A Course on

UNDERGROUND PROCESSING

CLARENCE A. MILLER* Carnegie-Mellon University Pittsburgh, PA 15213

THE EVENTS OF RECENT YEARS have brought increased attention to processes for recovering fossil fuels and minerals from underground formations. Higher prices for petroleum have caused the industry to give serious attention to more sophisticated recovery processes involving not just flow, as in conventional processes, but also heat and mass transport, various phase changes, chemical reactions, and interfacial phenomena. Underground coal gasification seems promising for the future. Research has increased on in situ processing to recover oil from oil shale and tar sands. Solution mining of uranium has begun in some locations.

The number of engineers working on such processes has increased rapidly in recent years, especially in research. As more extensive field applications develop, additional engineers will be required to design processes applicable to specific locations and to supervise production operations. Because flow, transport, and chemical reaction are involved in most of the processes, chemical engineers are well suited for this work and should be much in demand.

To acquaint chemical engineering students with this rapidly growing field and to provide them with pertinent fundamental information not ordinarily covered in a chemical engineering curriculum, I have developed a one-semester course in "Underground Processing." Although basically a graduate course, it is open to interested undergraduates who have had courses in fluid mechanics, transport phenomena, and thermodynamics.

GEOLOGICAL BACKGROUND

TABLE 1 IS AN OUTLINE of the course. The first section deals with geological background material. In contrast to the usual situation in a chemical plant, the "reactor" for an underground process is not built to the designer's specification



Clarence A. Miller received his B.A. and B.S. Degrees in chemical engineering from Rice University in 1961. After spending four years as an engineer with the Navy's nuclear power program in Washington, D.C., he undertook graduate studies at the University of Minnesota, receiving his Ph.D. Degree in 1969. He spent twelve years on the chemical engineering faculty at Carnegie-Mellon University and joined Rice University in September, 1981, as a Professor of Chemical Engineering. For the last several years his major research interest has been investigation of interfacial phenomena in enhanced oil recovery processes.

but is provided by nature. It is usually the result of geological processes which have occurred over periods of tens to hundreds of millions of years. As it is accessible only through a few widely spaced wells, details of how its physical structure and chemical composition vary with position are not known. Some understanding of its geological origin is useful in determining how effective various processes might be.

The difference in pore structure between sandstones and limestones, the two most common reservoir rocks for petroleum, provides an example of the importance of geology to an engineer. The pore space in a sandstone is basically that originally present between the individual sand grains just after deposition, although some decrease in pore size occurs over time due to compaction as the deposit is buried and due to precipitation of silica, calcium carbonate, or other substances on the surfaces of the grains and at their junctions. Also called cementation, the precipitation at junctions serves to bind the individual grains to-

^{*}Present address: Rice University, Houston, TX 77001.

[©] Copyright ChE Division, ASEE, 1981

gether to form a rock.

The situation is quite different for limestone rocks which, in the first place, are often formed by deposition of rather irregularly shaped particles consisting of shells or skeletal fragments of various marine creatures. Then too, some recrystallation after deposition is common in carbonate rocks. Since a density change is involved, porosity and pore structure are affected. Pore structure changes are also caused by cementation, which can be extensive, by dissolution of material in water flowing through the rock, and by fracturing, which occurs more easily than for sandstones. The overall result is a pore space much less regular than in a sandstone. Differences in pore size

TABLE 1

Course Outline for Underground Processing

- A. GEOLOGICAL BACKGROUND
 - 1. General geology
 - a. Plate tectonics theory
 - b. Formation and characteristics of sedimentary rocks
 - c. Age of rocks and the geological time scale
 - 2. Formation of fossil fuel and mineral deposits a. Origin of hydrocarbons in shale deposits
 - b. Relation to formation of other fossil fuels
 - c. Migration of petroleum from source rocks to traps
 - d. Formation of petroleum traps—sedimentary basins
 - e. Formation of mineral deposits by hydrothermal processes
 - f. Relation between plate tectonics and sites of fossil fuel and mineral deposits
- B. FLOW, TRANSPORT, AND INTERFACIAL
- PHENOMENA IN POROUS MEDIA
 - 1. Basic interfacial phenomena—interfacial tension, contact angles
 - 2. Interfacial phenomena in porous media—capillary pressure
 - 3. Single-phase flow in porous media-Darcy's Law
 - 4. Relative permeabilities and two-phase flow
 - 5. Conditions for trapping or mobilizing a residual phase
 - 6. Heat transport in porous media
 - 7. Mass transport, hydrodynamic dispersion
 - 8. Chromatographic transport
 - 9. Stability of displacement fronts in porous media
- C. DESCRIPTION OF UNDERGROUND PROCESSES
- 1. Petroleum recovery
 - a. Immiscible displacement, waterflooding
 - b. Polymer and surfactant flooding
 - c. Miscible displacement, carbon dioxide injection d. Thermal recovery processes
 - 2. Underground coal gasification
 - 3. In situ processes for oil shale and tar sands
 - 4. Solution mining of uranium

Because flow, transport, and chemical reaction are involved in most of the processes, chemical engineers are well suited for this work and should be much in demand.

and shape between rocks have a significant effect on displacement of one fluid by another, e.g., of oil by water, and are thus of great importance to the engineer.

Even when consideration is restricted to sandstone rocks, relatively minor differences in composition can be important for performance of certain processes. For instance, most sandstones contain some clay minerals although their primary component is silica. Clays can adsorb surfactant molecules and they can serve as sites of cation exchange between liquids in the pore space and the rock surface. Both these properties have a significant influence on enhanced oil recovery processes which employ surfactants. Indeed, failure to properly account for ion-exchange effects is believed to be the main reason for poor performance of at least one field test of the surfactant process. Thus, the amount of clay originally deposited with the sand is significant.

Finally, variation in depositional conditions with position and time can cause significant permeability variations within a petroleumcontaining rock. Injected fluids prefer to flow through high-permeability regions, largely bypassing regions of low permeability. In an extreme case, permeability barriers may exist between nearby wells in a formation, so that flow between the wells is minimal. Such a situation was found in a recent field test of an enhanced oil recovery process. Fortunately, it was discovered during preparations for the test, and process adjustments were made before the test was begun.

A brief overview of plate tectonics theory begins the course. Only some fifteen years old in its modern form, this theory has been the most exciting development in geology in decades because it has provided a unifying framework relating diverse results from many fields of geology. Then a rather extensive discussion of rock formation is given with emphasis on sedimentary rocks where oil, oil shale, and tar sands were formed and where they are found.

The next major topic is formation of fossil fuel and mineral deposits. As the result of extensive work by petroleum geologists and geochemists during the past thirty years, much has been learned about the origin of petroleum. Shale In contrast to the usual situation in a chemical plant, the "reactor" for an underground process is not built to the designer's specification but is provided by nature. It is usually the result of geological processes which have occurred over periods of tens to hundreds of millions of years.

is a sedimentary rock consisting mainly of small particles of clay minerals and other inorganic materials but also containing a few percent of organic material. If the original deposit forms under anaerobic conditions, the organic material is preserved and, on burial, undergoes chemical reaction to form a complex polymeric material known as kerogen. As burial depth increases, the temperature rises until, at some point, further reactions take place in which the kerogen releases hydrocarbon molecules in order to form a more compact structure consisting largely of multiple aromatic rings. Hydrocarbons so produced are the constitutents of petroleum.

With modern analytical techniques such as gas chromatography, the composition of organic material in shale has been measured as a function of depth in several locations. Such work has allowed the course of the reactions which generate hydrocarbons in shale to be followed. It has also shown that the same basic chemical process is responsible for formation of petroleum, coal, and oil shale. Differences in these materials are the result of differences in composition of the initial deposits. Oil shale is richer in organic material than most petroleum source rocks while coal forms from deposits which are primarily organic with only a few percent of inorganic material, just the opposite of shales. The differences in composition between the terrestrial organic material which forms coal and the marine organic material which is the source of most oil also lead to major differences in the distribution of reaction products, e.g., to generation of more methane and fewer longer-chain hydrocarbons in coal. Oil shales have never been subjected to temperatures high enough to cause appreciable hydrocarbon release. Effecting such release is the chief objective of oil shale processing. Two excellent summaries of current knowledge of fossil fuel formation are the recent books by Tissot and Welte [1] and Hunt [2].

Also covered in the course are "primary" migration of hydrocarbons from the shales where they form to nearby sandstones or limestones, a process which remains poorly understood, and "secondary" migration of oil within the reservoir rocks. Generally speaking, oil travels upward owing to gravitational effects until it reaches a "trap" where a low-permeability shale or some other permeability barrier precludes further upward movement. Several geological structures which can cause trapping are considered. So are salt dome formation and other geological conditions which can cause these structures to form. Some comments are made on the emerging picture of the connection between plate tectonics and oil formation.

Tar sands are oils which have been degraded after trapping by exposure to ground waters containing bacteria. The bacteria preferentially consume short-chain and paraffinic compounds. Depending on the amount of degradation, the remaining oil may be only slightly more viscous than the original oil, or it may be a "tar" with a viscosity of tens of thousands of centipoise or more.

Student assignments for this part of the course consist of: 1) a set of simple problems which provide a feeling for the magnitude of such quantities as the rate of plate motion over the earth's surface, the heat flux from the earth's surface, and the amount of water needed to increase the porosity of a limestone rock by dissolution, and 2) a short paper on some aspect of the geological part of the course. Topics selected by the students have ranged from discussion of certain geophysical and geochemical methods for locating oil and mineral deposits to a summary of the arguments given by the few geologists who have yet to accept plate tectonics theory. Most of the papers, however, have dealt with some aspect of the formation of fossil fuels in more detail than the class notes and lectures.

In summary, some knowledge of geology is essential to those working in underground processing. Experience has shown that the more one knows about formation properties, the better the chances of process success. Although engineers naturally interact with geologists, who have a detailed understanding of depositional conditions, in developing formation descriptions, the interaction is more productive if the engineer has some background in geology.

INTERFACIAL PHENOMENA, FLOW, AND TRANSPORT IN POROUS MEDIA

A LTHOUGH THE FORMATIONS which serve as sites for underground processes vary widely in structure and composition, they may all be considered porous media. Since interfacial phenomena control the distribution of immiscible fluids such as oil and water within a porous medium at the low flow rates common in oil recovery processes, the first step is a thorough discussion of interfacial tension and contact angles. A brief account of surfactants and their properties is included as well to provide a background for later consideration of surfactant processes for enhanced oil recovery.

In porous media interfacial phenomena are responsible for the pressure difference or "capillary" pressure between immiscible fluids. Variation of capillary pressure during slow displacement of one fluid by another is described. Emphasis is given to interfacial instabilities which lead to "Haines jumps," the rapid and irreversible final stage of displacement occurring in individual pores even when the overall rate of displacement is slow. As a result of these instabilities, capillary pressure behavior exhibits hysteresis, i.e., capillary pressure variation when water displaces oil is not simply the reverse of that when oil displaces water.

Next, single-phase and two-phase flow in porous media are discussed. Consideration is restricted to low flow rates where Darcy's Law applies, the usual situation in underground processing. An important topic is the mechanism of trapping of a residual phase when one fluid displaces another. Because of such trapping, water is usually able to displace only about half the oil originally present in a reservoir. Obviously, the conditions required to prevent trapping are of great interest. These amount to a sufficiently large ratio of viscous to interfacial forces, i.e., a sufficiently large value of the dimensionless capillary number $(\mu v / \gamma \phi)$, where μ and v are continuous phase viscosity and superficial velocity, γ is the interfacial tension between fluid phases, and ϕ is porosity.

After some coverage of heat and mass transport in porous media and hydrodynamic dispersion, chromatographic transport in porous media is considered. Introduction of the methods of chromatographic analysis is a key part of the course since they are used later in the analysis of oil recovery processes. The presentation consists

FALL 1981

of a sequence of examples of ever increasing difficulty, ranging from simple adsorption of a solute or its partitioning into a trapped fluid phase to immiscible displacement of one fluid by another (Buckley-Leverett analysis) to ion exchange phenomena to two-phase displacement processes with partitioning of various components between phases. The method of characteristics is used to solve the simpler examples and to illustrate how traveling concentration waves develop. Then the more complicated examples are treated by Helfferich's general scheme [3], which begins with the assumption that concentration waves occur.

Winding up this portion of the course is a discussion of the stability of displacement fronts in porous media. No matter how well a fluid can displace another from an individual pore, its effectiveness in a large-scale process is limited if the macro-

... the study of flow, transport, reaction, and interfacial phenomena in porous media is an excellent application of basic chemical engineering principles and one that has utility far beyond underground processing.

scopic front between displacing and displaced fluids is unstable. For in this case the injected fluid travels through the reservoir in channels, completely bypassing many pores containing the oil or other fluid originally present.

The lectures here deal first with instability in the form of viscous fingering which occurs, for example, during waterflooding of high viscosity oils. Then transport effects are discussed with stress given to their importance in thermal processes for oil recovery and in underground coal gasification.

Homework problems are assigned frequently throughout this part of the course as the basic material is by nature more quantitative than in the geological background section.

PROCESSES FOR FOSSIL FUEL RECOVERY

IN THE LAST PART OF THE course the major underground processes in use or being developed are described. More attention is given to petroleum recovery than to other processes, primarily because more is known about it. Waterflooding is considered first. Then polymer flooding, surfactant flooding, and miscible displacement, e.g., with high pressure carbon dioxide, are discussed. Basic physical mechanisms are stressed in lieu of details of processes performance. Simplified analyses using chromatographic transport methods are used to illustrate the main features of each process.

Because the chromatographic analyses employed assume that phase equilibrium and chemical reaction equilibrium are reached instantaneously, other methods are used for analysis of thermal oil recovery processes such as steam drive and underground combustion. In these processes the rate of heat transport from the reservoir to the surrounding formations is of great importance, and heat conduction terms must be included in the analysis.

Finite rates of chemical reaction are important in other types of processes. Examples are the use of acids to dissolve some of the rock near a well, thereby increasing permeability, and reverse combustion processes which are used in the initial stages of underground coal gasification and which are potentially of use in in-situ tar sand recovery.

Some aspects of a process are more important in underground than in ordinary processing. Clearly one highly desirable feature of an underground process is relative insensitivity to variations in formation properties since, as indicated above, detailed knowledge of such properties at all points in a formation cannot be obtained.

The linked-vertical-well method of underground coal gasification is used as an example to illustrate this point. Reverse combustion is used to "link" injection and production wells, i.e., to provide a high-permeability path between them. Once the link is complete air or oxygen can be injected at relatively low pressure with a high degree of assurance that, whatever the flow properties of the original coal, most of the injected gas will travel along the link where resistance to flow is low. This behavior has the highly desirable results that most injected gas participates in the main gasification reaction and that only a small amount leaks away to surrounding areas where its presence could be undesirable from an environmental point of view.

Student assignments here consist of some homework problems on waterflooding and surfactant flooding and a project involving a short paper on some feature of a particular underground process of interest to the student. Some of these papers have been basically literature surveys, while others have been analyses of certain processes using chromatographic transport methods.

CONCLUDING REMARKS

NO EXISTING TEXTBOOK IS suitable for the entire course. As a result, I have prepared notes for most parts. Some books and articles which have proved useful in this task and which are sources of further information for students are listed below [1-12]. The last part of the course on the processes themselves is, except for the discussion of waterflooding, based largely on journal articles which have appeared during the past few years.

In summary, the course provides an introduction to underground processing to acquaint students with opportunities in this area and with pertinent fundamental knowledge. The geological background material has been emphasized to a greater extent in this article than in the course itself because of its novelty and because the author believes that interaction between chemical engineering and geology may be fruitful in generating research ideas beyond the present topic. From a more traditional chemical engineering view, however, the study of flow, transport, reaction, and interfacial phenomena in porous media is an excellent application of basic chemical engineering principles and one that has utility far beyond underground processing.

REFERENCES

- 1. Tissot, B. P. and D. H. Welte, *Petroleum Formation* and Occurrence, Berlin, Springer Verlag, 1978.
- 2. Hunt, J. M., Petroleum Geochemistry and Geology, San Francisco, W. H. Freeman, 1979.
- 3. Helfferich, F., Soc. Petrol. Eng. J., 21, 51-62 (1981). "Theory of multicomponent, multiphase displacement in porous media."
- 4. Barnes, H. L. (ed.), Geochemistry of Hydrothermal Ore Deposits, 2nd ed., New York, Wiley, 1979.
- 5. Selley, R. C., An Introduction to Sedimentology, New York, Academic Press, 1976.
- Press, F. and R. Siever, *Earth*, San Francisco, W. H. Freeman, 1974.
- 7. Scheidegger, A. E., *The Physics of Flow Through Porous Media*, 3rd ed., University of Toronto Press, 1974.
- 8. Dullien, F. A. L., Porous Media—Fluid Transport and Pore Structure, New York, Academic Press, 1979.
- 9. Muskat, M., *Physical Principles of Oil Production*, New York, McGraw-Hill, 1949.
- 10. Craig, F. F., Jr., *The Reservoir Engineering Aspects* of Waterflooding, Dallas, Society of Petroleum Engineers of AIME, 1971.
- 11. Craft, B. C. and M. F. Hawkins, Applied Petroleum Reservoir Engineering, Englewood Cliffs, N.J., Prentice-Hall, 1959.
- Aris, R. and N. R. Amundson, Mathematical Methods in Chemical Engineering, Vol. 2, Englewood Cliffs, N. J., Prentice-Hall, 1973.

CHEMICAL ENGINEERING EDUCATION