Groundwater Restoration with In Situ Uranium Leach Mining

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INTRODUCTION

In situ leach mining of uranium (U) has developed into a major mining technology, which compares favorably with traditional open pit mining and underground mining for recovery of U ore. Since 1975, when the first commercial mine was licensed in the United States, the percentage of U produced in the United States by in situ mining has grown steadily from 0.6 percent to 10 percent in 1980. Part of the reason for this growth is that in situ mining offers less initial capital investment, shorter start-up times, greater safety, and less labor than conventional mining methods. There is little disturbance of the surface terrain or surface waters, no mill-tailings piles, and no large open pits. Also, many of the hazards of working in a U-processing plant are reduced.

In situ leach mining also has environmental disadvantages. During mining, large amounts of groundwater are circulated and there is some withdrawal from an area where aquifers constitute a major portion of the water supply for other purposes. When an ammonia-based leach system is used, the ammonium ion is introduced into an area where cation exchange on clays (and some production of nitrate) may occur. Also, injection of an oxidant with the leach solution causes valence and phase changes of indigenous elements such as As, Cu, Fe, Mo, Se, S, and V as well as U. Thus, other species along with uranium may be solubilized. Furthermore, the surrounding groundwater can become contaminated by escape of the leach solution from the mining zone.

Following mining, the water in the mined zone is left with an ionic strength that exceeds the natural baseline level for some constituents. In particular, much attention has been directed toward the high (relative to baseline) ammonium concentration, following leaching with an ammonium-based system, and toward the fate of heavy metals solubilized during the mining process. A question of major interest concerns restoration of the sites, since many of the mining intervals or horizons coincide with fresh groundwater. Restoration is necessary to reduce the amounts of undesired chemical constituents left in solution after mining operations have ceased and thus to return the groundwater to a quality consistent with premining use and potential use.

The questions and problems faced by the in situ mining industry, and by those who regulate it, are representative of those faced by any developing industry that has the potential for causing adverse environmental changes on a valuable resource. This chapter presents an overview of the in situ mining technology, including U deposition, mining techniques, and groundwater restoration alternatives. The latter part of the chapter covers the situation in South Texas. Economics and development of the industry, groundwater resources, regulation, and restoration activities are also reviewed.

URANIUM DEPOSITION IN SANDSTONE

The principal regions in the United States with potential U recoverable by in situ leach mining are the Wyoming Basins, the Colorado Plateau, and the Gulf Coastal Plain of Texas. The southern Black Hills and northeastern Colorado, within the Great Plains region, also contain sedimentary U deposits that may be amenable to in situ leach mining. The formation of a U deposit requires five factors: a source of U, a transporting

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media, a host rock, a trapping mechanism, and a preservation mechanism. The U in sandstone-type deposits is thought to have been derived from either granite or volcanic material that was deposited with, or later than, the sandstone. Oxygenated groundwater percolating through these units oxidizes and mobilizes the U and other minerals. The U-enriched groundwater moves downdip in the sandstone until it reaches an interface between oxidizing and reducing groundwaters. At this interface the U and other elements, including As, Se, V, Cu, and Mo, precipitate out of solution. Such deposits are called "roll-front" deposits. Typically they follow the general trend of the drainage channels. Individual ore bodies in sandstone lenses tend to be long and narrow but rarely exceed 100 m in length. These deposits are usually bounded above and below by impermeable mudstones or shales and are interspersed with clay stringers. Geologically and mineralogically, the deposits are similar. The ore is unoxidized, with the principal ore minerals being uraninite [$(U_{1-x}^{+4}, U_x^{+6})O_{2+x}$; ideally, UO_2] and coffinite [$U(SiO_4)_{1-x}(OH)_{4x}$]. Because of the mode of transport and deposition, many U deposits coincide with fresh groundwater aquifers (Ford, Bacon, and Davis Utah, Inc., 1979).

MINING TECHNOLOGY

When exploration indicates the existence of an ore body, a sampling, testing, and monitoring program is established to determine the extent and quantity of ore, baseline water quality, and optimal well pattern design. Observation wells are located in front of, in, and behind the ore body in the ore-bearing aquifer and in any aquifers directly above and below the ore horizon. Monitor wells are also drilled at the sides of the ore zone to encircle the production site and within the containment area to monitor excursions (Geraghty & Miller, Inc., 1978). After the necessary permits are obtained, production can begin.

In situ leach mining is that method by which the ore mineral, in the original geologic setting, is preferentially leached from the host rock by the use of specific leach solutions and the mineral recovered. The leach solution, or lixiviant, is introduced into the formation through injection wells. Once in the formation the leach solution oxidizes, complexes, and mobilizes the ore. The enriched, or pregnant, solution is then removed from the formation through production wells. At the surface the U in the pregnant solution is recovered by ion-exchange techniques. The residual U-barren solution from the ion-exchange operation is regenerated with suitable leaching chemicals and recirculated to the well field for reuse. To minimize the likelihood of migration of the leach solution from the production zone, the system is usually operated with more solution being withdrawn than injected. This excess withdrawal is referred to as the "bleed." It is disposed of through solar evaporation and deep-well injection.

Once a suitable loading has been achieved on the exchange resin, the ion-exchange column is removed from the production stream and the resin is cycled to the elution step. During elution, the resin is contacted with a chemical solution that

strips the U from the resin; the resulting product solution is termed the eluate, and eluted resin is recycled to the loading operation. The U is recovered from the eluate by precipitation. Following thickening and filtration, the final product is primarily ammonium diuranate, a yellow-colored precipitate or slurry termed *yellow cake*. The filter cake can then be dried and packaged for shipment (Larson, 1978).

During development of the in situ mining industry, leaching experiments were carried out with two major types of leaching systems. Acid-leaching systems use the sulfate of dilute sulfuric acid as the chelating agent with the U mobilized as the uranyl sulfate complex, $UO_2(SO_4)_3^{-4}$. In the presence of significant $CaCO_3$, large amounts of acid are required and there are problems with precipitation of $CaSO_4$. Generally, acid-leaching systems have not found significant in situ use. Alkaline, or carbonate, leaching systems have used carbonates and bicarbonates of ammonium and sodium to complex the U into the uranyl tricarbonate form, $UO_2(CO_3)_3^{-4}$. Many of the early commercial operations used an ammonium carbonate-bicarbonate leach with hydrogen peroxide as the oxidizing agent. The leach solution is made by adding from 0.5 to 2.0 g/L H_2O_2 and from 0.5 to 10.0 g/L of the NH_4-HCO_3-CO_3 species to the groundwater. Typical values for H_2O_2 are less than 1.0 g/L (Kidwell and Humenick, 1981), while those for the NH_4 species vary from 1 to 3 g/L (D. Tweeton, U.S. Bureau of Mines, personal communication, 1982). Oxidation results from the decomposition of H_2O_2 to water and oxygen. Uranium is recovered as $(NH_4)_4UO_2(CO_3)_3$. In the presence of certain clays, ammonium is exchanged for other cations. This exchanged ammonium is difficult to remove during restoration. To avoid the problem of ammonium restoration the industry has moved to the use of a sodium carbonate-bicarbonate leach with oxygen as the oxidizing agent. Although cation exchange does not present a restoration problem with this leach, the presence of sodium can cause dispersion of certain clays and resulting loss of permeability. It appears that this is less of a problem at a moderate rather than a high pH; however, research on leaching is continuing. Recently there has been interest in a potassium carbonate-bicarbonate leaching system. This system may avoid both the problems of difficult restoration faced by the ammonium system and potential loss of permeability faced by the sodium system. The primary obstacle standing in the way of the potassium carbonate-bicarbonate lixiviant is its expense. In late 1980, 1 kg-mole of potassium bicarbonate cost $31, whereas 1 kg-mole of ammonium bicarbonate cost $8 and 1 kg-mole of sodium bicarbonate cost $7 to $8 (U.S. Bureau of Mines, 1981). Researchers in petroleum engineering at the University of Texas at Austin have developed the use of a KCl preflush. This preflush satisfies the cation exchange sites with potassium before leaching and reduces the consumption of potassium carbonate-bicarbonate during leaching. Because the cost of KCl is relatively low (about $5/kg-mole), the cost of using potassium carbonate-bicarbonate is reduced. An additional benefit of the KCl preflush is better maintenance of permeability. Also, in laboratory experiments, the chloride preflush did not reduce U recovery. If the potassium lixiviant is used with a KCl preflush, it may be cost competitive with either the ammonium or sodium leaches (U.S. Bureau of Mines, 1981).

GROUNDWATER RESTORATION ALTERNATIVES AFTER LEACH MINING

A strategy for restoration is necessary to reduce the amounts of undesired materials left in solution after operations have ceased and thus return the groundwater to a quality consistent with pre-mining use and potential use. In situ U mining affects groundwater quality directly by the introduction of compounds with the leaching solution and indirectly by the release of materials such as heavy metals from dissolution of minerals present in the rocks. Ammonium is an example of a compound that may be introduced into an aquifer during the mining process. Although ammonium systems are no longer in general use by the U-mining industry, they were the primary leaching system used during the initial period of development. Ammonium may be innocuous in the subsurface, but since it may oxidize to nitrates, a mobile species for which an EPA drinking water standard of 10 mg/L of nitrogen has been established, much attention has been focused on the question of ammonium restoration. This topic is addressed later in this chapter. The question of the release of heavy metals and other materials during mining is still of major concern. Kidwell and Humenick (1981) investigated trace groundwater contaminants released during mining (in South Texas) and found that U was the major trace element to appear in significant quantity during groundwater sweeping (discussed below) of a leached area. They also found that leaching operations can rapidly solubilize Mo, but that its concentration declined to EPA limits within a few pore volumes of groundwater sweep if it was not recycled in the leaching solution. Arsenic was found to be released after a period of time, though in minor amounts, and V and Se concentrations were not found to be significant. Of major interest is their conclusion that groundwater sweeping may restore the production area, and the extent of the sweep depends on the U content and time needed to leach the ore but not on the strength of the oxidant used.

Groundwater restoration entails a range of techniques from in situ processes within the aquifer to surface-separation processes that treat the water at the surface for later reinjection to the former production zone or disposal by other means and finally to ultimate disposal methods that dispose of the water that is not feasible to treat. In fact, most strategies involve a mix of in situ techniques, surface-separation processes, and ultimate disposal methods to achieve restoration. A more detailed discussion of restoration methods is given in the report by Ford, Bacon, and Davis Utah, Inc. (1979) and by Charbeneau *et al.* (1981).

During the initial phase of restoration, water containing the highest concentrations of contaminants is produced from the mining zone. These first few pore volumes are usually disposed of by partial evaporation and subsequent deep-well injection. Following this initial phase of restoration, in situ methods may be applied. The simplest strategy is to suspend restoration activity in the aquifer after disposal of the first few pore volumes from the production zone. In this way the physicochemical actions at work within the aquifer, including reduction, neutralization, precipitation, and adsorption, may restore the groundwater naturally. Natural processes are important in all

restoration schemes because they provide the mechanism by which the aquifer is stabilized after the groundwater quality has been preliminarily restored and because they may reduce the need for extensive surface treatment. However, restoration based on natural processes has never been proposed by an operator nor approved by any state or federal agency. The mechanisms of natural restoration are complex, and the important physicochemical properties are highly variable. This makes it difficult to provide evidence and assurance of improvement in groundwater quality by natural mechanisms. Despite these uncertainties, natural restoration may yet prove to be adequate to protect the environment. A number of methods have been suggested to enhance in situ restoration. These include biological nitrification, bacterial precipitation, and chemical precipitation.

Associated with the in situ and surface-separation processes of restoration are the methods for removal of water from the aquifer—groundwater sweeping and groundwater recirculation. In a groundwater sweep, injection operations are curtailed and a number of wells are used as production wells, which draw water from the contaminated part of the aquifer (production zone) and draw cleaner water from the nonaffected part of the aquifer. This method may be conducted with surface separation and reinjection of the treated water into the aquifer at a point away from the production zone, or it may be conducted without recharge in order to draw exclusively from the undisturbed waters outside the production zone. Because there is no recharge, the fluid migrates uniformly to the production wells. After such a sweep, the water may be disposed of by deep-well disposal, by solar evaporation ponds, or by irrigation or runoff to surface flows.

Restoration by groundwater recirculation is achieved through the continued pumping through the same production and injection wells used in the mining operations, except that the direction of induced flow may change and the groundwater is reinjected without being reconstituted with the leaching solution (lixiviant). Recirculation may be conducted either as forward recirculation, in which the same wells are used as in the mining process for production and injection, or as reverse recirculation, wherein the mining injection wells are converted to production wells and the mining production wells are converted to injection wells. If a continued five-spot well pattern is used in the mining process, the major distinction between forward and reverse recirculation concerns the behavior of the boundary wells. With forward recirculation the lixiviant is pushed further away from the injection wells along the streamlines of the flow field. If the system is operated with a bleed, then the lixiviant will eventually be pushed into a production well and recovered. In the case of reverse recirculation the lixiviant is pulled back along the same streamlines to the former injection wells and recovered. There are trade-offs between forward and reverse recirculation, and a system may be designed so that only some of the mining wells operate and the system is a mix between forward and reverse recirculation. In any case, this continued circulation enhances the natural physicochemical processes referred to earlier. Additionally, production water could be processed or treated at the surface to reduce chemical concentrations to background levels and then reinjected into

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the aquifer to leach additional contaminants. Also, the processed solution could be constituted with compounds that enhance restoration before reinjection.

Surface-separation processes consist of pumping contaminated water from the underground production area and treating the water in a suitable treatment plant at the surface. The treated water may be used as recharge to the aquifer or otherwise disposed of depending on the quality characteristics of the final effluent from the treatment plant. The residuals from the treatment process must be disposed of properly to avoid undesirable pollution of surface waters or of the land. Because of the characteristics of the production water from in situ mining, physicochemical processes are applicable for the treatment of the contaminated water before its final disposal. These processes include reverse osmosis, electrodialysis, distillation, chemical precipitation, adsorption, ion exchange, foam fractionation, and freeze separation. A 1978 ranking of separation methods to produce a water satisfactory for recharge is presented by Ford, Bacon, and Davis Utah, Inc. (1979). Reverse osmosis and recharge with either deep-well disposal or solar evaporation of the treatment waste appears to be the most economically effective technique. The system costs ($/1000 gallons) for recharge/reverse osmosis/deep-well disposal and recharge/reverse osmosis/solar evaporation and pond disposal are $1.78 and $2.19, respectively. More recent information indicates that electrodialysis is now competitive with reverse osmosis (Garling, 1981). The separation processes have as their objectives the production of a water suitable for recharge to the aquifer. If recharge is not required to meet quantity demands from the aquifer, sweeping and deep-well disposal or sweeping followed by solar evaporation of the sweep water in properly designed and constructed ponds may be the preferred restoration scheme.

A major part of any restoration scheme is the choice of method or methods for ultimate disposal of the wastes. Two methods are available: (1) deep-well injection and (2) evaporation in ponds followed by solid-waste disposal of the sludge and residue. During restoration, at least the first few pore volumes containing the most heavily contaminated water are disposed of either through deep wells or evaporation in ponds. This is necessary because in situ methods are inadequate and surface-separation methods are infeasible to effect restoration of such water. A major advantage of deep-well disposal over surface-disposal methods is that the objectionable waste solution is completely removed from the environmentally sensitive biosphere. Deep-well disposal systems are limited to solutions with low-suspended solids and turbidity and solutions that will not plug the injection zone by the precipitation of solids in reactions between the waste and the solutions and solid matrix of the host aquifer. The solar-evaporation pond process reduces the volume of wastewater but generates a sludge residue that must be impounded. Solar-evaporation ponds are effective for the disposal of most pollutants from groundwater restoration. Solid-waste-disposal schemes are required for the solid wastes generated. The solid-waste-disposal system is required to collect, store, treat, handle, transport, place, and secure the objectionable materials.

IN SITU URANIUM MINING IN SOUTH TEXAS

The previous sections have suggested that U is deposited locally along with additional potential contaminants, that the in situ mining process solubilizes the U and other materials, and that a number of restoration options are available to return the groundwater to a state compatible with pre-mining use and potential use. It is important to realize that the technology is new and that large-scale restoration, while in progress, has not been fully achieved to date. Estimates as to the ease or difficulty of restoration are based on laboratory studies and small-scale prototype operations. At present, it appears that restoration is possible but that the amount of water and energy use depends on the restoration method(s) applied and on site-specific factors.

In the following sections the in situ U-mining technology as developed and practiced in South Texas is discussed. Along with industry development and the present status of restoration activities, both the groundwater resources and regulation are reviewed.

Development and Economics of In Situ Mining in South Texas

Uranium was first discovered in the Tordilla Sandstone of Karnes County in 1954. Western Karnes County was mined extensively for the U.S. Atomic Energy Commission (AEC) by a few private companies using surface mining. The first area mill was built in 1960 by the Susquehanna Company and the Western Nuclear Company. The mill was developed under a purchase agreement with the AEC for a quantity of uranium oxide to be sold at $9.90 per pound from 1960 to 1962, followed by another quantity from 1962 through 1966 to be sold at $8 per pound. This mill was one of 17 U concentrating and processing facilities in the United States at that time and had the smallest output. From 1967 to 1970 the AEC began a "buying stretchout" program, which did not include Texas, and the Susquehanna-Western mill closed temporarily. At that time AEC sales of uranium oxide were at about $6 per pound. As the demand for U by the federal government fell, sales to private companies began to increase. Exploration grew rapidly in 1968 in South Texas and throughout the United States. Surface-mining activity increased in 1969 and 1970 with three mills operating in Karnes and Live Oak Counties.

The development of in situ mining offered the prospect for production of similar quality but from deeper ore bodies. The process also posed less danger from radioactivity both during mining and afterward because the process did not entail removing the overburden, as in surface mining, and therefore did not leave behind radioactive tailings ponds. The first modern in situ U-leach mine was operated by Utah Construction and Mining Company (now Utah International, Inc.) at its Shirley Basin site in Wyoming. During 1961 to 1963, the company experimented with many techniques, particularly with regard to well-development procedures and leach solutions. From 1963 to 1969 in situ mining was the only method used by this company for U production. After 1969 the in situ leach operation was replaced by open-pit mining. The first industrial devel-

opment in Texas was in 1975 when Atlantic Richfield, U.S. Steel, and Dalco became partners in a pilot in situ U plant using the Dalco-developed process at the Clay West site near George West in Live Oak County.

The first state permit for commercial in situ mining of U in the United States was issued to Atlantic Richfield and partners for the operations at the Clay West site, effective January 1975. In October 1976 a second in situ site was permitted for the Burns Ranch site directly across the highway from the first site. At that time Texas led the nation in the production of uranium yellow cake produced by the in situ technique. In 1977 and 1978, both in situ and surface-mining operations developed and expanded, while exploration for deposits suitable for both types of operations continued to be sought. Comparisons between surface and in situ mines at this time showed that while conventional surface mines produced yellow cake for $11.83 per pound, in situ mining could produce it for $8.73. Also, lead time for the operational development of in situ mines was 2 to 3 yr, as opposed to 7 to 10 yr for surface mines. By 1980, 9 companies were mining at 25 sites by means of in situ technology, and 6 additional applications for permits had been filed. The location of the U district within the State of Texas is shown in Figure 12.1. The current distribution of in situ mines is shown in Figure 12.2, where in situ mines are found in five counties: Bee, Duval, Karnes, Live Oak, and Webb.

Demand for U slacked in 1979. The price climb had been dramatic: from $8 per pound in 1970, $10.50 in 1975, and $22 in 1976 to $43 and more for spot markets in 1978. Prices fell from $43.25 in December 1978 to $40.25 in December 1979, a 6 percent decline after the stable price high of 1978. The price continued to decline, reaching $31 in July 1980, and by September 1981 the price stood at $23.50 per pound.

Two developments accounted for much of the reduction in demand. One was the artificial demand created under the Long-Term Fixed-Commitment Enrichment Contracts, which, from their inception in 1973, caused a disparity between feed requirements that had existed years before and actual reactor needs (reflecting cancellations and deferrals). In 1979 the transition to the Adjustable Fix-Commitment Enrichment Contract was completed; it eliminated the higher, artificial demand by reducing feed requirements to more closely correspond to actual reactor needs. Another factor contributing to demand reduction was the Three Mile Island nuclear reactor accident. As a result of the accident, numerous reactor construction schedules were canceled or indefinitely deferred and start-up dates were delayed. The U.S. Nuclear Regulatory Commission ceased licensing, and the requirements for design change and retrofit remained uncertain. These developments together reduced U.S. uranium needs and delayed the dates to meet these needs. The full effects of lessened U.S. demand were not felt immediately owing to the unprecedented level of activity among non-U.S. buyers on the U.S. market; chief among them South Korea, Taiwan, Japan, Switzerland, and West Germany.

Meanwhile supply continued to grow with the momentum generated by contracts concluded during the price rise of 1974 to 1976. What was then perceived as a shortfall in supply accelerated exploration, development, and ambitious programs
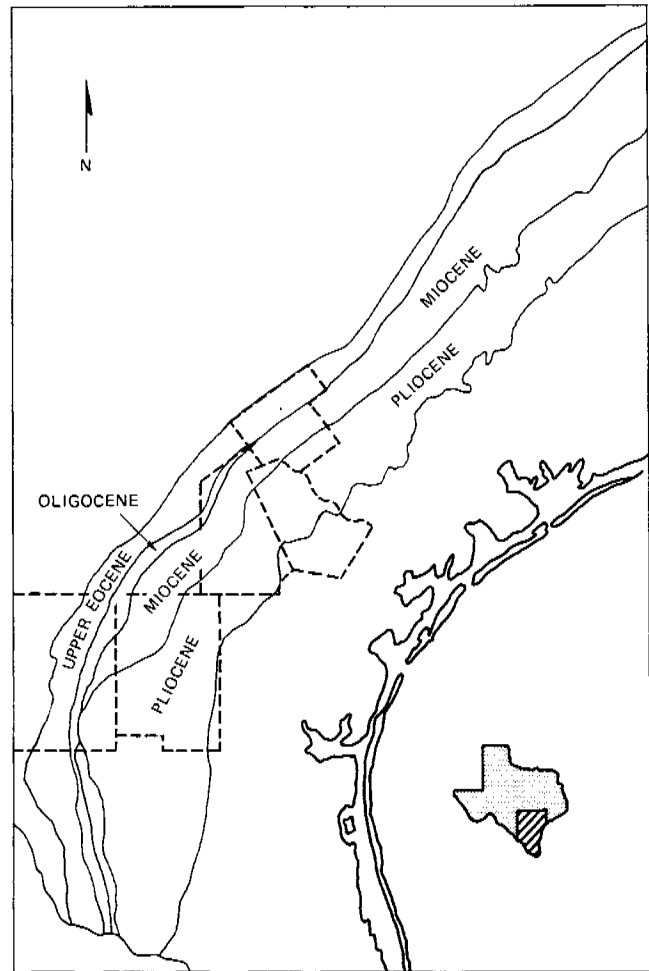

FIGURE 12.1   Geologic formations in South Texas (surface outcrops).

of expansion, not only in South Texas and the other U areas of the United States but in Africa, Canada, and Australia as well. Production during 1979, as throughout the history of the industry, exceeded consumption worldwide. Utility inventories grew steadily during the 1970s. Exploration in the United States as a whole declined 23 percent in 1979 in direct response to the drop in U prices. But activity in South Texas continued at its previous pace with in situ mining remaining the favored recovery method.

Groundwater Resources of the South Texas Mining District

The water resources of the South Texas U-mining district consist primarily of groundwater. This resource is highly variable in quantity and quality, and the impact of restoration on the resource is a question of major concern. In the South Texas region, U occurs in several different formations of the early- to mid-Tertiary clastic wedge of the coastal plain, including the Jackson Group (Eocene), Catahoula Formation (Oligocene-Miocene), Oakville Formation (Miocene), and Goliad Forma-
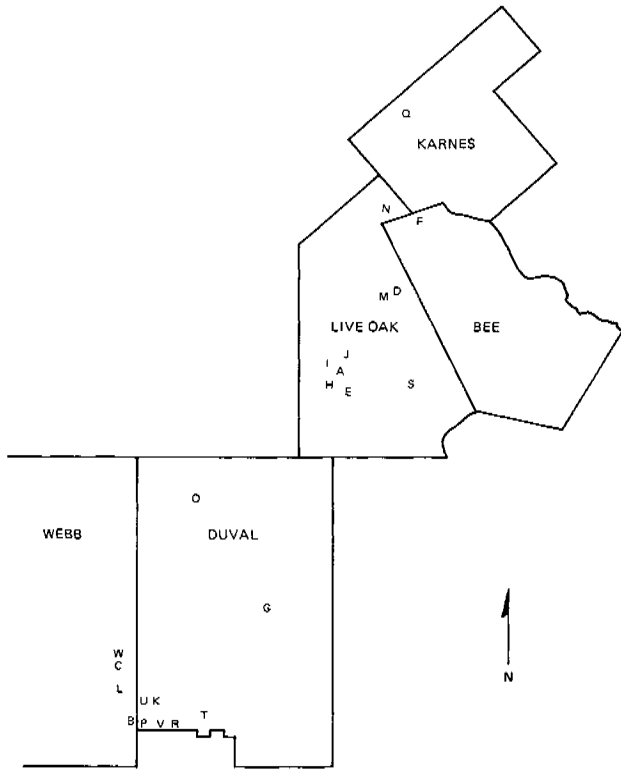
FIGURE 12.2 South Texas uranium mining district. The following reference letters refer to the mine and operating company: A, Moser, U.S. Steel Corp./N.M. Uranium, Inc.; B, O'Hern, Mobil Nufuels Corp.; C, Bruni, Wyoming Minerals Corp.; D, Lamprecht, Wyoming Minerals Corp.; E, Burns Ranch, U.S. Steel Corp.; F, Pawnee, Intercontinental Energy Corp.; G, Palangana, Chevron (Union Carbide); H, Clay West, U.S. Steel Corp./N.M. Uranium, Inc.; I, Boots/Brown, U.S. Steel Corp./N.M. Uranium, Inc.; J, Pawlik, U.S. Steel Corp.; K, Holiday-El Mesquite, Mobil Nufuels Corp.; L, West Cole, Tenneco Uranium, Inc.; M, ZamZow, Intercontinental Energy Corp.; N, Nell, Mobil Nufuels Corp.; O, Piedre Lumbre, Mobil Nufuels Corp.; P, Longoria, Uranium Resources, Inc.; Q, Hobson, Everest Minerals Corp.; R, McBride, Caithness Mining Corp.; S, St. Lucas, Everest Minerals Corp.; T, Las Palmas, Everest Minerals Corp.; U, Benavides, Uranium Resources, Inc.; V, Trevino, Conoco, Inc.; and W, Santonino (test), Urex, Inc.

tion (Pliocene). The different formations outcrop in belts trending roughly northeastward parallel to the coast, with younger rocks cropping out near the coast and successively older units farther inland (Figure 12.1). The Tertiary sediments in the area were deposited on the subsiding margin of the Gulf Coast geosyncline, and their regional dip is gently basinward (southeast) with some local reversal occurring along fault zones. The sequence of rocks in the region typically thickens in the direction of regional dip, until it encounters one or more belts of contemporaneous faults. The thickness of the Tertiary sediments increases greatly across these faults, coincident with a change from fluvial to coastal plain facies (Galloway, 1977).

Precipitation on the various outcrops on the coastal plain, and northwest of it, is the source of regional groundwater.

Nevertheless, a large part of that precipitation either runs off at the land surface or is dissipated by evapotranspiration. Only a small fraction of the precipitated water infiltrates the soil and the subsoil, moving downward to the water table and becoming part of the groundwater in storage. The general direction of movement of the groundwater flow in the area is southeastward, to the Gulf, from areas of natural recharge to areas of natural discharge. However, this general pattern is locally interrupted by concentrated pumping and local faults. Although the topographic relief is slight, there is evidence of topographic control of groundwater movement. The principal means of natural discharge are transpiration, evaporation, and interformational leakage, while pumped wells constitute the principal artificial means of groundwater discharge.

Using information developed by the Texas Water Development Board (1977), the groundwater availability in the U-mining area of the Gulf coastal plain is estimated to be about 100×10^6 m^3/yr. This quantity exceeds the present rate of water use of 50×10^6 m^3/yr for municipal, industrial, and agricultural users (Charbeneau et al., 1981). Water use is not expected to exceed 75×10^6 m^3/yr by the year 2000. Thus under present circumstances, there is not expected to be a water-quantity problem in this South Texas region in the near future.

Concerning questions of restoration, the original or baseline water quality is as important as the quantity of water. Strategies for aquifer restoration, after the U ore has been withdrawn, are developed with an understanding of the baseline water quality and of prospective users of waters of varying qualities. Dissolved chemicals are common to all subsurface waters. They result from the leaching of materials in the soils and rocks through which the water must pass to reach and flow in the aquifer. Most of the groundwater used for municipal supplies in the in situ area is fresh (0 to 1000 mg/L of dissolved solids), though some may be slightly saline (1000 to 3000 mg/L). Since in some cases the only water available may be slightly saline, it is used for human consumption even though the mineral content may be too high for certain manufacturing and industrial uses. The U.S. Environmental Protection Agency (EPA) has established maximum contaminant levels (MCL) for some constituents in public water systems. The EPA regulations set forth primary MCL values that are enforceable by law and secondary values that represent reasonable qualities for drinking water but are not federally enforceable. An examination of 165 wells in Duval County has shown that most of the water sampled does not meet the MCL values in all respects and that dissolved solids and chloride content are especially high. Again, however, this is the best water available and is currently being used for drinking with no obvious adverse effects (Shafer, 1974).

The major ions that normally constitute more than 90 percent of the total dissolved solids (TDS) in groundwater are the cations sodium, calcium, and magnesium and the anions bicarbonate, chloride, and sulfate (Freeze and Cherry, 1979). The exact chemical makeup is controlled by the source of water and the path it takes through the subsurface system. For the Gulf Coast aquifers, low-pH water near the outcrop dissolves $CaCO_3$. As this water moves downdip, the calcium is exchanged for sodium in clays and the sodium content increases. Bicarbonate

is the stable anion of the carbonate system at the predominant pH. As such, most waters in the Gulf Coast aquifers become sodium-bicarbonate waters downdip. As these waters continue to migrate it is common to find an increase in sulfate and particularly chloride concentrations. Baseline water-quality data for a number of South Texas in situ leach mining sites have been gathered by Humenick et al. (1978). These data show a TDS ranging from 700 to 2300 mg/L with highly variable cation and anion concentrations. For the cations, which are more important than the anions in terms of groundwater restoration (of ammonium), sodium is the major species accounting for 53 to 96 percent of the cations on an equivalents basis (sodium concentrations ranged from 11.1 to 30.6 meq/L). Calcium is second in importance, magnesium third. The major anions are bicarbonate and chloride.

Regulation of the In Situ Uranium Industry in Texas

In situ mining of U offers various cost and environmental advantages over surface mining but may create several types of environmental impacts if certain precautions and remedies are not followed. Among the most important of these impacts are the effects on groundwater. The state of Texas, through its policy to "conserve and develop natural resources" (stated in the Texas Water Code of 1977), has endowed various state agencies with regulatory authority over industries that develop natural resources. What sets in situ U mining apart from other kinds of in situ mining (for coal and lignite) and all surface mining, however, is that in situ U is regulated by the Texas Department of Water Resources, whereas the Texas Railroad Commission regulates all other in situ and all surface mining. This arrangement of regulatory authority over the mining industry is unique among states.

The Texas Railroad Commission (TRC) was created in 1890 by an amendment to the state constitution and charged with regulating rates and tariffs for a private, profit-making sector of the economy—the railroads. The Railroad Commission was the first regulatory agency in the state and gradually expanded its authority to cover other forms of transportation. In 1917 the Texas legislature declared pipelines to be common carriers like railroads, and thus the regulation of oil and gas transportation also came under TRC jurisdiction. When the state passed the Surface Mining and Reclamation Act in 1975 to control wastes resulting from activities associated with surface mining and the production of oil and gas, the logical agency to which the legislature turned for implementing was the TRC, which already had authority over the transportation aspects of oil and gas. Commercial in situ U mining came into production at this time. A 1977 amendment to the Surface Mining and Reclamation Act proposed to place all the regulation of in situ mining—that for coal, lignite, and U—in the hands of the TRC; as enacted in 1978, however, the amendment included coal and lignite but not U.

In situ mining of U has stood apart from oil, gas, surface mining, and in situ mining for other materials. Because of the Texas Water Quality Board's authority over wells, in situ U operators first came to this agency for permits to operate. In situ U mining has remained under the authority of the state water agency, which since 1977 has been the Texas Department of Water Resources (TDWR). The language of the state's water-quality policy was passed on from the Water Quality Act of 1967, which established the Texas Water Quality Board, to the TDWR in 1977. It reads: "to maintain the quality of water in the state consistent with the public health and the economic development of the state; . . . and to require the use of all reasonable methods to implement this policy." The implications for a mining industry regulated by the TDWR with its goal of maintaining water quality differ from the implications for all other mining industries in Texas, which are regulated by the TRC. The resulting situation has created a difference in the regulations faced by the U industry in Texas. Since the Surface Mining and Reclamation Act, land reclamation has been required for surface mining in Texas, but the mineral and energy-related industries regulated by the TRC do not have to restore groundwater affected by their operations. However, dating from the first permit issued in 1975, restoration guidelines have been included in the permit application, which the TDWR requires of the in situ U-mining companies.

The first permit, issued to Atlantic-Richfield in 1975, handled the subject of aquifer restoration in the following way. Values for the elements being checked were derived from analysis of the production area and water quality in the vicinity of production. These values were then used by the Texas Water Quality Board in light of EPA Interim Drinking Water Standards, the TWQB Hazardous Metals Board Order, and other pertinent data, to determine when adequate restoration had been accomplished. Restoration was to begin within 60 days after U production had ceased. Two conditions determine when the aquifer had been "substantially returned" to its original condition and quality: (a) if the concentrations of all hazardous metals in the baseline determination are less than or equal to those for freshwater, as determined by TWQB Board Order 70-0828-5, and all concentrations are returned to no more than 10 percent above original baseline values as determined above; (b) if the baseline concentrations of one or more heavy metals are significantly (at the discretion of the TWQB) in excess, according to Board Order 70-0828-5 for discharge to inland water, and the original water considered to have marginal potability, the prime parameter used for re-establishing the aquifer will be the total dissolved solids that must be returned to less than or equal to 10 percent above the original baseline concentration for the control grid. This permit as written in 1975 gives no schedule or deadline for when restoration should be completed. Schedules for restoration, suggested by the applicant in the permit application, do not appear until the 1979 permits.

Restoration requirements have evolved with the industry since this 1975 permit. Those for the 1980 permits are as follows: Upon completion of mining, the permittee notifies the TDWR district office and Executive Director and proceeds to re-establish groundwater quality in affected mine-area aquifers to levels consistent with values listed in the Restoration Table for the mine area. Production-zone values in this table are established by using the highest value for each parameter shown in the Baseline Water Quality Table of the permit in columns titled Mine Area Average and Production Area Average. The

overlying aquifer values are taken from the values for each parameter shown in the same table of the column titled Non-Production Zone Average. "In no instance will a Restoration Table value be established lower than the Baseline value nor higher than the Upper Limit value. Restoration values may be modified by the Commission in accordance with TDWR rules . . . by amendment to the permit if other water quality criteria warrant such action." Thus, in essence, the current restoration strategy is return to baseline.

The regulation of in situ mining by TDWR over the first 5 yr of production raises a number of critical issues both in terms of evaluating past performance and setting standards, or goals, for continued regulation in the future. The first and major issue concerns the flexibility of restoration standards. For the industry as a whole there is a question of whether the restoration strategy of return to baseline is too rigid in terms of the quantity of water and energy that will have to be used to attain this quality. From the point of view of the individual companies, the variability of restoration guidelines through time and from one site to another can give unfair advantages to some permit holders. Nevertheless, the state agency believes that restoration standards must be flexible and negotiable, because of the site specificity and evolving technology involved. Industry, too, realizes that the lack of fixed standards on which they can plan is an inevitable consequence of in situ U mining at this time.

A second regulatory issue that has been raised concerns scheduling and duration of restoration activities. As with flexible restoration standards, flexibility in scheduling and duration of restoration programs presents certain advantages to some industries. A longer duration period, for example, may be less costly for the company involved. Natural effects of water movement in the aquifer and the chemistry of heavy metals aid in restoration over time. The fact that the area water is out of use for a longer period is a disadvantage for other users and potential users in the area. However, the fact that less water is used for restoration over a longer period is a plus for other users. For most recent permits, each area is to be restored in a period of 3 to 4 yr. The principal difference among permits lies in the number of areas into which the permit is divided. The longest restoration period that has been proposed calls for restoring 16 areas, at 4 yr each, staggered over a period of 49 yr.

A third regulatory issue concerns coordination of policy for surface and in situ mining. As has been mentioned, the TDWR regulates in situ U mining in Texas, whereas surface U mining is regulated by the TRC. Restoration requirements differ for each. Regardless of the method of mining, the finished product sells on the market for the same price. Without coordination of regulatory policy between the TRC and TDWR, it is possible that companies regulated by one agency are subject to higher-cost restoration programs than are companies regulated by the other agency. Questions raised include: Should all companies mining U in Texas be subject to the same, or a similar range of, restoration costs? If it is in the interest of the state to promote one method of U mining over the other, which method should it promote, and why? Is groundwater protected to the same degree in surface and in situ mining?

A fourth regulatory issue concerns the evaluation of resto-

ration options and their impacts. A study of this issue would provide useful criteria for advocating the restoration method that offers the best long-term protection of the state's water resources. The Ford, Bacon, and Davis Utah, Inc. (1979) report addressed work in this area and could be used as a guide for making an evaluation of the various restoration options.

Restoration Activities in the South Texas Uranium-Mining District

During the early development of the industry most of the companies anticipated use of groundwater sweep with either deep-well disposal or evaporation-pond disposal to achieve restoration. It was expected that target restoration values could be achieved with production of only a few pore volumes from the sites. A review of recent restoration attempts is presented by the U.S Bureau of Mines (1981). Table 7 (page 35) of that report summarizes the results from thirteen tests, eight of them in Texas, with only one of them on a commercial site (for this site, operated by Intercontinental Energy Corporation, restoration is still ongoing). With production of from 0.5 to 15.2 pore volumes, when an ammonium lixiviant was used, the ammonium value remained far above the target value. In some cases the U concentrations approached target values; however, in most, they remained above target values. The data for the ongoing restoration at Intercontinental Energy Corporation's Pawnee property are shown in Table 12.1. Only the ammonium, molybdenum, and conductivity values remain substantially above target values after production of 2 pore volumes. After production of 15.2 pore volumes from a one-pattern field test at Wyoming Mineral Corporations Irigaray property in Wyoming, ammonium remained at 33 mg/L of N (target value less than 1 mg/L), while U (U_3O_8) was reduced to 1 mg/L (target value of 0.098 mg/L). These data show the extreme difficulty of ammonium restoration by groundwater sweep and suggest that heavy metals (particularly U) may require considerably more than a few pore volumes to return to target values.

In terms of the future development of the industry, the question of ammonium restoration is moot because operators have switched to nonammonium lixiviants. However, ammonium has been used at a number of commercial sites in South Texas, and these sites are yet to be restored to the permit's

TABLE 12.1 Data for Ongoing Restoration at Intercontinental Energy Corporation's Pawnee Site, Texas[a]

Parameter	Achieved	Target
NH_3-N	167 mg/L	0.01 mg/L
U_3O_8	2.4 mg/L	2.0 mg/L
TDS	911 mg/L	903 mg/L
Mo^{2+}	2.8 mg/L	1.0 mg/L
Cl^-	107 mg/L	250 mg/L
Ca^{2+}	80 mg/L	200 mg/L
Conductivity	1899 mho/cm	1310 mho/cm

[a]Leaching reagents, NH_3HCO_3 + H_2O_2; area involved, 75 ft × 250 ft; patterns involved, 5; restoration by groundwater sweep with reverse osmosis and spraying; pore volumes used, 2.

required values. The question of ammonium restoration has been researched at The University of Texas at Austin for a number of years. Recently, Charbeneau (1981) suggested the following relationship for the number of pore volumes (PV) required to restore ammonium to a level C_{NH_4} by groundwater sweep in an aquifer with anion concentration (C_A), cation exchange capacity (Q_V), and selectivity coefficient (K) for exchange between ammonium and sodium, which is assumed to be the predominant cation in the formation water:

$$PV = 1 + \frac{KQ_V C_A}{[C_A + (K - 1)C_{NH_4}]^2}.$$

This equation is based on an analysis of mass transport (without dispersion) with the conditions of electroneutrality, filled exchange sites, and a mass-action law formulation of cation exchange. For the South Texas region, C_A varies from 11 to 31 meq/L, Q_V from 3 to 24 meq/100 g (with a porosity of 0.24 and a material density of 2.55 g/cc, these correspond to Q_V values varying from 250 to 2000 meq/L), and from laboratory studies, the value of K is about 3.5 (see Charbeneau *et al.*, 1981). As an example, with C_A = 18 meq/L and Q_V = 650 meq/L, the number of pore volumes required to reduce ammonium to EPA drinking-water standards (10 mg/L of N; 0.71 meq/L) is 106. It should be apparent that in this example the water and energy use for ammonium restoration by groundwater sweep is excessive. The equation above, however, does suggest that water requirements can be greatly curtailed through use of recirculation with the injected fluid constituted with appropriate high ionic strength chemicals. An analysis of this type is presented by Charbeneau (1982); however, continued research is necessary.

Recently, a number of the companies have reviewed their options for groundwater restoration. The general trend is away from groundwater sweep with deep-well disposal toward recirculation with surface treatment of the waste stream before reinjection. This strategy, which involves minimal net water use, will probably become the standard for the industry if natural restoration methods are found to be insufficient. For the future of the industry, restoration research will probably focus on the fate of heavy metals. Research on leaching will continue with a search for an optimal lixiviant that is operationally effective in U recovery, will not affect the hydraulic properties of the host rock, and will not present significant restoration problems.

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