THE UNDERGROUND WATER LEACHING OF RADIONUCLIDES PRODUCED IN SOIL BY HADRONS CREATED IN HIGH ENERGY PROTON INTERACTIONS

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Summary

Soil samples collected from the NAL construction site were irradiated downstream from Target stations at the Argonne National Laboratory ZGS and Brookhaven National Laboratory AGS. Radiochemical analyses were performed to determine the radioactivities induced in the soil. Batch-type leaching experiments were then conducted to determine the distribution of the induced radionuclides between the soil and water. Of the radionuclides produced in the soil, only 3H, 22Na, 45Ca and 54Mn were detected in the leach waters. The results were used to determine possible contamination of underground reservoirs.

Irradiation

Glacial till removed from the vicinity of the NAL main ring extraction hall (sample 1) at beam level was placed downstream from the L-3 target station of the Argonne National Laboratory ZGS. Also, near surface and highly compacted clay and silt samples from the vicinity of the neutrino facility were exposed near the G-10 internal target of the Brookhaven National Laboratory AGS. The aluminum discs were attached to the soil samples as flux integrators. The hadron fluence was calculated using the 22Na production cross section for 27Al given by Cummin. The flux may be estimated, assuming different energy spectra as given in Table I.

<table>
<thead>
<tr>
<th>Shape Exposure at</th>
<th>AGS</th>
<th>ZGS</th>
</tr>
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<tbody>
<tr>
<td>E-2</td>
<td>2.9 E 7</td>
<td>4.3 E 4</td>
</tr>
<tr>
<td>Alsmiller2</td>
<td>3.2 E 7</td>
<td>4.7 E 4</td>
</tr>
<tr>
<td>E-1</td>
<td>4.2 E 7</td>
<td>6.8 E 4</td>
</tr>
</tbody>
</table>

The irradiation periods were approximately 30 days long.

Soil Activation

The approximate chemical composition of the glacial till is given in reference 3.

The γ activity in the soil was measured using a Ge(Li) γ-ray spectrometer. Other ß and X-ray emitters were isolated by chemical separation and counted in a NaI(Tl) γ-ray or liquid scintillation spectrometer. From the observed activities, the macroscopic cross sections, \( \Sigma n \), where \( n_i \) = number of target atoms/gram of soil and \( \sigma_i \) = excitation function in \( \text{cm}^2 \), were calculated using the Alsmiller spectrum and are listed in Table II as \( \text{cm}^2/\text{g} \) soil.

Earlier, the results for sample 1, had been described in a technical memorandum.

Leaching Experiments

Simulation of the leaching process by the underground water was done by mixing a known volume of NAL deep well water with a measured amount of irradiated soil. After stirring vigorously for one hour, the resulting slurry was centrifuged. The supernate was decanted and passed through a 0.45 μ Millipore filter. The gamma activity in the water was measured with a NaI(Tl) crystal. Tritium and other ß emitters were detected with a liquid scintillation spectrometer.

Results

3H : The leaching experiments indicate that all of the removable 3H exists in the form of water. Thus all of the tritium flows with essentially the same velocity as the ground waters.

22Na : Approximately 10-20% of the 22Na produced in the soil is immediately dissolved in the water. Additional leaches of the same sample indicate that only 2-4% of the activity remaining after the previous leach was removed by successive passes of water.

The leach waters were then processed with samples of non-irradiated soil to determine if the \(^{22}\text{Na}\) would re-exchange into the soil outside of the irradiation zone.

The average velocity of ion migration is less than that of the water because the \(^{22}\text{Na}\) ions subsequently undergo exchange processes passing through non-activated soil. Quantitatively the average velocity of the \(^{22}\text{Na}\) ions is \(1/2\) the velocity of the water. This is calculated from the porosity of the soil, the leachable fraction after the first wash, etc.

\(^{54}\text{Mn}\) : Less than 1% of the \(^{54}\text{Mn}\) produced in the soil was removed by the leach waters.

\(^{46}\text{Ca}\) : Approximately 5-10% of the \(^{46}\text{Ca}\) produced in the soil was removed by the leach waters.

**The Batch Process**

By a batch process we have measured the leachable fraction of the radionuclides due to high energy hadron spallation of typical NAL soils. This process is expected to force the soil to yield not less activity than it would normally yield to the slow and gentle flow of underground water.

**Conclusions**

The safe upper limits to fractions of radionuclides produced in NAL soils that may be carried away by underground waters have been measured. These fractions can be used in the design of shielding and of underground water flow controls to prevent objectionable contamination of domestic waters outside the NAL site.

**References**