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AN INVESTIGATION OF THE REGIONAL CHARACTERISTICS OF THE DEVONIAN SHALES FOR THE STORAGE/DISPOSAL OF RADIOACTIVE WASTES*

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ABSTRACT

Devonian-age shales underlie a large portion of the northeastern quadrant of the country and are abundantly thick and relatively near to the land surface within the Appalachian, Illinois and Michigan basins of that area. Although these thick accumulations of argillaceous sediments are best-known for their natural gas production and potential, they would also appear to be good candidate rocks for radioactive waste repositories as the shale is generally impermeable, and therefore, relatively free of circulating ground waters to disperse any waste emplacements.

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DISCLAIMER

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Introduction

Although the research, development and demonstration (RD&D) program in the United States for the permanent storage of high-level radioactive wastes has focused its efforts on bedded-salt formations, the potential suitability of other subsurface rock types, such as granite, basalt and argillaceous sediment, has long been recognized. With detailed investigations, these alternatives may, in fact, be shown to offer comparable physical, chemical and geological properties.

Under the Office of Nuclear Waste Isolation (ONWI), which is being administered by the Battelle Columbus Laboratory for the U.S. Department of Energy (DOE), work is being directed toward efforts to more fully investigate some of the most promising alternatives to rock salt. One of these alternatives, which is the subject of this paper, is being examined by the Oak Ridge National Laboratory as an integral feature of ONWI's Terminal Waste Storage Program. Although argillaceous rocks are widespread throughout the continental U.S., the Devonian shales that underlie a large portion of the northeast are particularly attractive for study.

Firstly, because the Devonian black shales are petroliferous, they are presently being extensively investigated for their natural gas production and kerogen conversion potential.[1] Since the early 1800's, high quality gas has been produced in modest amounts for local domestic and small commercial markets; oil production in these same general areas, however, has been primarily realized from siltstones and sandstones that overlay or feather into the black shales. The determination of the Devonian shale hydrocarbon potential is presently being carried out through DOE's Eastern Gas Shales Project, which is being administered by the Morgantown Energy Technology Center, Morgantown, West Virginia.[1] This work is providing extensive characterization data of these Devonian shales, which include statigraphy, mineralogy, porosity, permeability, structure, etc. Even though some shale characteristics that would be favorable for gas production might not be acceptable features for a radioactive waste repository, e.g.,

a high degree of fracturing, it is clear that most of these data would be directly applicable to the assessment of both potential uses. Consequently, near-term research results can be achieved for argillaceous rocks for the Terminal Waste Storage Program by using this available data base and technology that are being collected and developed specifically for these Devonian shales.

Secondly, the Devonian shales were selected for investigation by OWNI when earlier scoping analyses pointed to favorable conditions for more detailed study. This summarized the geologic and hydrologic characteristics of selected salt and impermeable sedimentary rock deposits, including argillaceous formations, for the purpose of identifying potentially suitable repositories for deep geologic disposal.[2] Many shale deposits in the U.S. could not meet the selection criteria established for the study. The primary reasons for this tentative rejection of shale formations included high seismic risk; relatively high permeability; inadequate thickness or excessive depth; interbedding with permeable strata of siltstones, sandstones and limestones; and extensive fracture and deformation due to regional tectonism. However, many shale deposits in the U.S. appeared to have characteristics that made them worthy of more detailed investigation. Three of these shale formations are located in the northeastern U.S. and include (a) the Devonian-Mississippian Ellsworth Shale in Michigan; (b) the Mississippian Coldwater Shale in Michigan; and (c) the Devonian Ohio Shale in central and northern Ohio. These shales lie in the basins being examined by the Eastern Gas Shales Project.

Characterization of Devonian Black Shales

Those argillaceous sediments of Upper Devonian and Mississippian age that are commonly referred to as black shales are found in twenty-six states in the U.S. and in extensive portions of Canada.

The terminology "Devonian black shale" is generally used in references to a sequence of contiguous fissile shales, mudstones and claystones, including any interbedded foreign strata such as siltstones and sandstones. Consequently, as would be expected, black shale formations can exhibit more or less a single color, or can show marked variations in coloration, and in either case, dark or black beds may or may not be present. The colorations found may vary from light shades of grays, greens and browns to dark and blackish values. In addition to similar age and clay mineral composition, an important feature common to all of the so-called black shales is their high organic content. While variations in coloration can be partially attributable to differences in the sources of the clastics and their nonclay constitutents, the colors shown by these argillaceous sediments are likely to also reflect the thermal maturity of the carbonaceous content. The black shales under investigation by the Eastern Gas Shales Project are located in the Appalachian, Illinois and Michigan basins. The areal extent of these three basins, and their corresponding massive argillaceous sediments, is a significant portion of the northeast quadrant of the U.S.

In the Appalachian Basin, which reaches from southwestern New York to northern Alabama, the Ohio and Chattanooga shales are the two best known shale formations. The Ohio Shale, which is often referred to as the Ohio Brown Shale, has been the dominant source of natural gas production in these three basins since the early 1800's, and consequently, the Ohio Shale and northern portions

of the Chattanooga Shale are the eastern shales of most interest for stimulated natural gas production. The western boundary of the Appalachian Basin corresponds to the western outcrop of the Ohio Shale. This north-south outcrop extends for the full length of the state and into Kentucky. There are three ascending Devonian shale members in the Ohio Shale sequence, which are known as the Huron, Chagrin and Cleveland Shales. The Devonian-Mississippian Bedford Shale and Lower Mississippian Berea Sandstone formations rest on the Ohio Shale. The Ohio Shale sequence is brownish to grayish-black in appearance and varies in thickness from about 400 ft in central Ohio to more than 4000 ft in the southeastern part of the state.

The Devonian black shales in the Illinois Basin which extends through major portions of Illinois, Indiana, Kentucky and Tennessee, are represented by the New Albany Shale sequence. In ascending order, the New Albany Shale consists of the Blocher, Sweetland Creek, Grassy Creek and Hannibal Shale members. The coloration in the sequence varies from greenish-gray to very dark browns and black. The brownish-black carbonaceous shale constitutes more than 80% of the New Albany in the southwestern part of the basin, but elsewhere its abundance declines to 20 to 40 percent. The thickness of the New Albany Shale sequence varies from about 100 ft near its outcrop at the Cincinnati Arch to more than 300 ft in extreme southwestern Indiana. The New Albany Shale both thickens and dips into the Illinois Basin. Compared to the Appalachian Basin, natural gas production from Indiana's New Albany Shale has been modest. Although gas has been produced from about ten small fields in the past, all but one have been abandoned. From these New Albany gas fields, little correlation has been observed between thickness of the black shale and the occurrence of gas, even though these are organically rich shales.

The Devonian black shales in the Michigan Basin consist of a three member shale sequence which is commonly referred to as the Antrim Shale. Although the Michigan Basin is almost entirely limited in areal extent to the southern peninsula of Michigan State, the basin and Antrim Shale extend a short distance into northern Indiana and the northwestern extreme of Ohio. In ascending order, the three members of the Antrim Shale are the Antrim, Ellsworth and Sunbury Shales. Antrim shale exhibits beds of both black organic rich deposits and greenish-gray calcareous shale and the Ellsworth consists of interbedded greenish-gray nonorganic shale and black carbonaceous shale. The Sunbury is a black organic shale that shows a high gamma trace, but this stratum is quite thin. The Antrim Shale sequence lies beneath the Mississippian Coldwater Shale formation, and in places where the thin black Sunbury Shale is absent, the Ellsworth cannot be readily distinguished from the overlying Coldwater. Although there have been reports of gas showings in the Antrim Shale sequence, there are no commercial wells in the Michigan Basin.

Terminal Radioactive Waste Storage Program

With the above introductory information and brief characterization of the Devonian black shales as background, the following is a summary statement of OWNI's ongoing and planned research program for the deep geologic disposal of radioactive wastes in these rocks. This research will focus on the argillaceous rock formations in the Appalachian, Illinois and Michigan basins, which are also considered to be promising for their hydrocarbon potential. Because of the

natural gas stimulation and kerogen conversion potential of the shales in these basins, the DOE and the oil and gas industry have been carrying out an active RD&D program to better understand the nature of the argillaceous deposits. While it is clear that some of the preferred rock characteristics for gas and in situ shale oil production, e.g., high permeability and organic content, are contrary to those rock qualities sought for a radioactive waste repository, it is recognized that a cooperative research effort between these two DOE programs can have a mutually beneficial and synergistic effect.

The ONWI-ORNL RD&D program pertaining to argillaceous sediments is in its initial research stage. The thrust of this research activity is to determine the suitability of using massive fissile shale, mudstone and claystone formations, which can be anticipated to show variable interbedding with siltstone, sandstone, limestone and dolomite, as a geologic setting for a terminal radioactive waste repository. At first, the results of this work will include lithological, mineralogical, geochemical, geophysical, hydrological and engineering investigations based on the published literature, the open files of the Federal and state Geological Surveys and the data base and technological advancements provided by the Eastern Gas Shales Project. During this first phase effort, no field studies or tests on the black shales are expected to be performed by ONWI-ORNL. This is because there already exists a large foundation of data and technology that is readily available from extensive field measurements made by others. For example, during the October 1978 Second Eastern Gas Shales Symposium, the number of core locations reported for the Eastern Gas Shales Project alone were 19 wells cored, 13 planned, and 29 proposed.[1]

Hence, this initial shale study will involve the analysis and interpretation of all the relevant data and information that can be assembled. These data have resulted from shale characterization work, which includes lithological, chemical, physical and mineralogical parameters, and well logging, core analyses and fracture and seismic determinations based on various geophysical and remote sensing techniques. It is intended that the results of this first phase investigation by ONWI-ORNL on shales will considerably advance our understanding of the Upper Devonian-Mississippian black shale sequences in the Appalachian, Illinois and Michigan basins in the following areas:

a. the statigraphy of individual strata and their interbasin correlations, including imperfections in the massive argillaceous sediments due to interbedded permeable sandstones, limestones, etc.

b. structure,

c. mineralogy, including the relationships between color, gamma logs, concentrations of uranium, thorium and other metals such as nickel and vanadium, and organic carbon content,

d. resource potential, with emphasis on locations exhibiting a low potential for natural gas stimulation, kerogen conversion and coal production,

e. the porosity, degree of impermeability and natural fracturing and hydraulic fracturing characteristics,

f. seismic risk and other tectonic considerations, e.g., folding, deformation, and

g. groundwater hydrology and the accounting of all boreholes.

It is anticipated that the results of this first phase effort will provide the means to develop criteria to be used in subsequent and more detailed investigations. Such criteria would be necessary for the identification of specific formations and geographical areas that merit further evaluation, which might involve well coring and logging, core analyses and other field determinations.

References

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