THE USE OF NATURALLY OCCURRING HELIUM TO ESTIMATE GROUND-WATER VELOCITIES FOR STUDIES OF GEOLOGIC STORAGE OF RADIOACTIVE WASTE

by

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ABSTRACT — In a study of the potential for storing radioactive waste in metamorphic rock at the Savannah River Plant near Aiken, South Carolina, the rate of water movement was determined to be about 0.06 m/yr by analyzing gas dissolved in the water. The gas contained up to 6 percent helium, which originated from the radioactive decay of natural uranium and thorium in the crystalline rock. The residence time of the water in the rock was calculated to be 840,000 years from the quantity of uranium and thorium in the rock, their rates of radioactive decay, and the quantity of helium dissolved in the water. The estimation of ground-water velocities by the helium method is more applicable to the assessment of a geologic site for storage of radioactive waste than are velocities estimated from packer tests, pumping tests, or artificial tracer tests, all of which require extensive time and space extrapolations.

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INTRODUCTION

Any study of the subsurface to demonstrate the safety and feasibility of storing radioactive waste in the geologic environment must include investigations in many subject areas; for example, rock properties, structural geology, hydrogeology, geochemistry, seismicity, and resource development. In all of these investigations, a degree of certainty is required that exceeds that required in most geologic siting studies. Thus, scientists pursuing these investigations must apply many techniques of measurement. Each different method that provides independent but coinciding results contributes to the certainty of the final understanding and conclusions.

The water velocity in the vicinity of a proposed geologic storage site is one, but by no means the only, subject of key interest. The hydrologic studies that are commonly made (Table 1)

* Work done under USDOE Contract No. AT(07-2)-1.
to predict the water velocity suffer because neither the time nor the space scale is comparable to the scale of interest in waste repository considerations. Tests of much longer duration are needed.

Studies of natural processes that have been under way for a very long time can be used to supplement the usual procedures for determining water velocity (Table 1). These studies are almost always of a geochemical nature.

Measurements to determine the age of ground water such as those that use tritium or carbon-14 may be quite helpful. However, because these radioisotopes decrease in amount with time since recharge, there is a limiting age beyond which they are not useful. Unfortunately, this limit is less than the age of interest necessary for containment of actinide elements. The use of an element or isotope whose concentration increased in amount with age would be desirable. Helium produced in situ by the radioactive decay of naturally occurring actinides is such an element. The helium content of water, therefore, provides a method for measuring both the age and flow rate of ground water. The following discussion will elaborate on the application of this method for studying the hydrology of buried crystalline rock at the Savannah River Plant (SRP).

This study was part of a broader study at SRP from 1961 to 1972 to evaluate the safety of storing radioactive waste in metamorphic bedrock. The bedrock is buried beneath 1000 feet
of Coastal Plain sediments at the plant site. This investigation was deferred in 1972 pending a more detailed evaluation of alternative concepts of waste storage and management.

HYDROLOGIC SYSTEMS AT THE SAVANNAH RIVER PLANT

The Savannah River Plant is located on the Atlantic Coastal Plain about 32 km southeast of the outcrop of the crystalline metamorphic rock at the Fall Line, which separates the Piedmont and Coastal Plain physiographic provinces (Figure 1). There are three separate geologic and hydrologic systems (Figure 2) at SRP: the Coastal Plain sediments (Miocene and Eocene sediments on Figure 2) in which water occurs in porous unconsolidated to semiconsolidated sand and clay; the crystalline metamorphic basement rock, consisting of chlorite-hornblende schist and hornblende gneiss with lesser amounts of quartzite where water occurs in small fractures; and a buried Triassic basin, consisting mostly of red consolidated mudstone with some poorly sorted sandstone, where water occurs in the intergranular space but is very restricted in movement due to the extremely low hydraulic conductivity.

The water in the Coastal Plain sediments is separated from the water in the crystalline metamorphic rock by a layer of dense clay or sandy clay, called saprolite. This layer is the residual weathering product of the crystalline rock from the period when the rock was last exposed to the atmosphere before being covered by Cretaceous sediment, perhaps 100 million years
ago. The gross separation of these two systems is shown by the following observations. Water in the Coastal Plain sediment contains about 30 mg/L of dissolved solids as opposed to about 6,000 mg/L for the water in crystalline rock. Pumping tests in the crystalline rock (up to a year in duration) show no indications of leakage from the sediments above. Water in the crystalline rock contain dissolved gases including helium. The presence and composition of the gases indicate that they do not escape to the sediments at a significant rate.

From water injection and removal tests on packed-off sections of rock, Marine (1966) concluded there are two types of fractures in the crystalline rock. The first type consists of minute fractures that pervade the entire rock mass but transmit water extremely slowly. Rock that consists only of this type of fracture is called "virtually impermeable rock." The other type of fracture is restricted to definite zones and has larger openings that transmit water at a faster rate. Rock that includes this type of fracture is called "hydraulically transmissive rock."

The hydraulic conductivity of both types of crystalline rock was discussed by Marine (1967, 1975) and representative values are $1.2 \times 10^{-5}$ meters/day (m/d) for virtually impermeable rock and $3.3 \times 10^{-2}$ m/d for hydraulically transmissive rock.

From the application of hydrologic principles to the geometry of the ground-water system in the crystalline rock, a flow path can be inferred. This flow path is supported by the piezometric
map (Figure 3) constructed from water levels in the wells that penetrate the crystalline rock. This flow path is 51-km long from the recharge area to Well DRB 6 near the center of the plant site.

HELIUM CONTENT OF WATER FROM THE CRYSSTALLINE ROCK

The water from the crystalline rock contains sufficient dissolved gas to effervesce when brought to the surface. This gas contains 91 to 96 percent nitrogen by volume (Table 2). However, the most significant constituent is dissolved helium. From the quantity of helium collected and the uranium and thorium content of the rock, a residence time for the water in the rock can be calculated.

In Table 2, there are wide differences in the amount of gas recovered and the percent helium in the gas between Wells DRB 3, DRB 5, and DRB 6 (Figure 3) on the one hand, and Wells DRB 2 and DRB 7 on the other. Wells DRB 2 and DRB 7 penetrate only virtually impermeable rock, whereas Wells DRB 3, DRB 5, and DRB 6 each penetrate one or more sections of hydraulically transmissive rock. The condition of the rock may affect the concentration of helium in the gas and the amount of gas dissolved in the water; however, these differences may better be explained by the methods of collection of the gas samples, which differed between the two groups of wells.

Wells DRB 3, DRB 5, and DRB 6 were pumped continuously, and a fraction of the water was passed through a water-filled plastic
bottle inverted in a tub of water. As the gas evolved from the water passing through, it displaced the water in the bottle. At a pumping rate of 78 L/m (Well DRB 6), the water that leaves the rock at a depth of 544 m takes about two hours to travel up the well and pass through the sample bottle. Wells DRB 2 and DRB 7 would not yield water continuously at a rate sufficient to pump. These wells were therefore pumped intermittently, drawing the water level down to the pump suction (187.5 m below surface) each time; whereupon the wells were left idle for periods that varied from one to eight days before being pumped again. During these idle periods, the dissolved gas and the helium had ample opportunity to come into equilibrium with their new temperature and pressure conditions. In Well DRB 2, for example, about four days elapsed between the time the water left the rock and the time that it passed through the sample bottle. The different methods and rates of obtaining water from the various wells may explain some of the differences in helium concentration and in the amount of gas recovered. Gas recovery was not considered complete because of the short period of time required for the water to pass through the inverted bottle. In addition, all elements probably did not evolve in the true proportion in which they existed in the dissolved state.

To overcome these difficulties, equipment was developed to collect water samples in a pressurized flask and evolve the gas in the laboratory. The pressure was approximately equivalent to
a depth of 274 m of water. The analyses of two such collections from Well DRB 6 are shown in Table 3. This table also shows the values calculated for Wells DRB 6 and DRB 7 by applying Henry's Law to the earlier data. The agreement of these calculated values for Well DRB 6 with the data from the pressurized sample collection bottles is considered good. The last column of Table 3 contains reference values for the amount of each gas dissolved in pure water in equilibrium with mean sea level air at 20°C.

This knowledge of the amount of helium in the water was used to calculate the minimum residence time for the water in the crystalline rock.

CALCULATION OF RESIDENCE TIME

The helium generation rate from uranium is reported (Levorsen, 1967) as $11.0 \times 10^{-5}$ mm$^3$/g U-yr, and that from thorium as $3.1 \times 10^{-5}$ mm$^3$/g Th-yr) at standard temperature and pressure. For the crystalline rock (Well DRB 6 data), the uranium and thorium contents average 1.5 and 8 ppm, respectively (Proctor and Marine, 1965). The helium generation rate from uranium is $1.5 \times 10^{-6}$ g U/g rock $\times 11 \times 10^{-8}$ cm$^3$ He/(g U-yr), or $16.5 \times 10^{-4}$ cm$^3$ He/(g rock-yr). The helium generation rate from thorium is $8 \times 10^{-6}$ g Th/g rock $\times 3.1 \times 10^{-8}$ cm$^3$ He/(g Th-yr) = $25 \times 10^{-4}$ cm$^3$ He/(g rock-yr). Combining the uranium and thorium data, the helium generation rate is $41 \times 10^{-4}$ cm$^3$/g rock-yr.
Assuming one m³ of crystalline rock with an average fracture and nonfracture porosity of 0.2% [0.08% for effective porosity of hydraulically transmissive rock (Webster and others, 1970); and 0.13% for intergranular porosity from laboratory measurements], there are \(2 \times 10^3\) cm³ of water in one m³ of rock. In Well DRB 6 (see Table 3) there is 0.50 cm³ of helium per liter of water, or \(0.50 \times 10^{-3}\) cm³ helium per cm³ of water, or 1.00 cm³ helium per m³ of rock. The average density of the crystalline rock is reported (Christl, 1964) to be 178 lb/ft³, or about \(2.9 \times 10^6\) g/m³; therefore, the helium accumulated in the crystalline rock is:

\[
\frac{1.00\;\text{cm}^3/\text{m}^3\;\text{rock}}{2.9 \times 10^6\;\text{g/rock/m}^3\;\text{rock}} = 3.45 \times 10^{-7}\;\frac{\text{cm}^3\;\text{He/rock}}{\text{g}\;\text{rock}} \tag{1}
\]

If the accumulated helium is divided by the generation rate for uranium and thorium given above, the result will be the minimum length of time necessary to accumulate the observed quantity of helium,

\[
\frac{3.45 \times 10^{-7}\;\frac{\text{cm}^3\;\text{He}}{\text{g}\;\text{rock}}}{41 \times 10^{-14}\;\frac{\text{cm}^3\;\text{He}}{\text{g}\;\text{rock-yr}}} = 840,000\;\text{yr} \tag{2}
\]

The length of flow path (Figure 3) from near the outcrop area through Well DRB 6 is inferred to be 51 km. Assuming flow along this path constitutes the residence time for the water, the velocity of the water is \(\approx 0.06\;\text{m/yr.}\)
Several assumptions involved in this calculation need explanation:

(1) The water contains 100% of the helium generated. This assumption cannot be true as some helium must reside in the rock where it was first generated. Table 4 shows helium analyses of the rock, and the uranium and thorium content of the same rock samples. A definite correlation exists between the uranium-thorium content and the helium content of the rock specimens. The present-day helium in the rock represents from 3 to 27 vol % of the helium formed in $2.9 \times 10^8$ yr, the potassium-argon age of the rock. The variation in three clay samples is somewhat greater. Assuming 0.2 vol % rock porosity, the amount of helium in water is $\sim 0.34$ nL/g rock at Well DRB 6, which represents about 7-1/2% of the helium in the rock and water. Thus, considering the helium in the water (0.34 nL/g rock), and the helium retained by the rock (4.2 nL/g rock), the total accumulation is 4.54 nL/g rock. Dividing this accumulation by the helium generating rate of $41 \times 10^{-8}$ nL He/g rock per year, 11 million years is required to generate the helium, rather than the 840,000 years shown in Equation 2 which is based on helium in water only.
(2) No significant changes have occurred in the rate of transfer of helium from rock to water during this 11-million-year period. Helium is assumed not to have accumulated in the rock and then been released recently to the water, either because the presence of water is recent or because of some change in the transfer mechanics or coefficients. The top of the crystalline rock is now about 198 m below sea level and the geologic-hydrologic history indicates it has probably been below sea level at least since Eocene time (~50 million years ago). The crystalline rock has probably always been saturated with water since the beginning of the Cretaceous deposition, even before it sank below sea level. However, the extent to which changes in the dissolved solids concentration in the water may alter the mechanics of helium release from the rock are not known.

(3) These computations assume that the helium was generated by the same type of rock as that in the area in which it is found. This is a very reasonable assumption. However, it is possible that a granite intrusion with a higher content of uranium and thorium may have contributed some of the helium. For example, assume the helium-generating rock contains 20 ppm uranium, a very unlikely value as this is the maximum value reported (Clark, 1966) for 242 analyses of silicic igneous rock from all over
the world. There is on the average four times more thorium than uranium in granite rocks (Clark, 1966). Therefore, the assumed helium-generating rock would contain 80 ppm of thorium. These concentrations would give this rock a helium-generating rate of $370 \times 10^{-14}$ cm$^3$ He per g of rock per year, and would still require 90,000 years to generate the observed accumulation of helium in the water.

(4) The calculation also assumes that the volume and density of the rock in which the helium was generated are equal to the volume and density of the rock in which it has accumulated, although these two volumes need not consist of the same kind of rock. If the volume of rock in which the helium was generated were smaller than the volume of accumulation (the volume of rock explored), a longer period of accumulation would be required. If the helium were generated in a large volume of rock and concentrated in the explored volume of rock, then a shorter time of accumulation would be indicated. However, the helium occurs in solution with water and not as a free gas. Laboratory and field studies (Carter and others, 1959) showed that helium is a highly satisfactory tracer of water if there is little opportunity for loss to the atmosphere. This finding would indicate
that helium in solution moves with the water and could not concentrate independently of the water.

(5) The previous calculations presuppose that no helium has been lost from the rock during the past 11 million years. If helium has been lost, then the length of time to accumulate the observed quantity of helium must be correspondingly greater. It would be nearly impossible to observe leakage directly because the high porosity, permeability, and the quantity of water naturally flowing in the Cretaceous sediments above would make the slight amount of helium leaking from the crystalline rock into those sediments undetectable. However, if any significant quantity of helium were leaking out of the rock, the observed quantity of helium could not accumulate.

CONCLUSIONS

During investigation of natural water velocity in geologic material of low permeability, the common field investigational techniques should be supplemented by geochemical and isotope analyses to explore the possibility of using natural water constituents to develop ground-water velocity information. One such analysis involves measuring the concentration of helium to determine ground-water velocities. This method has the advantage that the concentration of helium (the nonradioactive, stable, daughter
product of naturally occurring actinides) increases with increasing age of the water rather than decreasing in concentration as do radioactive isotopes.
REFERENCES


<table>
<thead>
<tr>
<th>Method</th>
<th>Hydraulic Conductivity</th>
<th>Gradient</th>
<th>Porosity</th>
<th>Regional Velocity</th>
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<tr>
<td>Laboratory measurements</td>
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<td></td>
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</tr>
<tr>
<td>Instantaneous injection (slug) tests</td>
<td></td>
<td></td>
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<tr>
<td>Drawdown or recovery tests</td>
<td></td>
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<tr>
<td>Aquifer tests using observation wells</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Two-well tracer test (from velocity measurement)</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Multiwell tracer test under natural gradient (from velocity measurement)</td>
<td></td>
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</tbody>
</table>

* Listed in order of decreasing extrapolatibility and increasing time for collecting test data.
## Table 2

Selected Analyses of Effervesced Gas from Wells in Crystalline Metamorphic Rock

<table>
<thead>
<tr>
<th>Well</th>
<th>Date of Collection</th>
<th>Water Removed, gals</th>
<th>Pumping Conditions at Time of Sampling</th>
<th>Water Conductivity, ccm</th>
<th>Number of Samples Averaged</th>
<th>Gas Evolved through Sampler, ml purgas/liter H₂O</th>
<th>Gas Composition, mol %</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>DBB 2</td>
<td>5/23/63</td>
<td>3300</td>
<td>Pumped at 2.6 gpm for about 1 hour each day for 8 days</td>
<td>4300</td>
<td>-</td>
<td>10.5</td>
<td>95.00 1.27 1.49 3.05 1.10 &lt;0.01 0.01 0.154</td>
<td>SRL</td>
</tr>
<tr>
<td></td>
<td>5/24/63</td>
<td>3500</td>
<td>-</td>
<td>4800</td>
<td>5.3</td>
<td>95.74</td>
<td>1.33 1.41 1.45 4.08 &lt;0.01 0.01 0.074</td>
<td>SRL</td>
</tr>
<tr>
<td>DBB 3</td>
<td>6/29 to 7/3/63</td>
<td>3500</td>
<td>Continuous at 2 gpm for 3 days</td>
<td>5300</td>
<td>7</td>
<td>0.82</td>
<td>94.98 1.32 1.75 1.16 0.77 &lt;0.01 &lt;0.01 0.014</td>
<td>SRL</td>
</tr>
<tr>
<td></td>
<td>5/1/63</td>
<td>229,000</td>
<td>Continuous at 2 gpm for 1 day</td>
<td>-</td>
<td>-</td>
<td>95.6</td>
<td>1.17 1.6 1.7 Trace 0 - -</td>
<td>Be Mines; hydrocarbons, 0.0 mol %; hydrogen sulfide, 0.0 mol %. SRL</td>
</tr>
<tr>
<td>DBB 5</td>
<td>5/7 to 5/10/63</td>
<td>26,000</td>
<td>Continuous at average of 6 gpm for 3 days</td>
<td>4000</td>
<td>6</td>
<td>0.90</td>
<td>95.96 1.45 2.21 0.17 0.18 &lt;0.01 &lt;0.01 0.012</td>
<td>SRL</td>
</tr>
<tr>
<td>DBB 6</td>
<td>5/10/63</td>
<td>840,000</td>
<td>Continuous at 20.5 gpm for 28 days</td>
<td>8800</td>
<td>8</td>
<td>0.60</td>
<td>92.35 1.05 5.28 0.26 0.74 0.04 0.11 0.11</td>
<td>SRL</td>
</tr>
<tr>
<td></td>
<td>1/24/63</td>
<td>1,020,000</td>
<td>Continuous at 20.5 gpm for 3 days</td>
<td>8000</td>
<td>8</td>
<td>0.60</td>
<td>91.00 1.10 6.00 0.30 1.00 0.2 0.4 0.056</td>
<td>SRL</td>
</tr>
<tr>
<td></td>
<td>1/22 to 1/23/63</td>
<td>990,000</td>
<td>Continuous at 20.5 gpm for 2 days</td>
<td>-</td>
<td>-</td>
<td>94.6</td>
<td>1.1 4.2 0.1 0 0 0.1 - -</td>
<td>Be Mines; hydrocarbons, 0.0 mol %; hydrogen sulfide, 0.0 mol %; Be Mines</td>
</tr>
<tr>
<td></td>
<td>1/23 to 1/24/63</td>
<td>1,020,000</td>
<td>Continuous at 20.5 gpm for 3 days</td>
<td>-</td>
<td>-</td>
<td>94.7</td>
<td>1.1 3.7 0.3 0.1 0.1 0.1 0.1 - -</td>
<td>Be Mines</td>
</tr>
<tr>
<td>DBB 7</td>
<td>6/17/63</td>
<td>4500</td>
<td>Pumped at 2.4 gpm for about 1 hour every 3 days over a period of 2 months</td>
<td>2800</td>
<td>14.00</td>
<td>91.32</td>
<td>1.18 0.26 5.87 1.37 &lt;0.01 &lt;0.01 0.036</td>
<td>SRL</td>
</tr>
<tr>
<td>Tuscaloosa Formation</td>
<td>Very Rare</td>
<td>Continuous; about 1000 gpm</td>
<td>Low</td>
<td>0.1</td>
<td>95.20</td>
<td>1.09 &lt;0.01 &lt;0.01 4.14 1.61 &lt;0.01 &lt;0.01</td>
<td>SRL</td>
<td></td>
</tr>
</tbody>
</table>

a. Total quantity of water removed from well from completion of packer tests to time of sampling.
### TABLE 3

**Gases Dissolved in Water Samples from Wells DRB 6 and DRB 7**

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
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<tbody>
<tr>
<td>Helium</td>
<td>0.50</td>
<td>0.57</td>
<td>0.036</td>
<td>0.036</td>
<td>0.58</td>
<td>0.06</td>
<td>0.00004</td>
</tr>
<tr>
<td>Argon</td>
<td>0.40</td>
<td>0.43</td>
<td>0.0066</td>
<td>0.16</td>
<td>0.45</td>
<td>0.64</td>
<td>0.375</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>18.0</td>
<td>20.9</td>
<td>0.546</td>
<td>12.8</td>
<td>13.3</td>
<td>25.6</td>
<td>13.7</td>
</tr>
<tr>
<td>Oxygen</td>
<td>0.50</td>
<td>0.79</td>
<td>0.006</td>
<td>0.19</td>
<td>0.29</td>
<td>0.58</td>
<td>7.14</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>0.071</td>
<td>0.011</td>
<td>0.0018</td>
<td>0.82</td>
<td>0.058</td>
<td>1.9</td>
<td>0.002</td>
</tr>
</tbody>
</table>

---

*a. Data from Handbook of Chemistry and Physics.*
TABLE 4
Thorium, Uranium, and Helium Contents of Selected Core Samples of Crystalline Metamorphic Rock

<table>
<thead>
<tr>
<th>Well</th>
<th>Depth, ft</th>
<th>Th, ppm</th>
<th>U, ppm</th>
<th>He, nl/g Measured</th>
<th>Calculated</th>
<th>Percent of Calculated He Measured</th>
</tr>
</thead>
<tbody>
<tr>
<td>DRB 6</td>
<td>1833.8</td>
<td>5</td>
<td>1</td>
<td>4.2</td>
<td>77</td>
<td>5</td>
</tr>
<tr>
<td>DRB 7</td>
<td>1845</td>
<td>13</td>
<td>2</td>
<td>49.3</td>
<td>181</td>
<td>27</td>
</tr>
<tr>
<td>DRB 8</td>
<td>1293.4-1294.8</td>
<td>13</td>
<td>2</td>
<td>47.0</td>
<td>181</td>
<td>26</td>
</tr>
<tr>
<td>P6R</td>
<td>1634.7-1635.8</td>
<td>19</td>
<td>5</td>
<td>22.7</td>
<td>321</td>
<td>7</td>
</tr>
<tr>
<td>PBR</td>
<td>1041</td>
<td>5</td>
<td>1</td>
<td>1.9</td>
<td>59</td>
<td>3</td>
</tr>
<tr>
<td>DRB 8d</td>
<td>905.19-906.61</td>
<td>13</td>
<td>5</td>
<td>109.0</td>
<td>277</td>
<td>39</td>
</tr>
<tr>
<td>DRB 8d</td>
<td>932.35-934.85</td>
<td>19</td>
<td>2</td>
<td>5.0</td>
<td>235</td>
<td>2</td>
</tr>
<tr>
<td>DRB 8d</td>
<td>964.62-965.52</td>
<td>7</td>
<td>2</td>
<td>31.4</td>
<td>127</td>
<td>25</td>
</tr>
</tbody>
</table>

a. Spark source spectrometry analyses by Lawrence Livermore Laboratories, Livermore, California.
b. Calculated production in $2.9 \times 10^8$ yr using Th and U analyses, and rates of $3.1 \times 10^{-9}$ nl He/(g Th) (yr) and $11 \times 10^{-2}$ nl He/(g U) (yr).
c. Does not include helium in water (<1% of calculated total).
d. Clay and rock materials from the saprolite region.
FIGURE 1. Location of the Savannah River Plant and Nearby Geologic Provinces
FIGURE 2. Generalized NW to SE Geologic Profile Across SRP
FIGURE 3. Piezometric Map of Water in Crystalline Metamorphic Rock and Inferred Flow Paths