Underground Coal Gasification: An LLNL Primer

I. Introduction

Underground coal gasification (UCG), wherein coal is converted to gas in-situ, moves the process of coal gasification underground. The produced gas, commonly known as synthesis gas or syngas is created by the same chemical reactions that occur in surface gasifiers. The gas is produced and extracted through wells drilled down into the coal seam, to inject air or oxygen to combust the coal in-situ, and to produce the coal gas to the surface for further processing, transport, or utilization (e.g., Figure 1). The process relies on the natural permeability of the coal seam to transmit gases to and from the combustion zone, or on enhanced permeability created through reversed combustion, an in-seam channel, or hydro-fracturing.

The syngas can be used to create many products (electric power, chemical feedstock, liquid fuels, hydrogen, synthetic gas). Application of UCG can increase the coal resource available for utilization enormously by gasifying otherwise unmineable deep or thin coals under many different geological settings. For example, a 300-400% increase in recoverable coal reserves in the U.S. is possible. For developing countries undergoing rapid economic expansion, including India and China, UCG also may be a particularly compelling technology.

The challenge of managing CO₂ emissions creates a strong drive towards pairing UCG with carbon capture and sequestration (CCS). The composition and outlet pressures of UCG streams at the surface are comparable to those from surface gasifiers; as such, the costs and methodologies for pre-combustion separation are directly comparable (e.g., Selexol at $25/ton CO₂). Conventional post-combustion and oxy-firing options may also be applied to UCG-driven surface applications. In addition, the close spatial coincidence of conventional
geological carbon storage (GCS) options with UCG opportunities suggests that operators could co-locate UCG and GCS projects with a high likelihood of effective CO₂ storage. There is also the possibility of storing some fraction of concentrated CO₂ streams in the subsurface reactor. While this appears to have many attractive features, there remains substantial scientific uncertainty in the environmental risks and fate of CO₂ stored this way.

The economics of UCG appear extremely promising. The capital expenses of UCG plants appear to be substantially less than the equivalent plant fed by surface gasifiers because purchase of a gasifier is not required. Similarly, operating expenses are likely to be much lower because of the lack of coal mining, coal transportation, and significantly reduced ash management facilities. Even for configurations requiring a substantial environmental monitoring program and additional swing facilities, UCG plants retain many economic advantages.

The growing level of worldwide interest in UCG is evidenced by the increasing number of workshops and consortia in recent years. In 2006, there were two workshops (Houston, Texas, U.S. and Kolkata, India); two new consortia, one commercial project set to deliver gas, and resurgence in published documents. Already, for 2007, numerous workshops have taken place or are planned (London, Brazil, Houston, and Canberra) and several commercial projects are ready to deliver gas. Companies in the U.S. (GasTech), India (GAIL), South Africa (Eskom), China (XinAo), Canada (Laurus), and Australia (Linc Energy, Ltd.) have announced projects that include both electric generation and coal-to-liquids. Renewed interest also has been driven by recent successful UCG pilots overseas, such as the Chinchilla operation in Australia, and more widespread knowledge of the 50+ year Uzbekistan UCG commercial operation, although published information about this operation is still limited.

II. UCG at Lawrence Livermore National Laboratory

Lawrence Livermore National Laboratory (LLNL) has played a pivotal role in the development of UCG. It conducted numerous UCG field tests in the 1970s and 1980s, pioneering site evaluation, process modeling and combustion technology. LLNL invented and successfully employed the “Controlled Retraction Injection Point” (CRIP) method, used in some of the Rocky Mountain field tests near Hanna, Wyoming, and in the European trials. This method, shown schematically in Figure 2, uses a horizontally drilled lined injection well where the lining can be penetrated at different locations for injection of an air or O₂/steam mixture. As the injection point is retracted along the well, the cavity in the coal seam gets longer and wider due to reaction of the coal wall with the hot gases.

More recently, LLNL has revisited process modeling and the issues associated with UCG siting, environmental management, and CO₂ management in combination with UCG. The Lab received funding last year from the U.S. Department of Energy to write a best practices report.
Figure 2. Schematic diagram of UCG using the CRIP method, surface processing for H₂ generation and reinjection of CO₂ for sequestration in the coal seam.

A. Computational Fluid Dynamic Models of UCG and Aspen Modeling of Surface Facilities

LLNL has developed a simplified Computational Fluid Dynamic (CFD) model of the UCG process. The model consists of an underground coal seam, approximated as a hollow tube with a given coal thickness. Steam and oxygen are fed through an injection well, and the product syngas is harvested via a production well located at a fixed distance from the injection well. Fluid mechanically, the cavity was treated as a k,ε turbulent zone while the coal was a laminar zone with a solid (carbon) present to account for heat conduction. A mass source of a combination of H₂O(l) and CH₄ in the coal zone simulated the influx of liquid water and the pyrolysis of coal. In fact, the coal zone in the calculation would in reality be a coke zone possessing porosity because of pre-pyrolysis of the coal.

Details for one modeled scenario are as follows. An approximately 1 cm thick thermal wave penetrates the coal ahead of the main combustion/reforming reaction wave, resulting in the pre-pyrolysis. It was assumed that 1 mol CH₄ is formed from 5 moles of coal modeled as CH₀.₈. The CH₄ in the model is transported by convection and diffusion toward the interface and therefore through the combustion/reforming reaction zone. It is important that this CH₄ and the reaction products produce a net mass flux at the interface, hence producing a so-called “blowing” boundary layer. Oxygen must be transported against this convective flux in order to reach the interface. Gaseous water in the case of a zero liquid water influx would also have to overcome this transport barrier to reach the interface. This case included a liquid water influx providing gaseous water from the coal-side to the interface. The importance of the transport of reactants through the blowing boundary layer requires a realistic gas velocity, such as 10 m/s. This in turn meant that the 10 meter long channel modeled was not sufficiently long to deplete the injected O₂. Gasification yields therefore are expressed per mol of consumed O₂ implying that a greater yield would be obtained in a longer channel that depletes the oxygen.

A comparison of gas product composition between model and experiment is shown in Table 1 (water added to make both water contents equal to 33 %). H₂ and CH₄ agree well, but...
compared to the field results for the Rocky Mt. Tests, the field burn showed much more CO$_2$ (and less CO) than the model. The high CO/CO$_2$ ratio in model reflects a high-temperature water-gas equilibrium (approximately 1400K) implying “frozen” kinetics, i.e. the water-gas shift kinetics are too slow at lower temperatures to reduce the ratio down to values closer to the experimental value. In reality with a longer channel and therefore longer gas residence time (residence time is only of the order 1 second here) the CO/CO$_2$ ratio should decrease. However, this is not considered too important because the surface plant will reduce this ratio very effectively in the catalytic membrane reactor.

### Table 1. Typical UCG gas compositions adjusted to 33 mol% water content.

<table>
<thead>
<tr>
<th>Component</th>
<th>UCG Model</th>
<th>Field Measurement*</th>
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<tbody>
<tr>
<td>H$_2$</td>
<td>27.2</td>
<td>27.3</td>
</tr>
<tr>
<td>CO</td>
<td>13.0</td>
<td>6.4</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>19.4</td>
<td>27.2</td>
</tr>
<tr>
<td>CH$_4$</td>
<td>7.4</td>
<td>6.4</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>33.0</td>
<td>33.0</td>
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* Average for day 40-50 from CRIP reactors at RM I3

The Aspen model of the surface facilities simulates gas cleanup, H$_2$ to CO balancing via the water-gas shift reaction, and subsequent utilization of the syngas for the production of power and chemicals, such as ammonia, methanol, or transportation fuels.

**B. Combustion, Air Emissions, and Hydrocarbon Kinetics Modeling**

Over the last 20 years, the LLNL combustion chemistry group has developed detailed chemical kinetic mechanisms for hydrocarbons. These mechanisms are used worldwide by industrial and university researchers for modeling the chemistry of fuels in internal combustion engines and the air emissions of toxic species from refinery burners. The hydrocarbons include alkanes up to iso-octane and n-heptane, aromatics including benzene and toluene, and cyclo-alkanes including methyl-cyclohexane. Additionally, LLNL’s detailed chemical kinetic models can treat the chemistry of oxygenated species such as formaldehyde, other aldehydes, plus selected alcohols, ethers, and methyl esters. The model also includes the chemistry of formation in stack emissions of toxic species such as benzo-a-pyrene, aldehydes, butadiene, and other species on the 1990 U.S. Clean Air Act list, and includes the nitrogen chemistry necessary for the prediction of the emissions of oxides of nitrogen.

The models have sufficient chemistry to represent all the key chemical classes of n-alkanes, iso-alkanes, cyclo-alkanes, alkenes, aromatics, PAHs, and oxygenated hydrocarbons. This capability is being adapted for UCG applications to represent the chemistry of volatile organic hydrocarbons coming off from the in situ burning of underground coal. The chemistry of the evolution of these volatile organic hydrocarbons can be followed as they react with air and steam to form carbon monoxide and hydrogen or possibly react to form undesirable products, such as air toxic species and NOx.
C. Environmental Issues in Underground Coal Gasification

LLNL has the capability to assist UCG operations at both the project planning and site assessment stages and during operations by providing the necessary tools and expertise to understand and minimize the environmental impacts of UCG, which include groundwater contamination and surface subsidence. The environmental threat posed to groundwater resources as a result of underground coal gasification (UCG) is comprised of three principal elements: the generation of contaminants within the burn chamber, enhanced vertical hydraulic conductivity of the rock matrix above the burn chamber as a result of collapse and fracturing, and buoyancy-driven upward flow of groundwater in the vicinity of the burn chamber toward potable water resources at shallower depths.

The complexity of UCG systems requires use of integrated hydrological, geochemical and geomechanical models that presently are unavailable commercially or in most private environmental consulting firms. LLNL has developed these models both through its previous involvement in UCG pilots and through needs to track the fate and transport of a variety of hazardous contaminants under conditions similar to those of UCG operations (Table 2, Figure 3).

Table 2: Coupled Processes Affecting UCG Contaminant Fate & Transport

<table>
<thead>
<tr>
<th>Coupled Processes</th>
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<tbody>
<tr>
<td>Thermally-driven upward flow of groundwater resulting from in situ burning of coal;</td>
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<tr>
<td>Buoyancy effects from fluid density differences reflecting gradients in dissolved</td>
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<tr>
<td>solids content and temperature of groundwater;</td>
</tr>
<tr>
<td>Partitioning of organic compounds and dissolved metals onto mineral surfaces during</td>
</tr>
<tr>
<td>solute transport;</td>
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<tr>
<td>Bioattenuation of organic compounds derived from coals that migrate into potable</td>
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<tr>
<td>water aquifers.</td>
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</table>

From this experience, LLNL has devised a suite of modeling tools that are appropriate to simulating the environmental consequences of UCG operations, including risk-based decision-making (RBDM) during UCG design stages or planning and design of environmental mitigation or remediation strategies for existing operations (Figure 4).
Figure 3. Example of flow and thermo-convection in a vertical cross section over time. The set of 6 figures corresponds to: the mesh, the concentration of a contaminant, the temperature field, the resulting stream function, vorticity, and the flow field (velocity). The bottom of each figure represents the roof of the seam. In the middle of roof, it is assumed that the burning chamber has “collapsed” resulting in a source of heat and contaminants. The figures show the development of convection cells (stream function and velocity) that result in an upward movement of the leaking contaminant (mushroom-like cloud in red in the upper row, second figure).
D. Carbon Management and Underground Coal Gasification

UCG combined with carbon capture and storage (CCS) has distinct advantages. Because UCG syngas reaches the surface at elevated pressure and high temperature, decarbonization may proceed at reduced cost, in some cases (e.g., water-shift separation or partial decarbonization through PSA), at extremely low cost. LLNL is studying both traditional and novel means of UCG decarbonization. UCG also provides opportunities for enhanced storage as well. In a conventional context, CO₂ may be stored in geological targets such as saline formations, depleted oil and gas fields, and unmineable coal seams (typically at a depth >800 m). In addition to conventional storage sites near UCG operations, CO₂ may be stored in the in-situ gasification zone. This has substantial advantages, including the creation of porosity and permeability, local engineering control, use of pre-existing wells, and exploitation of properties of coal that might lead to auto-closure (e.g., swelling and CO₂ sorption).

However, the gasification process also dramatically alters the coal seam and hydrological system in ways that are difficult to estimate or predict. As such, CO₂ injection into the gasification zone may carry substantial risks not thoroughly understood or studied in the field. LLNL is investigating the specific siting and integration aspects of UCG+CCS, with focus on the integrated systems engineering of the UCG process, surface treatment, and continuous injection stream. This work focuses on three components:

- Advanced simulation: Reactive transport simulation provides the main platform for integrating hydrological, geochemical, and geomechanical processes. These models are
underlain by integrated laboratory experiments, including co-contaminant effects (e.g., H$_2$S, SO$_x$, and NO$_x$ injection)

- Integration of monitoring data streams: LLNL places special emphasis in integration and joint inversion of geophysical, geochemical, and operational data streams to provide robust, quantitative information for decision making.
- Quantification of risk: LLNL has developed proprietary tools for carbon management, including source term definition, prediction, and modeling, GIS-based risk screening, and site characterization capabilities.

LLNL has delineated the critical concerns and research needed on the key processes and mechanisms of CO$_2$ storage risk. These include:

- T-P-D constraints for operation: The cavity temperature at a given pressure must be sufficiently low to avoid flashing or boiling of CO$_2$ at injection pressures. Similarly, the injection pressure must be sufficient to remain supercritical and prevent flashing.
- Geomechanical response: The injection pressure must exceed hydrostatic pressures to displace cavity water. This will prompt a number of geomechanical responses, such as fracture dilation, crustal uplift, and potentially inducing fracture.
- Ground-water displacement risk: Cavity injection above hydrostatic pressures will displace UCG zone brines into the coal seam and adjoining formation. This may flush VOCs or metals from the cavity into saline aquifers or coals.
- Geochemical response: CO$_2$ injection will form carbonic acid in the cavity, which may react quickly with the coal, rock, ash, or slag in the cavity. This could leach metals, sulfur, and other harmful species into the UCG zone water, further altering the local chemistry and increasing risk.
- CO$_2$ fate: Free-phase CO$_2$ would remain supercritical and buoyant, applying upward pressure on the cavity. The geomechanical, fault migration, and well-leakage risks may be greater than conventional storage due to the thermal stresses and shocks of heating and quenching.

E. Monitoring and Petrophysics

To date, monitoring of UCG projects and burns has been quite limited. No geophysical surveys have been deployed during UCG projects. Although the Chinchilla, Australia- project extensively sampled groundwater for chemistry and pressure, no other geochemical or operational monitoring occurred. As such, substantial uncertainty exists in subsurface process, engineering, environmental concerns, risk, and even footprint of operation.

LLNL has developed and utilized many geophysical and geochemical monitoring technologies. These include electrical resistance tomography (ERT); electromagnetic induction tomography (EMIT); use of smart casing; passive seismic monitoring (e.g., microseismic); and crustal deformation tools such as tilt-meter, InSAR, and GPS. This work naturally dovetails into the integrated inversion methodologies. Second, we have developed forward geophysical solvers. These may be used to understand and predict the acoustic, thermal, electrical, deformational, and gravitational transients caused by UCG.
LLNL also has laboratory facilities for measurements and experiments in rock physics and geomechanics, including measuring the effects of UCG burn-induced changes in stress fields on rock mechanical and physical properties and performing experiments simulating the effects of UCG on rock-fluid systems.

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