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WELL LOGGING WITH CALIFORNIUM-252

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### ABSTRACT

Californium-252 is an isotopic neutron source that has only recently become available for experimental well logging. One curie of  $^{252}\text{Cf}$  emits  $4.4 \times 10^9$  neutrons per second by spontaneous fission, 300 times the emission rate of any other one-curie radioisotopic source. Californium-252 has several other advantages as a high yield source for well logging: very small physical size, low gamma and heat emission, and expected low cost relative to other sources. A 50-millicurie  $^{252}\text{Cf}$  source fabricated at the Savannah River Laboratory was made available by the U. S. Atomic Energy Commission to the U. S. Geological Survey for a feasibility study on well logging. The nuclear and physical characteristics of this source and some of the health physics aspects of its use in the field are discussed.

The source was used to make epithermal neutron logs, which are compared with logs made with plutonium-beryllium and americium-beryllium sources in the same well. The high neutron flux available from  $^{252}\text{Cf}$  permitted the use of longer than usual spacing while maintaining a high count rate and excellent sensitivity. In addition, continuous activation logs were made utilizing a spacing of 5.5 feet from the source to detector. Aluminum-28 was identified as the chief radioisotope contributing to the log response. This new technique may provide a log more closely related to clay content than the natural gamma log. Stationary irradiation experiments were also carried out in boreholes, and sodium-24 and manganese-56 were readily produced and identified.

Suggestions for additional research on logging applications and problems resulted from this feasibility study. Potential well logging applications not investigated include the activation of temporary depth markers and the use of stable tracers that can be activated at the site or in the well. The high neutron yield of californium-252 will facilitate in situ activation analysis for many elements as an aid to exploration for oil, water, and other minerals.

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## INTRODUCTION

Californium-252 is an intense neutron emitter which has only recently become available for research in well logging. The Savannah River Laboratory, operated by E. I. du Pont de Nemours and Company for the U. S. Atomic Energy Commission, fabricated the source described in this article. The source was made available to the U. S. G. S. (U. S. Geological Survey) for in situ activation experiments directed toward mineral exploration and well logging. The mineral activation research was directed by F. E. Senftle of the U. S. G. S., Geologic Division, who also provided assistance with well logging. Well logging described in this report was carried out by personnel of the U. S. G. S. Water Resources Division research project on borehole geophysics under the supervision of W. S. Keys. The purpose of this research is to investigate the application of well logging to groundwater hydrology; however, most of the techniques and purposes are quite similar to applications in the petroleum industry.

The source, containing about 50 millicuries of  $^{252}\text{Cf}$ , was made available for a logging feasibility study, which was carried out at the Savannah River Plant in South Carolina. During the time of the study, most of the deep observation wells on the plant contained packers in preparation for a pumping test, so the logging was restricted to two adjacent wells on which only a limited amount of background information was available.

As a result of this feasibility study, the operational characteristics and limitations of new logging equipment were established. Source handling procedures were also tested, and sufficient data were collected to provide a basis for the planning of future logging investigations with  $^{252}\text{Cf}$ .

### Characteristics of Californium-252

Californium-252 is a trivalent actinide with an effective half-life of 2.65 years and emits  $2.34 \times 10^{12}$  neutrons per second per gram ( $4.4 \times 10^6$  neutrons per second per curie) by spontaneous fission. The average neutron energy of the fission spectrum (Figure 1) is 2.3 Mev (million electron volts). Gamma production rate is  $1.3 \times 10^{13}$  photons per second

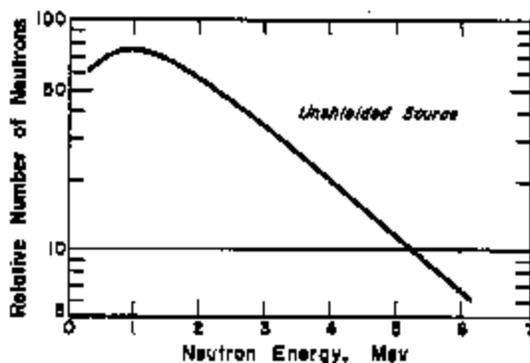


Fig. 1 Neutron energy spectrum of  $^{252}\text{Cf}$ .

per gram, exclusive of internal conversion X-rays. Gamma dose rate is  $1.4 \times 10^2$  R per hour per gram at one meter. The neutron dose rate is  $2.4 \times 10^3$  rem per hour per gram at one meter. The gamma spectrum is shown in Figure 2. Californium-252 emits  $1.9 \times 10^{13}$  alpha disintegrations per second per gram (5.11 Mev per disintegration).

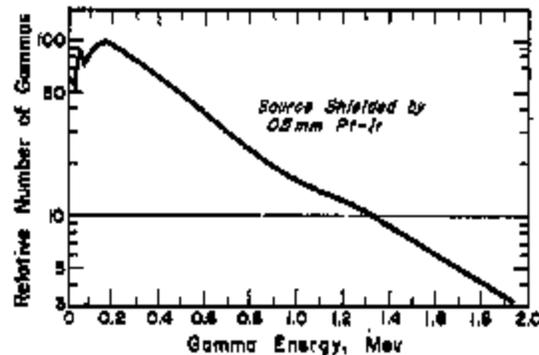


Fig. 2 Gamma energy spectrum of  $^{252}\text{Cf}$ .

Table 1 shows the relative yield and energy of one-curie neutron sources commonly used for well logging (Reinig, 1968):

Table 1

Yield of 1-Ci (curie) Neutron Source

<u>Source</u>	<u>Average neutron energy (Mev)</u>	<u>Yield (n/sec)</u>
$^{239}\text{Pu-Be}$	4.5	$2.0 \times 10^6$
$^{241}\text{Am-Be}$	~4	$2.0 \times 10^6$
$^{226}\text{Ra-Be}$	~3.6	$1.5 \times 10^7$
$^{252}\text{Cf}$	2.3	$4.4 \times 10^6$

The gamma dose rate for a  $^{252}\text{Cf}$  source with a flux of  $5 \times 10^{10}$  n/sec is 2.9 R/hr at 1 meter; a  $^{241}\text{Am-Be}$  source of the same yield has a gamma dose rate of 2.5 R/hr at 1 meter.

Total heat output from the isotope is 39 watts per gram; approximately 50% of this is due to alpha decay. Pressure created in the inner capsule by helium from alpha decay and fission gas buildup in a 50-millicurie  $^{252}\text{Cf}$  source will be 4.8 atm (70.7 psig) at 30°C (at infinite decay).

Source Description and Preparation

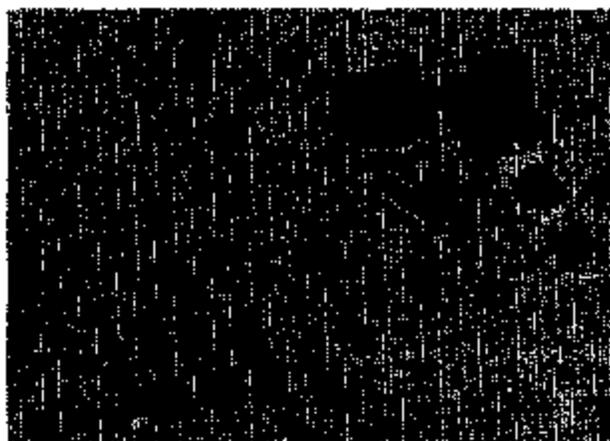
The source used in this study contained 90.1 microgram equivalents of  $^{252}\text{Cf}$  (approximately 50 millicuries) and had a neutron emission rate of  $2.1 \times 10^6$  neutrons per second on the calibration date (April 29, 1968). It is a point source of  $\text{Cf}_2\text{O}_3$  pelletized in a matrix of platinum metal, which is doubly encapsulated in stainless steel.

To purify californium, the tripositive elements were scavenged by ferric hydroxide, and then the actinides were extracted from 12M LiCl - 0.1M HCl to 0.61M tri-*n*-octylamine in diethylbenzene by a laboratory-scale Tramex process. Actinides were then stripped from the extractant with 8M HCl and dried in platinum ware. Organic residues were wet ashed with a mixture of concentrated HNO<sub>3</sub> and H<sub>2</sub>O<sub>2</sub>.

Californium was separated from residual salts by sorption on a column of di-2-ethylhexyl phosphoric acid (HDEHP) on a diatomaceous earth support from 0.10M HNO<sub>3</sub>. Californium was eluted from the HDEHP column with 4M HNO<sub>3</sub>, and the solids-free product was used to prepare the source by coprecipitation of Cf(OH)<sub>3</sub> with Fe(OH)<sub>3</sub>.

The hyperpure californium was carried on a minimum of 0.5 milligram of Fe<sup>3+</sup> as a hydroxide precipitate. After centrifugation and removal of the supernate, the precipitate was transferred to a cone of 1.5-mil-thick platinum which was hammer-welded to a thin (24 Brown and Sharpe gage) Pt-Ir wire to facilitate remote handling in a shielded facility. Figure 3 is an exploded view of the various source components described here.

Fig. 3 Exploded view of source components. Left to right. Pt cone, tube, and pellet on wire handle; inner capsule with pellet and seal plug; outer capsule with sealed inner capsule and seal plug.



After the precipitate was dehydrated under an infrared lamp and heated to 200°C in a furnace, the cone was folded into a thin tube and placed in a pelletizer by drawing the Pt-Ir wire to which the cone was attached through the pelletizer base. The source pellet was then formed by inserting a ram and applying pressure. The source was removed from the pelletizer and slowly heated to red heat (800°C) in a furnace to convert remaining hydrous oxide to Fe<sub>2</sub>O<sub>3</sub> and Cf<sub>2</sub>O<sub>3</sub>. At approximately 75% of theoretical density, the active volume of the source is about six cubic millimeters after pelletizing and heating.

The source pellet, with its wire still attached, was inserted into the primary capsule, which was sealed with silver braze alloy (flow point 855°C). This primary capsule was enclosed in an outer capsule, which was also sealed with silver braze alloy of a slightly lower flow point. The exterior dimensions of the outer capsule were approximately 1/2 x 3/8 inch.

Since this logging study, the source was sealed into a third capsule which was welded closed. Henceforth, primary and outer capsules will be sealed by welding.

After the primary capsule was sealed, and again after the outer capsule was sealed, the source was pressurized in a helium atmosphere at 30 psig for 30 minutes. Leak tests on the individual capsules in a helium leak detector whose lower detection limit is  $2.4 \times 10^{-9}$  cubic centimeters of helium per second indicated no leaks. Dummy sources prepared in the same manner were mounted, sectioned, and subjected to metallographic examination to assure that the method produced well-bonded seals. A dummy source was dropped onto a steel plate from heights of 30 and 66 feet. No significant damage occurred to the dummy source as a result of these tests.

A manganese-bath method was used to determine the rate of neutron emission from this source. Calibration of the system was validated with a  $^{252}\text{Cf}$  source previously calibrated at the National Bureau of Standards. The precision of the determination of the neutron emission rate is estimated to be approximately 5%.

This source was loaned to the U. S. G. S. under the U. S. Atomic Energy Commission's market evaluation program for  $^{252}\text{Cf}$ . If investigations such as this indicate a large future market for  $^{252}\text{Cf}$ , the cost of the radionuclide may be between \$0.50 and \$1.50 per microgram. At this price the estimated cost of a  $^{252}\text{Cf}$  source with a yield of  $5 \times 10^{10}$  n/sec is \$10,000.

#### LOGGING EQUIPMENT

All experiments were carried out with small logging equipment utilizing 2500 feet of 0.10-inch-diameter single conductor cable. This type of cable is used on most water well loggers; therefore, the equipment described can be readily adapted to almost any logger. Considerable equipment development was required to achieve a linear transmission of variable height pulses through several thousand feet of single conductor cable. This amplifier, which employs integrated circuits, will transmit variable height pulses through as much as 6000 feet of single conductor cable, but improvements are needed to achieve the linear pulse transmission required for pulse height analysis. Probes used for conventional neutron and gamma logging transmit shaped pulses of either positive or negative polarity. Surface equipment separates pulses according to polarity so that two radiation logs can be made simultaneously.

The first probe developed for in-hole gamma spectrometry with the  $^{252}\text{Cf}$  was 1-1/2 inches in O.D. and 4 feet long. A small-diameter probe is desirable because 2-inch-I.D. tubing is used in many water resources investigations. The NaI(Tl) crystal in this probe is only 3/4 x 3/4 inch, and has acceptable resolution for gamma peaks below 1 Mev, but the efficiency for detecting higher energy photons drops off rapidly. With this crystal, resolution for the 0.662-Mev cesium-137 peak is 10% FWHM (full width half maximum) through 2500 feet of cable and 11% through 6000 feet of cable. The dc power for both the high voltage supply for the photomultiplier tube and electronics is supplied from the surface. A 3/4 by 3-1/2-inch NaI(Tl) crystal was used for continuous gamma logging. However,

it was not employed for gamma spectrometry because our previous investigations had shown that a more nearly equant crystal provided better photopeak resolution.

Because the 3/4 by 3/4-inch NaI(Tl) crystal was found to have a very low efficiency for detecting high energy gamma photons, a 1-3/4 by 2-inch crystal was substituted during the  $^{252}\text{Cf}$  investigations. A probe 4 inches in diameter and 3 feet long was built and power for the photomultiplier and electronics was provided by separate batteries in the probe. The larger detector has superior efficiency for higher energy radiation and provides spectra comparable to that from laboratory equipment in the 0-3 Mev range needed for these experiments. Resolution for the cesium-137 peak through 2500 feet of cable was 7% (FWHM). Subsequent to the work at Savannah River, a 1-7/8-inch-O.D. probe containing a 1-3/8 by 2-inch crystal and a battery high voltage supply has been completed for use in small diameter holes. It provides adequate resolution of the 2.62-Mev photopeak of  $^{208}\text{Tl}$  in the thorium series.

Standard neutron logs were made with a 1 by 1-1/2-inch lithium iodide crystal enriched in lithium-6 in a 1-1/2-inch-O.D. probe. Energy discrimination was used to reduce sensitivity to gamma rays below 1.5 Mev, and a cadmium shield eliminated most thermal neutrons. Therefore, the sonde primarily detects epithermal neutrons along with some high-energy gamma rays.

The neutron sources were used and stored in 1-1/2-inch-O.D. stainless steel subs with double O-ring seals. The source subs are threaded to mate the bottom end of spacers of various lengths which attach to the sonde. The shortest spacers are made of Mallory 1000 metal for efficient shielding of gamma radiation. Longer stainless steel spacers are filled with water or paraffin for neutron moderation.

All of the downhole equipment used was axially symmetrical. No side collimation, decentralizers, or centralizers were used. It is doubtful if decentralized, side-collimated probes would have greatly altered the results.

The truck-mounted logging equipment and part of the probes were built by Well Reconnaissance Inc. and modified by project personnel. Logs are recorded on a Westronics 2-pen recorder and a Roberts magnetic tape recorder. Two multichannel spectrum analyzers were utilized for data acquisition: a Nuclear Data 128-channel analyzer with Polaroid photographs of scope presentation, and a Technical Measurements Corp. 400-channel analyzer with paper tape printout and X-Y plotter output.

The spectral logging equipment was calibrated with small radioactive sources at the surface. Cesium-137, cobalt-60, and thorite ( $\text{ThO}_2$ ) were used most because they provided peaks throughout the energy range under study. Figure 4 shows a downhole spectrum and an energy calibration with a  $^{60}\text{Co}$  source and detector at the surface. The major equipment problem encountered was gain shift due to temperature effects at the surface and in the hole. Gain shift was also caused by significant changes in count rate. Surface temperatures reached  $100^\circ\text{F}$  in contrast with downhole temperatures averaging  $75^\circ\text{F}$ . Spectral shift due to temperature changes can probably be reduced. However, shift due to drastic changes in radiation intensity

and non-linear energy response are still problems in much laboratory equipment (Heath, 1964). Gain shift in downhole equipment was very rapid until temperature was stabilized. Figure 5 shows two in-hole spectra of sodium-24 made one hour apart.

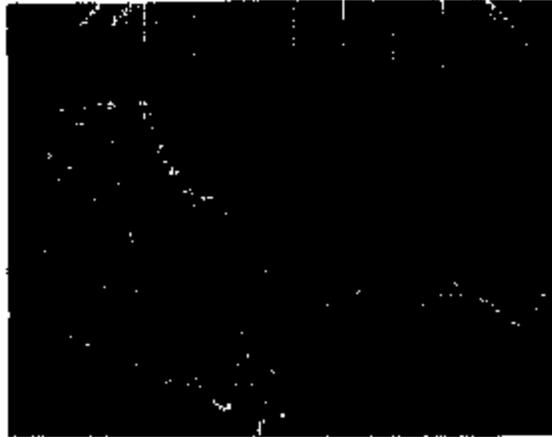


Fig. 4 Sodium-24 spectrum made at a depth of 995 feet in well DRB-3 (upper trace) and cobalt-60 spectrum used for energy calibration. Oscilloscope picture - the ordinate is counts per channel, and the abscissa is gamma energy increasing to the right.



Fig. 5 Two in-hole sodium-24 spectra on the 128-channel analyzer showing drift over one hour. The 1.37-Mev peak has shifted 1 channel due to drift in the probe, and the spectrum has shifted about 3 channels due to instability of surface electronics.

The drift due to downhole instability is one channel. The positioning error due to drift of electronics at the surface is about three channels. In-hole calibration was accomplished by using identified activation peaks; manganese-56 was readily identified and present on most in-hole spectra. The stainless steel spacers near the source became activated and were used

for surface calibration, and it was a simple matter to activate the casing both above and below the water table for in-hole calibration. The oscilloscope presentation of spectra is particularly useful for rapid qualitative examination in the field and for adjustment of gain. Digital printout was only used where quantitative data on peak heights or areas were required.

#### Source Handling and Shielding

A water-filled 55-gallon drum was used as a shield over the drill hole to minimize personnel radiation exposures because there was no prior experience with this intense neutron source. The drum was moved and positioned over wells by a vehicle with a tailgate lift, and logging was done through the shield. In retrospect, the need to perform all operations through the shield is questionable. Tables 2, 3, and 4 summarize the radiation intensities from the bare and shielded source, personnel exposure rates, and cumulative radiation exposure of personnel who handled the source during the logging experiments.

Very little additional exposure would result from lifting the made-up tool out of the shield and placing it directly in the well. Several problems were encountered because the water-filled shield is heavy and cumbersome. A swivel plate covering the hole in the bottom of the drum could not be operated without raising the drum. Therefore, locking chain clamps were occasionally used to hold the spacers and source sub while threaded connections were being made over the well. During one of these operations, a chain clamp released, and the source and spacers dropped into a deep well. After source recovery, the tailgate lift was equipped with a spring-loaded, sliding door that could be operated without moving the drum. Improvements in source handling procedures are necessary before any further logging is done with californium.

Table 2

Radiation Intensities from the 50-mCi <sup>252</sup>Cf Source

<u>Type of Radiation (unshielded)</u>	<u>30 cm</u>	<u>50 cm</u>	<u>1 meter</u>
Fast Neutrons (mrem/hr)	1700	900	230
Gamma (mr/hr)	150	90	15
<u>Type of Radiation (through field shield)</u>	<u>3 inches</u>	<u>6 inches</u>	<u>3 feet</u>
Total Neutrons (mrem/hr)	-	40	8
Gamma (mr/hr)	55	-	-

Table 3

Personnel Exposure Rates Encountered to Install  
50-mCi <sup>252</sup>Cf Source in Shield and During Use

	<u>Neutrons and Gamma</u> <u>(mrem/hr)</u>
Unshielded	
Body	1,025
Hands	22,000
Source in sub (changing tools)	
Body	50
Hands	200
To handle shield (at one foot)	
Body	50

Table 4

Radiation Exposure Accumulated by Personnel  
During this Feasibility Study

<u>Person</u>	<u>Gamma</u> <u>(mr)</u>	<u>Neutrons</u> <u>(mrem)</u>	<u>Total</u> <u>(mrem)</u>
A	55	75	130
B	15	60	75
C	15	50	65

NEUTRON REACTIONS IN WELL LOGGING

In order to relate the experiments described in this report to conventional neutron logging, a short review of the relevant neutron reactions is necessary. The moderation of fast neutrons by elastic scattering from hydrogen nuclei is well understood. It is the main process in conventional neutron porosity logging, which is widely used in the petroleum industry, and in neutron moisture logging, which is common in groundwater investigations. The same process is effective in producing the neutron porosity logs made with <sup>252</sup>Cf.

The various neutron reactions that produce gamma radiation are not widely understood and yet they form the basis for a number of techniques that are potentially useful for identifying elements in situ. Therefore, gamma-producing neutron reactions that might take place in well logging are described briefly in the following paragraphs. For a complete description consult the references by J. D. Owen, 1966 and R. L. Caldwell et al., 1966.

In general, gamma radiation with energies characteristic of the target nucleus are produced by the inelastic scattering of neutrons by nuclei and by the capture of neutrons. The probability that neutron reactions of these types will take place is defined as cross section and is expressed in barns ( $10^{-24}$  cm<sup>2</sup>). Neutron cross sections are a function of the type of reaction, the energy of the impinging neutron, and characteristics of the target nucleus.

The gamma rays or photons produced by neutron reactions are classified here as prompt, capture, and activation. Prompt gammas result from inelastic scattering of fast neutrons with energies above a threshold that is characteristic of the target nucleus. Prompt gamma radiation is only present during neutron irradiation and so is not a factor in the activation experiments at the Savannah River Laboratory. An example of this type of reaction is the 6.09-Mev gamma produced when a neutron above 6.46 Mev is inelastically scattered from an oxygen nucleus.

Neutron capture can occur with either fast or thermal neutrons. However, due to the 2.3-Mev average energy of neutrons from <sup>252</sup>Cf and the hydrogenous environment below the water table, thermal neutrons are probably most important in the Savannah River experiments. Capture gamma rays are emitted immediately after a neutron is incorporated in the nucleus. They die away within a few milliseconds after neutron irradiation is terminated and so were not measured in the experiments described. An example of the use of capture gamma rays in well logging is the identification of chlorine by neutron irradiation.

Neutron activation produces radioactive nuclides from stable isotopes. The emission of activation or delayed gamma rays begins with neutron irradiation and dies off after the neutron flux is terminated as a function of the half-life of the radioisotopes produced. The half-lives of isotopes produced and identified in well logging vary from a few seconds to many hours. Previous well logging investigations utilizing neutron irradiation and gamma spectrometry (Muench and Osoba, 1957, and Baker, 1957) applied only to the measurement of capture gamma rays.

The gamma activity that may be produced by neutron irradiation is related to the neutron flux used and to the nuclear characteristics of the parent and product nuclides by the following expression (Lyon, 1964):

$$A = N\sigma\phi \left( 1 - e^{-\frac{0.693}{T_{1/2}} t_1} \right)$$

where A = induced gamma activity (disintegrations/second)

N = number of target atoms present

$\sigma$  = cross section (cm<sup>2</sup>)

$\phi$  = neutron flux (neutrons/cm<sup>2</sup>/sec)

t<sub>1</sub> = irradiation time

T<sub>1/2</sub> = half-life of product nuclide

e = base of natural logarithms

Saturation is the highest activity that can be produced in a sample with a given neutron flux. When  $t_1 = 5T_{1/2}$ , an activity of 96.8% of saturation will be produced. At saturation activity,  $A = N\sigma\phi$ ,  $N$  can be calculated from the following relations:

$$N = \frac{N_{av} w k}{\text{Atomic weight}}$$

where  $N_{av}$  = Avogadro's number ( $6.02 \times 10^{23}$  atoms/mole)

$w$  = weight of element (grams)

$k$  = fractional isotopic abundance of target isotope

From these relationships it can be seen that if a product nuclide is identified, a quantitative analysis can be made from measurements of the gamma activity present. It is doubtful whether accurate quantitative activation analysis is possible in wells, but these same relations provide a basis for planning qualitative analysis for specific parent nuclides. Numerous tables provide data on gamma energies, half-lives, isotopic abundances, and cross sections in order to help plan activation experiments. Senftle and Hoyte, 1966, calculated counting rates for various nuclides in both a thermal and 14-Mev neutron flux after a 2-minute irradiation and a 5-second delay time (Table 5). With a smaller flux and shorter irradiation time, a sufficient gamma intensity for logging purposes can still be achieved.

Table 5

Counting Rates for Some Common Nuclides Easily Activated  
by Thermal Neutron Capture\*

Parent Nuclide	% Abundance	Daughter Nuclide	Counts per Second per Gram After 2-min. Irradiation	$T_{1/2}$	Energy of Major Gamma Peaks (Mev)
$^{27}\text{Al}$	100	$^{28}\text{Al}$	$2.7 \times 10^4$	2.3 min	1.78
$^{37}\text{Cl}$	24.5	$^{38}\text{Cl}$	$8.1 \times 10^2$	37.5 min	2.16, 1.63
$^{41}\text{K}$	6.88	$^{42}\text{K}$	$1.9 \times 10^2$	12.4 hr	1.53
$^{24}\text{Mg}$	11.2	$^{25}\text{Mg}$	$3.1 \times 10^2$	9.5 min	0.84, 1.02
$^{55}\text{Mn}$	100	$^{56}\text{Mn}$	$1.2 \times 10^4$	2.58 hr	0.84, 1.81, 2.13
$^{23}\text{Na}$	100	$^{24}\text{Na}$	$2.1 \times 10^2$	15.0 hr	1.37, 2.75
$^{109}\text{Ag}$	3.09	$^{110}\text{Ag}$	5.9	2.62 hr	1.26

\* Based on 10% counting efficiency, a flux of  $10^6$  n/cm<sup>2</sup>/sec and a normal nuclide abundance.

(After Senftle and Hoyte, 1966, with additional data from Goldman and Stehn, 1961).

In borehole activation experiments, irradiation and analysis must be planned to favor formation of the desired activation products. Fortunately, both energy and half-life can be used to produce a nearly unique solution to the identification of isotopes. Irradiation times should be selected to create the maximum activity possible in the element sought with the minimum possible activity in interfering elements. If a short half-life nuclide is sought in a matrix of long half-life nuclides, irradiation should be short. If a long half-life isotope is sought where a considerable background of short half-life isotopes exists, long activation is necessary, and spectral analysis should be delayed to allow decay of background radioisotopes.

Fast neutron reactions with silicon-28, silicon-29, and oxygen-16, which are present in large quantities in most rocks, may produce a background which would mask other elements (Senftle and Hoyte, 1966). The relatively low average energy of neutrons emitted by  $^{252}\text{Cf}$  reduces the background from fast neutron reactions.

#### LOGGING EXPERIMENTS

The Savannah River Plant of the U. S. Atomic Energy Commission was chosen for this feasibility study because at the time the U. S. Geological Survey did not have an A.E.C. license for the transportation and use of  $^{252}\text{Cf}$  in well logging. The field experiments described in this report were completed in two weeks during June and July, 1968. The plant is located on the Atlantic Coastal Plain about 20 miles southeast of the Fall Line where crystalline rocks crop out. These crystalline rocks, which are encountered at a depth of 900 to 1000 feet on the plant site, are predominantly schist and gneiss with lesser amounts of quartzite (Marine, 1967). Overlying the crystalline rock is a layer of saprolite which averages 50 feet in thickness and then unconsolidated to semiconsolidated sediments consisting predominantly of sand and clay. Exploratory drilling, logging, and hydraulic testing are being carried out to investigate the feasibility of storing radioactive waste in chambers excavated in crystalline rock. The saprolite effectively separates poor quality water in the crystalline rocks from potable water in the overlying sediments. Permeability of the crystalline rock is a function of the size and abundance of open fractures. Sand beds in the lower part of the sedimentary sequence are the producing aquifers.

Two holes were logged with the  $^{252}\text{Cf}$  source: DRB-3 which is 1088 feet deep and has 6-inch casing to 950 feet with the remainder open hole; and PIA which is 809 feet deep and has 6-inch casing to 795 feet. The casing of both wells was pressure cemented. The only lithologic information available was from driller's logs. Geophysical logs made by a commercial service company include a gamma ray and a neutron log of DRB-3 and a caliper and electric log of PIA.

### Conventional Neutron Logging

The first phase of this study was to compare conventional neutron logs made with other sources with logs made with  $^{252}\text{Cf}$ . Allen et al., 1967, suggest that an improved porosity log can be obtained by the use of longer spacing between source and detector and with sources having a yield of  $5 \times 10^8$  neutrons per second or greater. No porosity data were available for DRB-3 and only three analyses from FIA so it was not possible to test their theory quantitatively. However, the porosity ranges from about 35% in some sediment sections to less than 1% in the bedrock. Figures 6 and 7 suggest that californium does provide a qualitatively superior log under the conditions in hole DRB-3.

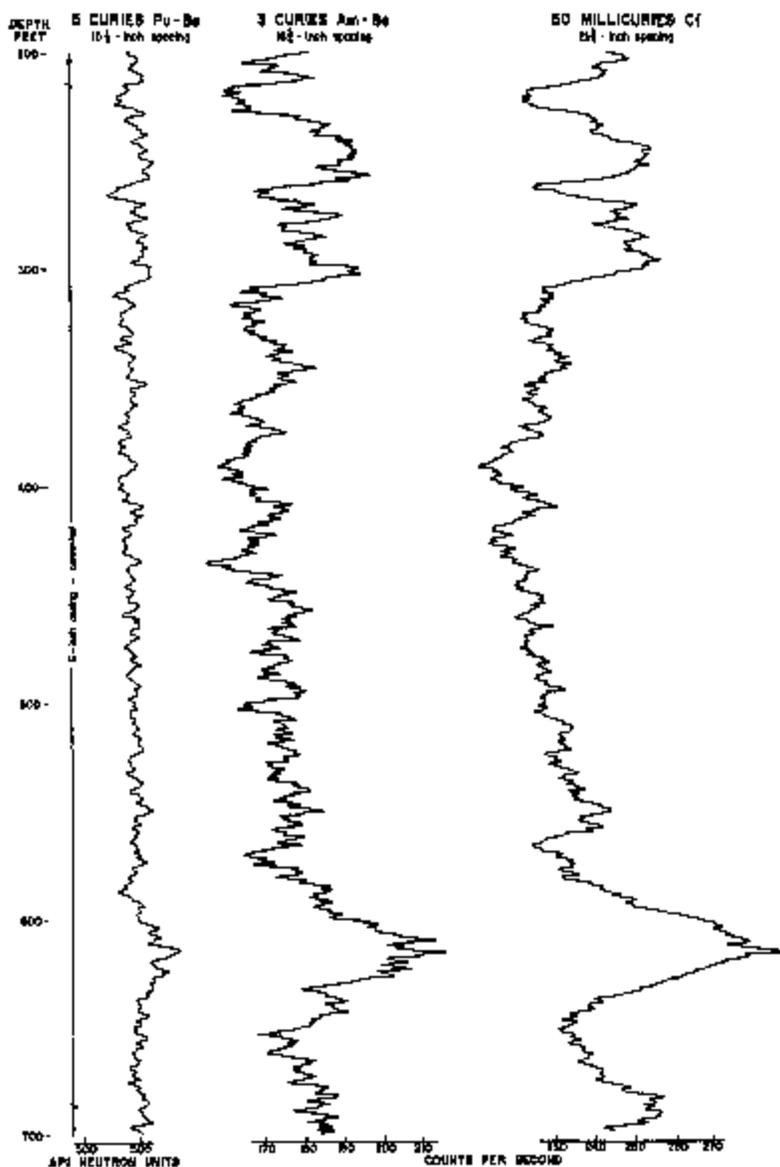


Fig. 6 A comparison of three neutron logs made with different sources in cased section of well DRB-3.

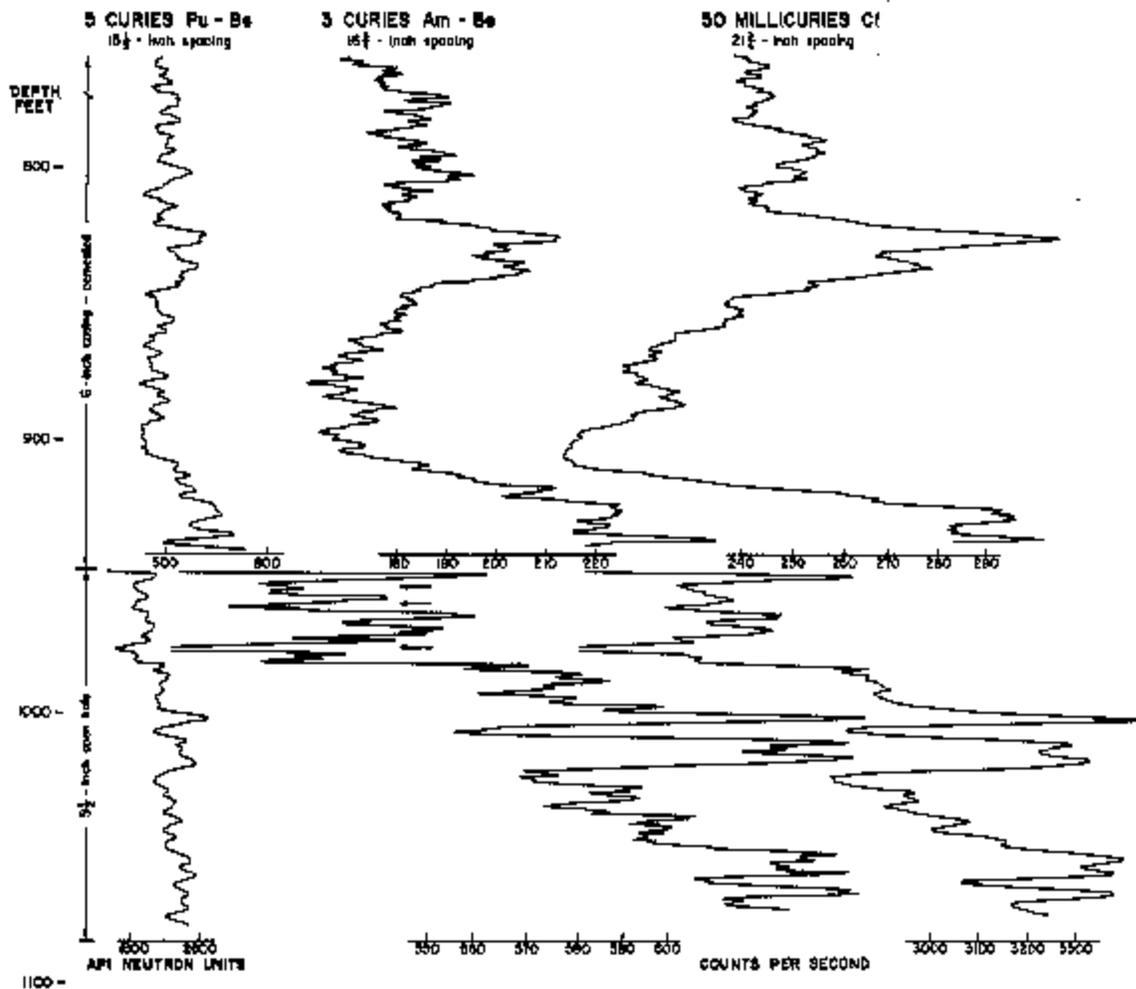


Fig. 7 A comparison of three neutron logs made with different sources in cased and open sections of well DRB-3.

The left log in these figures was made prior to this study, by a commercial service company that used a plutonium-beryllium source. The middle log was made with an americium-beryllium source as part of this study. The greater neutron yield from  $^{252}\text{Cf}$  and 21-3/4-inch spacing produced a log with the same basic character as the other two logs, but with a greater sensitivity to lithology, reduced statistical fluctuations, and lower sensitivity to variations in borehole diameter in the uncased portion where a caliper log was made. Major neutron log deflections due to borehole diameter changes are marked with arrows on the log made with  $^{241}\text{Am-Be}$  in Figure 7.

In the uncased part of the hole, the  $^{252}\text{Cf}$  neutron log was made at 1/10 of the sensitivity of the  $^{241}\text{Am-Be}$  neutron log. A log with the californium source 26-3/4 inches from the detector has deflections similar in amplitude to those shown on the  $^{241}\text{Am-Be}$  log, but the deflections are smaller than those seen on the  $^{252}\text{Cf}$  log where shorter spacing was used. In theory the probe having longer spacing should be more responsive to lithologic properties and less responsive to borehole effects. This appears to be the case with the logs in the present study but more back-

ground information is needed to definitely establish this relationship. In general, as the spacing was increased the log showed fewer deflections due to thin beds or borehole rugosity in the cased part of the hole. Table 6 gives the logging parameters for the three logs shown in Figures 6 and 7.

Table 6

Logging Parameters

Source	Neutrons per Second	Spacing (inches)	Sensitivity		Logging Speed (ft/min)
			Cased Hole	Open Hole	
5 Ci $^{239}\text{Pu-Be}$	$8 \times 10^6$	15-1/2	160 API units/in.	640 API units/in.	60
3 Ci $^{241}\text{Am-Be}$	$8.6 \times 10^6$	16-3/4	10 cps/in.	10 cps/in.	40
50 mCi $^{252}\text{Cf}$	$2.1 \times 10^8$	21-3/4	10 cps/in.	100 cps/in.	40

The count rates measured for the  $^{252}\text{Cf}$  log made with 21-3/4-inch spacing varied from about 200 cps (counts per second) to over 3000 cps, and were only slightly lower for 26-3/4-inch spacing. Allen and others, 1967, state that a count rate of 166 cps is acceptable in high porosity sandstones. The highest apparent porosities shown on the neutron logs of DRB-3 are in the clays, and the lowest are in the unfractured crystalline rocks. Radioactive tracer tests showed that the highest permeability in the bedrock of well DRB-3 was at about 978 feet where a high porosity is shown on the neutron logs (I. W. Marine, Personal Communication).

Continuous Activation Logging

After it was determined that less than one-minute neutron irradiation in the hole produced activation gamma intensities considerably above background, it was decided to attempt activation and logging on a continuous basis, of the gamma radiation produced. The CAL log (continuous activation log) should be clearly distinguished from the conventional neutron-gamma log, because prompt gamma photons from inelastic scattering or capture are an insignificant part of the radiation recorded on the CAL log. A spacing of 5-1/2 feet was used between the  $^{252}\text{Cf}$  source and a 3/4- by 3-1/2-inch NaI(Tl) crystal. When this probe was stationary in the hole or logging up the hole, radiation detected was only slightly above natural background. Therefore, with our equipment CAL logs can only be made with the sonde traveling down the hole. The logging speed and spacing determine the length of irradiation time and radioactive decay prior to counting. No energy discrimination was used for the CAL log. All equipment adjustments were exactly the same as those used to make the low-sensitivity gamma logs shown immediately to the left of the CAL log in Figures 8 and 9. The logging speed was maintained as constant as possible. The average for DRB-3 was about 5 fpm (feet per minute) and for PIA about 5.5 fpm. Rapid fluctuations in logging speed did not occur. The difference in logging speed between PIA and DRB-3 did not noticeably alter the character of the logs.

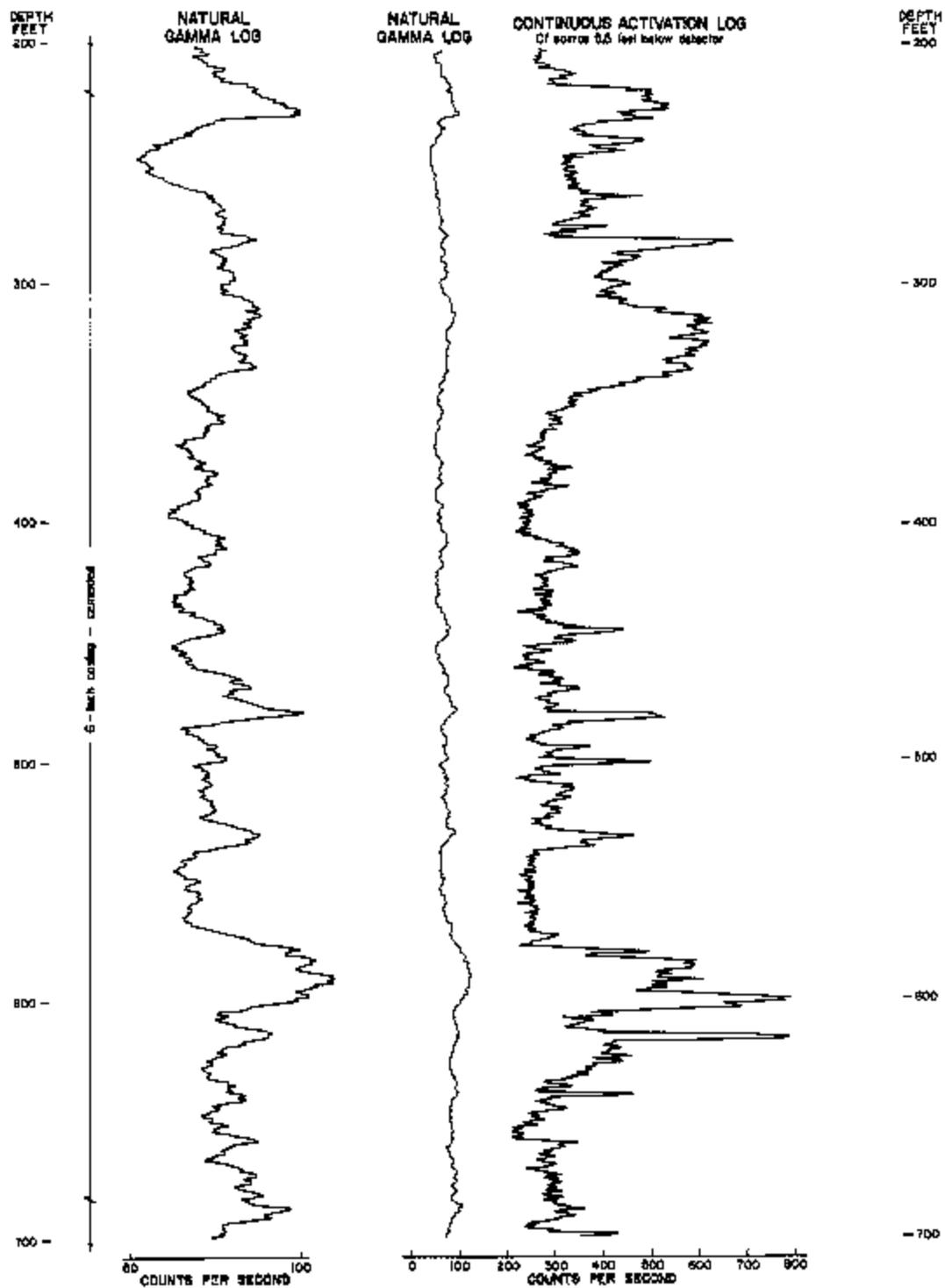


Fig. 8 Natural gamma log and continuous activation log made with the same equipment adjustments.

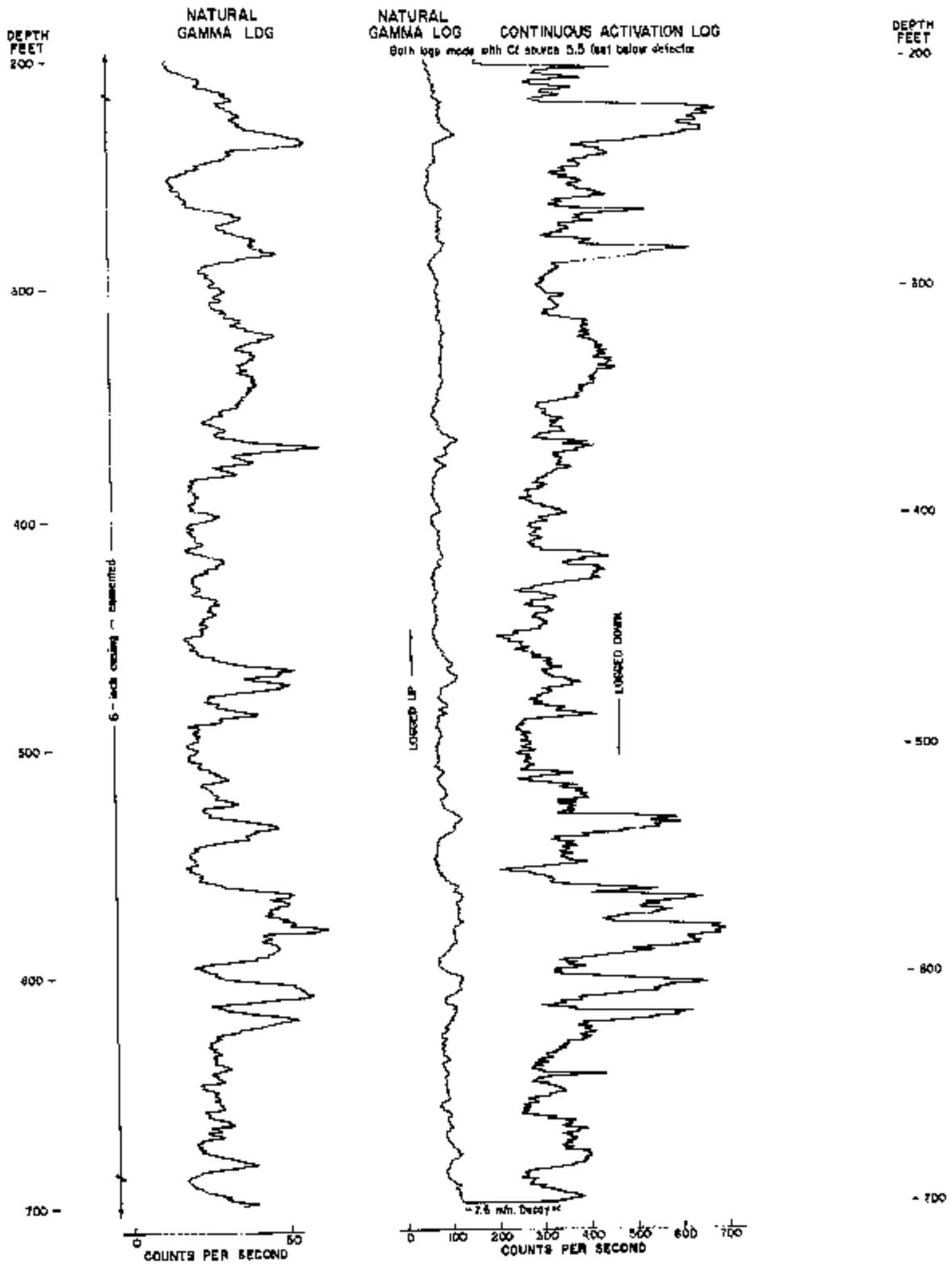


Fig. 9 Continuous activation logged down and natural gamma logged up after 7.5-minute decay. No change in equipment adjustments.

The use of larger  $^{252}\text{Cf}$  sources that will be available in the future will permit a faster logging speed during continuous activation. Speeds of 20 fpm, commonly used for pulsed neutron logging, are probably feasible with a larger source, and shorter spacing or energy discrimination to reduce background. Two detectors at different distances from the source will permit the calculation of half-life or will distinguish between activation products having different half-lives. Two detectors having discriminator windows set for different energy ranges will also provide more diagnostic information.

After the CAL log of PIA was made to a depth of 700 feet, the probe was maintained stationary in the hole for approximately 7-1/2 minutes at which time the radiation intensity was approaching normal background. A log was then made moving up the hole at the normal logging speed of 40 fpm but with no changes in sensitivity or positioning (Figure 9). The low-sensitivity natural gamma log produced with the source following the detector is almost exactly the same as a log made on the same settings without the source attached to the probe. Therefore, very little radiation from the vicinity of the source was reaching the detector. Count rates varied from 200 to 700 cps on the CAL log and from 70 to 130 cps on the gamma log made with the source attached.

For comparison purposes, a natural gamma log made on a more sensitive scale is shown on the left side of Figures 8 and 9. In general, in these wells the CAL log is similar to the natural gamma log, but the sediment beds are more sharply delineated partly because of the slower logging speed. Some thin sediment beds that are not apparent on the natural gamma log stand out as stratigraphic markers on the CAL log. Stratigraphic correlation is somewhat easier with the CAL log because of the delineation of thin beds. Apparently, an element or elements more abundant in the sediment beds with a higher content of radioactive material are activated by an intense neutron flux. Changes in porosity should have some effect on the character of the CAL log; however, no consistent relationship to the conventional neutron logs is obvious.

Because of similarity with the natural gamma log, it was initially believed that potassium-40 or a nuclide in the uranium or thorium decay series was being activated to produce the effects noted on the CAL logs. Field experiments with both the  $^{252}\text{Cf}$  and  $^{241}\text{Am-Be}$  sources showed that the gamma activity of uranium ore and thorite was increased by neutron irradiation, but the specific isotope responsible was not clearly identified. Bismuth-214 peaks did seem to be emphasized after an activation of 10 to 20 minutes. However, subsequent work has shown that most of the apparent  $^{214}\text{Bi}$  activity was due to the activation of aluminum-27 in the samples. Because spontaneous or neutron-induced fission of uranium does occur in nature, fission and the resulting products may be responsible for some of the increased gamma activity (Rankama, 1963).

### Stationary In-Hole Activation

In order to identify the radionuclides contributing to the CAL log, short irradiation experiments were performed at a number of depths in the two wells. Short-term activation was carried out with 5-1/2-foot spacing between the source and the 1-3/4 x 2-inch detector, so that the gamma spectra could be examined immediately after activation. A one-minute activation was found to produce a sharp peak of sufficiently high gamma intensity so that the detector could be accurately located. A one-minute irradiation produced count rates in the vicinity of the major photopeak that averaged 100 cpm per channel in contrast to a background of less than 10 cpm per channel at the same energy (Figure 10).

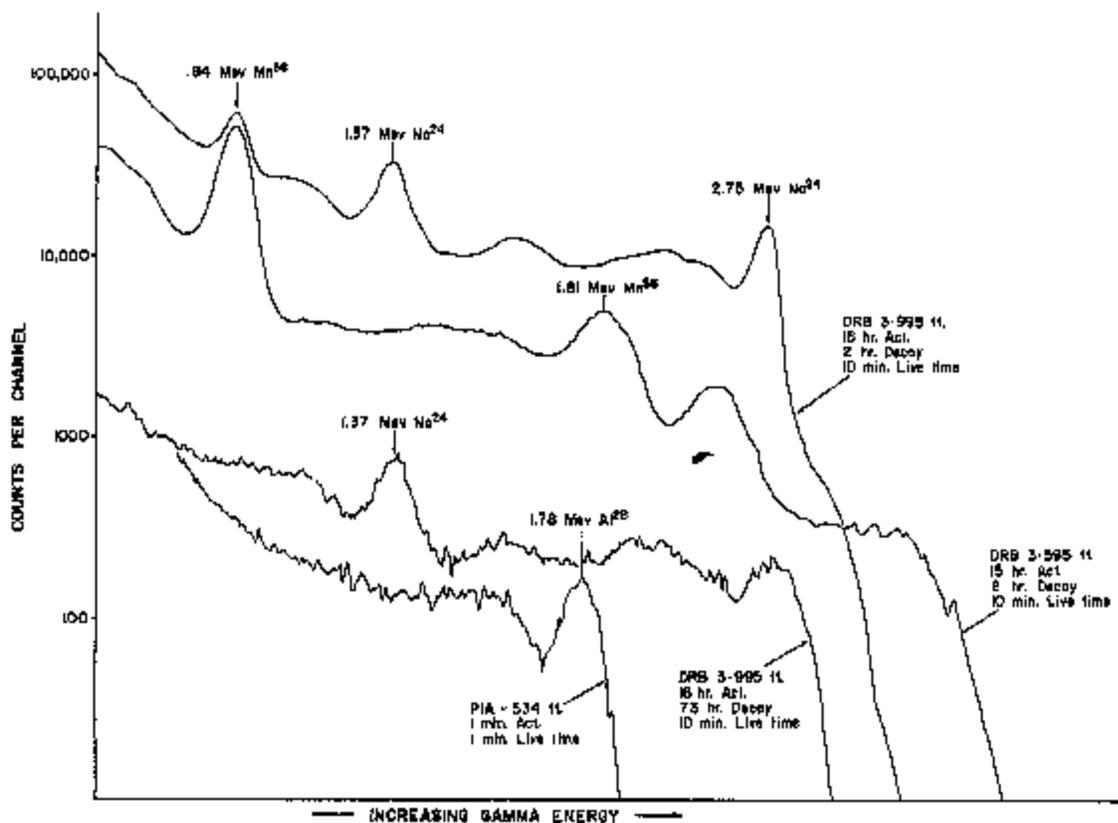


Fig. 10 A comparison of gamma spectra produced at different depths by stationary irradiation with californium-252.

Background was subtracted for  $T_d$  calculations. The depths of points to be activated were selected on the basis of the different radiation intensity shown on the CAL log. In general, the height of the major photopeak in the activation spectrum increased with higher total gamma intensity shown on the CAL log.

Figure 11 is a typical spectrum resulting from a one-minute activation in PIA compared with the natural background at the same point. The major photopeak has an average energy of 1.76 Mev based on several different spectra. At three different depths, several delayed spectra with one-minute live times were printed out with the last one six minutes after irradiation was terminated. Half-lives calculated on the 1.76-Mev photopeak were 2.1, 2.4, and 2.4 minutes. A table of isotopes arranged in ascending order based on their gamma energies (USAEC, 1960) shows that only aluminum-28 has an energy and half-life close to the calculated values. Recent data yielded an energy of 1.78 Mev and a  $T_{1/2}$  of 2.3 minutes.  $^{28}\text{Al}$  is produced by thermal neutron activation (215 mb) from the only stable isotope of aluminum,  $^{27}\text{Al}$ .  $^{28}\text{Al}$  is also derived from  $^{28}\text{Si}$  by fast neutron reaction with a cross section of 250 mb for 14-Mev neutrons. Laboratory experiments with a  $^{241}\text{Am}$  source, which has a higher average neutron energy than  $^{252}\text{Cf}$ , indicate that  $^{27}\text{Al}$  is readily activated and  $^{28}\text{Si}$  is not. Bismuth-214, which is present in the uranium decay series, has a peak at 1.77 Mev but has a half-life of 19.7 minutes.

Irradiation of 10 minutes or more in PIA produced a 0.84-Mev  $^{54}\text{Mn}$  photopeak that was not obvious on the one-minute activation spectra. Irradiation for the longer period did not significantly improve the  $^{28}\text{Al}$  spectra; therefore, one minute was used in order to reduce interference from the 1.81-Mev  $^{54}\text{Mn}$  peak.



Fig. 11 Background (lower spectrum) compared with spectrum produced by one-minute activation in hole PIA at a depth of 252 feet. The major photopeak represents aluminum-28.

The clay minerals are essentially hydrous aluminum silicates. Aluminum oxide is the second most abundant oxide in many clays (Rankama and Sahama, 1950). Clay tends to have a higher aluminum content than the parent igneous rocks. There is a significant increase in the percentage of aluminum and iron oxides and a decrease in quartz content with decreasing grain size from fine sand to clay (Pettijohn, 1949).

It appears that at least part of the similarity of the CAL log and the natural gamma log is due to higher content of both natural radioisotopes and aluminum in the finer-grained sediments. Differences between natural radioactivity and aluminum content can also be determined by comparing logs. Based on these preliminary studies it appears that the CAL log responds basically to the aluminum content of the sediments, and may be the clay log suggested as a possible future technique by Mardock, 1960. A clay and sandy clay unit separating the upper and lower aquifers is clearly defined on the CAL logs. In DRB-3 (Figure 8), the unit is between 580 and 620 feet, and in PIA (Figure 9), between 560 and 620 feet. Lenses of sandy clay in the upper aquifer are much more clearly defined on the CAL log than on either the natural gamma or neutron logs.

At the Savannah River Plant, it was convenient and safe to leave the  $^{252}\text{Cf}$  source positioned in a well overnight in order to investigate the possibility of activating long-lived radioisotopes. The gamma radiation present at the depth of irradiation was localized in a thin zone, had very high intensity, and persisted for several days. One overnight activation peak at a depth of 995 feet in the uncased portion of DRB-3 was measured periodically for three days. Figure 10 compares spectra after 2- and 73-hour decay. Periodic spectra were recorded on the 400 channel analyzer. At an energy of about 1.37 Mev, the background before activation averaged 25 counts per channel; 2 hours after irradiation was terminated, the activity was 35,000 counts per channel at this energy, and three days later, it was still over 800 counts per channel. Several energy calculations on the persistent peak near the center gave 1.38 to 1.39 Mev. Half-life calculations on this peak indicated an average of 13.5 hours; the higher energy peaks gave a  $T_{1/2}$  of 15 and 16 hours. Sodium-24 has major gamma peaks at 1.37 and 2.75 Mev, a  $T_{1/2}$  of 15 hours, and the most likely parent,  $^{23}\text{Na}$ , has a thermal neutron cross section of 530 mb. Two other possible parent nuclides,  $^{24}\text{Mg}$  and  $^{27}\text{Al}$ , have fast neutron capture cross sections of 180 and 120 mb, respectively, measured with neutron energies greater than 14 Mev. The identification of  $^{24}\text{Na}$  is quite positive, but the relative contribution from several possible parent nuclides requires more study. The gamma-ray spectrum catalogue (R. L. Heath, 1964) shows plotted spectra measured in the laboratory for most of the common radioisotopes so that unknowns can be tentatively identified by visual comparison if interfering isotopes are not present.

Figure 10 shows a rather prominent peak at about 0.84 Mev after 2-hour decay in both the cased and open hole. This peak is not present the following day because it has a much shorter  $T_{1/2}$  than  $^{24}\text{Na}$ . Because of the high  $^{24}\text{Na}$  activity it is necessary to subtract background in computing  $T_{1/2}$  for this peak. A half-life of approximately 2.5 hours is then obtained. Manganese-56 has a major peak at 0.84 Mev and a half-life of 2.6 hours. The thermal neutron cross section for the reaction  $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$  is 13 barns. The fast neutron cross section for the following reaction is 150

mb:  $^{55}\text{Fe}(n,p)^{55}\text{Mn}$ . Because the relative amounts of manganese and iron in the crystalline rocks are not known, the parent nuclide cannot be definitely identified. A comparison of activation with and without a thermal neutron shield might provide identification of the parent isotope by changing the dominant neutron energy.

Overnight activation in the cased portion of DRB-3 did not produce an identifiable sodium spectrum even after the  $^{55}\text{Mn}$  peaks from the casing had nearly decayed. The water in the bedrock contains from 518 to 740 ppm sodium, and the water in the sediments averages less than 2 ppm sodium. It is probable that the fluid column in DRB-3 has only minor changes in fluid quality; numerous trips with logging sondes and source recovery operations served to mix the fluid column. Shielding by casing or grout did not affect short-term activation of aluminum. For these reasons, it appears that activation in DRB-3 does indicate the relative sodium content of the formation waters; however, the sodium content of the rocks is not known. Activation of dry samples of the crystalline rocks produced a sodium spectrum of much less intensity than brine activation. No apparent sodium peaks were produced by neutron irradiation of dry sediment samples. In general, the sodium content of the matrix of sedimentary rocks should be relatively low so activation analysis would be indicative of sodium in formation waters. If the approximate sodium content of the rocks can be determined, neutron activation analysis provides a potential means for the semiquantitative determination of sodium chloride in the formation waters of both cased and open holes. In-hole activation analysis can provide a more positive identification of brine than the sometimes ambiguous neutron chloride logs.

#### FURTHER STUDY AND POTENTIAL APPLICATIONS

It is hoped that  $^{252}\text{Cf}$  sources with a yield of at least  $10^8$  n/sec will become available for logging research by private companies as well as government agencies in the near future. As a result of the feasibility study reported here, several recommendations for additional field investigations can be made. Continuous activation experiments should be done at various logging speeds and with source-to-detector spacings selected to emphasize isotopes of interest. Two detectors at different distances from the source may provide a means of distinguishing several short-lived radioisotopes. For this purpose accurately regulated and recorded logging speed is desirable. Experiments with the CAL log should also include the use of a single-channel analyzer in order to eliminate undesired radiation. In conjunction with energy discrimination, the multiscaling capability of most pulse height analyzers can be used to plot the decay of short-lived radioisotopes. A larger source would also be helpful for continuous activation logging. Further investigations of the activation of natural radioisotopes should be made to definitely establish their contribution to the CAL log. Continuous and stationary activation experiments should be performed with and without cadmium and boron shields around the source.

If gain shift problems due to changes in temperature and radiation intensity cannot be completely solved, an internal energy standard should be used. A spectrum stabilizer can then be locked on a preselected peak. Coincidence counting and solid state detectors may ultimately improve the

resolution of in-hole activation analyses. When equipment problems are solved, techniques developed in the laboratory for the analysis of multi-component spectra may permit semiquantitative activation analyses in boreholes. In order to attempt semiquantitative analysis, it will be necessary to utilize correction curves for borehole parameters such as those published by Rhodes and Mott, 1966. A decentralized, side-collimated detector should be used in order to reduce geometric variables. Future logging investigations with  $^{252}\text{Cf}$  should be carried out in a borehole where chemical and physical analyses of the core are available for comparison with log response.

Because of its relatively short half-life,  $^{252}\text{Cf}$  may not replace  $^{241}\text{Am-Be}$  and  $^{239}\text{Pu-Be}$  sources for conventional porosity and moisture logging. However, several other borehole applications are possible in addition to the activation analysis described here. The  $^{252}\text{Cf}$  source has several advantages for borehole investigations: small size, no maintenance required, and a stable predictable flux that requires no monitoring. Other applications will undoubtedly be found for a neutron source that combines an intense flux with very small physical size. Activation analysis of fluid in a hole can be accomplished by appropriate shielding of the gamma detector. Activation can be used to produce temporary depth markers in a hole or in a string of casing or drill pipe. Soluble or insoluble tracers, with energies and half-lives suited to the problem, can be activated in the well. A  $^{252}\text{Cf}$  source can be incorporated in a radioactive tracer ejector-detector sonde in order to activate a tracer in the well. In wells filled with brine, a segment of the fluid column can be activated and then followed with a gamma detector under injection or pumping conditions. Aluminum in drilling mud can also be easily activated in situ with  $^{252}\text{Cf}$  for measuring fluid velocities in boreholes (Wichmann, 1967). The use of activable tracers reduces the problems associated with obtaining short half-life tracers in the proper strength at the proper time.

In conclusion, downhole neutron activation analysis is feasible with isotopic sources and relatively inexpensive portable equipment. With further development, the techniques described here can provide information on the chemical nature of the environment in boreholes which can be useful in the development of petroleum, water, and other minerals.

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