DP-1328

662864

LOCATING END BOUNDARIES OF IRRADIATED ^{242}Pu TARGET CORES BY UNDERWATER DETECTION OF ^{252}Cf FAST NEUTRONS

T. R. HEROLD

W. F. AYRES

DU PONT

REG. U. S. PAT. OFF.

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, S. C. 29801

PREPARED FOR THE U. S. ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(07-2)-1

Printed in the United States of America
Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151

Price: Printed Copy $4.00; Microfiche $0.95

662864

DP-1328

Instruments
(TID-4500-R60, UC-37)

LOCATING END BOUNDARIES OF IRRADIATED ^{242}Pu TARGET CORES BY UNDERWATER DETECTION OF ^{252}Cf FAST NEUTRONS

by

T. R. HEROLD

and

W. F. AYRES

Approved by

D. E. Waters, Manager
Laboratory Operations and Services Division

Publication Date: August 1973

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, S. C. 29801

PREPARED FOR THE U. S. ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(07-2)-1

ABSTRACT

A method and instrument were developed to locate the end boundaries of irradiated ^{242}Pu target cores under water by detecting the spontaneous fission neutrons from irradiation-produced californium isotopes in the cores. The neutrons are detected in an air-filled collimator tube extending from near the core to the water surface. By this technique twenty-one targets were marked and cut under 4.5 m of water without releasing radioactive contaminants. A similar technique could be used to find and map neutron emitters in water or other neutron-moderating media.

CONTENTS

INTRODUCTION

During Savannah River Plant (SRP) campaigns to produce ^{252}Cf and other transplutonium isotopes,[1,2] tubular targets containing ^{242}Pu were irradiated in SRP reactors and shipped to Oak Ridge National Laboratory (ORNL) for processing and ^{252}Cf recovery. To fit the targets into SRP shipping casks, a method was developed for removing the aluminum ends without exposing the active core. The targets (designated Mark 18A) had the dimensions: 10.7-cm outside diameter, 0.26-cm wall thickness, 2.3 m long, with a 1.37-m active core. They were clad with aluminum on outer and inner surfaces for corrosion protection. It was necessary to cut the targets to within about 7.5 cm of each end of the active core. Figure 1 shows the linear dimensions of a typical target housing with the approximate position of the core material.

The intense radioactivity in the targets after exposure necessitated handling and cutting them under 4.5 m of water in the Receiving Basin for Offsite Fuels (RBOF).[3,4] Each target required individual inspection because of irregularities in core length and spacing.

Figure 2 is a view of the disassembly basin before it was filled with water. For testing, the target was placed inside a close-fitting, two-piece, hinged housing of stainless steel that was designed to hold it for cutting. The housing with the target was then positioned on the cradle shown in Figure 2 with the fuel support channel removed. The core location was determined with the top half of the housing opened in order to mark the core and position the target. The housing was then closed over the target and securely fastened. A neoprene lining on the inner housing wall secured the target while the ends were cut off with a power band saw.

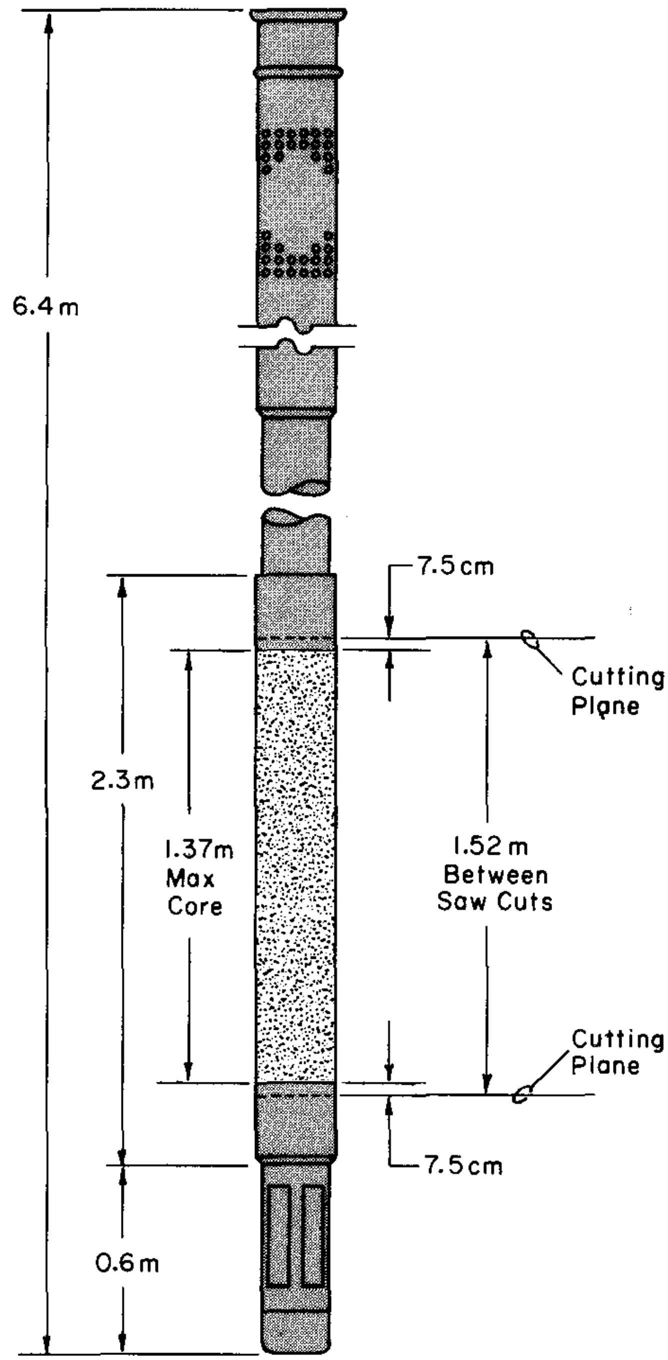

FIGURE 1 Mark 18A Target Housing

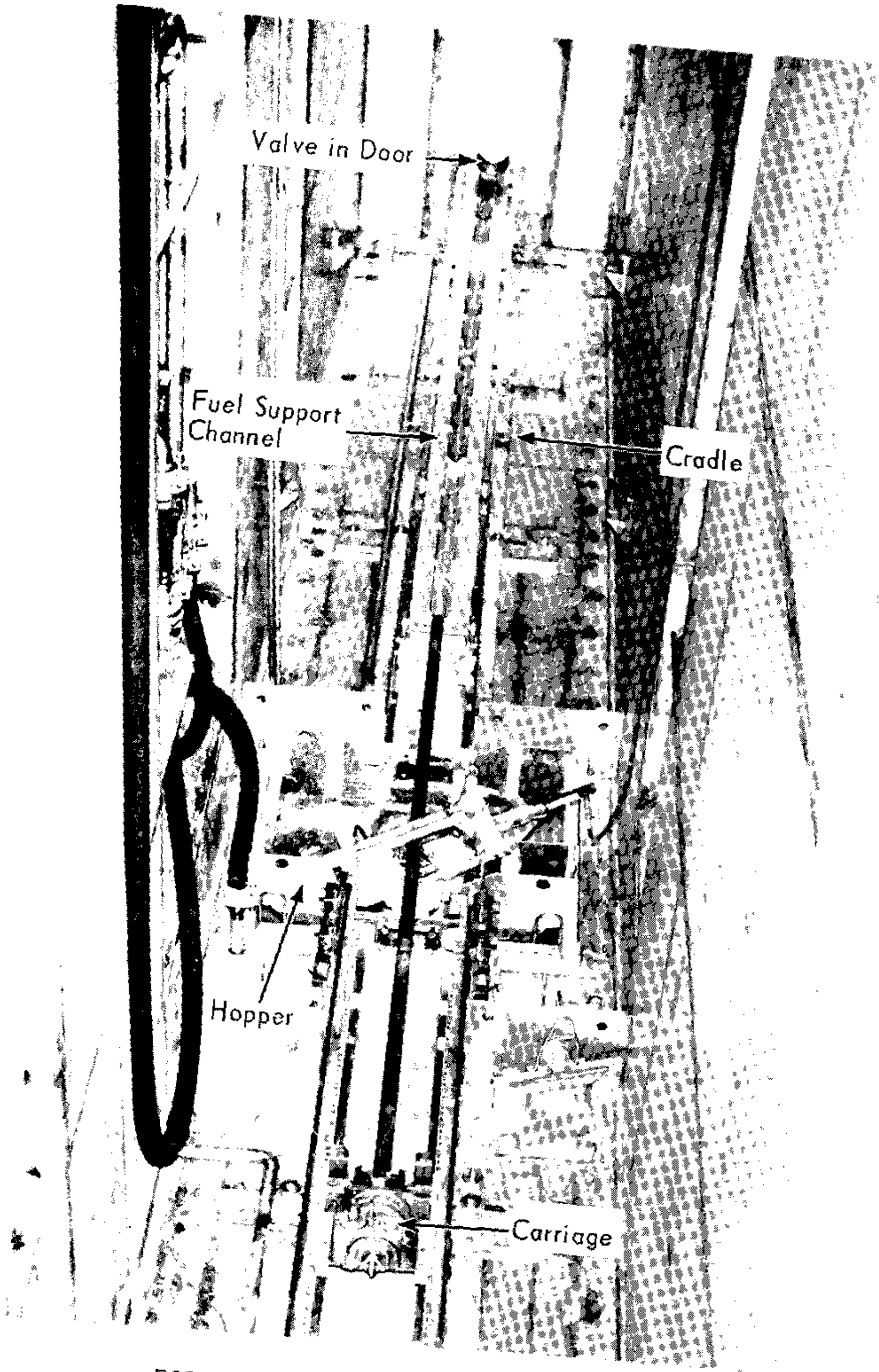

FIGURE 2 Disassembly Basin

POSSIBLE DETECTION TECHNIQUES

The detection techniques that were considered included spontaneously emitted gamma and Cerenkov radiation and fast neutrons.

GAMMA

An ionization chamber was considered initially. The chamber would be operated under water and arranged to view gamma radiation from the irradiated target through a narrow lead collimator slit. However, this technique was impractical because too much bulk shielding would be required to reduce the high gamma background from the Disassembly Basin floor, table, cradle, etc., to a tolerable level at the chamber and also to provide the collimator slit. The electrometer and high voltage supply required with this type of detector would also complicate the readout instrumentation from the lightweight, hand-held package that was desired. The long cables required for signal and high voltage connections between chamber and electrometer could also adversely affect electrometer operation due to flexure and high moisture conditions.

CERENKOV

Detection of Cerenkov radiation[5,6] produced by the irradiated ^{242}Pu targets was also considered. This radiation is generally considered to be the electromagnetic shock wave that arises from a charged particle moving through a medium at a speed greater than that of light within the medium. Thus Cerenkov radiation does not appear until a given charged particle exceeds a threshold energy that ranges from 750 kev for beta particles in water to 500 Mev for protons in water.[7] The main objections to this technique were that (a) any hydrogenous detection medium coupled to a photomultiplier tube for observing relativistic beta particles would be glutted with response to proton recoils from the intense neutron flux, and (b) any available light sensor would not have sufficient sensitivity to locate the actual core boundaries, and the apparent boundaries would possibly change with time out of the reactor. Another objection was that possible "feathering" of the target material during manufacture by extrusion would not contain sufficient material to produce discernible Cerenkov radiation, but if cut through by the saw would thoroughly contaminate the facility.

It was recognized that Cerenkov radiation from the irradiated targets would provide a valuable, qualitative check on whatever measurement technique was used to locate the core boundaries.

NEUTRON

Detecting spontaneous fission neutrons emitted from irradiation-produced californium isotopes in the ^{242}Pu targets provided the simplest, most reliable method for locating the target cores. The difficulty of obtaining the required resolution of ±1 cm with any slow neutron technique was recognized and fast neutron techniques were examined. With the targets immersed in water, any fast neutron measurement would necessarily be made either in the immediate vicinity of the target or at some distance from the target through an appropriate collimator. The latter approach was selected because the fast neutron detectors available would not operate properly in the intense field at the target.

One of the oldest and most widely used methods for detecting fast neutrons in the presence of gamma radiation involves (n,p) scattering.[8-10] By this method, a neutron gives all or part of its energy to a proton, and the ionization and excitation produced by the proton in a counter gas make neutron detection possible. The main advantages of using (n,p) scattering are the relatively large cross section in the Mev range and the operating characteristics of a proton-recoil proportional counter that permit detecting fast neutrons in a relatively intense gamma field. Gamma rays can be rejected because the pulses due to gamma-produced secondary electrons are smaller than most of the pulses produced by fast neutrons.

The most feasible approach was to use a proton-recoil proportional counter in detecting streaming neutrons from the core through an air-filled collimator.

FAST NEUTRON DETECTOR

DESCRIPTION

The block diagram in Figure 3 shows the fast neutron detector consisting of a neutron counter and associated electronics. The neutron counter is a commercially available model* with a polyethylene wall lining and argon-methane filling. The vendor added an inner end cap of polyethylene to provide end-on sensitivity. Better directional response would be obtained by eliminating the polyethylene wall lining, but this modification was not readily available from the vendor. The counter is 2.5 cm in diameter by 15 cm long with a stainless steel cathode.

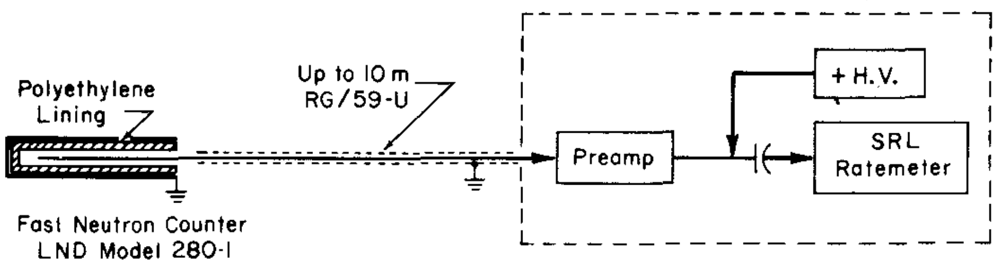

FIGURE 3 Block Diagram of Fast Neutron Detector

The preamplifier and Savannah River Laboratory ratemeter were described in a previous report.[11] The preamplifier circuit utilizes two silicon transistors in a low noise design and typically provides 50-mv output pulses from ^{252}Cf neutrons (average energy ≃ 2.2 Mev). The general-purpose ratemeter features silicon solid-state design throughout and provides direct readout by a panel meter accompanied by audio signal. The panel meter indicates counts per minute on 10^2, 10^3, 10^4, 10^5, or 10^6 linear ranges, and from 10^2 to 10^6 cpm on a logarithmic scale. The ratemeter cabinet is 15 by 25 by 15 cm.

A miniature high-voltage power supply mounted inside the ratemeter cabinet provides polarizing voltage for the neutron counter. The preamplifier was also mounted inside the ratemeter cabinet for this application. Figure 4 is a schematic diagram, and Figure 5 is a photograph of the complete instrument.

* Model 280-1, available from LND, Inc., 3230 Lawson Blvd., Oceanside, New York 11572

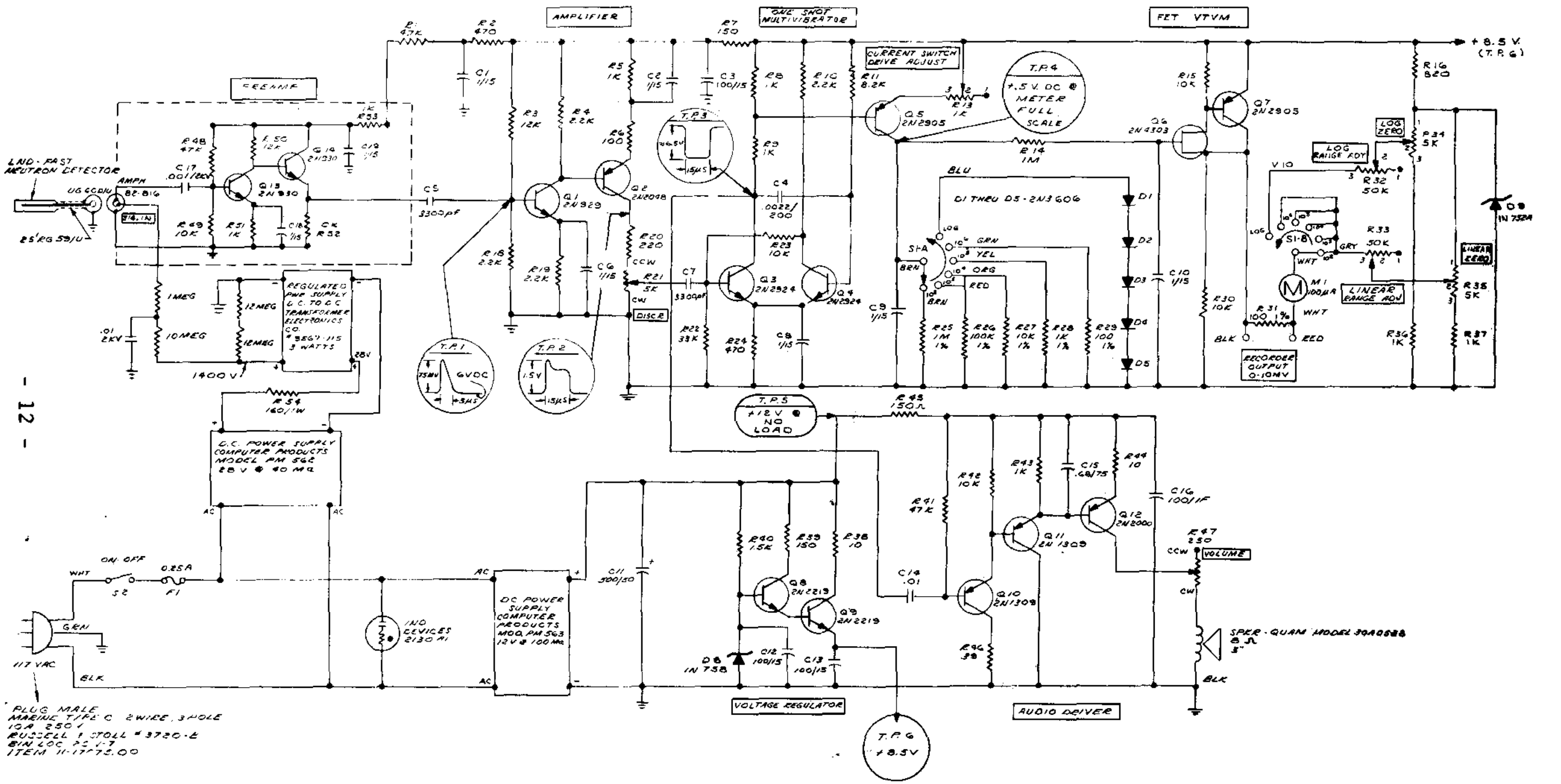

FIGURE 4 Schematic Diagram of Fast Neutron Detector

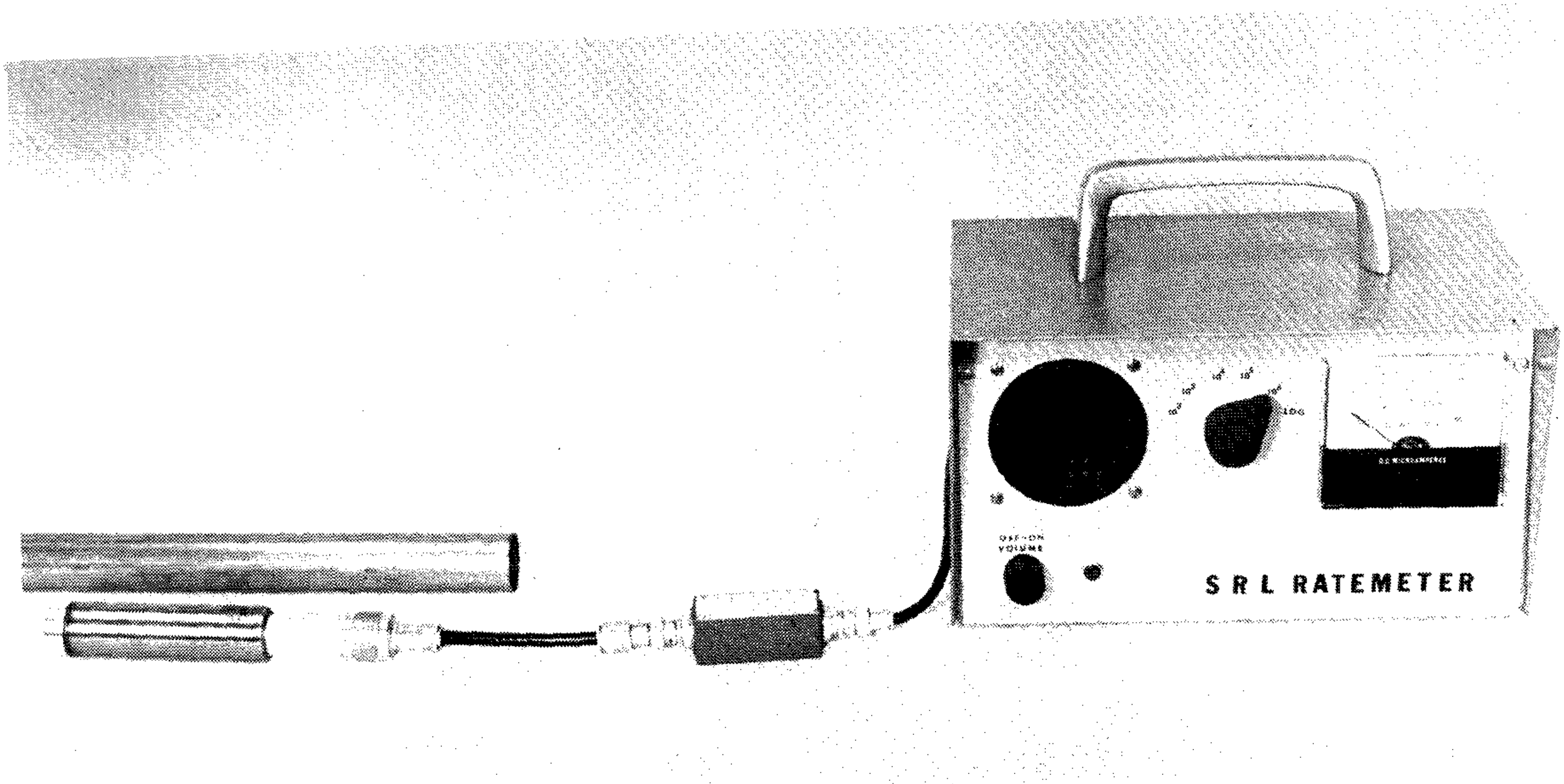

FIGURE 5 Photograph of Fast Neutron Detector

THEORY OF OPERATION

Figure 6 illustrates a straight, cylindrical, air-filled collimator immersed in H_2O moderator with a point source of fission neutrons, S, placed at one end and the neutron counter, D, at some distance, X, from the source in the collimator. In this case, most of the neutrons reach the counter by direct flight, or streaming, from the source.[12] Additional neutrons scatter into the counter from a region one to two slowing down lengths from the source.[13] The slowing down length of fission neutrons in H_2O is about 5.7 cm.[14] Simon and Clifford[15] studied the behavior of fast neutrons streaming through long air ducts in shielding material, and from their work we infer that the scattered neutrons at D amount to only a few percent of the total. Thus, this technique should yield good resolution when viewing point sources, line sources, or the core boundary in an irradiated target.

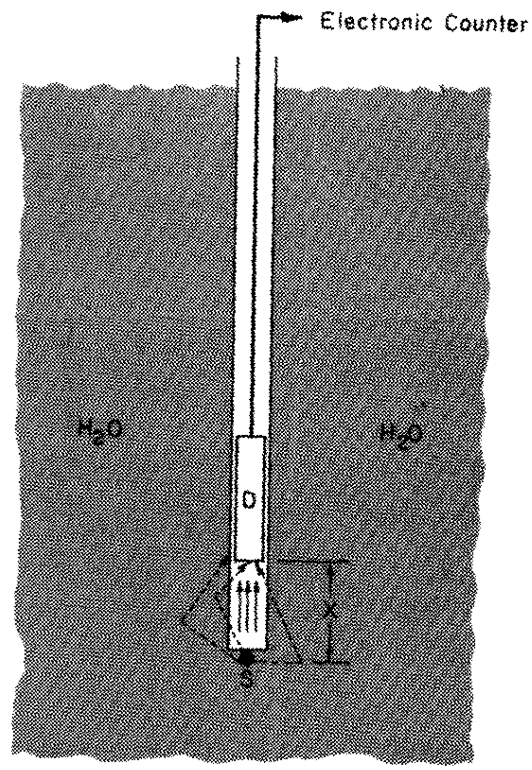

FIGURE 6 Air-Filled Collimator in H_2O Moderator

LABORATORY TESTS

For laboratory tests, the neutron counter was arranged as shown in Figure 3 and placed inside a metal tube with a cap welded to one end for watertightness. The air-filled tube was then lowered into a large tank of water. A source was placed on a horizontal mount in the water to simulate a core boundary. The source was approximately 3 mm in diameter with a volume of about 6 mm^3 and contained 33 μg of ^{252}Cf. The source was in a housing 9 mm in diameter by 35 mm long. This source strength was chosen for the tests because a typical irradiated core was expected to contain about 40 μg of $^{252}Cf/cm^2$. A line source was not available for the laboratory tests.

Figure 7 shows response curves obtained by moving the counter and air-filled tube in discrete steps from left to right over the source. Curves were obtained for different source-to-counter spacings, as shown. In all tests, the air-filled tube was positioned less than 0.5 cm above the horizontal mount. The curves indicate that the minimum source-to-counter spacing for good resolution is about 10 cm. At the 10-cm spacing, a ±0.5-cm deviation from the source-to-counter centerline produced a ±10% change in count rate, and a ±1.0-cm deviation (claimed as the measurement accuracy in detecting core interface) produced a ±25% change in count rate. At the 10-cm source-to-counter spacing, the count rate at 10-cm horizontal displacement from the source falls from 1.35×10^4 to about 1.6×10^3 cpm, or to about 12% of the peak value. Because the tests were performed with an essentially point source instead of a line or large area source, the resolution deduced from the curves shown in Figure 7 for a given geometry is probably optimum.

Neutrons up to two slowing down lengths away from the source, or about 11.4 cm, could contribute a few percent of the total count rate by scattering into the counter. The experimental curves from the point source indicate that the scattered contribution for the 10-cm source-to-counter spacing would be less than 10%, and could be further reduced by increasing the source-to-counter spacing. Similar experimental curves from a line or large area source would possibly indicate a somewhat greater scattered contribution.

The laboratory tests showed that the core boundary in an irradiated target would not produce a step reduction in neutron count rate, but would produce a rapid fall-off in count rate that would asymptotically approach the background level and require interpretation of the scattered contribution to the count rate from about two slowing down lengths beyond the core boundary.

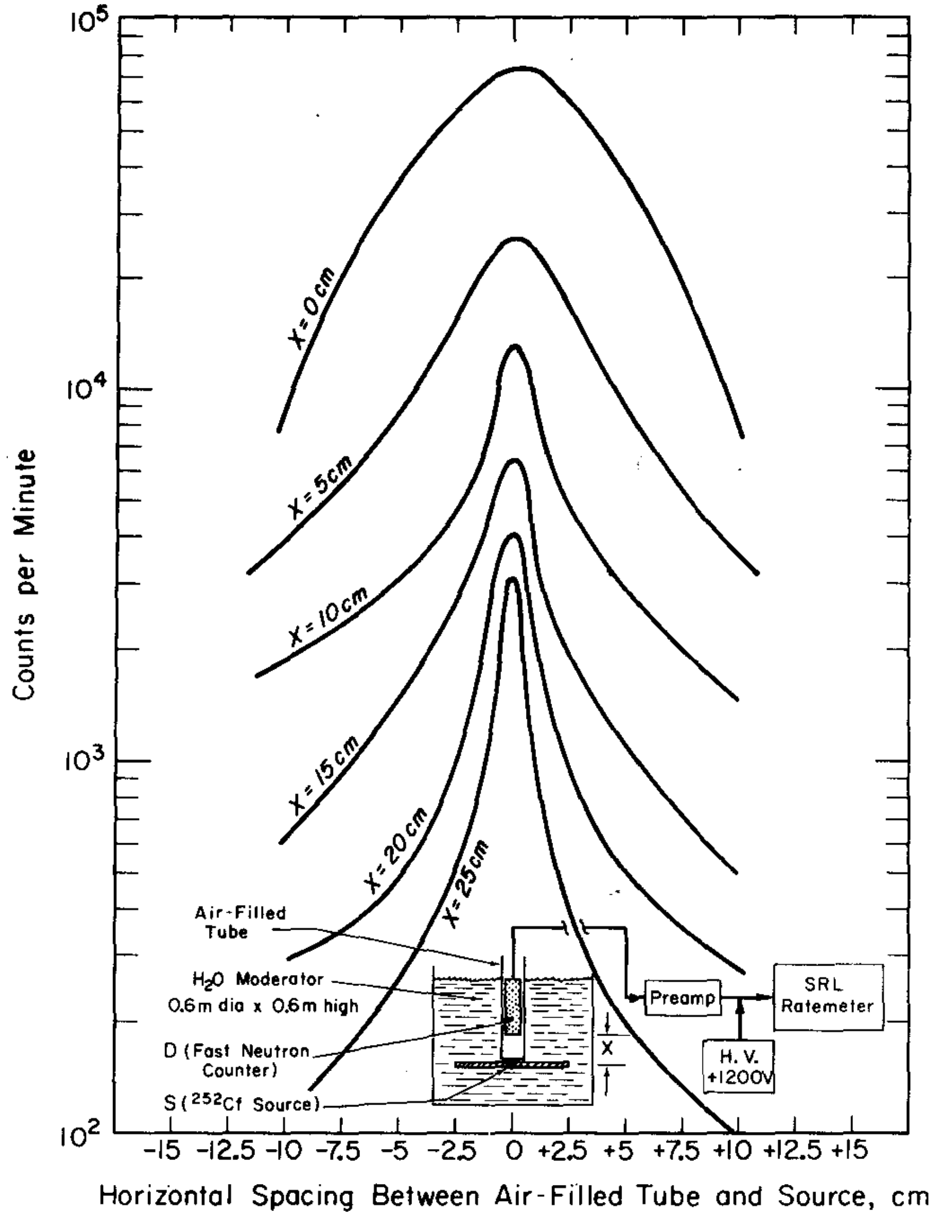

FIGURE 7 Counter Response to ^{252}Cf Source

RBOF TEST RESULTS

A movable platform in the Disassembly Basin at RBOF contained and transported the irradiated target in the hinged, cylindrical case previously described that replaced the fuel support channel (Figure 2). With the case opened, an air-filled pipe about 3 cm in inside diameter by 5.5 m long was centered about 1 cm above the target core. The pipe was suspended from a bracket on the basin railing with provision for height adjustment and lateral movement. A lead plug about 2.8 cm in diameter by 6.4 cm long was inserted in the pipe ahead of the neutron counter to shield it from the intense gamma radiation emitted from the target. The lead plug is essentially transparent to neutrons. During the initial checkout, a movable, close-fitting, lead shield about 3 cm thick by 35 cm long was installed around the pipe to further shield the neutron counter from gamma radiation. The sleeve was positioned with a chain hoist at whatever height the neutron counter was placed. A ratemeter discriminator setting of about 0.5 Mev further reduced the sensitivity to gamma radiation and neutron scatter.

All other neutron-emitting materials were moved at least 3 m away from the neutron counter. The height of the neutron counter was then adjusted for a full-scale ratemeter reading on the 10^6 cpm scale, and ranged between 8 and 14 cm above the target. The position of the lead sleeve was then varied to obtain the lowest count rate, thereby indicating maximum side-shielding against gamma radiation. A calibrated rod was placed alongside the target to obtain accurate measurements relative to the target end. The platform supporting the target and hinged case was then moved until the counter was over one end of the target where the ratemeter reading was recorded. Ratemeter readings were obtained at about 7.5-cm intervals along the target by moving the platform. A typical scan covered about 180 cm of target. A plot of ratemeter readings versus distance along the target was obtained, and a typical plot is shown in Figure 8. The core length was assumed to be within the region defined by tangent lines drawn from the inflection points to the baseline. The inflection points generally corresponded to about 25% of the maximum count rate. The shaded areas of Figure 8 are thought to derive from neutrons scattered into the counter from about two slowing down lengths beyond the core boundaries.

After the core was located, the core boundaries were rotated under the counter to check for runout or irregularities arising from the extrusion process. The targets were then marked and cut by locating the center of the core from the endpoints and measuring 76 cm on each side of the center to obtain the 1.52 m maximum length that will fit into the ORNL shipping cask.

The core position in all targets was verified by visual observation of Cerenkov radiation with the basin in darkness. The typical core lengths indicated by Cerenkov radiation were about 2 to 3 cm shorter on each end than those detected with the neutron detector.

The fast neutron technique was used to mark and cut twenty-one targets without releasing radioactive contaminants to the RBOF disassembly basin. Similar techniques could be used to find lost neutron sources in water or other liquids by sealing a directional counter in an air-filled tube with about 10 cm of air space in front of the counter. The bare counter could be used to find neutron sources in air.

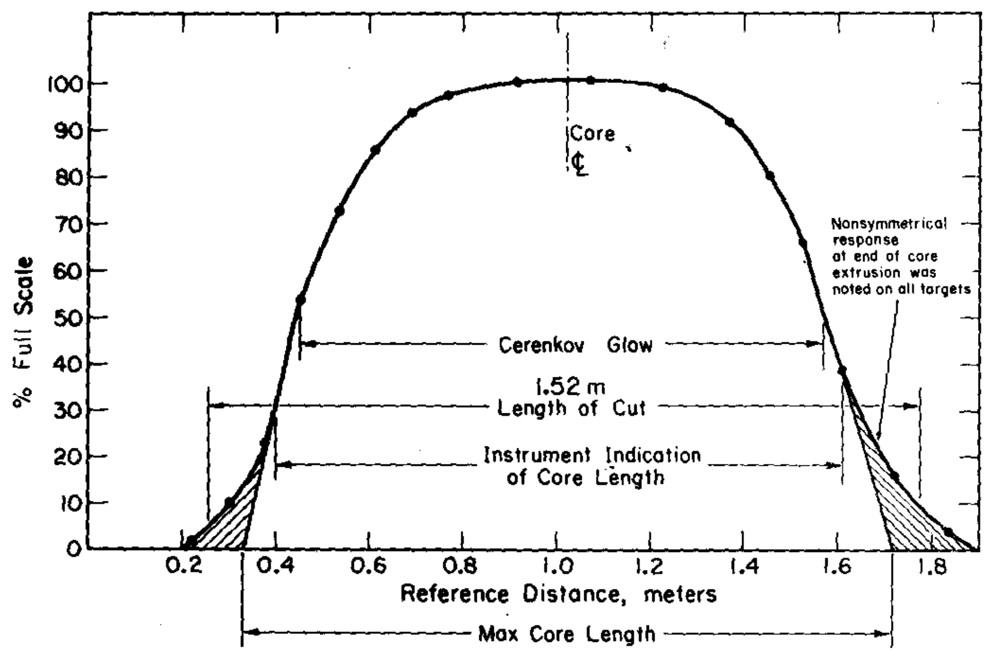

FIGURE 8 Typical Response Curve from RBOF Tests

REFERENCES

1. W. P. Overbeck, C. H. Ice, and G. Dessauer. *Production of Transplutonium Elements at Savannah River.* USAEC Report DP-1000, E. I. du Pont de Nemours and Co., Savannah River Laboratory (1965).

2. J. L. Crandall. "Californium-252 Neutron Sources." Paper presented at *ANS National Topical Meeting on Application of Californium-252,* Austin, Texas, September 11-13, 1972.

3. J. H. Owen, Compiler. *Processing of Power Reactor Fuels.* Nineteenth Quarterly Progress Report, April 1 to July 1, 1962. USAEC Report DP-782, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S.C. (1962).

4. F. C. Locke. *A Basin Facility for Examining Irradiated Nuclear Fuel.* USAEC Report DP-922, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S.C. (1964).

5. P. A. Cerenkov. "Visible Glow of Pure Liquids Under the Influence of X-Rays." *Compt. Rend. Acad. Sci. U.R.S.S. 2,* 451 (1934).

6. I. Frank and I. Tamm. "Coherent Visible Radiation of Fast Electrons Passing Through Matter." *Compt. Rend. Acad. Sci. U.R.S.S. 14,* 109 (1937).

7. A. H. Snell, Editor. *Nuclear Instruments and Their Uses.* Vol. I, p 171, John Wiley and Sons, Inc., New York (1962).

8. P. W. Reinhardt. "A Portable Instrument for the Detection of Fast Neutrons in the Presence of Gamma Radiation." *Phys. Rev. 71,* 836 (1947).

9. G. S. Hurst, R. H. Ritchie, and H. N. Wilson. "A Count-Rate Method of Measuring Fast Neutron Tissue Dose. " *Rev. Sci. Instr. 29,* 981 (1951).

10. E. B. Wagner and G. S. Hurst. "Advances in the Standard Proportional Counter Method of Fast Neutron Dosimetry." *Rev. Sci. Instr. 29,* 153 (1958).

11. T. R. Herold. "Nuclear Instrumentation for Curium and Californium Processing." *IEEE Transactions NS20* (No. 1), 232 (1973).

12. J. B. Marion and J. L. Fowler, Editors. *Fast Neutron Physics* (Part I - Techniques). pp 781-783, Interstate Publishers, New York (1960).

13. Ibid., p 777.

14. S. Glasstone and M. C. Edlund. *The Elements of Nuclear Reactor Theory*. p 183, D. Van Nostrand Co., New York (1952).

15. A. Simon and C. E. Clifford. "The Attenuation of Neutrons by Air Ducts in Shield." *Nucl. Sci. and Engr. 1*, 156 (1956).