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CHATTANOOGA SHALE: URANIUM RECOVERY BY *IN SITU* PROCESSING

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CHATTANOOGA SHALE: URANIUM RECOVERY BY *IN SITU* PROCESSING

ABSTRACT

The increasing demand for uranium as reactor fuel requires the addition of sizable new domestic reserves. One of the largest potential sources of low-grade uranium ore is the Chattanooga shale -- a formation in Tennessee and neighboring states that has not been mined conventionally because it is expensive and environmentally disadvantageous to do so. An *in situ* process, on the other hand, might be used to extract uranium from this formation without the attendant problems of conventional mining. We have suggested developing such a process, in which fracturing, retorting, and pressure leaching might be used to extract the uranium. The potential advantages of such a process are that capital investment would be reduced, handling and disposing of the ore would be avoided, and leaching reagents would be self-generated from air and water. If successful, the cost reductions from these factors could make the uranium produced competitive with that from other sources, and substantially increase domestic reserves.

A technical program to evaluate the processing problems has been outlined and a conceptual model of the extraction process has been developed. Preliminary cost estimates have been made, although it is recognized that their validity depends on how successfully the various processing steps are carried out. In view of the preliminary nature of this survey (and our growing need for uranium), we have urged a more detailed study on the feasibility of *in situ* methods for extracting uranium from the Chattanooga shale.

INTRODUCTION

The increase in future energy requirements in the United States, coupled with the possibility that a larger fraction of that energy must be produced by nuclear power plants, has led to uncertainties concerning the long-term adequacy of domestic uranium reserves. Continuing ERDA projections have shown sharp increases in the demand for uranium during the next 25 years.¹ Such estimates are based on specific but reasonable sets of assumption concerning such factors as population growth, per capita power consumption, types of reactors to be constructed, etc.

Undoubtedly, enough uranium can be found to meet this demand, since it is so widely distributed -- although at low concentrations -- in such materials as shales, granites, and seawater.² The problem is, rather, in producing it at a reasonable cost as our richer reserves become depleted. If we compare the projected requirements with current estimates of low-cost reserves³ and potential resources⁴, some future problems become obvious.

The uranium industry considers it necessary (because of lead time in exploration, construction of new facilities, and sales contracting) to maintain an 8-year forward reserve of available ore. This is reflected in the minimum production base curve of Fig. 1. Based on this projection, an 8-year reserve could not be maintained for more than another 10 to 15 years. To maintain such a reserve it would be necessary to either expand low-cost reserves through continued successful exploration, to use more costly resources, or to increase imports.

Most of the low-cost uranium now being mined in this country is from sandstones in New Mexico, Utah, Colorado, and Wyoming. Some is being recovered from the leaching of copper ores, and the rest as a by-product of manufacturing wet-process fertilizer from phosphate deposits. As these sources are exhausted, leaner deposits from the same places will be mined at increasingly higher costs. Mine and gaseous diffusion plant tailings will also be reworked. These conventional resources are shown in Fig. 2.⁵ The large mass of uranium in the Chattanooga shale is not a low-cost resource^{*} if mined with conventional mining methods. The shale only contains 60 ppm uranium, about 5% of the concentration in ore presently being processed. The few published cost estimates (Appendix B) have given the general impression that the black shales are a "last resort" resource of uranium. However, if a low-cost extraction method can be developed, the shales could be upgraded to ores, which would substantially augment domestic uranium reserves.

The Chattanooga and related Devonian shales cover extensive areas in the eastern and central United States. Their geology, mineralogy, and chemistry are discussed in Appendix A. In addition to uranium, they have a relatively high content of organic material, and have in the past been considered as potential oil shales. Such factors as oil yield on pyrolysis, oil quality, and

* It is considered an "indicated submarginal resource"; i.e., a known deposit not now minable economically.

Fig. 1. Projected requirements for nuclear power.¹

Fig. 2. Available domestic uranium at conventional mining costs.⁵

formation geometry do not make this an attractive possibility, although the organic content has important implications for uranium recovery. The recoverable oil content of the shale averages $39 \text{ m}^3/\text{Gg}$ (8 gal/ton).

The shales vary widely in kerogen and uranium content, depth, and thickness, and are not well explored in all areas. Various estimates have been made of the amounts of potentially recoverable fuels, but the resource is generally agreed to be very large. Duncan and Swanson⁶ estimate that the shales contain $5 \times 10^{11} \text{ m}^3$ (3×10^{12} bbl) of oil. Swanson⁷ and Hickman and Lynch⁸ estimate that the central part of the Chattanooga formation contains 4.7 Tg (5×10^6 tons) of uranium.

The central (and best-explored) area of the shale covers parts of Tennessee, Kentucky, and Alabama (Fig. 3). It is a shallow formation, typically about 46 m (150 ft) deep, and the richest part, with which we will be concerned, is a bed 4.6 m (15 ft) thick.

SHALE PROCESSING

From existing cost estimates (discussed in Appendix B), it appears that uranium cannot be recovered from the Chattanooga shale by conventional mining at present or extrapolated future market prices. However, we believe that an *in situ* technique, to be discussed in the following sections, offers potential economic (and environmental) advantages.

In Situ Techniques

The recovery of uranium by *in situ* leaching of ores has been studied for about 20 years⁹ and is currently being used for some low-grade deposits. The process involves extracting oxidized uranium from a relatively permeable host rock. The problem in applying such methods to the Chattanooga shale is that it is an impermeable, reducing formation, which means that additional processing would be required to provide the necessary permeability and oxidation state. Such conditions might be produced by a three-stage sequential recovery process:

- Fracturing
- Retorting
- Pressure leaching

Fig. 3. Central portion of the Chattanooga shale formation.

In the first stage of the process, permeability would be developed by explosive fracturing, in which one of three types of explosive charges (point, line, or sheet) would be set in the shale bed. An array of point charges could be emplaced by directional drilling; line charges by horizontal drilling; sheet charges by hydraulic fracturing and propping (Figs. 4 and 5).

The second stage involves igniting and retorting* the shale to remove as much of the liquid and gaseous hydrocarbons as possible (Fig. 6).

Stage three is begun by flooding the formation with water (Fig. 7). Excess heat from retorting is extracted from the rock, reducing its temperature to the optimum for leaching. At this point, air or oxygen is added to the fluid circulated through the shale ash (Fig. 8). The remaining pyrite is oxidized, producing sulfuric acid. Uranium is dissolved in hot, oxidizing acid solutions. The leach liquor is pumped to a surface recovery plant, and its uranium separated by solvent extraction or ion exchange.

Technical Program For Process Development

From the preceding scenario, it is evident that to successfully extract uranium from the Chattanooga shale, we will have to further develop *in situ* techniques for the solution mining of low-permeability formations. A complex process will be required, for which existing technology is not adequate. The Lawrence Livermore Laboratory is in a favorable position to develop such a process since it is already studying coal gasification and shale oil extraction. The principal problem areas in process development are:

- Rock mechanics and fracturing
- Retorting
- Ash properties
- Leaching
- Fluid control
- Environmental impact

Each of the above areas will be discussed in more detail in subsequent sections.

* The primary purpose of retorting is not to recover shale oil. Rather, it is to:

- Increase permeability
- Heat the formation for leaching
- Remove organic matter to minimize reagent consumption

Fig. 4. Hydraulic fracturing and propping along bedding planes.

Fig. 5. Explosive fracturing.

Fig. 6. Igniting and retorting.

Fig. 7. Water flooding with steam production.

Fig. 8. Pressure leaching step for uranium dissolution.

ROCK MECHANICS AND FRACTURING

To remove uranium from shale by solution mining, we must first develop an inexpensive, efficient fracturing technique to increase permeability. Presumably, this would involve drilling a pattern of vertical holes to the Gassaway member (the richest portion of the shale), since lateral access is very limited.

Field experience in fracturing this type of formation is scanty, although a variety of hydraulic and explosive fracturing techniques are available. A hydraulic fracturing process for shale, in which the void volume is created by fracturing the shale from cased drill holes perforated with horizontal slots, has been developed by the Oak Ridge National Laboratory.¹⁰ The Pumpkin Valley member of the Conasauga shale was fractured at depths of 200 to 300 m (700 to 1000 ft). The results showed that horizontal or conformable sheet fractures with diameters up to about 120 m (400 ft) could be produced. The process was used on an experimental basis to dispose of intermediate-level radioactive wastes.¹¹

Some attempts have also been made to increase the permeabilities of western oil shales. A series of tests was carried out in the Green River formation using a variety of fracturing methods, including hydraulic and explosive fracturing.¹²⁻¹⁴ It was reported that ignition, horizontal retorting, and some oil recovery were possible.

The composition of the Chattanooga shale differs from that of the shales mentioned above, so existing data on the effectiveness of various techniques are of only limited value. Blair¹⁵ has measured a few of the physical properties of the shale which relate to fracturing behavior (Table 1), although the shale member sampled was not specified. Properties are likely to vary considerably with position in the formation, and many additional measurements will be necessary.

Table 1. Physical properties of Chattanooga shale.

Apparent specific gravity, g/ml	2.53
Compressive strength, GPa	0.834-0.848
Impact toughness, m/m ²	1.4 -4.2
Scleroscope hardness, scleroscope units	42-44
Young's modulus, GPa	11.2-17.3
Modulus of rigidity, GPa	5.7
Poisson's ratio	0.13-0.19
Specific damping capacity	1200
Longitudinal bar velocity, m/s	2591

Determination of the directional fracturing properties is particularly important because the Gassaway member is relatively thin. It is desirable that most breakage should be in horizontal planes. Vertical fractures down through the Dowelltown member and into the underlying limestone could cause some loss of fluids in subsequent processing steps.

Only a few qualitative observations on fracturing behavior are available at present. Breakage is generally conchoidal, except along joints and bedding planes.¹⁶ It has been observed that freshly mined shale from the Gassaway member fractures readily along the laminae.¹⁷ However, the 3-m-thick bed of claystone in the Dowelltown member immediately below the Gassaway is poorly laminated and nonfissile.¹⁸ Weathered material from outcrops is quite fissile, presumably because of rapid, preferential oxidation of the organic-rich black laminae.

It is possible that some additional void space must be provided before the permeability of the shale can be sufficiently increased by fracturing. Drilling, reaming, and mining techniques for accomplishing this should be considered.

RETORTING

Burn Front Control

After fracturing, the shale is retorted by igniting the rubble and injecting air or oxygen to support combustion and force the burn front evenly forward. The direction and advance of such a burn front will require careful

control. Bypassing unretorted shale must be minimized, both horizontally and vertically. Some process variables that can be used for such control are the injection well and fracture patterns, the ignition timing and the input rates, and composition and distribution of the injected gas.

Oil Recovery

As the burn front advances, the organic material in the shale, kerogen, decomposes to yield shale oil, gas, and carbonaceous residue (char). The oil produced by the retorting process flows into collector wells at the bottom of the rubble zone and is pumped to the surface.

Chattanooga shale has a higher carbon/hydrogen ratio than western oil shales (Appendix A), which results in a lower oil yield on retorting in relation to total organic content. However, the substantial amount of residual char is not without value, in that it can be burned to supply retorting heat. This implies that a greater fraction of the kerogen will be converted to retort gas, relative to the western shales. It is necessary, then, to consider the possibilities for using this fuel product.

Gasification

The more highly aromatic nature of the Chattanooga shale kerogen suggests that gasification should be explored as an alternative means of removing hydrocarbon values. As in the gasification of coal, the oxidation of the high carbon content of the shale

can provide the energy for the steam-carbon reaction

Such reactions can be carried out *in situ*, but it is likely that an oxygen plant would be required, and that additional surface processing facilities would be required for the production of methane for pipeline gas by shift conversion

and methanation

An extensive technical and economic analysis will obviously be required to decide whether such a process can be applied economically to Chattanooga shale. For the present discussion, we will simply assume that organic material will be recovered as oil without special treatment.

Hydrogasification

Hydrogasification experiments by Schultz¹⁹ showed that gas produced from Chattanooga shale had a higher energy content than that from Green River oil shales, and that the yield of standard gas produced per volume of oil assayed was about twice as high. There are not enough data to properly extend Shultz's batch experimental conditions [977 K (704°C) and 14.8 to 20.7 MPa (2000 to 3000 psi)] to an *in situ* operation with different pressures, temperatures, residence times, and particle sizes.* However, based on Shultz's calculations and the current price of hydrogen, we have roughly estimated that it would cost \$40/mg (\$36/ton) for hydrogen to process Chattanooga shale using his method. Since this does not appear close to being an economical process, it will not be considered further.

Permeability

Laboratory pyrolysis experiments on unconfined Chattanooga shale have shown that internal pressures developed by volatile hydrocarbons cause the shale to split along bedding planes (Fig. 9). If this also occurs *in situ*, it will increase the permeability of the rubble, exposing a greater surface area to leaching solutions. However, it is also possible that the retorting of confined shale may cause swelling, which could reduce permeability. Thus, while the removal of a substantial amount of kerogen should lead to an increase in porosity, it is not clear that a corresponding increase in permeability will result.

* The available evidence is that, for *in situ* operations, short residence times and low temperatures favor higher yields of high molecular weight products. It was also observed that the effects of particle size were negligible between 0.044 and 4 mm (5 to 325 mesh).²⁰

Fig. 9. Chattanooga shale samples before retorting (left figure) and after retorting (right figure).

Steam Production

After retorting, the formation will be at an estimated temperature of 836 K (538°C). Water will then be injected to reduce the temperature to about 448 K (175°C) (experimental leaching has been done at that temperature).²¹ If possible, we will use the energy transferred from the shale to the steam in on-site operations. After condensation, it will be reinjected to conserve water. It might also be possible — by injecting air with the water — to oxidize the char and recover the resultant heat of combustion in the steam.

Future Work

We suggest retorting shale in the laboratory to determine oil, gas, and spent shale properties as a function of temperature, burn rate, input gas composition, and rubble size. Oil properties that should be investigated include yield, viscosity, fractionation products, and sulfur content. Gas properties include chemical composition and energy content. Spent shale properties include remnant char and pyrite contents, ash strength, permeability, and stability during flooding.

LEACHING PROCESSES AND URANIUM EXTRACTION

Acid Pressure Leaching

Acid pressure leaching refers to a process in which water and oxygen or air are circulated through a sulfide ore formation. The injected fluid reacts with the sulfides to produce a hot, oxidizing sulfuric acid solution, which then leaches the desired mineral from the rock. Several investigators have considered using pressure leaching to extract uranium from sulfidic ores.²¹⁻²⁷ Pollara et al.²¹ for example, have developed a leaching method whose specific purpose is to extract uranium from Chattanooga shale. Their study was designed for an above-ground process and covered the following conditions:

- Particle size — 0.04 to 4 mm
- Pressure — 0.17 to 1.0 MPa of oxygen
- Temperature — 273 to 473 K
- Time — 5 to 200 min

The process made use of the 11% pyrite content of the shale to generate sulfuric acid. The acid dissolved the uranium which was later recovered from solution by solvent extraction.

One of the problems with the process is that retorting the shale to remove oil also has the effect of reducing the amount of pyrite. For example, in an early experiment on Chattanooga shale, it was found that retorting in the temperature range 844 to 916 K removed almost all the oil but also reduced the sulfur content from 8.6 to 5.6-6%.¹⁷ This reduction in pyrite decreases the amount of sulfuric acid that can be produced, which, in turn, may decrease the amount of uranium recovered in the leaching process.

Although roasting the shale can adversely affect uranium recovery,³⁰ under some conditions controlled retorting does not necessarily decrease the solubility of the uranium in the leaching phase. Studies on the recovery of uranium from coal ash indicate that reactions in which insoluble uranium compounds are formed do not proceed rapidly below 1000 K.³¹ This important question evidently requires further experimental study. It is also necessary to consider the extent to which silicates might be converted to soluble metal oxides at high temperatures. (Such conversions would result in more alkaline leaching solutions.)

Another aspect of the pressure leaching process that has not been sufficiently examined is the leaching of coarser material for longer times. It is known that the mineral grains of the shale are cemented together by organic material.^{28,29} As this material is dissolved by oxidizing acidic solutions, the rock should progressively decompose, allowing access to uranium remaining in the interiors of large ore fragments. Considerable work remains to be done, however, in determining uranium extraction rates as a function of particle size, time, solution temperature, acid strength, oxygen partial pressure, and retorting conditions. The possibility of precipitate formation and clogging of fissures should also be examined.

Carbonate Leaching

Pressure leaching processes with carbonate solutions have been used in some low-grade uranium-bearing formations that have enough natural permeability.³²⁻³⁶ The solution compositions vary, but are usually made alkaline with ammonia and contain an oxidizing agent. Uranium is converted to a soluble carbonate complex by:

The uranium is later recovered either by precipitation as insoluble uranates

or by hydrogen reduction to insoluble UO_2

A carbonate process should be considered as an alternative to acid pressure leaching. It has the advantage that such solutions would be compatible with the underlying limestone. However, it has the disadvantages that reagents would have to be added at increased cost, and that they would be consumed by reactions with residual pyrite and organic material after retorting.

Uranium Extraction

Solvent extraction appears to be the most satisfactory means of removing uranium from shale leach liquors,^{21,26,37} although recovery by ion exchange should also be considered. These processes will need to be reviewed and revised, since it is likely that solutions produced *in situ* will differ in composition from those previously studied.

FLUID FLOW AND RECOVERY

The successful recovery of uranium from a shale mine depends, in part, on whether we can design an efficient flow system for the fluids, with minimal leakage losses. Such a system must take into account the varied effects of drilling, fracturing, retorting, and leaching on permeability. One way to control the fluid might be to direct flow from the outside of a mine field in toward a central production hole. Figure 10 shows a conceptual schematic of a small central portion of such a field, in which injection and injection are done from holes on the perimeter, and unfractured rock beyond provides the necessary container.

Fig. 10. Central portion of drill pattern and mine field.

ENVIRONMENTAL IMPACT

To determine what effect an extraction process will have on the environment, we will first have to define the location, geology, and hydrology of a site. But wherever the site, and whatever its physical details, our major concern will be to prevent the leach liquors used in the process from contaminating ground waters. The flow of leach liquors outside the immediate fracture zone is likely to be very slow, but the proximity of drainage features such as aquifers, faults, and outcrops should be considered. The rate of neutralization of leach solution leaking through cold, raw shale should also be determined. At the end of the processing operation, the spent liquor could be disposed of safely and economically by reinjecting it into the shale. In the event of downward leakage, the acidic liquor would be neutralized by the thick underlying limestone, a process that is also known to occur naturally.³⁸

Considerable demands will be made on local water supplies, although the anticipated disposal rate is a "worst case" estimate, and is likely to be reduced during process development.

Surface effects will be limited to a slight warming trend and subsidence, neither of which are expected to be significant. Burwell¹² observed that localized ground surface temperatures rose as much as 10 K, 15 months after termination of a burn at a similar depth. In the present project, however, it is anticipated that the formation will be cooled considerably after retorting, so surface effects will be small. Subsidence should be minimal, since only a fraction of the thin shale member is to be removed.

Finally, the effect that the process might have on the quality of air and the safety of wildlife must be considered. Sulfur compounds will be produced in retort gas and steam, but will be removed prior to venting and flaring. Cooling ponds will be fenced and covered to protect wildlife. Some population growth will result from the project, to an extent which has not yet been evaluated.

APPENDIX A

SHALE PROPERTIES

Geology

The Chattanooga shale is a late Devonian marine sediment underlying several southeastern and midwestern states.^{7,8,39} Because of its obvious potential as a large source of uranium, extensive investigations were carried out on the shale between 1944 and 1955. The information from these studies is summarized and discussed by Conant and Swanson¹⁸ and Brown.⁴⁰

Extensive exploratory drilling was done in the formation in 1953.⁸ Drill core samples were taken, and an adit was driven into an outcrop at Sligo Bridge near Smithville, DeKalb County, Tennessee. Tonnage lots of the Gassaway member were taken for retorting and leaching experiments.

The structure of the formation is shown in Fig. A1, as illustrated by Kehn⁴¹ and Hickman and Lynch.⁸ It is seen to be relatively shallow in this central portion, typically covered by about 46 m (150 ft) of overburden, mostly cherty limestone. The lower part of this "chert," where unweathered, is very hard, and diamond drilling is required. The roof is generally good, and conventional mining of the shale would be feasible if it were economical. The upper, or Gassaway, member of the formation is richest in uranium and is about 4.6 m (15 ft) thick. The lower, or Dowelltown, member is composed of an upper claystone unit and a lower unit of relatively thin, lean, black shale. Near the top of the Dowelltown member is another persistent feature, a 30-mm bed of bentonite.⁴² The underburden is about 60 m (200 ft) of Leipers limestone, of Ordovician age.⁴³ This is a material composed of about 75% carbonates and 25% clay.³⁸

The shale is generally a uniform, well characterized formation, and the Youngs Bend area in DeKalb County, Tennessee has been suggested as the most favorable site for a conventional mine.^{5,8} However, additional geological reconnaissance is required in the new context of an *in situ* process. Site selection will also require information on existing permeability, joint structure, and hydrology. The proximity of the formation to water tables, aquifers, wells, and outcrops must be considered since the earth is to be used as a container for pyrolytic and chemical reactions.

Fig. A1. Stratigraphy of the Chattanooga shale formation in the Young Bend area, Tennessee.^{8,41}

A possible location for an experimental mine is the Flynn Creek crypto-explosive feature,¹⁸ an area of about 8 km² (3 mi²) where the shale has a thickness of up to 60 m (200 ft). This local thickening is, however, entirely in the leaner Dowelltown member.

Mineralogy and Chemistry

The shale is a very fine-grained rock formed by slow sedimentation in a quiet, shallow inland sea.^{16,44,45} The bottom muds and water were at most times reducing, a favorable environment for the precipitation of uranium from solution.⁴⁶⁻⁴⁸ The uranium remains associated with the clays and organic materials upon which it was originally sorbed. No specific uranium mineralization is present, confirming the lack of redistribution.

The mineralogical composition of the shale, according to Bates and Strahl,¹⁶ is given in Table A1. This is representative of samples of the Gassaway member taken from the Sligo adit. The chemical composition of this same material, as determined by Wilson et al.,¹⁷ is given in Table A2.

Table A1. Mineralogical composition of the Gassaway member of the Chattanooga shale (Bates and Strahl).¹⁶

<u>Mineral</u>	<u>Composition</u> <u>%</u>
Quartz	22
Feldspar	9
Illite + minor kaolinite and muscovite	31
Carbon	13.6
Total organic matter	16-22
Pyrite and marcasite	11
Chlorite	2
Iron oxides	2
Tourmaline, zircon, apatite	1

Table A2. Chemical analysis of the Gassaway member of the Chattanooga shale (Wilson, et al.).¹⁷

Component	Amount, %	Component	Amount, %
Uranium	0.0089	P ₂ O ₅	1.1
Hydrogen	1.6	K ₂ O	2.2
Sulfur	8.63	Na ₂ O	0.39
Carbon	13.6	CaO	1.69
SiO ₂	45.7	MgO	1.17
Fe ₂ O ₃	10.4	Mo	0.027
Al ₂ O ₃	11.1	Ge	None
L.O.I.	23.3 ^a		

^a Loss on ignition determined at 1223 K (950°C), 1 hr.

The kerogen of the Chattanooga shale differs from that in western oil shales in that it has a higher C/H ratio, a consequence of the substantial contribution of humic terrestrial organic materials to its formation.⁴⁵ It would be more properly referred to as a carbonaceous or coaly shale than as an oil shale. In fact, thin lenses of coal are not uncommon in the shale. Table A3 illustrates the approximate relationship between the compositions of coal and some shale kerogens.^{28,49,50}

Table A3. Carbon-hydrogen-oxygen atomic ratios in coal and shales.

<u>Formation</u>	<u>Composition</u>	<u>Reference</u>
Green River shale	CH _{1.53} ^O .056	49
Chattanooga shale	CH _{1.0} ^O .273	28
Wyoming coal	CH _{.858} ^O .208	50

APPENDIX B

COSTS

Introduction

An economic evaluation of the *in situ* multicomponent extraction process that has been outlined is important in that its implementation depends not only on the resolution of technical problems but on achieving substantial cost advantages over other sources of uranium, including conventional mining of the shale.

The available cost estimates for conventional mining have been updated for purposes of comparison. Then a modeling and cost estimation exercise for *in situ* processing was undertaken. Its purpose was to investigate whether uranium might be produced at a competitive price, with reasonable ranges of processing parameters.

Conventional Mining Costs

The formation outcrops extensively around the Nashville basin, and it would, therefore, be possible to gain access to the shale without drilling, although this would greatly limit the amount of available material.

Costs for an experimental mine in the Chattanooga shale were estimated by Hickman and Lynch⁸ (Table B1). The range of costs arises from the choices of beds to be mined; the costs do not include subsequent processing and uranium recovery. Inflation to 1975 costs was estimated with the Nelson cost indexes.⁵¹

Table B1. Conventional mining costs for Chattanooga Shale.

<u>Cost</u>	<u>1967</u>	<u>1975</u>
\$/Mg shale	1.49-4.41	2.00-5.95
\$/ton shale	1.35-4.00	1.82-5.39
\$/kg U ₃ O ₈	24.75-73.33	33.38-98.89
\$/lb U ₃ O ₈	11.25-33.33	15.17-44.95

An exhaustive process design and cost analysis for the extraction of uranium from the shale was made by Bieniewski, et al. in 1971.⁵ This process featured room-and-pillar mining, crushing, grinding, and acid leaching. Uranium was to have been recovered from the leach solution by solvent extraction. In this model, U₃O₈ could have been produced at a selling price of \$153/kg (\$69/lb) in 1969. That would be about \$200/kg (\$90/lb) in 1975 dollars.

Project Model

We have assumed that a central processing plant will be built that is capable of producing 3.6 Mg (4 tons) of U_3O_8 per day, for 20 years. This is equivalent to the output of a conventional plant of intermediate size.⁵²

To estimate the cost, we also assumed that the shale will be processed in 30-day modules, each having 1/12 of the total annual area. Two weeks would be allowed for retorting and two for leaching. This requires that all fractured units in the module be retorted simultaneously at a burn rate of about 2.7 m (3.8 ft) per day.

Simplified flow sheets for the retorting and uranium extraction phases of this model are shown in Figs. B1 and B2. A summary of the basic processing parameters that have been assumed is given in Table B2.

The parameters of Table B2 have been used to estimate production costs and a selling price for uranium. The uranium content of the shale has been assumed to be 60 ppm, although Wilson¹⁷ found 90 ppm in the Gassaway member. The values of other metals that may be recovered from the shale liquor process stream have not been considered, although such a potential exists. Molybdenum, for example, is three times as abundant as uranium in the shale. It is also present in higher concentrations in leach liquors, and competes with uranium in ion exchange extraction. Other elements found in such solutions include nickel, chromium, cobalt, copper, gallium, scandium and vanadium.

Since it is anticipated that uranium will be the most valuable product extracted from the shale, the oil, gas, and steam have been considered as by-products. A byproduct credit from oil sales has been used in computing the selling price for uranium. Although energy from retort gas and steam could perhaps be economically used on site to provide process heat and power for pumping operations, no credit for such values has been allowed in the cost estimates. The more straightforward assumption has been made of purchased power. However, the magnitude of the available product energy has been considered.

The energy produced in steam by water flooding after retorting was calculated for the estimated mass of shale processed, and from the temperature change and shale specific heat given in Table B2. This does not include the steam produced by flash cooling the leach liquor.

In a trial pyrolyzation under conditions approximating Fischer assay, gas was produced at LLL from 28 m³/Gg (6.9 gal/ton) Chattanooga shale samples at a rate of 38 m³/Mg (1224 scf/ton), with a caloric value of 48 MJ/m³

Fig. B1. Retorting phase.

Fig. B2: Uranium extraction phase.

Table B2. Physical data for model.

SHALE DATA	
2.000E+01 PROJECT LIFETIME, YEARS	1.250E-01 AIR CONSUMPTION IN COMPLETE RETORTING, CU METRES/KG SHALE
1.314E+06 ANNUAL U3O8 PRODUCTION, KG	1.590E+05 AIR CONSUMPTION IN RETORTING, CU METRES/HR AT STP
5.000E-01 OVERALL U FRACTION RECOVERED	8.000E-01 COMPRESSOR EFFICIENCY
3.300E+01 SHALE OIL GRADE, CU METRES/GIGAGRAM	4.800E+01 CALORIC VALUE OF RETORT GAS, MJ/CU METRE
2.000E-01 OVERALL OIL FRACTION RECOVERED	1.000E-01 OVERALL GAS ENERGY RECOVERY FACTOR
6.000E+01 AVERAGE U CONCENTRATION IN SHALE, PPM	1.783E+14 TOTAL ANNUAL ENERGY AVAILABLE IN GAS, J
3.714E+10 ANNUAL SHALE MASS PROCESSED, KG	8.360E+02 AVERAGE BED TEMPERATURE AFTER RETORTING, K
2.300E+00 SHALE DENSITY, G/ML	
1.297E+03 SHALE AVERAGE HEAT CAPACITY, J/KG-K	
1.615E+07 ANNUAL SHALE VOLUME PROCESSED, CU METRES	
4.570E+00 SHALE BED THICKNESS PROCESSED, METRES	
3.534E+00 ANNUAL SHALE AREA PROCESSED, SQ KM	
7.500E+01 AVERAGE FRACTURE DIAMETER, METRES	
4.871E+03 AREA PER HOLE, SQUARE METRES	
7.254E+02 TOTAL ANNUAL HOLES DRILLED	
3.062E+02 RADIUS OF 30 DAY PROCESSING FIELD, METRES	
1.000E-03 AVERAGE FRACTURE THICKNESS, METRES	
3.000E+00 NUMBER OF FRACTURES IN BED	
1.060E+04 FRACTURE VOID VOLUME, CU METRES/YEAR	
2.000E-01 WEIGHT FRACTION OF KEROGEN IN SHALE	
7.428E+09 TOTAL MASS OF KEROGEN IN PROCESSED SHALE, KG	
1.300E+00 KEROGEN DENSITY, G/ML	
RETORTING	
3.000E-01 FRACTION OF IN SITU KEROGEN BURNED	
LEACHING	
	2.467E+07 ANNUAL WATER CONSUMPTION, CU METRES
	4.480E+02 LEACH SOLUTION TEMPERATURE, K
	1.869E+16 TOTAL ANNUAL ENERGY AVAILABLE IN STEAM, J
	2.816E+03 TOTAL PROCESS WATER INPUT, CU METRES/HR
	6.523E+02 SPENT LIQUOR DISPOSAL RATE, CU METRES/HR
	5.341E+02 LEAKAGE LOSSES FROM SHALE BED, CU METRES/HR
	1.202E+03 FLASH COOLING LOSS, CU METRES/HR
	3.778E+05 FLASH HEAT LOSS, MJ/HR
	3.608E+03 POND INPUT, CU METRES/HR
	4.280E+02 POND EVAPORATION LOSS, CU METRES/HR
	9.651E+05 POND HEAT LOSS, MJ/HR
	3.180E+03 RECOVERY PLANT THROUGHPUT, CU METRES/HR

(1286 Btu/scf). Although the relation of such a pyrolysis experiment to an underground retorting is not known, it is likely that the caloric value and actual recovery will be lower. The available energy was therefore estimated at 10% of the total. The recycle of retort gas may be advantageous, but it was not considered in the present model.

Capital Investment

Bieniewski⁵ has estimated how much it would cost to build a plant in which uranium would be recovered from shale leach liquors. This has been used for convenience in computing costs, although it is not certain that the process used is the most economical. The total plant cost has been scaled linearly (by a multiplying factor of 4.45) for an increase in output from 0.81 Mg/day (0.9 tons) to 3.6 Mg/day (4 tons). It was also multiplied by a factor of 1.3 to account for cost inflation from 1969 to 1975.

Other capital investment costs have been estimated using the Lang method⁵³ and a cost factor of 4.74 for a fluid process. This is based on the delivered costs of process equipment, other than that used in the recovery plant. Capital costs are summarized in Table B3.

Table B3. Capital cost estimates.

RETORTING EQUIPMENT		1.200E+05 TOTAL COST OF OIL RECOVERY PUMPS
3.000E+01 RETORT COMPRESSORS REQUIRED		3.674E+03 LENGTH OF 12.7 CM (5 IN.) PIPING, METRES
5.300E+03 AIR FLOW, CU METRES/HR/COMPRESSOR AT STP		1.808E+05 PIPES CAPITAL COST
6.610E+04 ESTIMATED COST PER RETORT COMPRESSOR		9.000E+04 POND CONSTRUCTION
1.983E+06 TOTAL COMPRESSOR CAPITAL COST		
2.798E+01 OIL FLOW TO STORAGE, CU METRES/HR		OTHER
5.000E+00 REQUIRED OIL STORAGE CAPACITY, DAYS		2.766E+05 INSTRUMENTATION, AT 10% OF EQUIPMENT COSTS
3.358E+03 REQUIRED OIL STORAGE CAPACITY, CU METRES		2.119E+07 TOTAL PLANT COSTS, BASED ON DELIVERED PROCESS EQUIPMENT
1.989E+05 STORAGE CAPITAL COST		2.119E+06 STARTUP COST, AT 10% OF OTHER FIXED CAPITAL COSTS
1.500E+04 OIL DEMIST EQUIPMENT		1.000E-01 CONTINGENCY, FRACTION OF OTHER FIXED CAPITAL COSTS
5.000E+04 SULFUR PLANT FOR RETORT GAS CLEANUP		2.119E+06 CONTINGENCY CAPITAL COSTS
2.000E+04 RETORT GAS FLARE FACILITY		2.471E+05 LEASE COST, \$/SQ KM/YEAR
LEACHING EQUIPMENT		8.732E+05 TOTAL ANNUAL LEASE COSTS
8.375E+05 CAPITAL INVESTMENT, 1969 U RECOVERY PLANT		1.746E+07 TOTAL 20 YEAR LEASE COSTS
4.445E+00 SCALE FACTOR FOR INCREASED PLANT SIZE		1.000E-01 WORKING CAPITAL, FRACTION OF TOTAL CAPITAL INVESTMENT IN PHYSICAL PLANT
1.300E+00 COST INFLATION FACTOR, 1969-1975		2.119E+06 WORKING CAPITAL, \$
4.840E+06 CAPITAL INVESTMENT, U RECOVERY PLANT		4.289E+07 TOTAL CAPITAL INVESTMENT
4.642E+04 ESTIMATED TOTAL COST FOR POND PUMPS		
9.802E+05 TOTAL LEACH COMPRESSOR COSTS		
6.210E+04 TOTAL COST, SPENT LIQUOR REINJECTION PUMPS		

Operating Costs

Operating costs are summarized in Table B4. Drilling and fracturing are assumed to be contracted operations. The estimated drilling and casing cost listed is for a 0.15 m (6 in) hole with 0.13 m (5 in) casing and a final inside diameter of 0.10 m (4 in). The depth is 46 m (150 ft). The total costs for hydraulic fracturing include fluid, equipment, and labor. The fracturing explosive is assumed to be a modified ammonium nitrate-oil slurry with a density of 1.45 and a cost of \$0.66/kg (\$0.30/lb). The calculated cost is for the quantity required to fill the void volume created by hydraulic fracture.

Table B4. Operating cost estimates.

1.150E+06 ANNUAL OPERATING COSTS, U RECOVERY PLANT	3.000E-02 WATER COST, \$/CU METRE
3.000E+03 DRILLING AND CASING, COST PER HOLE	7.402E+05 TOTAL ANNUAL WATER COST
2.176E+06 TOTAL ANNUAL DRILLING, CASING COSTS	8.000E+05 TOTAL ANNUAL LABOR COST
2.650E+02 HYDROFRACTURE COST, \$/CU METRE VOID VOLUME	5.000E-02 ROYALTY FRACTION OF ANNUAL PROFITS
2.809E+06 TOTAL ANNUAL HYDRAULIC FRACTURE COST	6.348E+05 TOTAL ANNUAL ROYALTIES
1.017E+07 TOTAL ANNUAL EXPLOSIVE COST, \$	2.119E+05 TOTAL ANNUAL INSURANCE
2.994E+10 ENERGY REQUIRED FOR AIR COMPRESSION, J/HR	5.000E-02 STATE-LOCAL TAX RATE, FRACTION OF PHYSICAL PLANT CAPITAL INVESTMENT
8.333E+00 ELECTRICITY COST, \$/GJ	2.500E-01 STATE-LOCAL ASSESSMENT RATE
2.186E+06 AIR COMPRESSION COSTS, \$/YEAR	1.324E+05 TOTAL ANNUAL STATE-LOCAL TAXES
1.960E+04 SOLUTION PUMPING COSTS, \$/YEAR	6.970E+06 ESTIMATED INDIRECT COSTS, AND CONTINGENCY
4.411E+06 TOTAL ESTIMATED PUMPING COSTS, INCLUDING RETORT, \$/YEAR	3.020E+07 TOTAL ANNUAL OPERATING COSTS

The rate of air consumption in retorting was computed from the estimated fraction of the total kerogen consumed and the air required for complete retorting.⁵⁴ The energy required was taken to be that necessary to compress atmospheric air to hydrostatic pressure, divided by a compressor efficiency.

The rate of leach liquor flow was derived from the plant output and the estimated concentration of uranium in solution. The flow would be collected by 12 pipelines extending to the perimeter of the 30-day processing module. Pipeline size and liquid pumping costs were computed from this flow rate and

the number of lines required. The energy costs for pumping fluids through the shale bed are not readily computed, because of unknown and variable permeability. They were, therefore, estimated to be equal to the sum of the other pumping costs discussed above.

It is assumed that spent leach liquor will be recycled to take advantage of its sulfuric acid content, but that it will ultimately be left in the formation. No disposal costs are incurred. It is estimated that the volume of process water ultimately consumed in this way will be equal to the total volume occupied by kerogen. Shale with an oil assay of $33 \text{ m}^3/\text{Gg}$ (8 gal/ton) has a kerogen content of about 20%.⁴⁵ The kerogen density is taken to be 1.3. It has been assumed that water will be purchased.

Royalties on mineral products have been computed as an operating cost, at a fraction of the gross annual profits:

$$M = B \cdot N \cdot I (1 + F), \quad (\text{B1})$$

where

M = annual royalties

B = royalty fraction of annual profits

N = rate of return on investment

I = total capital investment

F = federal income tax rate

Labor costs have been approximated on the basis of four shifts per day of ten men each, at an annual cost per man of \$20,000. An additional one-third was allowed for supervisory costs. Insurance has been included at an annual rate of 1% of fixed plant costs. One-third of the total of other operating costs has been added for indirect costs, such as plant overhead, administration, distribution, marketing, and process development.

Selling Price Calculation

The selling price for U_3O_8 was calculated from Eq. B2. Straight-line depreciation and internal financing have been assumed.

$$P = [(A + 0.05(I - W - L) + NI(1 + F) - V(1 - F)) / T] \quad (\text{B2})$$

where

P = selling price, \$/kg

A = annual operating costs

I = total capital investment

W = working capital
 S = state-local tax rate
 R = assessment rate
 N = rate of return
 F = federal income tax
 T = annual kg U_3O_8 produced
 V = annual gross oil sales
 L = lease costs

A posted price for U.S. exempt crude in May 1975⁵⁵ of \$73.59/m³ (\$11.70/bbl) was used to estimate V, the oil product value.

The input and results of this selling price calculation are summarized in Table B5.

Table B5. Uranium selling price estimate.

2.000E-01 RATE OF RETURN ON INVESTMENT	1.804E+07 ANNUAL GROSS OIL SALES
4.800E-01 FEDERAL TAX RATE, FRACTION OF GROSS SALES	2.639E+01 U3O8 SELLING PRICE, \$/KG
7.359E+01 OIL SELLING PRICE, \$/CU METRE	1.197E+01 U3O8 SELLING PRICE, \$/LB

Cost Sensitivities

An analysis was made of the sensitivity of the computed selling price to changes in several of the assumptions of the model. The variables examined were uranium recovery, oil recovery, oil selling price, drilling costs, and fracture diameter. The results of this study are shown in Figs. B3-B7. It is evident that reasonable changes in individual parameters do not cause prohibitive increases in the U_3O_8 selling price. However, despite this apparent latitude in processing costs, unfavorable combinations of several factors could exist. It does not appear possible to make a more definitive cost statement until a technical development program has resolved some of the key problems that have been outlined.

Fig. B3. Sensitivity of the U_3O_8 selling price to overall fractional uranium recovery.

Fig. B4. The sensitivity of the U_3O_8 selling price to the oil fraction recovered.

Fig. B5. The sensitivity of the U_3O_8 selling price to the price of oil.

Fig. B6. Sensitivity of U_3O_8 selling price to drilling costs.

Fig. B7. Sensitivity of the U_3O_8 selling price to average fracture diameter.

Discussion of Costs

In addition to uncertainties in production cost estimations, comparison with the selling prices of uranium from other sources is difficult. The current uranium market is unstable. Prices are rising rapidly, and extrapolations are uncertain. As an example, a plot of contract prices for future delivery is shown in Fig. B8.⁵⁶ More recent data have continued to show increasing price levels.

Uranium reserves are arbitrarily divided, for assessment purposes, into classes according to production costs. The conventional definition of low-cost reserves has been \$17.64/kg (\$8/lb), although depletion and inflation are also making it more useful to consider \$22/kg (\$10/lb) and \$33/kg (\$15/lb) ores in this category. According to this classification, *in situ* processing could transform much of the vast beds of black shale in the eastern U.S. into low-cost reserves.

In comparing costs of *in situ* recovery with other methods, environmental effects and costs must also be considered. These factors alone are probably sufficient to prevent conventional mining of the shale.⁵⁷ For example, it is very unlikely that strip mining would ever be acceptable in the area occupied by the shale, because of the existing depth of overburden. In addition, it can be calculated that room-and-pillar mining, as outlined by Bieniewski,⁵ even after refilling the mined volume with tailings, would produce solid wastes equivalent to a bed 3.3 m (11 ft) thick over the whole of the area mined.

The large, long-term requirement for uranium means that its mining and processing will remain a large-scale industry, with high capital requirements. However, *in situ* processing, if feasible, would offer two major advantages: creation of substantial new ore reserves, and a means of exploiting them with lessened environmental impact.

Fig. B8. Contract prices for future delivery of U_3O_8 .

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226-250	8.00	551-575	13.50
251-275	9.00	576-600	13.75
276-300	9.25	601-up	*
301-325	9.75		

*Add \$2.50 for each additional 100 page increment from 601 to 1,000 pages; add \$4.50 for each additional 100 page increment over 1,000 pages.