RADIOLOGICAL ASPECTS OF URANIUM-SOLUTION MINING

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(Received March 12, 1982; revised and accepted May 3, 1982)

ABSTRACT


Ore bodies which were once uneconomical to mine by conventional methods such as tunneling or open pits, have become attractive due to the development of non-conventional techniques, involving considerably less capital expenditure. Technologies such as uranium-solution mining or in-situ leaching, have enabled commercial scale mining and milling of relatively small ore pockets of lower grade, and may make a significant contribution to overall U.S. uranium supplies during the next twenty years.

Commercial-size solution-mining production facilities have operated since late 1975. Solution mining involves the pumping of groundwater, fortified with oxidizing and complexing agents into an ore body, leaching and solubilizing the uranium in situ, and then pumping the solutions to the surface where they are fed to a processing plant. Processing involves ion exchange, precipitation, calcining and packaging operations.

This paper presents an overview of the solution-mining process and the health-physics monitoring programs developed over the last four years at several production facilities as a result of the radiological character of these processes. Although many radiological aspects of the process are similar to that of conventional mills, conventional-type tailings as such are not generated. However, liquid and solid waste materials may be generated and impounded. The quantity and radiological character of these by-products are related to facility specifics.

Some special monitoring considerations are presented which are required due to the manner in which radon gas is evolved in the process and the unique aspects of controlling solution flow patterns underground. An overview of the major aspects of the health physics compliance and monitoring programs that have been developed are discussed.

INTRODUCTION

Much interest has developed in alternative, i.e. non-conventional methods of uranium extraction in recent years. Ore grades considerably below the classical economic cutoff of conventional techniques, i.e. underground and open-pit mining, have become very attractive. This is due primarily to the

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0167-5583/82/0000—0000/$02.75 © 1982 Elsevier Scientific Publishing Company
lower capital expenditure requirements and the reduced manpower intensiveness of these non-conventional methods.

Solution mining, or in-situ leaching and by-product recovery techniques (uranium associated with copper and phosphoric acid production) have received considerable attention and financial commitment from some of the major U.S. uranium producers and energy companies. Production facilities generating poundage from these technologies are presently on stream in the U.S. using ore-grade feed to milling processes as low as 4–8 ppm uranium (Wyoming Mineral Corporation, 1977).

The majority of solution-mining activity in the U.S. at present is associated with a large uranium roll front along the South Texas Gulf Coast and in the Great Basins of Wyoming. The common denominator of these geologic settings is that the ore has been deposited by contact with reducing geochemical environments in shallow fluvial sandstone formations, confined by non-porous shale or mudstone layers above and below the uranium-bearing units.

STUDY POPULATION

All of the solution-mining facilities comprising the scope of this paper used alkaline leach chemistries and ammonium bicarbonate as the complexing agent during the period of study. Development and progression of these facilities proceeded in a step-by-step fashion as a general function of wellfield size and corresponding processing plant equipment capacities and flow rates.

Initial circulation of lixiviant for assessing general geochemical characteristics of the ore body usually began with 5–10 wells and a processing flow rate of ≥ 25 gallons (95 l) per minute (known as a "test" plant).

Sometimes an intermediate stage of development was used. Typical process flows were 100 gpm (380 lpm) with several dozen to approximately 100 wells ("R&D" plant).

The final stage of development is the commercial-scale production facility, with process flows in the range of 400–3000 gpm (1520–11,400 lpm) and associated wellfield of several dozen areas ("Production" plant).

TABLE I

<table>
<thead>
<tr>
<th>Study population</th>
<th>Number</th>
<th>Study period (Approximate total operational years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Test</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>R&amp;D</td>
<td>2</td>
<td>3.5</td>
</tr>
<tr>
<td>Production</td>
<td>3</td>
<td>8</td>
</tr>
</tbody>
</table>
Table 1 presents the number and associated operational years of each development stage from which radiological monitoring data has been accrued.

**PROCESS COMPONENTS**

The solution-mining process consists of five major operational components: (1) ore body; (2) wellfield; (3) lixiviant; (4) uranium-recovery process; and (5) waste-impoundment mechanisms.

**Ore body**

At the present, uranium deposits amenable to solution mining are associated with relatively shallow aquifers, about 200–500 ft (60–150 m) subsurface, confined by non-porous shale or mudstone layers. Uranium was transported to the present locations as soluble anionic complexes by the natural movement of oxygenated groundwaters.

![Diagram of uranium deposit](image_url)

**Fig. 1. Redox interface.**

![Diagram of vertical section](image_url)

**Fig. 2. Vertical cross section of typical uranium roll front deposit.**
Uranium deposition occurred in areas where the groundwater conditions changed from oxidizing to reducing. This produced a roll-front deposit with uranium concentrated at the interface between the oxidized and reduced sandstones. This interface is commonly known as the Redox Interface (Fig. 1). A vertical cross section of a typical uranium roll-front deposit showing the basic solution mining approach to in situ uranium recovery is depicted in Fig. 2.

Wellfield

The wellfield provides the means by which leach solution, known as the lixiviant, is circulated through the ore body to recover uranium. Therefore, the wellfield design is crucial in maximizing the effectiveness of lixiviant confinement and utilization. The principal considerations are well spacing, injection/production well orientations and well completion methods. Figure 3 presents well-completion considerations and Fig. 4 illustrates a typical plant/wellfield layout.

Well spacing and orientation is influenced by the hydrologic characteristics of the formation which limit the rate and efficiency of lixiviant circulation. Well-completion techniques contribute to vertical confinement and vertical sweep efficiency of the lixiviant through the mineralized zone. The ultimate number of injection and production wells comprising a wellfield is established by the desired rate of mining and the geohydrologic characteristics of the aquifer, in conjunction with the well patterns.

Fig. 3. Well completion.
The lixiviant reverses the chemical conditions which led to uranium deposition and thus solubilizes uranium as it is circulated through the mineralized formation. It consists of groundwater fortified with an oxidant and an anionic complexing agent. The oxidant converts uranium from the +4 (reduced) to the +6 (oxidized) valence state, making it amenable to complexation and solubilization. Typically, in alkaline leach-solution mining, the lixiviant composition is 0.25–1.0 g l⁻¹ H₂O₂ and 1.0–5.0 g l⁻¹ HCO₃⁻ at a slightly alkaline pH. Gaseous oxygen is also used in lieu of H₂O₂ as the oxidizing agent. An example of the basic mobilization chemistry in situ is presented below. Other lixiviant chemistries are also used involving, e.g., sodium/magnesium carbonates and acid leach methods.

Oxidation: U⁴⁺O₂ + H₂O₂ → U⁶⁺ + H₂O
Leaching: UO₂ + (NH₄)₂CO₃ + H₂O → (NH₄)₂[UO₂(CO₃)] + H₂O₂

Uranium-recovery process (Fig. 5)

The uranium-recovery process consists of four basic process circuits: (1) lixiviant/orpersion circuit; (2) resin transfer circuit; (3) elution/precipitation circuit; and (4) product drying and packaging.

In the lixiviant/orpersion circuit uranium is extracted from the recovered lixiviant by adsorption onto anionic resin beads. The lixiviant is then refortified and reinjected into the mineralized formation. Some provision for
calcium control may also need to be incorporated to reduce calcium-carbonate precipitation in the fortified lixiviant which, if not removed, could plug up the wells and reduce formation permeability. The degree of calcite precipitation is site-specific and is related to the geological formation and lixiviant chemistry chosen. More will be said about this calcite by-product later.

The lixiviant/sorption circuit and the elution/precipitation circuits are interconnected by means of a resin transfer system only. In the elution/precipitation circuit uranium is chemically stripped from the resin and precipitated from solution. The uranium precipitate, ammonium diuranate (ADU) or uranyl peroxide, depending on the precipitation chemistry, may then be conveyed to a product drying/packaging facility where it is calcined to obtain the final UO$_2$ powder. Some facilities take the precipitate through a dewatering step only. The "final product" may therefore be an intermediate slurry rather than a UO$_2$ powder. The barren supernate is refortified and recycled through the elution/precipitation circuit.

Waste-impoundment mechanisms

Various amounts of liquid and/or solid wastes may be generated by these processes. Large volumes of liquid waste may need to be impounded resultant from over recovery in the wellfield and for process chemistry control. Over recovery, i.e. recovery of a 1–5 percent greater volume than is
reinjected into the formation, is necessary to maintain a net inward movement of groundwater into the mineralized zone for solution control in situ. This results in continuous liquid bleeds from process which are impounded at the surface in large holding ponds. These holding evaporation ponds are typically equipped with chlorinated polyethylene liners, depth gauges and underground leak-detection systems. The radiological character of the impounded liquid wastes is site specific but typically includes small amounts of residual uranium and Radium 226 concentrations on the order of 500–3000 pCi l⁻¹ (18–110 Bq l⁻¹). Volumes are controlled via enhanced evaporation techniques. Ultimate deposition of these fluids involves chemical treatment to reduce parameters to near baseline values and reinjection into the aquifer during site restoration and decommissioning activities.

Some of these facilities also generate a solid waste which must be removed from process and similarly impounded at the surface. Depending on site specific formation characteristics and lixiviant chemistries, variable amounts of a calcite (CaCO₃) precipitate may be formed in the process. The degree of precipitation is site specific and related to the local importance of calcium chemistry. As mobilized radium will follow the calcium chemistry in the process, this by-product, if important at a particular site, will invariably contain the majority of mobilized Radium 226 locked up as radium carbonates and sulfates, coprecipitated with the calcium carbonates. These precipitates must be removed from the process to prevent well plugging and reduction in formation permeability. They are typically impounded at the surface in ponds similar to the liquid waste evaporation ponds, in tanks, or may be drummed as produced. Although the radioactivity per gram of this material is typically less than conventional mill tailings, and is almost exclusively uranium and Radium 226, nonetheless the material must be considered as low specific activity (LSA) waste. In the facilities studied, the radiological character of this material typically involved several hundred ppm uranium and 300–3000 pCi g⁻¹ (11–110 Bq g⁻¹) of Radium 226.

**RADIONUCLIDE MOBILIZATION**

A relatively small portion of the uranium daughter products in the ore body are actually mobilized by the lixiviant. The vast majority of secular equilibrium radionuclides remain in the host formation. Table II presents typical expected concentration ranges for the facilities studied in the processing plant feed stream as well as the reforffited tails being returned to the ore body. However, it should be noted that such values are probably process specific and may also be facility age dependent.

It appears that the Thorium 230 will equilibrate and very little is actually removed by the process. The majority of the mobilized Radium 226 (80–90 percent) which is 5–15 percent of the calculated equilibrium radium in the host formation, follows the calcium chemistry in the process and becomes locked up as radium carbonates/sulfates in the calcite slurry bleed stream.
TABLE II

Radionuclide concentrations in process streams (Bq l⁻¹)

<table>
<thead>
<tr>
<th></th>
<th>Th 230</th>
<th>Ra 226</th>
<th>Pb 210</th>
</tr>
</thead>
<tbody>
<tr>
<td>Loaded lixiviant</td>
<td>54–93</td>
<td>11–150</td>
<td>&lt;1.1</td>
</tr>
<tr>
<td>Barelixiviant</td>
<td>48–81</td>
<td>1.9–7.4</td>
<td>&lt;1.1</td>
</tr>
</tbody>
</table>

Variable amounts of calcium carbonate by-products are precipitated as a function of individual ore body geochemistries and process strategies and are bled from the process to a temporary impoundment reservoir. The Radium-226 content of this by-product is typically in the range of 300–3000 pCi g⁻¹ (11–110 Bq g⁻¹) of CaCO₃. After additional processing to remove residual uranium values, the ultimate deposition of this material is determined on a case-by-case basis with the appropriate regulatory authorities. However, the total activity per gram of this material is considerably less than what one would find in conventional mill tailings, as the radioactivity is almost exclusively due to the aforementioned radium and possibly several tens of ppm uranium.

It appears that little, if any, Lead 210 is mobilized as the lead carbonate complexes formed in situ are virtually insoluble in the alkaline lixiviant processes studied.

In addition to the species just discussed, variable amounts of Radon-222 gas are brought up from underground dissolved in the lixiviant. This subject will be treated in some detail in a later section of this paper.

RADIOLOGICAL CONSIDERATIONS

The health-physics controls and monitoring requirements of uranium-solution mining processes are associated with four broad categories:

1. Hydrologic/groundwater controls and monitoring of solution flow patterns underground.
2. Environmental impact monitoring—natural uranium decay series.
3. Waste management and control programs—including bleed solutions, calcite slurries and potentially contaminated equipment.

The following is a detailed explanation of each category.

HYDROLOGIC/GROUNDWATER CONSIDERATIONS

The geologic siting of the ore body, i.e., relatively permeable sandstone confined by shale or mudstone and the subsequent control of groundwater
movement are basic to the mining operation and to environmental control. Injection and recovery well-flow rates must be balanced so that chemical solutions do not migrate out of the mining zone.

As has been previously discussed, the radiological character of the solution returning from underground is composed of low-level concentrations of radioactive species. The average uranium concentration itself is typically less than 0.05 weight percent. Environmentally speaking, the need to control migration is as much a chemical control need as a radiological one.

To detect solutions that have potentially migrated out of the mining zone, monitor wells must be placed in the geologic unit being mined and in other geologic units that must be protected. These wells, encircling the wellfield within a few hundred yards (meters) of the mining unit, are used to detect potential migrations from the wellfield so that expedient detection and corrective actions can be taken to draw back the lixiviant into the mining unit. Additionally, trend wells are often drilled into the mining zone between the monitor wells and the mining unit. Trend wells are used to monitor solutions and detect potential migrations sooner. Should upper and/or lower aquifers exist that need to be protected, monitor wells are drilled into the first aquifer above and/or below the mining unit. Figure 6 depicts a monitor-well control grid of a typical production unit.

Monitor wells are typically sampled on a biweekly basis, but daily if

![Monitor-well grid pattern]

Fig. 6. Monitor-well grid pattern.
potential trends are observed. Examples of typical monitor-well analysis parameters used as indicators include uranium, Radium 226, chloride, ammonia, pH, conductivity/TDS (Total Dissolved Solids) and others.

In general, migrations from the mining zone would be the result of wellfield (hydrologic) imbalance, the presence of previously undetected faults or fractures resulting in undesirable communications between aquifers, or previously unidentified well-completion problems (cracks in casing, incomplete cementing, etc.). Once a possible migration is indicated, mining unit and trend monitors would be used to determine the extent of the migration, if any, by observing water level and chemistry changes.

Under routine operating conditions, a bleed stream, which is taken at the processing plant, is partially conveyed to a water reservoir. This provides for a net inflow of groundwater into the wellfield aiding in hydrologic control.

However, in the event of migration, several corrective action procedures are available to contain and correct migration of leachate. These methods may be applied to a few wells within a single production cell, or to the entire wellfield as the situation dictates. Examples are described as follows:

**Overpumping:** This method involves adjusting pumping so that the rate of flow into the injection wells is exceeded by the flow from the recovery wells. The net result is a general inward movement of native water.

**Reordering:** This is a variation of overpumping in that different ratios are applied to different areas in the wellfield. Hence, the inward movement of native water may be emphasized at one point or another. Reordering may further include direct pumping from one part of the field to another.

**Reducing injection:** This is the second way to adjust the ratio of recovery flow to injection flow. At the same time, it reduces the amount of leach solutions introduced into the production zone in the vicinity of the wells concerned.

**Ceasing pumping:** This method stops both the injection and recovery flows. Exclusive of the effects of natural forces (e.g. natural migration of groundwater, which are orders of magnitude smaller) this should arrest the further migration of leach solution beyond the established boundaries.

**Beginning restoration:** Restoration essentially means returning natural groundwaters to conditions which are acceptable for the long term to the appropriate regulatory agencies. It is usually the last step prior to reclaiming the site although it is an alternative available as a migration-corrective action procedure.

Restoration techniques involve pumping residual fluids from the wellfield and applying conventional water-treatment processes. The fluids are chemically treated on the surface to concentrate contaminants into a small volume brine which can be disposed of as a chemical waste. The clean water may be returned to the aquifer or discharged. The radiological constituents of the brine, primarily Radium 226, can be removed via standard treatment methods (e.g. barium-sulfate precipitation) and disposed of as a very small volume LSA (Low Specific Activity) waste.
URANIUM-SOLUTION MINING AND RADON GAS

A great deal of information has been published over the years on radon gas and its daughter products associated with occupational exposure in underground mines and potential environmental impacts from surface tailings impoundments.

In solution-mining processes, although the characters are the same, the relative roles they play are different. It appears that the majority of radon which is released at the surface is not, as in a conventional situation, a result of "slow" on-surface decay of radium over time. The radon is brought to the surface dynamically, dissolved in the lixiviant returning from underground. Just as dynamically, that portion of the total-dissolved radon which is above the solution’s saturation value, is released when encountering atmospheric pressures and temperatures. The fact that the vast majority of radon released results from dynamic plant operation rather than from surface impoundments has been quantified and verified in Brown and Smith (1980).

Although radon is considered minimally soluble in water at standard conditions, the physical and geochemical environments in situ apparently enhance the amount of gas the fluids can carry by several orders of magnitude. An empirical model has been developed to measure the radon release rate (curies yr⁻¹) and was applied to a commercial in-situ leach facility (Brown and Smith, 1980). Preliminary data for this facility indicated a source term of 300-400 curies yr⁻¹ (1.11 x 10¹⁵ - 1.48 x 10¹⁵ Bq yr⁻¹) at a total recovery flow of 800 gallons min⁻¹ (3040 l min⁻¹). This value is very site-specific and is probably related to ore grade, formation characteristics and other factors. On a curie/lb, U₃O₈ recovered basis, this is approximately 50 percent of the model mill case described in the U.S. NRC GEIS for Uranium Milling (U.S. Nuclear Regulatory Commission, 1980a). Table III presents the relevant measured radon and radium values for various process parameters used in this assessment.

The gas will come out of solution in the early stages of the process as it becomes exposed to the surface environment. Depending on the limitations

<table>
<thead>
<tr>
<th>Stage/location</th>
<th>Radium-226*</th>
<th>Radon-222*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Process solutions (Circulating)</td>
<td>70 → 500</td>
<td>8.3 x 10⁷ → 2.2 x 10⁷</td>
</tr>
<tr>
<td>Calcite in clarifiers</td>
<td>800 → 3000**</td>
<td>n.a.</td>
</tr>
<tr>
<td>Liquid-evaporation ponds:</td>
<td>600 → 900</td>
<td>Equilibrium assumed</td>
</tr>
<tr>
<td>Near surface</td>
<td>900 → 1300</td>
<td>Equilibrium assumed</td>
</tr>
<tr>
<td>Bottom sludge</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Values in pCi/l except ** which is pCi/g CaCO₃; n.a. is not applicable.
of climate on the design of a particular facility, "sources" may be large surface areas, out-of-doors surge ponds, out-of-doors enclosed surge tanks, in-plant surge tanks and absorption columns, or combinations thereof. Monitoring is therefore necessary for both environmental impact assessment, as well as to demonstrate that occupational exposure limits are not exceeded.

ENVIRONMENTAL-IMPACT MONITORING

The monitoring programs required to quantify airborne effluents at a solution-mining facility are not atypical of the environmental requirements for conventional mills and, of course, must be consistent with the requirements of U.S. NRC Regulatory Guide 4.14 (U.S. Nuclear Regulatory Commission, 1980b). Typical considerations in this regard include:

1. Radon within the restricted area — source-term measurements — typical locations would include surge ponds and tanks, in-plant tank local exhaust systems, if applicable.
2. Radionuclide particulates at site-boundary locations, specifically for uranium (natural), Thorium 230, Radium 226 and Lead 210 (if drying/calcining steps are involved), and passive monitoring for Radon 222.
3. Isokinetic type sampling of calciner scrubber exhaust for uranium.
4. Soil and vegetative sampling at site boundary locations for the same species as for air-quality monitoring.
5. Water-quality monitoring of surface waters, if applicable.

IN-PLANT HEALTH-PHYSICS PROGRAMS

Many authors in recent years have described, in detail, the health-physics monitoring and compliance programs associated with conventional uranium mining and milling (International Atomic Energy Agency, 1976; Federal Radiation Council, 1967; Rock et al., 1971; American National Standards Institute, 1973). The radiological program needs of a solution-mining processing plant are very similar and, in many cases, identical to those of a conventional mill, i.e. systems and procedures designed to minimize and accurately assess potential employee exposure to airborne oxide dusts and surface contamination, and include: (1) airborne uranium monitoring in appropriate process areas, primarily calcining and packaging areas including combinations of grab sampling, breathing zone sampling and continuous monitoring techniques; (2) surface area contamination surveillance and control; (3) respiratory protection programs; (4) bio-assay programs; (5) training and procedural development; (6) internal audit mechanisms; and (7) radon/ radon daughter monitoring.

Frequencies and details of program implementation are, of course, a function of applicable regulations, individual facility license conditions, and necessary good health-physics practices.
External radiation-exposure monitoring is absent from this list. Historically, there has been a minimum of concern with external gamma exposure in uranium milling. Typically, exposure potential of significance is restricted to close proximity to the ore body itself or to final product concentrates for extended periods of time.

In solution mining the former concern is, of course, not applicable and, as is the case with all mills in general, final-product storage areas are by their nature, security/control areas removed from the mainstream flow of personnel. However, at these facilities, Radium 226 build-up may occur in resin columns, sand filters, calcite clarifiers, etc., resulting in potential beta/gamma exposure during the cleaning of these systems.

All seven of these elements would represent the core of a solution-mining plant's programs as well. As described previously, solubilized radon will be brought up dynamically from underground with the uranium-bearing solution. A portion of the dissolved gas may be released within the first few process areas, primarily ion-exchange surge tanks and/or at the tops of the absorption columns themselves. If, as in the case of facilities in the warm Texas Gulf Coast, out-of-doors large surface-area surge ponds can be used, it is possible that very little gas remains available by the time the solution reaches in-plant areas. On the other hand, severe Wyoming winters prohibit exposure of the solutions to the atmosphere out-of-doors and may be piped directly from enclosed wellfield surge tanks to in-plant recovery tanks. Local exhaust systems may be required to remove the gas from in-plant vessels before it becomes an occupational concern. Monitoring is therefore required to evaluate engineering and health-physics needs within general plant areas, and particularly prior to entrance into enclosed tanks which are subject to significant radon daughter build-up.

In any individual facility the level of effort required, both in terms of monitoring frequencies and ventilation needs, must be evaluated on a site-specific basis and can change within a facility as design or operational conditions change. The mechanics of radon solubilization in situ are not yet fully understood, but may involve a complex combination of factors including the geochemical characteristics of the ore body, lithographic texture of the sandstone, depth of, temperature and pressure in the ore zone, as well as the details of the lixiviant chemistry itself.

To bring to a close the subject of occupational exposure, the following is an explanation of the relationship between radon and its daughters at these facilities. Under differing circumstances and conditions, both radon gas itself as well as its daughter products, can be important actors in occupational health-physics programs. A great deal of information exists in the literature which supports the view that the daughters of radon are considerably more serious actors than the parent in contributing to respiratory track dose (Coleman et al., 1956; Holaday et al., 1957; Jacobi, 1964; Allshuler et al., 1964; Schiager and Dahl, 1968). Extensive radon and radon daughter monitoring at these facilities has indicated that severe disequilibrium can be
encountered between radon and its daughters at both ends of the continuum. Situations were observed in which significant concentrations of radon gas existed (range of $10^{-7} - 10^{-4} \mu Ci/\text{ml}^{-1}$) in the absence of any significant levels of radon daughters in general plant areas. Conversely, situations were identified (e.g. within enclosed areas, etc.) in which tens of working-level concentrations of radon daughters were measured in the absence of significant radon-gas concentrations. Ventilation conditions apparently play a large role in determining this relationship. Monitoring considerations, therefore, should include both the measurement of radon daughters (for accurate assessment of potential dose commitments) as well as radon gas (for identification of source-term locations to implement necessary engineering controls, e.g. local exhaust systems).

CONCLUSION

This paper has presented an overview of uranium-solution mining processes and associated major radiological aspects and monitoring considerations. Admittedly, the purpose was to present an overview of those special health-physics considerations dictated by the In Situ Leach technology, as well as to point out similarities with conventional mill programs. It is believed that the technology has some potential environmental advantages. Non-conventional uranium-recovery techniques, such as solution mining, can play a significant role in complimenting worldwide uranium supplies during the next several decades.

REFERENCES


