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A GAMMA-PHOTONEUTRON METHOD FOR LABORATORY STUDIES OF SOIL WATER

J. C. Corey, R. H. Hawkins, and R. F. Overman

Savannah River Laboratory
E. I. du Pont de Nemours and Company
Aiken, South Carolina 29801

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DISCLAIMER

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SOIL BRIEF

The gamma-photoneutron method measures heavy water content of laboratory soil columns nondestructively. Measurements are not affected by soil density, soil texture, light water content, or type of clay. Infiltration and displacement of solutions are readily followed with the method.

J. C. Corey, R. H. Hawkins, and R. F. Overman, Savannah River Laboratory, E. I. du Pont de Nemours and Company, Aiken, South Carolina 29801.

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ABSTRACT

The gamma-photoneutron method is unique among nuclear methods used to study water movement in soil because the heavy water (^2H_2O) content of laboratory soil columns can be measured independently of bulk density of soil, soil texture, light water (^1H_2O) content, and type of clay mineral. The method requires a radioactive source that emits gamma photons in excess of 2.23 Mev and a neutron detector. The neutron detector measures photoneutrons produced when the deuterium nuclei in the gamma radiation field split into a photoneutron and a proton. The number of photoneutrons detected is linearly related to the heavy water content of the soil column.

Additional Key Words for Indexing: water content measurements, nuclear methods, ^{208}Tl, deuterium, heavy water.

A GAMMA-PHOTONEUTRON METHOD FOR LABORATORY STUDIES OF SOIL WATER[1]

J. C. Corey, R. H. Hawkins, and R. F. Overman[2]

Gamma transmission is usually used to measure nondestructively the water content changes in laboratory soil columns. The primary limitation of this approach is its sensitivity to soil density changes during experimentation that appear as water content changes to the experimenter. To overcome this limitation combined transmission measurements of gamma beams from ^{137}Cs and ^{241}Am have been used. The gamma-photoneutron method reported herein is proposed as an alternative to the dual gamma method to overcome the influence of soil density changes on water content measurements. The proposed technique is more versatile than the dual gamma method because the interface between displacing and displaced solutions along a soil column can be measured easily and rapidly, and water content of soil columns can be measured independently of soil density.

The gamma-photoneutron method is based on the interaction of gamma rays having an energy greater than 2.23 Mev (3) with deuterium present as heavy water (^2H_2O). This interaction produces photoneutrons and protons. The production of photoneutrons by gamma bombardment of deuterium has long been recognized, but has only recently been applied to soil water studies. Haskell and Hawkins (4)

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[2]Research Physicist and Research Chemists, respectively.

proposed the use of photoneutrons from heavy water to qualitatively follow water movement through soil under field situations. Hawkins and Horton (5) used the method to evaluate the reliability of a bentonite layer to stop percolating water. The production of photoneutrons has also been used to determine the deuterium content (2) of water enriched in deuterium, and has been proposed for use (6) as a neutron source of limited energy in reactor-associated research.

This study investigates the suitability of the gamma-photoneutron method for quantitative laboratory studies on soil water.

METHOD

Source Preparation

Very few radionuclides (6) with the required high energy gamma radiation have a sufficient half-life to be practical as gamma sources for neutron production from deuterium. In this study, the gamma source was radiothallium, ^{208}Tl, a member of the decay chain of radiothorium, ^{228}Th. ^{208}Tl reaches secular equilibrium with ^{228}Th approximately three weeks after pure ^{228}Th is separated from other radionuclides (8). The ^{228}Th used for the source of ^{208}Tl was purchased from the Radiochemical Centre, Amersham, Buckinghamshire, England as a crystalline nitrate with a quoted specific activity of 100 curies/g. The neutron background of this crystalline source was quite high [5.8×10^4 neutrons/(sec)(curie)] due to (α,n) reactions with elements having atomic numbers less than 45. Electrodepositing the ^{228}Th from a molten cesium chloride-lanthanum chloride mixture into a liquid bismuth cathode produced matrix elements with atomic numbers greater than 50 and reduced the neutron background to 3.3×10^3 neutrons/(sec)(curie). Reducing

the neutron background of the system reduced the counting errors in the detection of photoneutrons.

Experimental Apparatus

The 1.4-curie electrodeposited ^{208}Tl source (4.3 curies of ^{228}Th) was encapsulated in a stainless steel cylinder 3.0 cm in diameter and 3.5 cm long. This source was placed at the end of a lead collimator (Fig. 1) by a remote handling system and then surrounded with 25 cm of lead for personnel protection, inasmuch as radiation from the encapsulated source exceeded 60 r/h. The collimator had a slit 0.6 cm high and 5.0 cm wide. A hole 5.7 cm in diameter and perpendicular to the slit was cut through the collimator 9.43 cm from the source. Soil-filled glass tubes 5.7-cm in outside diameter and 5.28-cm in inside diameter were positioned in the hole in reproducible geometry. Surrounding the soil column was a paraffin moderator containing two ^{10}B-lined thermal neutron detector tubes. The moderator thermalized (13) the fast neutrons produced by the interaction of the high energy gamma photons with deuterium nuclei. The thermal neutron detector tubes were operated at 700 volts using a stable high voltage supply. Other necessary electronic components were a linear amplifier, single channel pulse height analyzer, scaler, and precision timer.

Theoretical

A number of interactions between soil, heavy water, and radiation occur in the gamma-photoneutron method. Gamma photons are attenuated by soil and water, gamma photons interact with deuterium nuclei producing photoneutrons, and these neutrons are attenuated by soil and water. The following calculations illustrate the influence of these processes on the feasibility of the method and indicate why a high flux of gamma photons is necessary.

The attenuation of gamma photons by wet soil is determined using the following equation (10):

$$\frac{I}{I_o} = \exp - (\rho_s \mu_s + \rho_w \mu_w)x \qquad [1]$$

where I = beam intensity after passage through soil (counts/min)

I_o = unattenuated beam intensity (counts/min)

ρ_s = density of soil (g/cm^3)

μ_s = mass attenuation coefficient of soil for gamma photons (cm^2/g)

ρ_w = water content (g/cm^3)

μ_w = mass attenuation coefficient of water for gamma photons (cm^2/g), and

x = thickness of sample (cm)

The mass attenuation coefficient of water for a 2.62 Mev gamma photon is 0.042 cm^2/g and of soil is 0.038 cm^2/g (7). The gamma photons undergoing greatest attenuation are those passing through the diameter of a cylindrical column (5.28 cm). Therefore, maximum gamma attenuation, using equation [1], for a water content of 0.35 g/cm^3 in a column with a soil density of 1.5 g/cm^3, is

$$\frac{I}{I_o} = \exp - (1.5 \times 0.038 + 0.35 \times 0.042)\ 5.28 = 0.68$$

The attenuation of neutrons produced from deuterium nuclei in the soil column is calculated using

$$\frac{I}{I_o} = \exp - (\rho_s K_s + \rho_w K_w)y \qquad [2]$$

where K_s = mass attenuation coefficient of soil for 0.2 Mev neutrons (cm^2/g)

K_w = mass attenuation coefficient of water for 0.2 Mev neutrons (cm^2/g), and

y = average distance a neutron has to travel to leave the column, approximated by the column radius (cm).

Using $\rho_s = 1.5$ g/cm^3, $\rho_w = 0.35$ g/cm^3, $K_s = 0.0293$ cm^2/g (12), $K_w = 0.3381$ cm^2/g (12), and $y = 2.64$ cm (the column radius), then the attenuation of neutrons produced within the column can be calculated using equation [2] to give

$$\frac{I}{I_o} = \exp - (1.5 \times 0.0293 + 0.35 \times 0.3381)\ 2.64 = 0.65$$

The number of neutrons escaping from the soil tube is approximated by

$$N_E = \sigma G_s n A \qquad [3]$$

where N_E = number of neutrons escaping/min

$\sigma = 1.4 \times 10^{-27}$ cm^2, cross section for the reaction (1,11)

G_s = minimum number of gamma photons reaching the glass cylinder

i.e. $G_s = \dfrac{\text{area of slit}}{\text{surface area of sphere}^3} \times$ number of photons/curie source strength (curies) x attenuation by soil

n = number of deuterium atoms present

i.e. n = volume of cylinder (cm^3) in beam x water content (g/cm^3) x deuterium concentration in water x $\dfrac{\text{moles deuterium/mole of heavy water}}{20 \text{ g/mole}} \times 6.025 \times 10^{23}$ atoms/mole, and

A = attenuation of the neutrons (0.65)

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[3] Radius of sphere = collimator length (9.43 cm) + radius of column (2.85 cm).

The total number of escaping neutrons produced by a 1.4-curie ^{208}Tl source in a soil column 5.28 cm in diameter containing 0.35 g/cm^3 of ^2H_2O is calculated using equation [3] to be 84.11 x 10^4 neutrons/min, as shown below:

$$G_S = \frac{0.6 \times 5}{4\pi(12.28)^2} \times 2.22 \times 10^{12} \times 1.4 \times 0.68 = 33.46 \times 10^8$$

$$N_E = 1.4 \times 10^{-27} \times 33.46 \times 10^8 \times 13.1 \times 0.35 \times 1.0 \times \frac{2}{20} \times 6.025 \times 10^{23} \times 0.65$$

$$= 84.11 \times 10^4 \text{ neutrons/min}$$

The number of neutrons detected depends on geometry and counting efficiency factors. These factors are difficult to predict, but are readily obtained experimentally. When the ^2H_2O content was 0.35 g/cm^3 in an experimental column, the neutron count rate was 3448 counts/min. This observation permits one to calculate that the geometry and counting efficiency combine to reduce the observed count rate to 0.41% [(3448/841,100) x 100] of the calculated number present. Such reductions indicate the necessity for the high gamma flux used in this experiment because 3.1 x 10^{12} gamma photons produced only 3448 counts from the soil column when it contained 0.35 g/cm^3 of ^2H_2O.

Laboratory Measurements

Because a theoretical approach to the number of neutrons produced and counted requires knowledge about the geometry and counting efficiency, an experimental calibration curve for the specific geometry of the experiment was obtained and used rather than a calculated one. Three soils, a montmorillonitic soil from Texas (Houston Black clay) and two kaolinitic soils from South Carolina (Gilead sandy clay and Lakeland loamy sand) differing in both

texture and mineralogy, were wet with varying amounts of ^1H_2O and ^2H_2O and packed to varying densities into glass cylinders 5.28 cm in inside diameter and 15 cm in length (Table 1). The cylinders were placed in the apparatus, and the photoneutrons counted during five minutes were recorded.

RESULTS

The neutron count rate increased linearly as the ^2H_2O content increased in the samples (Fig. 2). The linearity of the data demonstrates that the procedure can determine ^2H_2O content accurately over a wide range of soil densities (1.09 to 1.79 g/cm^3), soil textures, and light water contents (0 to 100% ^1H_2O). The linear equation of best fit is $Y = 4502.8 + 363.96\ X$ with a correlation coefficient of 0.998, where Y is counts per five minutes and X is the volume percent of ^2H_2O.

DISCUSSION

The proposed method for soil-water studies is expensive and can only be justified for special laboratory studies. The radioactive gamma source requires a large initial investment, and heavy water ($28.00 per lb) must be used as one of the fluids in all experiments conducted with the method. No accurate estimate can be made of the cost of a suitable gamma source because such sources are not commercially available at the present time.

The gamma-photoneutron method has two physical limitations. First, the method does not measure the water-filled porosity with respect to the soil, i.e., if the soil is shrinking away from the walls of the container, the method will not reveal the extent of

this contraction; rather the method measures the water-filled porosity with respect to the container. The second limitation is the large counting error at low heavy water contents. The use of more intense sources would reduce this counting error but would create additional shielding and source-handling problems. The use of columns with diameters larger than those used in the current study would also increase the count rate.

The number of candidates for a suitable gamma source in the gamma-photoneutron method is limited because the method depends upon gamma photons with an energy in excess of 2.23 Mev. The radioactive source must also have a reasonable half-life to overcome calibration corrections. Using one year as the minimum half-life for a suitable source, only ^{208}Bi and ^{106}Ru-^{106}Rh are suitable candidates. Unfortunately ^{208}Bi is not readily made in nuclear reactors, and the yield of high energy gamma photons from ^{106}Ru-^{106}Rh is less than 2%.

Short-lived radionuclides which are in secular equilibrium with long-lived members of a decay chain are suitable sources. The most suitable gamma source in this category is ^{208}Tl. ^{208}Tl, a member of the radioactive decay chains beginning with ^{232}Th or ^{232}U (Figure 3), could be obtained using the parent source ^{228}Th, ^{228}Ra, or ^{232}U. ^{228}Th was chosen as the parent source because it was readily available, had a sufficiently high specific activity, and a half-life of 1.91 years. ^{228}Ra would be preferred to ^{228}Th because it has a half-life of 5.7 years, but it is not readily available at the present time. Because of the half-life of ^{228}Ra, a gamma radiation hazard does not develop as rapidly following separation of ^{228}Ra as it does with ^{228}Th. Using ^{228}Ra the source could be readily fabricated during the period of low radiation hazard without using the remote handling facilities. The remote handling required for ^{228}Th adds considerably to the cost. The parent source ^{232}U has a half-life of 72 years, and can be made in nuclear reactors by irradiating ^{231}Pa [^{231}Pa (n,γ) ^{232}Pa (β) ^{232}U].

A burnup of 0.213 g of ^{231}Pa would produce 3.89 curies (0.186 g) of ^{232}U. Any one of the three parents would be a suitable method of obtaining ^{208}Tl, the final choice should be based on economic considerations.

APPLICATIONS

The proposed gamma-photoneutron method is very suitable for fundamental investigations of soil water movement because the measurements are linearly related to the $^{2}H_2O$ content and independent of bulk density, light water content, and soil type. The method is particularly well suited to miscible and immiscible displacement studies. The interface between oil and heavy water, air and heavy water, or water and heavy water at single or multiple locations in a system can be readily measured in one- and two-dimensional model studies. Because the technique is unaffected by differences in soil density, it is superior to the presently used gamma absorption method (10) for infiltration studies with swelling soils where soil density changes are likely. By placing growing plants in deuterated water solutions, plant physiologists might easily measure the conducting volume of plant stems and translocation rates through plants.

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LIST OF TABLES AND FIGURES

Table 1—Calibration Columns

Water content, g/cm³	^2H_2O in water, %	Density of dry soil, g/cm³
Lakeland loamy sand		
0.14	100	1.74
0.09	100	1.70
0.03	100	1.60
0.00	0	1.66
Gilead sandy clay, kaolinitic		
0.35	100	1.73
0.34	75	1.70
0.34	50	1.70
0.34	25	1.69
0.33	0	1.67
0.27	100	1.79
0.15	100	1.51
0.07	100	1.41
0.00	0	1.36
0.00	100	1.42
0.07	100	1.45
0.15	100	1.47
0.27	100	1.79
0.35	100	1.75
0.09	50	1.44
0.15	50	1.49
0.28	50	1.78
0.34	50	1.68
0.36	0	1.78
0.07	100	1.48
0.17	100	1.68
0.22	100	1.49
0.35	100	1.74
Houston black clay, montmorillonitic		
0.38	100	1.28
0.32	100	1.27
0.24	100	1.18
0.24	75	1.18
0.23	50	1.16
0.22	25	1.09
0.25	0	1.24
0.13	100	1.32
0.00	0	1.20

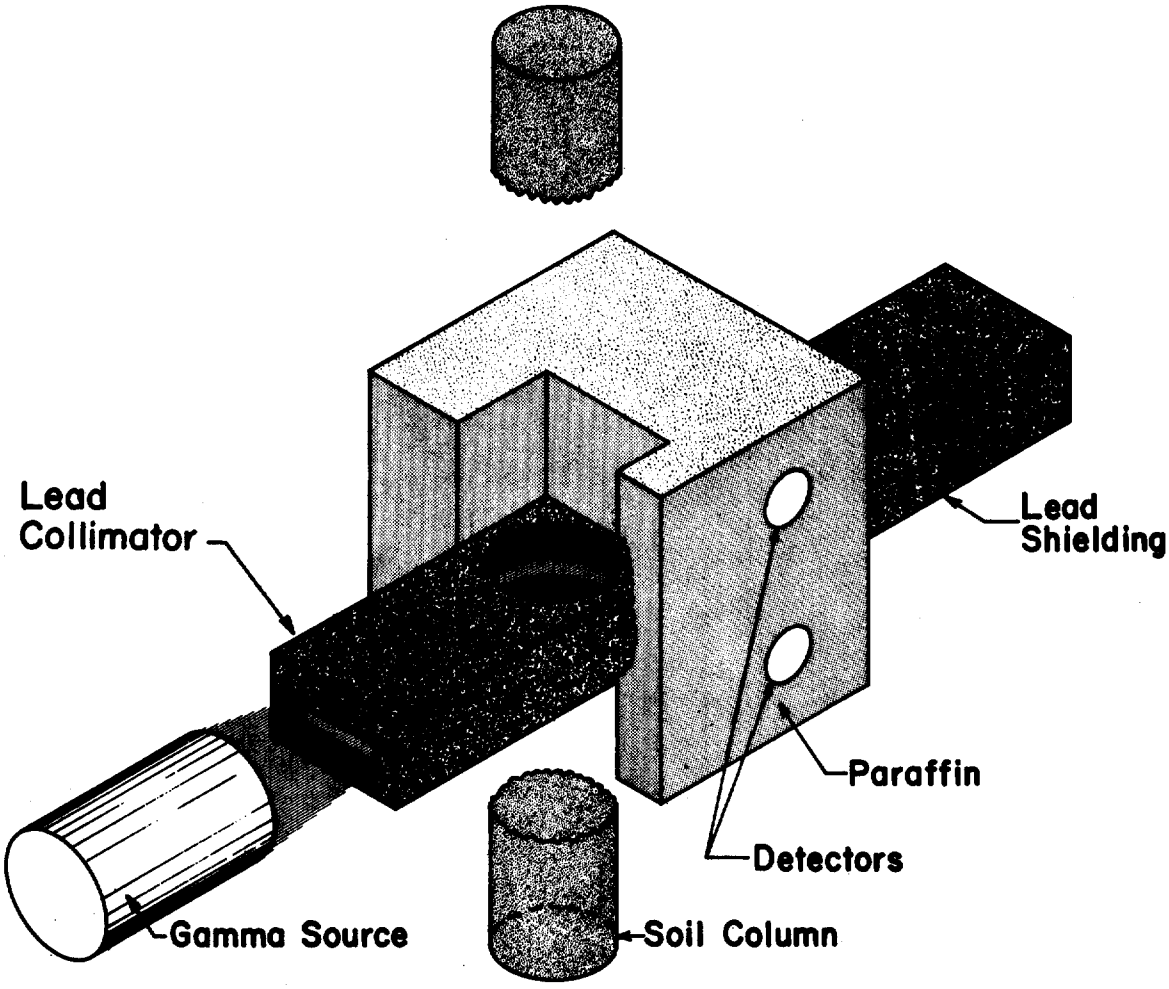

FIG. 1 SCHEMATIC DRAWING OF EXPERIMENTAL APPARATUS USED IN CALIBRATION EXPERIMENT

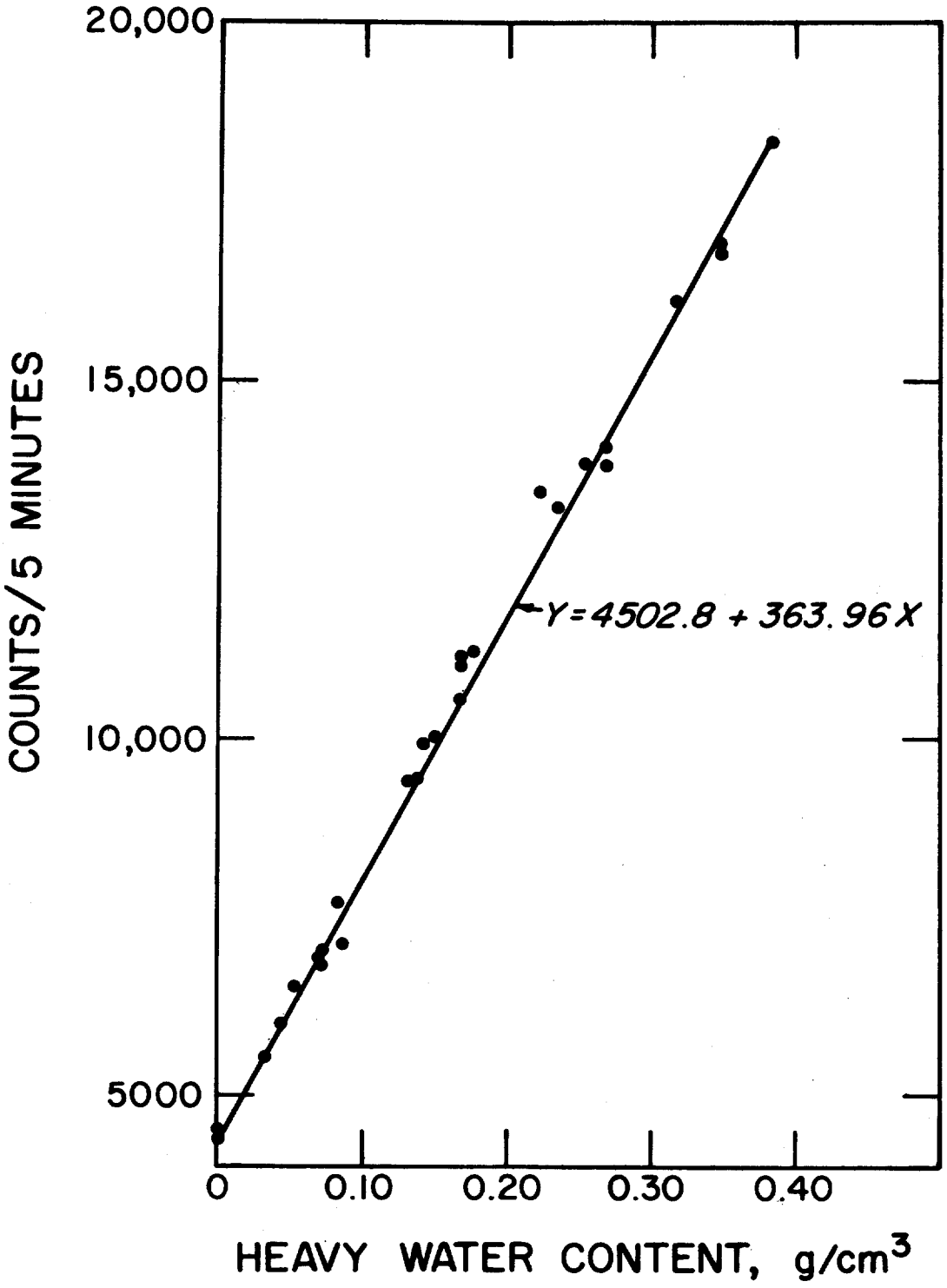

FIG. 2 CALIBRATION CURVE USING THE SOILS AND ^2H_2O WATER CONTENTS LISTED IN TABLE 1

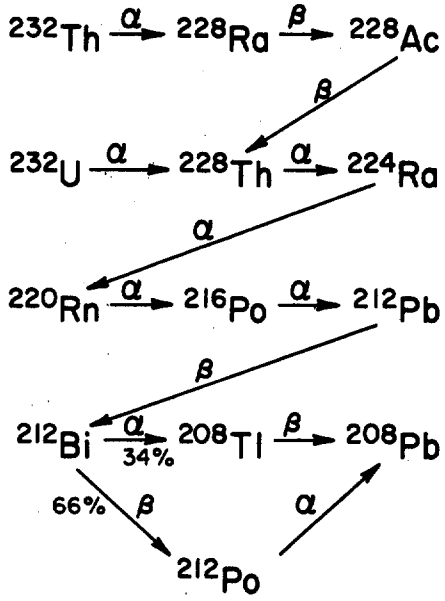

FIG. 3 DECAY CHAIN CONTAINING ^{208}Tl