Fuel-Flexible Gasification-Combustion Technology for Production of H₂ and Sequestration-Ready CO₂

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ABSTRACT

It is expected that in the 21st century the Nation will continue to rely on fossil fuels for electricity, transportation, and chemicals. It will be necessary to improve both the process efficiency and environmental impact performance of fossil fuel utilization. GE Global Research (GEGR) has developed an innovative fuel-flexible Unmixed Fuel Processor (UFP) technology to produce H₂, power, and sequestration-ready CO₂ from coal and other solid fuels. The UFP module offers the potential for reduced cost, increased process efficiency relative to conventional gasification and combustion systems, and near-zero pollutant emissions including NO_x. GEGR (prime contractor) was awarded a Vision 21 program from U.S. DOE NETL to develop the UFP technology. Work on this Phase I program started on October 1, 2000. The project team includes GEGR, Southern Illinois University at Carbondale (SIU-C), California Energy Commission (CEC), and T. R. Miles, Technical Consultants, Inc.

In the UFP technology, coal/opportunity fuels and air are simultaneously converted into separate streams of (1) pure hydrogen that can be utilized in fuel cells, (2) sequestration-ready CO₂, and (3) high temperature/pressure oxygen-depleted air to produce electricity in a gas turbine. The process produces near-zero emissions and, based on process modeling with best-case scenario assumptions, has an estimated process efficiency of 68%, based on electrical and H₂ energy outputs relative to the higher heating value of coal, and an estimated equivalent electrical efficiency of 60%. The Phase I R&D program will determine the operating conditions that maximize separation of CO_2 and pollutants from the vent gas, while simultaneously maximizing coal conversion efficiency and hydrogen production. The program integrates lab-, bench- and pilot-scale studies to demonstrate the UFP technology.

This is the eleventh quarterly technical progress report for the Vision 21 UFP program supported by U.S. DOE NETL (Contract No. DE-FC26-00FT40974). This report summarizes program accomplishments for the period starting April 1, 2003 and ending June 30, 2003. The report includes an introduction summarizing the UFP technology, main program tasks, and program objectives; it also provides a summary of program activities and accomplishments covering progress in tasks including lab-scale experimental testing, pilot-scale assembly, and program management.



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EXECUTIVE SUMMARY

This is the eleventh quarterly technical progress report for the Vision 21 UFP program supported by U.S. DOE NETL (Contract No. DE-FC26-00FT40974). This report summarizes program accomplishments for the period starting April 1, 2003 and ending June 30, 2003. The report provides a description of the technology concept and a summary of program activities and accomplishments covering progress in tasks including lab-scale experimental testing, pilot-scale fabrication and assembly, engineering and modeling analyses, and program management.

In the UFP technology, coal/opportunity fuels and air are simultaneously converted into separate streams of (1) pure hydrogen that can be utilized in fuel cells, (2) sequestration-ready CO₂, and (3) high temperature/pressure oxygen-depleted air to produce electricity in a gas turbine. The process is highly efficient relative to conventional electricity producing technologies and produces near-zero emissions. The Phase I R&D program will determine the operating conditions that maximize separation of CO_2 and pollutants from the vent gas, while simultaneously maximizing coal conversion to electricity efficiency and hydrogen production. The program integrates lab-, bench- and pilot-scale studies to demonstrate the UFP technology.

Work conducted in the eleventh quarter of Phase I has focused on accelerating assembly of the pilot plant, with additional experimental analysis being conducted on the lab scale system.

The lab-scale effort has included TGA experiments to evaluate and quantify the kinetics of OTM reduction and OTM speciation as a function of temperature. This information will provide key kinetic parameters for integration in process and kinetic modeling of the system. The residence times of solids in the pilot-scale system will be set based on kinetic modeling results to ensure that sufficient reaction has occurred.

The pilot-scale assembly effort has continued, with the development of a detailed system layout. Additional progress was made designing and testing the second-stage superheaters, designing and initiating construction of the air pollution control systems, testing the slurry feeding system and conducting a detailed safety analysis. In addition, methods were developed for estimating bed heights, estimating start-up times, removing water from product gases and sampling bed solids.

Modeling work conducted in the current reporting period has focused on development of an ASPEN process model of the pilot-scale system to assist in determining initial operating conditions for system shakedown.



INTRODUCTION

Electricity produced from hydrogen in fuel cells can be highly efficient relative to competing technologies and has the potential to be virtually pollution free. Thus, fuel cells may become an ideal solution to many of this nation's energy needs if one has a satisfactory process for producing hydrogen from available energy resources such as coal, and low-cost alternative feedstocks including biomass, municipal solid waste, sewage sludge, and others.

This Vision 21 UFP program addresses a novel, energy-efficient, and near-zero pollution concept for converting a conventional fuel (coal) and opportunity fuels (e.g., biomass) into separate streams of hydrogen, oxygen-depleted air, and sequestration-ready CO₂. The technology module encompassing this concept will be referred to as the *Unmixed Fuel Processor (UFP)* throughout this report. When commercialized, the UFP technology may become one of the cornerstone technologies to fulfill Vision 21 energy plant objectives of efficiently and economically producing energy and hydrogen from coal with utilization of opportunity feedstocks.

The UFP technology is energy efficient because a large portion of the energy in the input coal leaves the UFP module as hydrogen and the rest as high-pressure, high-temperature gas that can power a gas turbine. The combination of producing hydrogen and electricity via a gas turbine is highly efficient, meets all objectives of Vision 21 energy plants, and makes the process product flexible. That is, the UFP module will be able to adjust the ratio at which it produces hydrogen and electricity in order to match changing demand.

The Phase I Vision 21 UFP program is primarily being conducted by General Electric Global Research (GEGR), under a Vision 21 contract from U.S. DOE NETL (Contact No. DE-FC26-00FT40974). Other project team members include Southern Illinois University at Carbondale (SIU-C), California Energy Commission (CEC), and T. R. Miles, Technical Consultants, Inc. The UFP project integrates lab-, bench- and pilot-scale studies to demonstrate the UFP technology. Engineering studies and analytical modeling are being performed in conjunction with the experimental program to develop the design tools necessary for scaling up the UFP technology to the demonstration phase. The remainder of this section presents objectives, concept, and main tasks progress of the UFP program.

Program Objectives

The primary objectives of the UFP program are to:

- Demonstrate and establish the chemistry of the UFP technology, measure kinetic parameters of individual process steps, and identify fundamental processes affecting process economics.
- Design and develop bench- and pilot-scale systems to test the UFP technology under dynamic conditions and estimate the overall system efficiency for the design.
- Develop kinetic and dynamic computational models of the individual process steps.
- Determine operating conditions that maximize separation of CO₂ and pollutants from vent gas, while simultaneously maximizing coal/opportunity fuels conversion and H₂ production.
- Integrate the UFP module into Vision 21 plant design and optimize work cycle efficiency.
- Determine extent of technical/economical viability & commercial potential of UFP module.



UFP technology

The conceptual design of the UFP technology is depicted in Figure 1. The UFP technology makes use of three circulating fluidized bed reactors containing CO_2 absorbing material (CAM) and oxygen transfer material (OTM), as shown in Figure 1. Coal and some opportunity fuels (5-10% by heat input) are partially gasified with steam in the first reactor, producing H₂, CO and CO₂. As CO₂ is absorbed by the CO₂ sorbent, CO is also depleted from the gas phase via the water-gas shift reaction. Thus, the first reactor produces a H₂-rich product stream suitable for use in liquefaction, fuel cells, or turbines.

Gasification of the char, transferred from the first reactor, is completed with steam fluidization in the second reactor. The oxygen transfer material is reduced as it provides the oxygen needed to oxidize CO to CO_2 and H_2 to H_2O_2 . The CO₂ sorbent is regenerated as the hot moving material from the third reactor enters the second reactor.



Figure 1. Conceptual design of the UFP technology.

This increases the bed temperature forcing the release of CO_2 from the sorbent, generating a CO_2 -rich product stream suitable for sequestration.

Air fed to the third reactor re-oxidizes the oxygen transfer material via a highly exothermic reaction that consumes the oxygen in the air fed. Thus, reactor three produces oxygen-depleted air for a gas turbine as well as generating heat that is transferred to the first and second reactors via solids transfer.

Solids transfer occurs between all three reactors, allowing for the regeneration and recirculation of both the CO_2 sorbent and the oxygen transfer material. Periodically, ash and bed materials will be removed from the system and replaced with fresh bed materials to reduce the amount of ash in the reactor and increase the effectiveness of the bed materials.

Project Plan

The tasks planned for the UFP project are summarized in Table 1. These tasks are being conducted over approximately three-year period that started October 1, 2000. The success of the UFP program depends on the efficient execution of the various research tasks outlined in Table 1 and on meeting the program objectives summarized above.



PROGRAM PLANNING AND MANAGEMENT

Program planning activities have focused on meeting the objectives of the program as stated previously. GEGR has made use of several GE methodologies to obtain desired results and systematically conduct program design, construction and testing activities. Methodologies utilized in this program include New Technology Introduction (NTI) and Design For Six Sigma (DFSS). The NTI program is a detailed and systematic methodology used by GE to identify market drivers, and continually ensure that the program will meet both current and future market needs. The NTI program is also strongly coupled with the DFSS other quality and programs, providing structure to the design process and ensuring that the design accomplished through regular program reviews, detailed design reviews. market assessments, planning and decision tools, and specific quality projects aimed at identifying system features and attributes that are critical to quality (CTQ) for customers.

The project team meets weekly to assess progress, distribute workload, and identify and remove potential roadblocks. An expanded NTI Table 1. Main Tasks of the UFP program.

ruote i. main	rusius of une off program.		
Task	Task Description		
Lab-Scale	Design & assembly		
Experiments –	Demonstration of chemical		
Fundamentals	processes		
Task 1	Sulfur chemistry		
	Bench test facility design		
	Subsystems procurement&		
Bench-Scale Test	assembly		
Facility & Testing	Bench test facility shakedown		
	Reactor design testing		
Tasks 2 & 3	Parametric evaluation		
	Fuel-flexibility evaluation		
	Pilot operation support		
	Opportunity fuels resource		
Engineering &	assessment		
Modeling Studies	Preliminary economic assessment		
-	Kinetic & process modeling		
Task 4	Integration into Vision 21 plant		
	Pilot plant control development		
	Process design		
	Subsystems		
	specification/procurement		
Dilot Diant Dogian	Reactor design & review		
Assombly 8	Reactors manufacture		
Demonstration	Components testing		
Demonstration	Pilot plant assembly		
Tasks 5 6 8 7	Operational shakedown		
1 asks 5, 6, 6 1	modifications		
	Operational evaluation		
	Fuel-flexibility evaluation		
	Performance testing		
Vision 21 Plant	Preliminary Vision 21 module		
Systems Analysis	design		
Task 8	Vision 21 plant integration		
	Economic & market assessment		
Project Management	Management, reporting, &		
Task 9	technology transfer		

project team that includes senior management and other expert personnel also meets biweekly to gauge progress and ensure that adequate company resources are allocated and technical issues resolved to allow steady progress toward program objectives.

Program management activities also involve continuous oversight of program expenditures. This includes monthly review of actual expenditures and monthly projections of labor, equipment, contractor costs, and materials costs.

Technology transfer and networking with experts in the advanced power generation field is an important and ongoing part of project management. Team members continue to seek out



opportunities to present the UFP technology and progress at technical conferences. During the eleventh quarter, one paper was submitted for the Twentieth Annual International Pittsburgh Coal Conference to be held in Pittsburgh, PA from September 15-19, 2003. Earlier this year, an abstract was also submitted for the Twelfth International Conference on Coal Science (ICCS), to be held November 2-6 in Cairns, Queensland, Australia.

During this quarter, additional results from the experimental facilities were obtained, analyzed and used to assess operating characteristics of the system. The laboratory-scale activities are being conducted by SIU in Carbondale, IL, while the bench-scale and pilot-scale systems are located at GEGR's test facility in Irvine, CA. Significant progress was made toward the assembly of the pilot-scale system located at Irvine, CA.

EXPERIMENTAL

LABORATORY-SCALE TESTING

The primary objective of Task 1 is to perform a laboratory-scale demonstration of the individual chemical and physical processes involved in GEGR's fuel-flexible UFP technology. Specific objectives of Task 1 include:

- Support bench- and pilot-scale studies;
- Assist in process optimization and engineering analysis;
- Identify key kinetic and thermodynamic limitations of the process; and
- Verify the process parameters at laboratory scale.

Work conducted in the eleventh quarter of this program has focused on a kinetic investigation of the oxygen-transfer material (OTM) reduction and speciation reactions using a thermogravimetric analyzer (TGA) as well as initiating a test matrix of high-temperature, fluidized bed experiments to validate the TGA results. Details of the fluidized bed experiments will be provided after the test matrix has been completed.

The objective of the TGA experiments is to generate data that can be used to evaluate different kinetic mechanisms and derive kinetic constants. TGA experiments were conducted using a Perkin-Elmer TGA-7 thermogravimetric analyzer with a TAC 7/DX control box upgrade driven by Pyris software. OTM samples (~12 mg) were preheated under a N₂ atmosphere (heating rate 10°C/min) to the desired temperature (700-900°C). This temperature was then maintained as a reducing gas (a mixture of CO and H₂ in N₂) was fed at a flow rate of 30 ml/min. Pressurized gas cylinders of N₂, CO and H₂ were used to feed the reducing gas mixture. The gases were dried using a molecular sieve moisture trap before being fed to the TGA.

TGA experimental results include the weight change of a sample as a function of time. This weight change can be directly related to the extent of the reaction conversion, since oxidized OTM (OTM-O) has a different molecular weight than reduced OTM (OTM-R). Reaction stoichiometry dictates that a weight loss of 10% corresponds to complete reaction from OTM-O to OTM-R. The extent of conversion [$\alpha(t)$] was calculated using the formula below:



$$\alpha(t) = \frac{m_0 - m(t)}{m_0 - m_{10\%}} \tag{1}$$

Where:

 m_0 is the initial mass, m(t) is the mass at time t, and $m_{10\%}$ is the mass corresponding to complete conversion (10% mass loss).

The Avrami-Erofe'ev method was used to compare the kinetics of the isothermal solid-state reactions taking place in the TGA. The method is based on an equation describing nucleation and growth processes:

$$\alpha = 1 - \exp(-\beta t^{m}) \tag{2}$$

$$\ln(-\ln(1-\alpha)) = \ln\beta + m\ln t \tag{3}$$

Where:

- α is the extent of conversion at any given time, t
- β is a constant, partially depended both on nucleation frequency and rate of grain growth *m* is a constant associated with the geometry of the system

Plots of equation (3) yield lines with slopes m (the linear region of such plots is generally for α values between 0.15 and 0.50). The value of m is indicative of the specific solid-state kinetic mechanism, as described in Table 2. Results from these tests are summarized in the Results and Discussion section.

$1 - (1 - \alpha)^{1/3} = kt$ $\alpha = 1 - (1 - kt)^3$	m=1.07; Equation for phase-boundary-controlled reaction (surface reaction) for a sphere	(4) (5)
$-\ln(1-\alpha) = kt$ $\alpha = 1 - \exp(-kt)$	m=1; Equation for first-order reaction	(6) (7)
$[-\ln(1-\alpha)]^{1/2} = kt$ $\alpha = -\exp(-k^2t^2) + 1$	m=2; Avrami-Erofe'ev equation for phase change model	(8) (9)

TABLE 2. Selected solid-state reaction rate equations.

BENCH-SCALE TESTING

The objectives of the bench-scale testing task are to demonstrate the technical feasibility of the UFP technology and aid in developing modeling tools and pilot plant equipment design. The bench-scale system is also intended to provide data on individual UFP reactor modes to aid in pilot plant design and testing. Bench-scale testing was not conducted in the eleventh quarter to allow accelerated progress on the pilot-scale system. Testing will be resumed as needed to further investigate key behaviors next quarter.



RESULTS AND DISCUSSION

LABORATORY-SCALE TESTING RESULTS

TGA experiments were conducted at a range of reducing gas compositions. In each test, $90\% N_2$ was fed, with the remaining 10% varying from all H₂ to all CO and various mixtures between. Selected results are provided below.

The reaction time scale varies widely (particularly for lower temperatures) for reduction by CO and by H_2 , as shown in Figures 2 and 3. Note the difference in time scales, as at 700°C, complete reduction by CO is achieved after 30 minutes, while reduction by H_2 is complete after only one minute. The figures also indicate that increased temperatures reduce the time required to achieve complete conversion for any reducing gas. For example, at the expected pilot-scale operating temperature for OTM reduction (~900°C), the reaction time-scale difference reduces as complete reduction by CO is achieved in about three minutes, while reduction by H_2 is achieved in less than half a minute. Previous bench-scale data have shown similar behavioral trends.



Figure 2. Conversion degree as a function of time for a 90% N_2 , 10% CO mixture at a variety of temperatures.

Figure 3. Conversion degree as a function of time for a 90% N_2 , 10% H_2 mixture at a variety of temperatures.

Preliminary kinetic analysis suggests that the initial reduction by CO (up to 50% conversion) is best described by the first-order reaction model. The observed average m-value was 0.9, close to the value of 1.0 predicted by the first-order model. For initial reduction by H_2 , the average mvalue was 1.7, and analysis suggests that the Avrami-Erofe'ev phase change model (m=2) best describes this data.

Mixtures of CO and H₂ were also evaluated, and the results are shown in Figures 4 and 5. Results indicate that the Figure 4 mixture $(5.7\%CO, 4.3\% H_2)$ requires less time to achieve complete conversion (~5 minutes) than the Figure 5 H₂-dominant (2%CO, 8%H₂) mixture (~25 minutes).

These results suggest that conversion time is not linear with %H₂. Similar results were reported for the bench-scale system. Kinetic analysis indicated that the initial reduction behavior of both mixtures was best described by the Avrami-Erofe'ev phase change model. The Figure 4 mixture had an average m-value of 1.6, while the H₂-dominant mixture (Figure 5) had an average m-value of 1.15. Additional analysis of data to confirm stated observations is continuing.



of time for a 90% N_2 , 5.7% CO, 4.3% H_2 mixture at a variety of temperatures.

Figure 5. Conversion degree as a function of time for a 90% N_2 , 2% CO, 8% H_2 mixture at a variety of temperatures.

Using k-values from the derived kinetic expressions along with temperatures, reaction data were used to derive preliminary activation energy values. Due to some perturbations associated with low temperatures (<780°C), activation energies were derived using only data from temperatures between 780-900°C, as recommended by Tokuda (1979).

ENGINEERING AND MODELING STUDIES

Process Modeling

The objectives of the Process Modeling task are to develop models for the UFP technology, validate them using experimental data, and apply the models to assist in the design and operation of the pilot-scale system. In addition, process models will be used to make meaningful comparisons of the performance of the UFP technology relative to competing technologies.

Modeling work conducted in the eleventh quarter has focused on development of an ASPEN process model of the pilot-scale system to assist in determining initial operating conditions for system shakedown. The key process variables were identified during a series of six-sigma workout sessions. Table 3 lists these input variables, along with their respective limiting values. The targets for output values are also listed. This information formed the foundation of the ASPEN model. The limiting values and targets were defined based on available models, correlations, experimental results, or program objectives, as shown in the notes in Table 3.



targets for output values.				
Input Variables	Limits	Output Values	Targets	
Coal feed (lb/hr)	2-100 ^a	Solids transfer rates (kg/hr)	100-1000 ^b	
% Water in slurry	20-50 ^b	R1 outlet temp. (°C)	150-800 ^e	
Steam flow R1 (lb/hr)	185-320 ^a	R2 outlet temp. (°C)	900-950 ^e	
Steam flow R2 (lb/hr)	155-270 ^a	R3 outlet temp. (°C)	$1000-1150^{\rm f}$	
Air flow R3 (lb/hr)	178-378 ^a	% H ₂ (R1)	Maximize ^g	
Steam temp. R1 & R2 (°C)	400-900 ^c	% CO (R2)	Minimize ^g	
Air temp. R3 (°C)	400-900 ^c	% CH ₄ (R1)	Minimize ^g	
R2 solids split ratio	1:1, 1:2, 1:3 ^d	% CO ₂ (R2)	Maximize ^g	
Carbon burnout (%)	10-90 ^d	% O2 (R3)	Minimize ^g	
Solids transfer temp. (°C)	400-900 ^b			
Solids transfer steam flow (lb/hr)	50-70 ^b			

TABLE 3. ASPEN process model parameters:	Limiting values of key input variables and				
targets for output values.					

Source of estimate:

- a. Fluidized bed correlations
- b. Experimental results (Cold-flow model and slurry pump testing)
- c. Instrumentation limits
- d. Preliminary information from mass & energy balance
- e. Bench-scale experimental results
- f. Bench-scale experimental results (min) and phase diagram (max)
- g. Program objectives

An ASPEN based model was developed for the UFP pilot-scale system. In this model, three reactors are interconnected with solids transfer ducts, with coal and steam fed to the first reactor, steam fed to the second reactor, air fed to the third reactor, and auxiliary steam fed to the solids transfer ducts to entrain and transport bed materials between reactors. Unit operations unique to ASPEN include the use of a virtual coal decomposer (because ASPEN does not recognize coal as a component), a separator unit (to separate the solids and the gases exiting each reactor), a mixer (to add steam to the solids being transferred between reactors), and a solids splitter (to divide the solids stream exiting Reactor 2 into R1 and R3-bound components according to the specified split ratio). The ASPEN flow diagram is shown in Figure 6.

Two constraints were placed on convergence. The first is the R1 product gas temperature. Based on the optimized performance achieved during bench-scale testing, the R1 product gas temperature target was 750°C with a tolerance of $\pm 5\%$. The solids transfer rate from R2-R1 is adjusted when the R1 product temperature goes out of this range.

The second constraint is the temperature of the product gas from Reactor 3. This temperature is limited to $1150^{\circ}C \pm 5\%$ at the initial shakedown stage, but may be increased later as data is collected relative to the state of OTM with temperature in the R3. The flow rate of air fed to R3 is adjusted when the R3 product temperature goes out of range.



Figure 6. ASPEN process flow diagram, showing three main reactors and associated mixers and splitters.

Since the operating range of the input variables listed in Table 3 is so large, and due to the number of input variables, the initial focus of the process model was to identify the impact of the coal feed rate and the % water in the slurry on system performance. With this in mind, other variables were held constant for initial evaluations. Selected results of modeling efforts are provided below.

The product gas concentration of R1 is a key measure of overall system performance. Figure 7 shows the selected ASPEN model results for R1 product gas composition as a function of coal feed rate. H₂ constitutes the majority of the R1 product gas composition, and there is a comparatively small amount of CO_2 slip. The amount of CO is generally low, but increases slightly at higher coal feed rates (to 2-3%).



Figure 7. R1 product gas concentration as a function of coal feed rate at different slurry water concentrations.

Figure 8 shows the trends in R2 product gas composition for different coal feed rates and coalwater slurry compositions. As depicted in this figure, CO₂ is the predominant component (~60-70%), followed by H₂ (~20-30%) and CO (~2-5%). CO and H₂ concentrations decrease slightly with increasing coal feed rates. The water concentration in the slurry does not seem to affect the product gas concentration. The significant amount of H₂ slip suggests that the amount of char transferred from R1 to R2 could be in excess of that needed to reduce the OTM present in R2. This implies that higher coal conversions in Reactor 1 or increased amount of OTM in R2 may be necessary to minimize CO and H₂ slip from R2 product gas. Additional modeling test runs will be conducted to further investigate this behavior and minimize CO and H₂ slip.





Preliminary conclusions based on this analysis include:

- \circ Carbon burnout in R1 and amount of OTM in R2 need to be optimized to minimize CO and H₂ slip from R2 product gas.
- OTM oxidation in R3 is not impacted by coal feed rate or water percentage in coal slurry.
- Increased OTM reduction can be achieved with high coal feed rates and low water percentage in coal slurry, although this also results in higher solids transfer rates.
- Lower coal feed rates result in higher CAM decomposition in R2.

The modeling work is continuing to provide insight into pilot-scale operation, and additional results will be reported in the next quarter.

PILOT PLANT ASSEMBLY

The assembly of the pilot plant has continued in the eleventh quarter. Although delays in obtaining a construction and operating permit have prevented the system from being assembled as a single unit, work has progressed on individual components. In addition, the majority of all equipment and instrumentation is currently on site and awaiting permit approval. A summary of key activities and accomplishments is provided below.

Safety Analysis

(H)

A detailed safety and hazard analysis has been conducted for the pilot plant, following design for Six Sigma (DFSS) methodologies. A failure mode and effects diagram was developed to identify

potential hazards, their causes and effects, as well as possible mitigation steps that could be taken to minimize their likelihood or severity. In addition, the safety and emergency shutdown system has been designed, including the shutdown state of every piece of energized equipment and a detailed understanding of the path the venting gases will take. The criteria for different levels of shutdown and alarms have also been quantified. Standard operating procedures are currently being written and reviewed for every major piece of equipment, including decision trees for various types of system malfunctions.

Second-Stage Superheaters

The second-stage superheaters have been designed to provide an increased degree of superheat after the primary boiler/superheater product gas has been split, with the flow to each reactor and solids transfer leg metered and measured. Thus, a secondstage superheater will be used for each steam flow, as instrumentation for monitoring flow is not readily available at the required temperatures and pressures. The second-stage superheaters consist of an electric furnace, which contains a metal coil, as shown in Figure 9.

Key design elements include the following:

- 46kW Furnace
- o Coil:
 - $-\frac{3}{4}$ " OD x 0.062" Wall Inconel Tubing
 - 168ft Extended Length
 - -18" Coil OD
 - -1" coil spacing (wall to wall)
- 4" Refractory insulation
- 1" Coil to Heating Element Gap (wall to wall)
- 1000°C Heating Element

The length of the coil and the size of the furnace were specified based on detailed heat transfer analysis.

Coal Slurry Feed System

During the eleventh quarter, experimental investigations have continued for the coal slurry feeding system. In an initial atmospheric coal slurry test (50% water), the slurry flow rate was found to be linear over the pump operating range, and offset only slightly from the flow rate of water for the same pump settings. These results are shown in Figure 10. Initial testing of the pump discharging into a pressurized vessel was conducted with water for simplicity and clarity. The testing showed a marked departure from flow linearity at increased pressures (Figure 11). Investigation showed that the source of the problem was a fluttery pressure relief valve that was malfunctioning and relieving pressure at lower pressures than the manufacturer specified.



Figure 9. Schematic diagram of second-stage superheater: electric furnace and heating coil.

Although the potential for developing plugs in the slurry line had initially prompted the use of a pressure relief valve, safety analysis showed the potential for relieving the reactor pressure through this relief valve. Thus, an alternate scenario has been investigated, and includes the use of a pressure switch to protect the pump from overheating/overpressurizing the line in the event of a plug preventing flow to R1. Once this instrumentation has been installed, testing will proceed with water to validate the flow profile, and then with coal slurry to calibrate the offset from the water flow rate.



Figure 10. Flow rate of coal and water at increasing slurry pump speeds.

Figure 11. Water flow rate at increasing slurry pump speeds for three pressures.

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Auxiliary Systems

Condensation of Water from Product Gas

The UFP process makes use of a large amount of water as steam for fluidization of two of the reactors and also for bed solids transfer. The majority of this water must be removed from the product gas to provide greater mass balance accuracy, to prevent flowmeter malfunction due to

uncontrolled condensation, to avoid excessive load on the afterburner and to prevent equipment damage or malfunction in the product gas analyzers. The product gas from R1 and R2 has higher water concentrations because steam, and not air, is used as the fluidizing gas for these two reactors. The R3 product gas water content comes primarily from the steam used for solids transfer, since air is used as the fluidizing gas.

Water will be removed from the bulk of the product gases via a set of mist eliminators, one for each product line. The mist eliminators were selected as the most cost-effective means of water removal after a detailed review of available methods and equipment. As shown in Figure 12, mist eliminator units are vessels containing a fine mesh that will trap 99.9% of the water droplets that form upon expansion of the water-



Figure 12. Schematic diagram of mist eliminator for water removal from product gas.



containing product gas. The condensed liquid accumulates at the bottom of the vessel and will be drained as needed, while the dry gas passes through the mesh and continues to the afterburner. The use of secondary sample conditioning equipment for the slipstream of product gas sent to the continuous analyzers is currently under evaluation, and shakedown testing of the mist eliminators will evaluate their effectiveness and the consequent need for additional water removal prior to gas analysis.

Bed Height Measurement

Since the UFP includes three circulating fluidized beds, the ability to accurately monitor the bed height of each reactor is extremely important. Bed height can provide indications of inadequate solids transfer rates between reactors and accumulation of coal ash in the system. A method of continuously monitoring bed height has been developed and was previously tested in the cold flow model. It is based on a derivation of the equation that estimates pressure drop across a bed of solids: $(\Delta p/\rho_f)=\Delta h$, where Δh is the pressure-head loss in units of length of the flowing fluid. Figure 13 shows a diagram of R2 with the locations for differential pressure measurements. The formula for calculating the estimated bed height is also provided in the figure.

The cold flow model tests of this method showed excellent agreement with actual bed height measurements. The ability to monitor bed height will aid in characterizing UFP performance, providing key indicators of the functionality of the solids transfer between reactors.



Figure 13. Locations of differential pressure measurements for bed height estimation.



Startup Time

An assessment has been made to estimate the time required to bring the reactors up to temperature and pressure. During start-up operation, the reactor beds will initially be fluidized with air until the bed temperature exceeds the steam saturation temperature. The bed will be heated from room temperature to 300°C in approximately 10 minutes. Next, the R1 and R2 beds will be fluidized with superheated steam (R3 will continue with air fluidization).

The energy balance on the solid matrix is the key to estimating the heat-up time. It was assumed that the solid thermal conductivity could be disregarded. This assumption was confirmed by the calculation of the Biot number (Bi), which is the ratio of thermal resistances in the interior of the solid particle over its surface. Since the calculated Bi was less than 1, the dominant resistance lies at the surface, and the bulk thermal conductivity can be neglected as a rough approximation.

A set of equations was programmed in MathCAD software to generate the estimated heating time as a function of the desired temperature approach. This estimation method was

benchmarked against data from a similar system currently in operation at GEGR, and the results showed matching trends, although the UFP calculation underpredicted the heat-up time slightly.

Sampling of Bed Solids

A method was developed to allow sampling of bed solids while the pilotscale system is operating. Removal of solids from a high-temperature, highpressure system is admittedly complex, but it will provide valuable information on the state of the bed materials in each reactor. After detailed analysis, the solids transfer ducts were identified as the location to obtain solids samples. In order to extract a sample, purge sampling lines, maintain system pressure and prevent condensation in the sample, a preliminary valve sequence has been developed, as shown in Figure 14.

Step A, purging the sampling line between valve 1 and the solids transfer duct will ensure that a representative sample is obtained. Purging the sampling system, Step B, ensures that the sample is recovered under an inert atmosphere, preserving the



Step	A	В	С	D	E
alve #	Purge line	Purge line	Collect	Purge line	Isolate
1	0	8	0	Ø	\otimes
2	8	0	0	0	\otimes
3	Ø	0	0	0	\otimes
4	0	0	0	0	\otimes
uration of the step	AFAP	1 min	AFAP	2-3 min	n/a

Valve is open





oxidation state of OTM. The sample is collected during Step C, and the sample is effectively dried in Step D. After the system is isolated (Step E), the sample can be removed and sent for analysis, providing information on the effectiveness of the OTM and CAM bed materials and the extent of their utilization in the process.

Air Pollution Control System

The final construction drawings were completed for the emissions control system. The design of the quench was completed to allow cooling of the product gas from the afterburner before the gas is fed to the scrubber, which is very temperature-sensitive. Figure shows 15 the AutoCAD drawing for the layout of the integrated afterburner, quench and scrubber control system. A dimensioned version of this scaled drawing was used for construction. which was initiated in the eleventh quarter and is now almost complete.

The final length of the afterburner was set to allow a



Figure 15. Air pollution control system: afterburner, quench and scrubber.

residence time of 1 second, in order to ensure complete destruction of CO and any organic hydrocarbons. The scrubber was located after the afterburner to ensure that all sulfur compounds are in an oxidized state, in accordance with the design specifications for the type of scrubber selected.

System Layout

A detailed three-dimensional model of the system has been developed using AutoCAD to aid in system assembly. This model makes use of the actual dimensions of system components, and has been used to assess clearances and accessibility. This information will be of key importance in assembling the pilot plant.

The framework for the reactors and the scaffolding for the system have been designed and manufactured and are awaiting permit approval for assembly. Figure 16 is to-scale drawing showing the layout of the pilot-scale system in relation to the control room and bench-scale system.

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* drawing is to scale

Figure 16. Layout of pilot-scale system.



CONCLUSIONS

Work conducted in the eleventh quarter has focused on accelerating assembly of the pilot plant, with additional experimental analysis being conducted on the lab-scale system.

The lab-scale effort has included TGA experiments to evaluate and quantify the kinetics of OTM reduction. This information will provide key kinetic parameters for integration in process and kinetic modeling of the system. The residence times of solids in the pilot-scale system will be set based on kinetic modeling results to ensure that sufficient reaction has occurred.

The pilot-scale assembly effort has continued, with the manufacturing of the frame and scaffolding, the testing of the slurry pump at pressure, the delivery and initial testing of the second-stage superheaters, and other activities. A detailed safety analysis has been conducted, and is the start of ongoing efforts to ensure that each system component can be operated safely, and the potential hazards have been identified and mitigated where necessary.

Modeling work conducted in the current reporting period has focused on development of an ASPEN process model of the pilot-scale system to assist in determining initial operating conditions for system shakedown.

FUTURE WORK

Additional lab- and bench-scale testing is planned to provide further insight into the rates and mechanisms of char burnout, CO_2 release and OTM reduction processes. Other continuing work on UFP technology development will include the assembly and initial shakedown testing of the pilot-scale system, which will feature three fully integrated circulating, fluidized bed reactors. In addition, progress will be made on modeling tasks in support of pilot-scale system operation. Integral to all these efforts is the continuing analysis of the economics and competitiveness of the UFP technology based on experimental and theoretical findings. These tasks will aid in ensuring that the UFP system will meet the needs of the power generation industry both efficiently and economically.

Task 1 Lab-Scale Experiments – Fundamentals

Task 1 activities will continue to include testing using the lab-scale high-temperature, highpressure reactor and furnace. Kinetic tests involving coal, char, steam, air and combinations of oxygen-transfer material and CO_2 absorber material will be conducted. These experimental efforts will be closely coupled with the ongoing modeling efforts to ensure that the experiments will provide information useful in model validation. In addition, TGA experiments will be conducted to evaluate the kinetics of OTM reduction in the presence of CAM, which is thought to provide a beneficial effect.

Task 2 Bench-Scale Facility – Design/Assembly This task has been completed.



Task 3 Bench-Scale Testing

As needed future testing activities will focus on simulating pilot plant operation with regard to solids sampling and bed heat-up during startup operations. These tests will provide information on the feasibility of selected solids sampling designs. Effective sampling of the solid bed materials during pilot plant operation will provide valuable information on pilot plant performance. Testing the heat-up time of the small bench-scale bed will provide information that will be useful in validating the thermal model of bed heat-up. Additional bench-scale tests will be conducted to identify optimized operating conditions and characterize of bed material performance and ash behavior. Results of these tests will be used along with lab-scale results to modify and validate kinetic and process models, as well as provide inputs for economic evaluation efforts.

Task 4 Engineering and Modeling Studies

Process and kinetic models will be further developed and validated using results from testing activities. These models will also be used to provide information for pilot plant design efforts, such as setting solids recirculation rates. Ongoing economic assessments will continue to gauge the economic feasibility of the process, at different scales and considering competing technologies with additional costs associated with emerging CO_2 regulations.

Task 5 Pilot Plant Design and Engineering

This task has been completed.

Task 6 Pilot Plant Assembly

Assembly of the pilot plant has been delayed due to Air Quality Management District (AQMD) permit delays (despite early submittal of the UFP permit application). The permit to construct is expected soon, and the majority of the system components are already on site and awaiting final assembly. A plan will be developed for conducting shakedown testing of subsystems as they are installed, with special attention devoted to the safety and emergency shutdown systems and their integration with all equipment.

Task 7 Pilot Plant Demonstration

After the pilot plant is assembled, extensive shakedown testing will be conducted, with modifications made as needed. The operational evaluation of the UFP technology will then proceed, followed by performance testing to identify the optimum H_2 yield that can be achieved with thorough analysis of the experimental data. A fuel flexibility study will be conducted to assess the impact of blending biomass fuels with coal.

REFERENCES

Tokuda, M., Yoshikoshi, H., Ohtani, M., Transactions ISII, Vol. 13, 1979, p. 357.



LIST OF ACRONYMS AND ABBREVIATIONS

CAM CO₂ Absorber Material California Energy Commission CEC CTQ Critical to Quality DFSS Design for Six Sigma General Electric Global Research GEGR IGCC Integrated Gasification Combined Cycle NETL National Energy Technology Laboratory New Technology Introduction NTI Oxygen Transfer Material OTM Oxidized OTM OTM-O OTM-R Reduced OTM P&ID Process and Instrumentation Diagram R1 Reactor 1 R2 Reactor 2 R3 Reactor 3 SIU-C Southern Illinois University - Carbondale TGA ThermoGravimetric Analyzer Unmixed Fuel Processor UFP U.S. DOE United States Department of Energy