

Research on

IN SITU PROCESSING

T. F. EDGAR
R. S. SCHECHTER
*University of Texas
Austin, Texas 78712*

THE DISPARITY BETWEEN ENERGY supply and demand in the United States has emphasized the need to utilize in situ technology for recovery of fossil fuels and minerals. This technology offers a way to extract reserves which are not technically or economically feasible to recover using conventional mining and may provide a means of significantly increasing domestic fuel and feedstock production in the near future, thus reducing dependence on foreign oil. In situ processing combines extraction and conversion into a single step, and the various in situ processes (oil shale, coal, oil sands, uranium and other minerals) share a number of desirable features:

- Many of the health and safety problems of conventional mining are avoided; underground labor is eliminated altogether.
- Solid waste pollution is avoided since undesirable solid material is left underground.
- The amount of surface equipment for mining and processing is reduced, yielding economic leverage.

These positive features must be weighed against potential problems, such as subsurface pollution of ground water, low recovery efficiencies, and subsidence.

The development of the emerging in situ technologies is interdisciplinary in scope, involving exchange of information among the fields of engineering, earth sciences, chemistry, physics and environmental science. Chemical engineers are playing a leading role because the concepts used to design and operate chemical plants can be fruitfully applied to subsurface processing. In this article we shall indicate the types of research investigations chemical engineering graduate students can carry out, emphasizing two interdisciplinary projects at the University of Texas at Austin. These two projects deal with underground gasification of lignite, directed by T. F. Edgar, and in situ leaching of uranium, directed by R. S. Schechter.

The ultimate goal of the research is to develop a set of laboratory and computer-based tools which would allow site evaluation based on field and laboratory measurements of the mineral and associated overburdens.

APPLICABLE RESOURCES AND COMMERCIAL EXPLOITATION

THE DOMESTIC COAL SUPPLY is, of course, extremely large and will be the major resource for intermediate term energy production. In situ recovery of coal will increase the recoverable coal supplies to about 40% of the national resource estimate (10% is recoverable using conventional mining). Western coal can be successfully gasified in situ, but Eastern coals cannot be recovered easily because of their swelling characteristics. Texas lignite is the resource studied in the UT-Austin research; it is estimated that nearly 20 billion tons of lignite lie at surface mining depths (down to 90 meters), but approximately 35 billion tons lie between depths of 90 and 600 meters in seam thicknesses between 1.5 m and 4.5 m, which are technically feasible ranges for underground coal gasification (UCG) [A1]. There have been ten private sector and government-sponsored field tests, two of which are being performed in Texas. Demonstration-scale (e.g., 25 MW equivalent of electricity or greater) tests have been performed in the USSR with air injection, and several U.S. tests of this magnitude are planned for the early 1980's, using steam/oxygen injection to produce a medium Btu gas.

At the present time, in situ uranium leaching provides only a small fraction of domestic uranium requirements (less than 5%) but because of activity particularly in Texas and Wyoming the proportion can be expected to increase significantly in the future. A number of companies are now mining on a commercial scale in Texas. In 1979 there were 16 permits issued to five companies for approximately 1500 production acres. Seven addi-

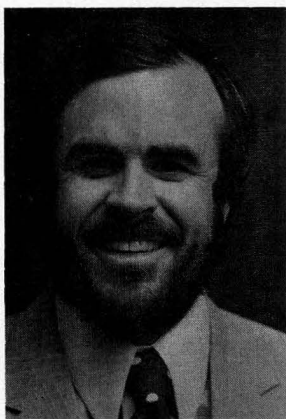
© Copyright ChE Division, ASEE, 1980

tional permits were being processed for an additional 2000 acres of production [A2].

Despite this high level of industrial interest in both in situ gasification and uranium leaching, significant problems, especially environmental, still remain to be resolved. Failure to resolve them adequately will result in a severe curtailment of these rapidly emerging technologies.

RESEARCH APPROACH AND SCOPE

THE PURPOSE OF THE UT-AUSTIN research on in situ processing is to develop the necessary theory and understanding of the underground process so that commercialization can be facilitated for a wide range of mineral deposits. The ultimate goal of the research is to develop a set of laboratory and computer-based tools which would allow site evaluation based on field and laboratory measurements of the mineral and associated overburdens. Such an evaluation must of necessity be semi-quantitative since there are never enough data available about the subsurface to predict accurately all physicochemical aspects of the recovery process. Laboratory screening is preferred because field testing is uneconomic for every potential site; an engineering and geological design



Thomas F. Edgar is Associate Professor of Chemical Engineering at The University of Texas at Austin, where he has been a faculty member since 1971. His research interests include chemical process control and coal combustion and gasification. He earned his B.S. Ch.E. from the University of Kansas and Ph.D. in chemical engineering from Princeton University. He is editor of the technical journal *In Situ*. (L)



Robert S. Schechter is the E. J. Cockrell, Jr. Professor of Chemical and Petroleum Engineering at The University of Texas at Austin. His research interests are mainly concerned with surface and interfacial phenomena with particular emphasis on tertiary oil recovery and in situ uranium leaching. He is a graduate of Texas A&M University (BS) and the University of Minnesota (Ph.D.). Professor Schechter was elected to the National Academy of Engineering in 1976. (R)

The research is strongly oriented toward developing mathematical models suitable for design and scale-up and for optimization of operation.

basis, which presently is incomplete, must be developed before economic application of the technology can be realized.

The research is strongly oriented toward developing mathematical models suitable for design scale-up and for optimization of operation. This entails development of a fundamental understanding of the dominant physical and chemical processes occurring in the subsurface. However, the research has pointed to the need for certain process improvements, and efforts are now being made to effect these improvements.

In order to develop successful techniques for scale-up from the laboratory to the field, both the coal and uranium projects have maintained close liaison with field personnel and other researchers in the field through biannual research review meetings. Each project has a review board made up of professionals from industry and government laboratories who critically evaluate the research results presented at these meetings.

At the University of Texas we are fortunate to have strong departments of chemical engineering, petroleum engineering, geology, and environmental engineering in order to pursue this research; there have been as many as ten staff and twenty students from the above departments involved in the coal and uranium projects. Annual support for the projects combined has been more than \$400,000/year; funding agencies have included the Department of Energy, Environmental Protection Agency, Department of HEW, National Science Foundation, Office of Surface Mining, Department of the Interior, Texas Petroleum Research Committee, and private companies.

UNDERGROUND COAL GASIFICATION RESEARCH

IN UCG, DRILLED HOLES ARE used to access the coal seam. After the coal seam is ignited, the permeability of the seam must be enhanced before actual gas production. This can be done by using a technique called counter-current or reverse combustion, which pyrolyzes a narrow channel of high permeability between two boreholes, thus allowing directional control over gas flow. After this "link" between boreholes is achieved, co-current combustion can be employed for gas production [A3].

Underground coal gasification systems represent a departure from conventional chemical reactors in that the internal geometry varies over time, and their boundaries are not fixed as in surface equipment. A number of specific models are needed to describe quantitatively the behavior of an underground coal gasification process. Such models can be subsequently used for process design and control. A model is needed to predict the chemical composition of the product gas as a function of the injected gas composition and rate as well as how product gas composition might vary with coal type, seam thickness, and water intrusion rate. There is a need to be able to predict the fraction of coal that will be recovered with a particular well pattern and to quantify the linking pattern. Also important are predictions of water influx and the potential of roof collapse and their effects on gas composition and sweep efficiency. Other predictive models can be used to quantify the environmental impact, such as subsidence and aquifer contamination. These physical and chemical models can be coupled with process economics models so that system design and operation can be optimized. The major emphasis of the chemical engineering research has been on predicting

Underground coal gasification systems represent a departure from conventional chemical reactors in that the internal geometry varies over time, and their boundaries are not fixed as in surface equipment.

product gas composition [A4] and sweep efficiency [B1], [B2] for a range of coals and geological conditions.

There has been some success in using mathematical models for interpreting field and laboratory data for in situ gasification. However, these models have employed of necessity tremendous simplifications in order to realize meaningful results for such a complex system [A5]. The in situ gasifier contains such components as a fixed bed, a channel, and a porous medium [B3]. The geometry of the gasifier may be irregular due to burning of the coal and resulting roof collapse. Complex heat transfer, mass transfer, and momentum transfer phenomena occur simultaneously in different parts of the system. It is clear that "global" mathematical models are not practical for the purpose of simulation; the challenge is to determine which model elements are crucial for

matching the process performance. Even simplified models are often characterized by moving boundaries, split boundary conditions, and stiff differential equations.

A useful one-dimensional conceptual model of UCG during co-current or forward combustion involves the existence of several distinct reaction zones, namely oxidation, reduction and pyrolysis/drying [A3], [A4]. These are the same zones that occur in moving-bed surface gasifiers. One way to simulate UCG in the laboratory is to operate a combustion tube charged with pulverized coal similar to that used in petroleum recovery studies [B4], [B5], [B6]. Field gasification temperatures can be in excess of 1300°C, which creates extreme experimental difficulty in tube design; typical oil combustion tubes are operated at less than 800°C, well within the limits of steel alloy materials. There is some disagreement about the ultimate usefulness of such apparatus for simulating the three-dimensional UCG process, but a combustion tube does provide the opportunity to study one level of scale-up. Modeling of such a system also implies reaction kinetics sub-models for oxidation [B7], [B8], gasification [B9], and pyrolysis [B10], [B11], [B12], which can be developed for a given coal using a variety of microreactors.

It can be argued that the basic reaction unit in UCG is a coal block rather than a coal particle, which would emphasize intra-particle heat and mass transfer effects [B13]. Self-gasification of the coal block by moisture transpiration and cracking and gasification of pyrolysis products [B14], which must diffuse through the block, also occur. In a three-dimensional interpretation of UCG, sweep efficiency and burning rates may be controlled by consolidated block transport phenomena [B15]. Coal block experiments can illuminate the effects of gas hydrodynamics, coal properties, and block transport phenomena on channel growth [B16], [B17]. Gas composition probably is only secondarily controlled by block gasification, although more field and laboratory experiments and mathematical modeling for a variety of sites are necessary before definitive conclusions can be reached.

Modeling of environmental effects, such as subsidence and subsurface water pollution, is relatively immature in development. Their predictability is problematic because of the lack of information on geohydrology and rock behavior as well as chemical, physical, and biological processes which occur in the subsurface. Natural

renovation of polluted groundwater has been reported in several U. S. field tests.

IN SITU URANIUM LEACHING RESEARCH

THE OBJECTIVES OF IN SITU leaching of uranium are to dissolve all the uranium-bearing minerals in place and to transport the dissolved uranium back to the surface in the fluid injected into the ore-bearing formation. In its simplest form, the chemical process consists of: (1) oxidizing the uranium-bearing species and (2) putting the uranium in solution in a chemical form which can be recovered at the surface. Both of these steps are to be accomplished while minimizing the effect of those reactions between the leach solution and the ore-bearing formation which are detrimental to the extraction process and minimizing the damage to the subsurface environment. The final stage of the mining process is to remove those pollutants introduced into the formation during the extraction stage. This is crucial since the zones which can be mined are in aquifers.

The modeling of the production stage requires that the rates of heterogeneous reactions of oxidant with uranium bearing minerals and with all the other minerals which compete for oxidant be established. This is a somewhat challenging step since there are often four or more minerals, each competing for oxidant. Furthermore, the rates cannot be studied independently since only studies on carefully preserved field samples are meaningful.

In a study of acid leaching kinetics, Tatom found [C1] that many of the heterogeneous reactions are initially mass transfer controlled so that during the early stages of leaching, the overall reaction depends on the local fluid velocity. As the quantity of unoxidized mineral decreases, there exists a point, different for each mineral, at which the intrinsic surface kinetics control the rate of reaction. Thus the reaction rate models tend to be more complex than that proposed by Galichon [C2], [A6] and represent an interesting interplay between mass transfer and surface kinetics.

Models for the chemical reactions which take place during carbonate leaching are now being developed [C3]. This research has the additional goal of comparing the reaction rates in ammonium carbonate solutions with those in potassium carbonate solutions. Ammonium carbonate has been the main ingredient in the leach solution used by most of the companies operating in Texas. However, many companies are now reluctant to use

this compound because of the severe requirements for NH_4^+ removal after mining which have been imposed by state and national regulatory agencies [A2].

Sodium carbonate is sometimes used but in many cases will damage the formation permeability so that fluid injection becomes virtually impossible. Potassium carbonate, which does not damage the formation permeability, has not been used because of its cost. However, the University of Texas research team has developed a procedure which utilizes a preflush that will reduce the cost of subsequently leaching with potassium carbonate by a factor of about four. This process [A7], for which patents are pending (assigned to the Bureau of Mines), is now being readied for field testing.

A second aspect of the research has been the development of restoration procedures. The initial work focused on the recovering NH_4^+ cations from the formation at the completion of the production stage. This is essentially an ion exchange process [A8], [A9], complicated by the existence of multiple substrates having different free energies of exchange, interlayer mixing, axial dispersion, etc., [C4]. Process modeling indicated the desirability of utilizing special fluids of high ionic strength to flush ammonium; however, in some cases the ammonium cation was strongly adsorbed and difficult to remove.

An interesting example of the type of problem which has been considered is the one-well test to evaluate the restoration parameters. This test involves the injection of relatively small volumes of leach solution and the subsequent production of a larger volume of fluid. Two important parameters—the cation exchange capacity and an average free energy—are to be determined by history matching the effluent composition. A study of this procedure revealed that this goal cannot be feasibly accomplished, and alternate techniques were proposed [A10], [C5]. Before this study, such tests were routinely performed, and in many cases, erroneous values of the parameters were adopted.

Finally, all the detailed models for production and restoration must be imbedded into a simulator that can accommodate an arbitrary well pattern, formation heterogeneity and anisotropy, variable injection and production volumetric rates and transient solution and solid phase compositions. All these requirements have not yet been satisfied, but Bommer [A11], [C6] has reported a relatively comprehensive two-dimensional simulator which

is now being used by several operators to design optimum systems.

In conclusion it should again be stressed that the most difficult problems are environmental in nature. There are many rules and regulations at the national, state and local levels which must be satisfied. Some of these rules are in conflict with others [C8]. Studies such as those now being conducted by Chen [C7] will help regulatory agencies make rational decisions with regard to those standards which should be strictly enforced and those which can be relaxed to some extent without altering the quality of the water. □

A. LITERATURE CITED

- A1. Edgar, T. F., Kaiser, W. R. Humenick, M. J. and Charbeneau, R. J., "Environmental Effects of In Situ Gasification of Texas Lignite," Report to Environmental Protection Agency, August, 1980.
- A2. Whittington, D. and Taylor, W. R., "Regulations and Restoration of In Situ Uranium Mining in Texas," Proceedings of the South Texas Uranium Seminar, Society of Mining Engineers of AIME (1979).
- A3. Gregg, D. W. and Edgar, T. F., "In Situ Coal Gasification," *AICHE J.*, 24, 754 (1978).
- A4. Natarajan, R., Edgar, T. F. and Savins, J. G., "Prediction of Product Gas Composition for UCG," Proc. Sixth Underground Coal Conversion Conference, Afton, Oklahoma, July, 1980.
- A5. Edgar, T. F., "Analysis and Modeling of Underground Coal Gasification Systems," FOCAPD Engineering Foundation Conference, Henniker, N. H., July, 1980.
- A6. Galichon, P., Breland, W. M., Cowley, A. H. and Schechter, R. S., "Chemical Factors in In Situ Uranium Leach Mining," *In Situ*, 1 (2), 125 (1977).
- A7. Tweeton, D., Guilinger, T. R., Breland, W. M. and Schechter, R. S., "The Advantages of Conditioning on Orebody with a Chloride Solution Before In Situ Uranium Leaching with a Carbonate Solution," SPE Preprint No. 9490, 55th Annual Technical Meeting, Dallas, Texas, Sept., 1980.
- A8. Hill, A. D., Walsh, M. P., Breland, W. M., Humenick, M. J., Silberberg, I. H. and Schechter, R. S., "Restoration of Uranium In Situ Leaching Sites," *Soc. of Petroleum Engr. J.*, 20, (1980).
- A9. Walsh, M. P., Humenick, M. J. and Schechter, R. S., "The Displacement and Migration of Ammonium Ions from Uranium In Situ Leaching Sites," in *In Situ Uranium Mining and Groundwater Restoration: W. J. Schlitt and D. A. Shock eds., Proc. of the New Orleans Symp., Soc. of Mining Engr. of AIME (1978).*
- A10. Kabir, M. I., Lake, L. W. and Schechter, R. S., "Evaluation of the One Well Uranium Leaching Test: Restoration," SPE Preprint 9486, 55th Annual Technical Conference, Society of Petroleum Engineers of the AIME, Dallas, Texas, Sept. (1980).
- A11. Bommer, P. M., Schechter, R. S., "Mathematical Modeling of In Situ Uranium Leaching," *Society of Petroleum Engineering Journal*, 19, p 393 (1979).

B. THESES—UNDERGROUND COAL GASIFICATION

- B1. Dinsmoor, B. (1979), "The Modeling of Channel Systems in Underground Coal Gasification."
- B2. Johnson, C. (1980), "A Three Dimensional Numerical Simulation for Underground Coal Gasification."
- B3. Galland, J. (1974), "Analysis and Modeling of Underground Coal Gasification Systems."
- B4. Westbrook, D. (1976), "Design of a Combustion Tube for Experimentation on In Situ Gasification of Texas Lignite."
- B5. Cook, R. (1980), "Combustion Tube Studies of Texas Lignite Using Steam-Oxygen Injection."
- B6. Ponnampuruma, J. (1980), "Combustion Tube Studies of Air Gasification of Texas Lignite."
- B7. Tseng, H. (1981), "Pore Size Effects on Oxidation Kinetics of Texas Lignite Char."
- B8. Hsia, S. (1977), "Oxidation Kinetics of Texas Lignite."
- B9. Bass, E. (1980), "Gasification Kinetics of Texas Lignite Char and Steam."
- B10. Cadwell, J. (1978), "Pyrolysis Properties of Texas Lignite Under Conditions of In Situ Gasification."
- B11. Matteson, M. (1979), "Pyrolysis Kinetics of Texas Lignite."
- B12. Athans, M. (1980), "Sulfur Compounds Produced During Pyrolysis of Texas Lignite."
- B13. Tsang, T. (1980), "Modeling of Heat and Mass Transfer During Coal Block Gasification."
- B14. Hunt, C. (1979), "Cracking of Low Heating Rate Pyrolysis Products Obtained from Texas Lignite."
- B15. Wellborn, T. (1980), "Measurement of Linear Burning Rates for Consolidated Lignite Cores."
- B16. Cornwell, J. (1982), "Channel Gasification of Lignite Blocks."
- B17. Triplett, K. (1980), "Analysis of Flow Patterns in Underground Coal Gasification Using Tracers."

C. THESES—URANIUM LEACHING

- C1. Tatom, A. (1980), "An Investigation of the Factors Determining the Rate of Uranium Leaching in Acid Solutions."
- C2. Galichon, P. (1976), "In Situ Leaching of Uranium Ore."
- C3. Guilinger, T. (1981), "Optimal Formulation of Carbonate Lixiviants for Uranium Solution Mining."
- C4. Walsh, M. (1979), "Investigation of the Fate of Ammonia from In Situ Uranium Solution Mining."
- C5. Kabir, M. (1981), "In Situ Uranium Mining Reservoir Engineering and Aspects of Leaching and Restoration."
- C6. Bommer, P. (1979), "A Streamline-Concentration Balance Model for In Situ Uranium Leaching and Site Restoration."
- C7. Chen, J. (1980), "The Migration of Low pH Solutions in Groundwater."
- C8. Shiao, S. (1980), "Environmental Aspects of Uranium In Situ Leaching."