generation in a carbon-constrained world.

- Prepare and disseminate results of the technical and policy studies through reports, papers, and workshops; and bring together industry, government and the public for analyzing energy options, policy and implications.

- Develop and validate modeling and simulation tools for reducing and eliminating greenhouse gas emissions from power generation, drawing from expertise in high-performance computing and multi-physics, multi-scale modeling. To produce predictive quantification of energy options requires formal verification and validation processes that rely on consistency between experimental observations and simulation results to make predictions and quantify uncertainty. Consequently, the Recipient shall organize this project around the principle of validation, integrating a consistent and systematic interpretation of experiments, models and simulations to:
  - Explain the experimental and simulation data sets together in order to quantify the ranges of behavior exhibited by both.
  - Quantify uncertainty and communicate the sensitivities and sources of uncertainty in ways that are accessible to both policy makers and technology decision makers.
  - Incorporate the legal and policy factors that influence final outcomes.

**Task 1.0 – Project Management**

**Subtask 1.1 – Project Management and Planning**

The Recipient shall update and revise the Project Management Plan within 30 days after award and manage project activities in accordance with the plan. The Project Management Plan will be updated as necessary.

**Subtask 1.2 – Briefings and Reports**

The Recipient shall monitor and coordinate the technical and financial activities of the project and will prepare and deliver reports and briefings as outlined in Sections D and E below.
**Task 2.0 – Technology Transfer and Outreach**

This task will focus on the dissemination of project results, on industrial, academic and public outreach, and on implementing the External Advisory Board (EAB) recommendations. Specifically, this task includes:

- Disseminating project results through the publication of reports, peer-reviewed/law-reviewed papers, trade-journal articles, and/or the participation in workshops/meetings.

- Hosting EAB meetings to obtain continued feedback on the Recipient’s research, technology transfer, and outreach efforts. The EAB shall meet formally on an annual basis with the potential for other informal discussions as needed. The EAB shall provide input on the selection of future ICSE-sponsored tasks/projects and, together with DOE, provide annual review of ongoing tasks/projects.

- Pursing industrial and public outreach opportunities to promote an improved understanding of technical, practical, policy, economic, and social challenges associated with utilization of domestic coal resources. These opportunities include hosting technology transfer workshops for several research areas with participants from industry, the EAB, the Department of Energy, other government agencies, and other interested parties. Activities may also include visiting relevant industrial sites, publishing results in trade journals, and hosting a public outreach meeting, as requested by the EAB.

- Working with librarians at the University of Utah’s Marriott Library to transition the repository from the DSpace platform currently housed on Institute servers to a stand-alone digital collection housed in the Marriott Library. This will ensure continued access to the materials currently housed in the repository related to the University of Utah’s coal research including project-related technical reports; presentations by ICSE faculty and students; presentations at ICSE-sponsored meetings; publications; reports; and relevant literature.

**Task 3.0 – Power Generation “Retrofit”: Oxy-Coal**

The ultimate objective of this task is to produce predictive capability with quantified uncertainty bounds for pilot-scale, single-burner, oxy-coal operation. This validation research brings together multi-scale experimental measurements and computer simulations, and to facilitate this integration, regular meetings between the oxy-coal simulation and experimental teams will be held to encourage continued feedback and refinement on measurement, data reduction and uncertainty quantification techniques. Particular attention is focused on ignition, coal flame stability, ash partitioning and ash deposition under pulverized oxy-coal conditions, with a smaller companion effort on application of circulating fluidized beds to oxy-coal combustion conditions. This predictive tool forms the basis for application to full-scale, industrial burner operations. This thrust area will include the following subtasks:

**Subtask 3.1 – Oxy-Coal Combustion Large Eddy Simulations**

The long-term objective of this subtask is a) to expand high-performance simulation tools to quantitatively predict the ignition and stability of oxy-coal burners for the express purpose of understanding the mechanisms involved in oxy-coal ignition and stability for retrofitting power
boilers and industrial furnaces for CO\textsubscript{2} capture, and b) to perform verification, V/UQ of the numerical and modeling error associated with this mechanism and intended use of the simulation tool.

The objective for Phase 3 is to wrap-up the high-performance simulation tool developments of the past years and to apply the technology to the new data coming from the University of Utah’s 100 kW oxy-fuel combustor (OFC) under Subtask 3.2. Specifically, the large-eddy simulation (LES) tool will be used together with the appropriate subset of data collected from Subtask 3.2 to jointly explore the physical mechanisms controlling the ignition and stability of oxy-coal flames under firing conditions being considered for CO\textsubscript{2} capture applications.

The Recipient shall:

- Define a selected subset of the OFC tests for LES simulations.
- Quantify uncertainties in scenario parameters, model parameters, numerical outputs, and experimental outputs for V/UQ (to be conducted in partnership with the experimental team conducting the tests under Subtask 3.2).
- Identify and publish the dominant mechanisms for oxy-coal ignition and flame stability.

Subtask 3.2 – Near-Field Aerodynamics of Oxy-Coal Flames with Directed Oxygen and Minimum Flue Gas Recycle

The objectives of this sub-task are to:

- Conduct oxy-coal combustion tests with a new Illinois Bituminous coal, using the existing co-axial burner, and compare coal jet ignition behavior for this coal to that already determined for a Utah Bituminous coal.
- Determine potential oxygen injection strategies leading to minimum flue gas recycle for retrofit in existing boilers, with emphasis on possible application to the ongoing Alstom 15MW BSF and Industrial Scale Burner Facility (ISBF) tests.
- Collaborate with the diagnostics team to implement non-intrusive diagnostics that elicit data suitable for validation of simulations with uncertainty quantification.

To meet the first objective the identical burner used in previous work shall be operated under oxy-coal combustion conditions as close as possible to those for which data on coal-jet ignition was obtained in previous work under this project. Using an Illinois Bituminous coal, the effect of partial pressure of O\textsubscript{2} in the primary jet shall be determined under constant aerodynamic flow conditions. Probability density functions of flame standoff distance shall be compared to those obtained previously for a Utah Bituminous coal, and the effects of coal composition shall thereby be explored. As in previous work, effects of secondary oxidant PO\textsubscript{2} and preheat temperature shall also be determined.

To meet the second objective work shall continue using the segregated directed O\textsubscript{2} configuration initiated in Phase 2. Data acquisition shall focus on photo-imaging techniques, as described for Phase 2.

To meet the third objective the Oxy-Fuel Combustor (OFC) team shall continue to work with the diagnostics team (Subtask 3.3) to operate the combustion facility, make necessary changes in the design of the furnace and otherwise assist in the implementation of advanced diagnostic measurements, using techniques described in Subtask 3.3.
Subtask 3.3 – Advanced Diagnostics for Oxy-Coal Combustion

During Phase 3, the Recipient shall address the challenges associated with making Particle Image Velocimetry (PIV) measurements in large-scale coal flames. The investigators shall build upon the Phase 2 development efforts carried out in the bench-top pulverized coal burner to perform a series of simultaneous PIV measurements and high-speed video emission imaging in the 100 kW OFC for a broad range of oxy-coal combustion conditions. The PIV measurements will provide detailed velocity data, and the high-speed video emission imaging will provide simultaneous temperature mapping. As a result, simultaneous 2D velocity maps will be provided, with corresponding temperature information based on emissions in the visible range for oxy-coal flames under various combustion conditions. This high-fidelity experimental data will be used for validation, verification and uncertainty quantification of high-fidelity advanced oxy-coal combustion simulations of ignition and stability of oxy-coal flames.

Subtask 3.4 – Oxy-Coal Combustion in Circulating Fluidized Beds

The oxy-fired pilot-scale circulating fluidized bed (CFB) will continue to be used to study operational impacts of variations in oxygen concentration, in-bed heat removal and external heat removal (from the solids recycle stream). A key objective is the development of multiple oxy-coal CFB data sets at pilot-scale that provide data for model validation, as well as an understanding of the impact of key process variables on bed temperature, bed agglomeration, solids recycle rate, and sulfur capture. Both SO₂ and SO₃ measurements will be made at a location suitable for SO₃ quantification (highly temperature sensitive). Axial profiles of SO₂, NOₓ, O₂, CO and CO₂ as well as temperature will be measured from the bed, through the freeboard, and before and after the cyclone. Solid samples will be taken of bed material and entrained solids at multiple locations, including the baghouse. A key distinction for the Phase 3 effort will be the addition of in-bed sorbent capture of sulfur and the impact operational parameters will have on sulfur partitioning as well as other measured variables. The Phase 2 work only considered native capture by coal ash species. Our bench-scale data has indicated significant challenges in achieving capture levels normally observed in air-fired combustion with limestone sorbents. The investigators have observed a strong temperature effect that will be explored and optimized in the pilot-scale studies.

Subtask 3.5 – Single-Particle Oxy-CO₂ Combustion

This subtask will focus on both pulverized-coal and fluidized-bed systems, with the following two objectives:

- The goal of this work is to investigate the role of oxy-combustion conditions on char oxidation. The work will utilize the framework of SKIPPY to perform a scoping study over a range of conditions and develop a model, which can be incorporated into the simulations. SKIPPY includes mass-transfer limitations and surface kinetics but is not suitable for incorporation into the LES simulations. However, the model can be used like a screening test to evaluate the important rate parameters for char oxidation under the conditions of interest. The Team will use existing literature data and on-going experimental results, obtained from a separately funded Sandia National Laboratory effort, for validation.

- For fluidized bed conditions, the effect of high-CO₂ environments on sulfur behavior, and in particular sulfur capture, from coal and coal char combustion in oxy-fired fluidized
beds will be explored in a bench-scale single-particle fluidized-bed reactor. Both SO$_2$ and SO$_3$ behavior will be determined. A first-generation single-particle reaction model will be further refined for use in obtaining kinetic rate information from these experiments, and to also provide a foundation for the implementation of a more simplified particle model for CFD simulations.

**Subtask 3.6 – Ash Partitioning Mechanisms for Oxy-Coal Combustion with Varied Amounts of Flue Gas Recycle**

This task focuses on two scales, a 100kW OFC laboratory combustor (Subtask 3.6a) and a drop-tube furnace (Subtask 3.6b).

The objectives of the 100kW studies (Subtask 3.6a) are to:

- Determine mechanisms governing effects of varied amounts of oxy-combustion flue gas recycle on ash partitioning mechanisms in a 100kW downflow combustor.

- Co-ordinate and integrate results with those from Subtask 3.6b and together provide validation data for sub-models, predicting size-segregated ash particle composition as functions of surrounding environments representative of oxy-coal flames with flue gas recycle amounts ranging from 30% to 0% (or whatever minimum value can be safely achieved).

To meet the first objective the Recipient shall continue the testing initiated under Phase 2. The screening tests completed under Phase 2 shall be complemented by tests under at least two flue gas recycle rate conditions. As in Phase 2, the Recipient shall withdraw exhaust gas samples under conditions of varying amounts of flue gas recycle. Exhaust samples will allow particle size distributions to be determined using low-pressure impactors, an aerosol particle sizer (for particles 0.6µm – 20µm), and a scanning mobility particle sizer (for particles 0.01µm – 0.6µm). The low-pressure impactors shall also yield size-segregated composition data.

To meet the second objective, the tests shall be conducted with the same coals that shall be used in the drop-tube studies of Subtask 3.6b, and under practical conditions that can be related to those used in the drop-tube studies of Subtask 3.6b

Subtask 3.6b is investigating the formation of fine ash under oxy-coal conditions and supports the work being performed in the pilot-scale unit (Subtask 3.6a). The work utilizes a drop-tube furnace to control the temperature and gas environment that a single particle of coal experiences. Fine particles are collected via a low-pressure impactor and analyzed with two on-line instruments: a scanning mobility particle sizer (SMPS) and aerosol particle sizer (APS). The combination of these two instruments and purchased coupling software, allows us to investigate the range of particles from 14 nm to 20 microns. Previous work has shown that this range of particles captures a large percentage of the ash. The Recipient shall complete experiments at a range of wall temperatures and oxygen/nitrogen, oxygen/carbon dioxide environments.

In Phase 3 the Recipient shall explore additional coal types, which are being utilized by the pilot-scale team (Subtask 3.6a). The experiments shall utilize the new drop-tube reactor developed under Phase 2, to allow operation at higher temperatures, and to ensure complete burnout of the coal, which has been a problem in the data gathered in the old system.
The data gathered to date have indicated a complicated relationship between wall temperature and oxygen concentration, which changes the particle temperature. A simplified drop-tube model will be developed to better understand the heat transfer in the system. In addition, the Recipient shall begin to develop a simplified nucleation and condensation model of particle formation and particle coagulation to implement in the simulation code.

**Task 4.0 – Power Generation “Retrofit”: Gasification**

The long-term objective of this thrust area is to provide simulation tools for industrial entrained-flow coal gasifiers that will predict: heat transfer by radiation and convection, coal conversion, soot formation, synthesis gas composition and slag behavior with quantified uncertainty.

Producing quantifiable answers to the questions posed by society about gasification systems depends on being able to put reliable error bounds on the results of large-scale simulations. In such simulations, many sources of uncertainty exist, including parameters in governing equations, boundary conditions, simplifications to physical models, and approximations in numerical methods. V/UQ of the gasification simulations will be an integral part of the gasification simulation and application effort. This effort will include a preliminary assessment of the potential variability of a simulation model's computed output given inaccuracies in both the model and in the experimental data.

This thrust area includes the following subtasks:

**Subtask 4.1 – Entrained-Flow Gasifier Simulation and Modeling**

In Phase 2, this subtask focused on developing the first LES simulation of a pressurized entrained-flow gasifier and on validating this with experimental data from a lab-scale entrained-flow gasifier operated at Brigham Young University in the 1980s. In Phase 3, a verification, validation, and uncertainty quantification campaign shall be carried out using data collected from the CANMET gasifier. This task will include critical evaluation of the recent CANMET sponsored-gasifier data along with any future collected data during this phase. Collaboration with CANMET is required for ascertaining estimates of scenario parameter uncertainty and measurement uncertainty. This collaborative effort will be facilitated by Prof. Kevin Whitty. In tandem, the Simulation Team will work to quantify numerical uncertainty and possible key model parameters. A parametric sensitivity test will be performed to determine the set of active variables that most contribute to the uncertainty. Once parameters are selected, a surrogate model (at the least a linear surrogate) will be constructed, and a consistency search will be performed to determine the uncertainty of the predictive capability. Methods of surrogate refinement will be explored including the use of higher order (greater than first order) surrogates and the potential use of Krigging.

Development of the University of Utah’s high-performance simulation tools with Direct Quadrature Method of Moments (DQMOM) will continue including refinement of coal physics models and incorporation of newly acquired models from either literature or other subtasks from this project. Additionally, work is required for assuring algorithmic stability and ensuring tight coupling between the coal and gas phase.

**Subtask 4.2 – Subgrid Mixing and Reaction Modeling**

The One-Dimensional Turbulence (ODT) model has shown the ability to provide quantitative agreement with Direct Numerical Simulation (DNS) data for complex reacting flows including
extinction and reignition. A variant of this model will be extended to include particle transport and reaction for coal combustion and gasification. This will provide high-fidelity data at a fraction of the cost of DNS for use in identifying and validating models.

As part of this effort, the open-source Cantera package will be extended to include detailed treatment for the thermodynamic properties of coal. This will provide a common, open standard for our various simulation codes to deal with thermodynamic properties for coal. Included in this will be the ability to do multiphase chemical equilibrium calculations. Validation will be performed on single-particle coal combustion data.

Subtask 4.3 – Radiation Modeling

The long-term objective of this research is a) to expand the radiative heat-transfer algorithms in the University of Utah LES codes to include coal particles under entrained coal gasification conditions, and b) to implement reverse Monte Carlo ray tracing (RMCRT) for radiative heat transfer in LES for high-pressure pulverized coal gasification. Heat transfer in entrained-flow gasifiers is dominated by radiative heat transfer. Current methods for computing radiative heat transfer in LES codes (including the University of Utah LES code - ARCHES) depend heavily on discrete ordinates methods (DOM) for radiative transfer computations. DOM computations are currently consuming more than half of the computational time for a pulverized-coal combustion simulation. The RMCRT method traces a series of rays through the domain and uses their intensities to calculate the radiative flux divergence of the system. Because each ray's intensity is independent of all other rays, RMCRT scales to massively parallel systems better than the DOM. Furthermore, RMCRT handles physical phenomena inherent with participating media better than the DOM. This will lead to faster computation time while simultaneously increasing accuracy.

In Phase 2, the RMCRT algorithm for single processor application and for parallelization on thousands of processors was identified, coded and tested. Ray tracing performance on single processors was significantly improved over previous implementations. The algorithm for achieving efficient performance on thousands of processors requires significant development in the Uintah Computational Framework (the ARCHES platform). This development is outside the scope of this award, but will occur during the Phase 3 time frame under separate funding. This development will allow the parallelization needed for RMCRT, which will be accomplished in Phase 3. During Phase 3 the general algorithm will be extended to include increased accuracy by including and improving the existing DOM radiation properties models for coal particles under entrained-flow gasification conditions. During Phase 3 further validation studies will be performed to quantify and potentially reduce the uncertainty in the accuracy of the particle radiation models and properties.

Subtask 4.4 – Char and Soot Kinetics and Mechanisms

This subtask addresses reactivity of coal chars as well as formation and reactivity of soot formed during coal gasification.

Pressurized coal gasification. Three additional coals of different rank will be studied (Illinois #6 hvb bituminous, Utah hvc bituminous, and Indiana Upper Freeport medium volatile). The Illinois #6 coal is being studied in the gasifier at the University of Utah. This is a unique opportunity to collect fundamental gasification data for the same coal as used in a pilot-scale gasifier. These additional coals will allow for correlations to be made for pressure-dependent swelling properties at the high heating rates expected in gasifiers, as well as for char gasification kinetics.
A pressurized flat-flame burner has largely been operated with CO as the fuel, with a small amount of H_2 to help burn the CO. We will add extra H_2 to increase the steam concentration in order to obtain steam-char gasification rates. Submodels for pyrolysis, swelling, and char gasification will be extended to treat more coals. Correlations for kinetic and swelling coefficients will be made based on new data obtained at high heating rates as well as for data in the literature.

**Soot formation and gasification.** Soot formation experiments will focus on actual coal tars and will be largely conducted at elevated pressure. The best coal tar surrogate identified from the atmospheric secondary pyrolysis experiments seemed to be biphenyl. This compound will now be the only surrogate examined at elevated pressure. Data will be used to include pressure effects into the soot model previously developed at atmospheric pressure. Gasification experiments of soot in a pressurized TGA will be initiated on collected samples from the pressurized flat-flame burner. Size and porosity characteristics of soot at various pressures and various times will be analyzed with small angle X-ray scattering at Argonne National Laboratory's user facility. This information will help the Recipient explore early soot and char formation, and it is a critical component of the development of an improved char structure model. In addition, this data will provide the probability distribution function that indicates the spatial distribution of the carbon, hydrogen, nitrogen, sulfur, and oxygen atoms in the bulk structure of the soot/char components.

**Subtask 4.5 – Slag Formation and Slag-Wall Interactions**

This subtask will terminate at the end of Phase 2.

**Subtask 4.6 – Acquisition of Validation Data in an Entrained-Flow Gasifier**

This subtask will expand work initiated during Phase 2 focused on quantifying performance of the University of Utah’s entrained-flow gasifier and evaluating uncertainty in these measurements. The range of conditions tested will be expanded to higher pressures (20+ atm), and the slurry feed rate will range as high as 50 kg/hr (1.2 mton/day). Wall temperature will be measured at a minimum of four locations simultaneously, and the post-quench synthesis gas composition (min. H_2, CO, CO_2, CH_4, C_2H_4, H_2S) will be measured. In order to provide more detailed, quantitative data for subsequent validation studies, two new activities will be initiated in Phase 3. These are described below.

**Subtask 4.6a - Characterization of injector spray behavior.** A new system will be built to allow visualization and measurement of injector performance. The system will comprise a pressure chamber with windows and a port for mounting the injector in the same fashion as it is in the gasifier. Initially, water and air will be used to simulate slurry and oxygen. Spray angle will be measured, and we will attempt to quantify droplet size distribution and droplet intensity using high-speed photography or video. Performance will be measured as a function of water flow rate, air flow rate and system pressure. Performance observed in the spray chamber will be compared to performance observed in the gasifier.

**Subtask 4.6b - Development of a probe for in-situ measurements.** A new probe will be developed to measure gas composition at different locations within the gasifier. The probe will be mounted to one of the six sampling ports along the length of the gasifier and will be designed to traverse radially from the wall to the centerline of the reactor. The technical feasibility of simultaneous sampling of condensed-phase material will also be considered.
Task 5.0 – Chemical Looping Combustion Reactions and Systems

The ultimate objectives of this task are to develop a new low-cost carbon-capture technology for coal through chemical-looping combustion (CLC) and to transfer this technology to industry through a numerical simulation tool with quantified uncertainty bounds. The specific research targets for these tasks are to quantitatively identify reaction mechanisms and rates, explore operating options with a laboratory-scale bubbling bed reactor, identify process modeling economics and demonstrate and validate simulation tools for a pilot-scale fluidized bed. In Phase 3, the CLC task will primarily focus on CuO/Cu₂O but include iron-based carriers in Subtask 5.1 and 5.2 using data from the literature for kinetics and verification. It will also include chemical looping with gasification products. This task comprises the following subtasks:

Subtask 5.1 – Process Modeling and Economics

The major contribution of this subtask to the chemical looping task is to develop process models for Chemical Looping with Oxygen Uncoupling (CLOU). Copper carrier options are being investigated for the CLOU process. These process models will build upon the preliminary ASPEN® models for CLOU developed previously in Phase 2 and incorporate custom models with specific kinetics within the air and fuel reactors. These kinetics will be derived from the complementary thermogravimetric analysis (TGA) and laboratory fluidized-bed tasks (Subtasks 5.3 and 5.4). The preliminary mass and energy balance ASPEN modules developed in Phase 2 will be replaced by more sophisticated expressions of kinetics for given residence times and reactor sizes. In particular the plug-flow treatment of air and fuel reactors will be augmented with the well-stirred reactor treatment which will require the use of population balances, including an allowance for diffusional and heat-transfer effects. These rate expressions are critical to the development of the process model. The ultimate objective of this subtask is to develop the models such that reactor size, material use, and other economic factors can be minimized, and CLOU can be compared to CLC. For the CLC case, existing kinetic data will be used for iron-based carriers.

Subtask 5.2 – HPC process simulation of pilot-scale fluidized bed and moving-bed CLC

The Phase 3 CLC process simulation objective is the development of a new capability for using high-performance computing (HPC) for moving-bed CLC applications. Moving beds have the potential for better utilization of the metal oxide carrier in the CLC process. The Ohio State research group under the direction of Professor Fan have studied these systems, which involve iron-based carriers, and have experimental data for simulation validation. To accomplish this task, the Recipient shall build on the CLC HPC simulation work performed in Phase 2 for dense two-phase flow systems. Trade-offs between modeling approaches and simulation tools will be studied. Quantification of uncertainty in the HPC simulation tools for moving-bed CLC process applications shall be accompanied with V/UQ work for experimental moving-bed configuration with no reaction using data from literature.

The Recipient shall continue the analysis of the HPC tools for applications to CLC in fluidized-bed configurations initiated in Phase 2. Once the moving-bed simulation capabilities are developed, an optimization study comparing the applicability, advantages and disadvantages of both fluidized-bed and moving-bed systems shall then be performed. The Recipient shall specifically focus on optimizing particle residence time for comparing the two flow configurations with particular attention on reducing the energy penalty and improving conversion. The optimization space and its uncertainty will be explored using simple non-reacting configurations.
Subtask 5.3 – Laboratory-Scale CLC Studies

During Phase 3, the CLC Team will continue evaluation of copper-based carrier performance using laboratory-scale reactors, focusing specifically on the high-temperature regime where oxygen uncoupling (CLOU) occurs. Evaluation will progress from characterization of fundamental chemical behavior of copper carriers towards more industrially relevant aspects including long-term performance (capacity, kinetics and avoidance of agglomeration) over many oxidation and reduction cycles.

Research during Phase 2 confirmed that pure copper oxides are unsuitable as carriers and that the copper must be supported on a substrate. In Phase 3, various supports, including titania and alumina, and CuO loadings to 50 wt%, will be experimentally evaluated. Based on experience with the single bed lab-scale reactor, a design for a larger 10 kWth, dual-bed, continuous flow bench-scale CLOU CLC system will be developed.

Subtask 5.4 – CLC Kinetics

The CLC Team will continue to collect experimental TGA data, augmented with mass spectroscopy data to monitor both the solids and the gases, developing kinetics for copper metal/copper oxides and will continue to assess the statistical uncertainty of the kinetic parameters. In addition during Phase 3, the subtask will focus on 1) the decomposition kinetics and oxidation kinetics of copper-based carriers on supporting materials, 2) the reduction kinetics with methane, and 3) the reoxidation of oxygen carriers with air. The focus of the entire effort is on quantitative assessment of uncertainties for validation purposes.

Task 6.0 – Underground Coal Thermal Treatment

The long term objective of this task is to develop a transformational energy production technology by in-situ thermal treatment of a coal seam for the production of substitute natural gas (SNG) and/or liquid transportation fuels while leaving much of the coal’s carbon in the ground. In this process the coal seam is heated indirectly with some thermal source to produce light gases and tars, as opposed to traditional underground coal gasification where the combustion and gasification reactions provide the heat for pyrolysis. Subsequently the heated formation will be used to inject CO2 captured from other surface processes and thus serve as a CO2 storage reservoir for sequestration. This process converts coal to a high-efficiency, low-GHG emitting fuels. It holds the potential of providing environmentally acceptable access to previously unusable coal resources. By increasing the abundance of cleaner fuels from coal, the proposed technology has the potential for significantly lowering fuel prices relative to other energy sources, and reducing the carbon footprint of coal utilization, thus providing the economic and environmental drivers for displacing imported energy sources.

The Recipient shall integrate the experiments, models and simulations of Phase 2 to create a laboratory scale demonstration of the technology. The Recipient shall also develop process models, obtain process thermo-chemical parameters, and develop CO2 absorption isotherms from this laboratory test facility. In addition, the Recipient shall develop simulation tools for heating, pyrolysis, and CO2 adsorption for the laboratory facility. Data collected from the facility shall be used by the Recipient for V/UQ.
Task 7.0 – Mercury Control

This task will terminate at the end of Phase 2.

Task 8.0 – Strategies for Coal Utilization in the National Energy Portfolio

Coal-fired power plants increasingly face opposition both from litigation and from regulatory uncertainty. At the same time, coal is certain to be a paramount resource, even in a carbon-constrained world, as the nation’s population and power demands continue to expand. Thus, a critical question is the legal and socio-economic position that coal will assume as we move toward climate regulation. In Phase 3, this task will address that question by analyzing regulatory approaches to promoting carbon capture and sequestration (CCS) technology as well as emerging legal challenges to deploying CCS technology.

Subtask 8.1 – Regulatory Promotion of Emergent CCS Technology

It is increasingly apparent that regulatory uncertainty is one of the factors that stand as a roadblock to commercial-scale deployment of CCS technology. Unless the regulatory landscape, including questions of liability, becomes more transparent and definite, investors are likely to remain reticent to back CCS. Especially for an emergent technology such as CCS, which would increase the price of electricity in a system that rewards the lowest-cost fuels, regulatory uncertainty is problematic. Our current climate change regulatory assessment is focused on these questions at a broad level, assessing where regulatory uncertainty—or lack of regulation—creates impediments to CCS.

Another regulatory problem made clear by the climate change regulatory gap assessment, however, is that unlike many other emergent technologies, regulation to directly promote CCS deployment remains somewhat lacking. Federal research and development funds clearly back the technology, but sometimes research alone is insufficient. In other sectors, regulation that directly promotes given technologies is comparatively prevalent, and strong.

This task will build on our current climate change regulatory gap assessment to analyze this more discrete question. Does CCS require direct regulatory promotion to develop on a commercial scale and, if so, what type of such market intervention is most appropriate? The task will juxtapose case studies of other instances where government has promoted energy technologies or innovations. Using these case studies as an analytic frame, the task will extract and analyze which models of technology-forcing regulation tend to work best in the energy industry. Are subsidies necessary? Mandates? Guaranteed prices? Hybrids of different approaches? Based on that assessment, and the lessons learned from the broader regulatory gap assessment, the task will then dissect which of these regulatory models would work best to promote increased use of CCS.

Subtask 8.2 – Emerging Legal Issues for CCS Technology

The legal framework for carbon sequestration continues to evolve, and the implications of these developing legal and regulatory issues will be critical elements of evaluating when and how large-scale carbon capture and sequestration (CCS) technology can be deployed. This task will focus on critical emerging issues, such as pending regulations under the Safe Drinking Water Act and the attendant permitting implications, recently proposed Council on Environmental Quality guidelines for greenhouse gases (recognizing sequestration as an activity requiring
evaluation under the National Environmental Policy Act (NEPA), but exempting federal land management agencies from this requirement), developing NEPA compliance issues for sequestration, the impacts of the Endangered Species Act and the laws governing cultural protection on sequestration, the effect of actions taken by the Environmental Protection Agency under the Clean Air Act on sequestration, the impacts of climate change legislation on sequestration efforts, and evolving federal energy and public land use policies insofar as they impact utilization of CCS technology. This task will build on analysis of federal and state laws relevant to sequestration completed under the Phase 2 climate change regulatory gap assessment.

**Task 9.0 – Validation/Uncertainty Quantification for Large Eddy Simulations of the heat flux in the Tangentially Fired Oxy-Coal Alstom Boiler Simulation Facility**

The ultimate objective of this task is to produce predictive capability with quantified uncertainty bounds for the heat flux in commercial-scale, tangentially-fired, oxy-coal boilers. Data from the Alstom Boiler Simulation Facility (BSF) for tangentially fired, oxy-coal operation will be used for sub-scale validation. The results of the BSF simulations will be applied as appropriate to Alstom’s 350 MWe oxy-coal demonstration design. This capstone project brings together Alstom’s DOE project for measuring oxy-firing performance parameters in the BSF with this University of Utah project for LES and V/UQ. The Utah work will include V/UQ support with measurements in the single-burner facility where advanced strategies for O₂ injection can be more easily controlled and data more easily obtained.

Task 3.0 provides the background for this capstone project by focusing on ignition, coal flame stability, ash partitioning and ash deposition under pulverized oxy-coal conditions. In Task 3.0 the methods for combining measurement and simulation with V/UQ are being demonstrated in the study of ignition and flame stability. Under Task 9.0, these methods will be extended to temperature and heat flux data and move the application to the larger scale BSF.

One of the outcomes of this task is to work with an industrial partner (Alstom) to transfer LES and V/UQ technologies to industrial application.

This task forms the basis for bridging the Phase I through III university research on clean and secure energy from coal to industrial/commercial full-scale applications. This task will include the following subtasks:

**Subtask 9.1 – LES simulation and V/UQ for heat flux in Alstom oxy-coal-fired BSF**

The objective of this subtask is to work directly with Alstom Power to apply the LES and V/UQ strategies developed under this research program to the Alstom BSF and 350 MWe oxy-coal demonstration design. The long-term objective of this research is a) to expand high-performance simulation tools to quantitatively predict the temperature and heat-heat flux in oxy-coal burners for retrofitting power boilers and industrial furnaces for CO₂ capture, and b) to perform verification, validation and uncertainty quantification of the numerical and modeling error associated with this intended use of the simulation tool. The work to be conducted under Phase 3 will take a large step towards this objective by bringing together the DOE funded research at both the university and at Alstom.

In conjunction with Alstom Power, the Recipient shall focus on three activities: simulations of the BSF, application of knowledge to Alstom’s 350 MWe oxy-coal demonstration design, and technology transfer.
At the BSF scale, the Recipient shall:
• Identify selected BSF tests for LES simulations to be used for the V/UQ analysis.
• Obtain adequate uncertainty in the measured data for V/UQ analysis.
• Determine the parameters affecting heat-flux variability in oxy-coal operation of a tangentially fired furnace from the combined experimental and simulation study.
• Establish a simulation matrix for the BSF to explore the uncertainty space from among the most sensitive of the uncertainty parameters.
• Complete a formal V/UQ analysis of the BSF.

At the 350 MWe scale, the Recipient shall:
• Summarize the lessons learned from the BSF simulations that would apply to Alstom’s design of a 350MWe oxy-coal demonstration.
• Perform preliminary LES simulations of the 350 MWe oxy-coal demonstration design.

With respect to the transition of knowledge, the Recipient shall:
• Transfer LES technology.
• Transfer V/UQ technology.

Subtask 9.2 – LES simulation and V/UQ for heat flux in subscale UofU oxy-coal-fired OFC

The objective of this subtask is to extend the simulation work being performed on the OFC under Subtask 3.1 to the temperature and heat flux applications applicable to this Task 9.0 capstone task. Simulations of the OFC appropriate to temperature and heat flux validation of the BSF will be performed by working with the experimental team under Subtasks 9.3 and 9.4. This simulation and validation work will be the underpinning for V/UQ of the larger scale BSF work of Subtask 9.1.

The Recipient shall:
• Work with the experimental and diagnostics teams in Subtasks 9.3 and 9.4 to define selected experiments and simulations of the OFC to be conducted for V/UQ of temperature and heat flux in a firing strategy applicable to full scale tangential firing for the single burner oxy-coal OFC.
• Quantify uncertainties in the scenario parameters, model parameters, numerical outputs, and experimental outputs from the selected set of simulations and experiments relating to temperature and heat flux. This will be accomplished by working closely with the diagnostics team of Subtask 9.3 and the experimental team of Subtask 9.4.
• Complete the V/UQ simulations and analysis for temperature and heat flux in the OFC as identified above.
• Provide an assessment to Alstom and DOE of the OFC V/UQ and apply knowledge from this subtask into Subtask 9.1, as appropriate.

Subtask 9.3 – IR camera diagnostics & V/UQ for Temperature measurements in UofU OFC

The objective of this subtask is to develop and perform diagnostics for the OFC during its operation to be used for high-resolution model validation. Validation will be performed on both
radiative flux and flame temperature based on infra-red (IR) imaging, as opposed to the
temperature maps based on images obtained with emissions in the visible ranges performed
with the velocity measurements in Subtask 3.3. To accomplish this subtask, the Recipient shall
design, fabricate, and calibrate multiple wide-angle radiometers (2π steradians). The
radiometers will first be calibrated with a black-body radiation source and then installed in the
OFC reactor in cooperation with Subtask 9.4. In addition, the Recipient shall measure
temperatures inside the OFC with a newly purchased high-speed infra-red (IR) camera. The
Recipient shall develop data-reduction methods to correlate pixel intensity to temperature. This
camera will first be calibrated with a black-body radiation source and then mounted on the OFC
reactor in cooperation with Subtask 9.4.

Once these diagnostic capabilities are installed in the OFC, they will be used as part of the
experimental verification and validation campaigns, as described in Subtask 9.4. The
radiometer and IR camera results from these tests will require data reduction and analysis, and
the investigators shall work closely with the investigators in Subtask 9.2 and 9.4 to integrate
their measurement results with the simulation results and other experimental measurements.

Subtask 9.4 – Heat flux profiles of UofU OFC using advanced strategies for O₂ injection

The objective of Subtask 9.4 is to obtain validation data of heat flux and temperature profiles for
axial flame simulations developed under Subtask 9.2, and to make comparisons between oxy-
and air- firing configurations. Data shall be obtained using diagnostic tools developed under
Task 9.3.

The Recipient shall:

• Meet with Alstom Power to determine which burner configurations among those listed
  below, would be of practical interest:
  
    o “Standard” oxy-coal co-axial burner configuration, as used in previous work on
      oxy-coal flame ignition.
    o “Advanced” oxy-coal multi-annular co-axial burner configuration, as used in a
      Subtask 3.2 that explored directed oxygen injection to minimize flue-gas recycle.

• Modify OFC to accept the diagnostic tools developed under Subtask 9.3.

• Develop and implement a zero-flow experiment (in collaboration with 9.2 and 9.3) for
  experimental verification of temperature and heat-flux measurements and validation of the
  heat transfer simulation sub-model.

• Measure heat-flux profiles and temperature profiles for oxy-coal co-axial flames of practical
  interest (as determined by 9.1) above, as validation experiments for Subtask 9.2. Diagnostic
  tools developed under Subask 9.3 shall be used. Heat-flux and temperature profiles for oxy-
  firing shall be compared to those for air firing.

D. DELIVERABLES (Phase 3)

The Recipient shall provide reports in accordance with the enclosed Federal Assistance
Reporting Checklist and the instructions accompanying the Checklist. The following Topical
Reports are due at the completion of each of the listed Tasks.
• Topical Reports
  - Task 3.0
  - Task 4.0
  - Task 5.0
  - Task 6.0
  - Task 8.0
  - Task 9.0

Topical reports are to be submitted electronically in pdf format directly to our Document Control Office at FITS@NETL.DOE.GOV following the format and instructions provided in Attachment 4 – Federal Assistance Reporting Checklist.

In addition to the reports identified on the Reporting Checklist, the Recipient shall provide the following deliverables directly to the NETL Project Officer by e-mail or other mutually agreed upon method:

• Updated Project Management Plan
• An improved high-performance simulation tool for simulating a pilot-scale oxy-coal combustor.
• V&V/UQ study of the CANMET gasifier with quantified uncertainty.
• A process model for comparison of different oxygen carriers and the CLC versus CLOU processes.
• V/UQ analysis of the BSF.
• At a minimum - eight papers related to the clean coal utilization for power generation.
  The papers will be prepared for publication in trade journals, peer-review journals, peer-reviewed conference proceedings, or law journals.

E. BRIEFINGS (Phase 3)

The Recipient shall prepare detailed briefings for presentation to the Project Officer at the Project Officer’s facility located in Pittsburgh, PA or Morgantown, WV. Briefings shall be given by the Recipient to explain the plans, progress, and results of the technical effort on an annual basis. DOE may substitute attendance of meetings at NETL with Recipient participation in external project/merit reviews. The Recipient shall provide and present a technical paper(s) at the DOE/NETL Annual Contractor’s Review Meeting, as necessary, held at the NETL facility located in Pittsburgh, PA or Morgantown, WV, or at an alternate location mutually agreed upon by the NETL Project Officer and the Recipient.