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CALIFORNIUM-252 PROGRESS

NUMBER 7, APRIL 1971

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DPST-Cf-252-7

CALIFORNIUM-252 PROGRESS, REPORT NUMBER 7 (U)
April 1971

by

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DPST-Cf--252-7

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Available to the public from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Rd., Springfield, VA 22161.

Numerous requests for the purchase of californium-252 have been received from both domestic and foreign groups since the announcement of the availability of californium-252 for sale at \$10 per microgram.

The first sale of californium-252, a 1-milligram encapsulated source, was to the University of Kentucky. The source was encapsulated by the AEC because no commercial encapsulator could meet the delivery date required by the University of Kentucky.

The first bulk californium-252 sold was purchased by Gamma Industries, a division of Nuclear Systems, Inc. The 100 micrograms purchased will be used to encapsulate individual sources.

Five domestic commercial encapsulators have expressed to the AEC their desire to be considered a potential encapsulator of californium-252 sources (see page 4). Gamma Industries has informed the AEC that they expect to offer sources as large as 5 milligrams for sale by June 1, 1971. Other companies interested in encapsulating californium-252 are urged to inform the AEC's Savannah River Operations Office of their plans.

The continuing interest in developing uses for californium-252 neutron sources in many fields of study is evident by the addition of three new groups (see page 5) to the market evaluation program, bringing the total number to 32. Fifteen universities have executed agreements to receive californium-252 sources under the university loan program with six other proposals being processed. Also, two summer institutes will use californium-252 in training programs to familiarize college and university science professors with neutron sources.

Encouraging results continue to be reported by californium-252 evaluators. Republic Steel reports test analyses showing that californium-252 activation analysis of manganese in steels is as accurate as that of conventional analysis methods, but takes less than half the time.

Columbia Scientific Industries reports that early evaluations of discrete sample and on-stream activation analysis with californium-252 indicate that the method is sufficiently sensitive to monitor several ores, including silver and copper.

Larger californium-252 sources are being requested in the market evaluation program. Several multimilligram sources have been approved for loan, and loans for several others are being negotiated. Gulf Energy and Environmental Systems, Inc. is considering the use of a 10-milligram source to expand their program to include pollution control studies. Evaluators are proving that large sources can be handled and are desirable for many applications.

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Californium-252 NEWS

First Sales of Californium-252 Made

Numerous requests for the purchase of californium-252 have been received by the AEC since availability of the isotope for sale was announced. The requests have been received from both domestic and foreign groups wanting the material for such diverse uses as research, mineral exploration, analytical studies, and reactor startup.

The first californium-252 sold was to the University of Kentucky. The one milligram of californium-252 was in the form of an encapsulated source. The material was encapsulated by the AEC because no commercial encapsulator could meet the required delivery date of the University of Kentucky. The price of the source, including californium-252 and encapsulation charges, is estimated at approximately \$13,500.* Encapsulation charges are determined on an individual basis using facility throughputs and may change on short notice. The charge does not include all costs an industrial organization may have, such as taxes, interest, and profit.

The University of Kentucky plans to use the source in numerous projects including the analysis of lunar samples obtained during the Apollo 14 mission. The source is also expected to be used for graduate courses and for producing short-lived tracers in the university medical center.

Gamma Industries, a division of Nuclear Systems, Inc., purchased the first quantity of bulk californium-252, 100 micrograms. Gamma Industries plans to offer the first complete commercial californium-252 encapsulation program in the United States. The firm

* The exact charge will be determined after final calibration prior to delivery.

plans to supply sources as large as 5 milligrams beginning June 1, 1971. Gamma Industries expects to also offer neutron radiography services and equipment in addition to sealed sources.

Californium-252 Featured at ANS Meeting on Neutrons

Applications of californium-252 will be discussed in many papers at the American Nuclear Society's Topical Meeting on "Neutron Sources and Applications" in Augusta, Georgia, on April 19-21, 1971.

Over eighty papers were submitted to the meeting. Invited rapporteurs will summarize the submitted papers in twenty presentations in sessions entitled "Neutron Sources," "Medical and Forensic Uses of Neutrons," "Exploration and Protection of Natural Resources," and "Industrial Uses of Neutron Sources." All the papers (both invited and submitted) will be published in the proceedings of the meeting.

The proceedings, *Neutron Sources and Applications* (Document Number CONF-710402), may be purchased from the National Technical Information Service, U. S. Department of Commerce, 5285 Port Royal Road, Springfield, Virginia 22151.

The meeting is sponsored by the Savannah River Section and the Isotopes and Radiation Division of the ANS, the Savannah River Operations Office of the U. S. Atomic Energy Commission, and the Southern Interstate Nuclear Board.

Encapsulation Services for Cf-252 Discussed

Representatives of thirteen companies met with the Savannah River Operations Office on January 28 to discuss commercial encapsulation services for californium-252. The agenda of the meeting included reviews of current orders for californium-252 sources, procedures for purchasing bulk quantities of californium-252, the current system by which the AEC provides encapsulation services for californium-252, and possible alternative systems. Comments by encapsulators on the systems were requested. Following these presentations, private meetings and tours of the Savannah River californium-252 facilities were conducted.

Of the thirteen companies at the meeting, those domestic companies having notified the AEC of their desire to be considered a potential californium-252 encapsulator are listed at right. Other companies desiring to be added to this list should inform the Savannah River Operations Office, providing the name, address, and telephone number of the person to contact in their organization regarding californium-252 services.

Potential Californium-252 Encapsulators

Gamma Industries Division
Nuclear Systems, Inc.
Post Office Box 2543
2255 Ted Dunham Street
Baton Rouge, Louisiana 70821
Attention: Mr. H. Richardson, Chairman

General Electric Company
Vallecitos Nuclear Center
Vallecitos Road
Pleasanton, California 94566
Attention: Mr. J. I. Sweeney, Manager-Marketing
Irradiation Processing Operation

Sanders Nuclear Corporation
95 Canal Street
Nashua, New Hampshire 03060
Attention: Mr. H. E. Rexford

NUMEC
Apollo, Pennsylvania 15613
Attention: Mr. W. L. Frankhouser

Monsanto Research Corp.
Nuclear Products Department
Station B, Box 8
Dayton, Ohio 45407
Attention: Marketing Manager

MARKET EVALUATION PROGRAM EXPANDS TO 32 PARTICIPANTS

The AEC's market evaluation program has gained three more participants. These participants will evaluate the application of californium-252 in the following fields:

- The National Aeronautics and Space Administration at the Goddard Space Flight Center, Greenbelt, Maryland, will evaluate 3 micrograms of californium-252 in a capture gamma method to analyze the Cytherean (Venus) surface, in determining water contaminants at remote stations, and for instrument calibration.
- The Department of the Army at the Army Materials and Mechanics Research Center, Watertown, Massachusetts, will evaluate the application of a 5-milligram californium-252 source in a subthermal neutron system used to radiograph certain composite materials. The source will also be used for activation analysis of various construction materials, such as silicon nitride and ceramic compounds.
- The Commission of the European Communities in its Joint Research Centre, Ispra, Italy, will evaluate

25 micrograms of californium-252 as a neutron interrogation source for determining the fissile material content in output solutions from reprocessing plants and in scrap material from nuclear fuel fabrication plants.

The addition of these groups to the program brings the number of evaluators to 32 (see table inside back cover).

The market evaluation program is aimed at assisting the AEC in determining the potential market for californium-252 in various applications. In this program, the AEC is lending californium-252 sources to a limited number of potential users that are interested in evaluating californium-252 in promising applications.

Other proposals for participation in the market evaluation program have been received, and contracts are being negotiated on some of them.

HANDLING GUIDE IS TEACHING AID

The *Guide for Fabricating and Handling Californium-252 Sources* is a valuable educational aid in teaching students and in training inexperienced employers who will be working with californium-252 or engaged in fabricating and reprocessing californium-252 sources. The guide was compiled by the AEC to assist users and fabricators of californium-252 sources. It provides a single-volume ready reference covering topics such as radiation characteristics, radiation control, shielding requirements, and contamination control for californium-252.

Universities which receive a californium-252 source through the university loan program are given a copy of the guide, which can be used:

- As supplemental reading for classroom work.
- In planning health physics procedures associated with an experiment.
- In planning routine handling techniques for sealed sources.
- In determining shielding requirements for experiments and storage containers.

Although written for users of californium-252, the contents of this document are applicable to neutron sources in general. Therefore, any neutron source

user or person entering the field of neutron source application without an extensive background in radiological techniques and precautions would find it useful.

Copies of the guide may be purchased for \$3.00 from:

National Technical Information Service
U. S. Department of Commerce
Springfield, Virginia 22151

Amendment to DOT Shipping Regulations is Proposed by AEC

The USAEC has recommended to the U. S. Department of Transportation (DOT) that the existing DOT regulations be amended to limit the quantity of californium-252 in special form that may be shipped in Type A packaging to no more than 2 curies. The limit in Type A packaging for californium-252 in normal form would remain at 0.001 curies. If the proposed amendment is adopted, DOT regulations would require that quantities of californium-252 in excess of 2 curies be shipped in suitable Type B packaging. To give ample notice to those who might be shipping more than 2 curies, the AEC recommended that this amendment be adopted as soon as possible, but not be made effective before July 1, 1971.

Summer Training Programs to Use Californium-252

College and university science professors will have the opportunity to use californium-252 neutron sources in the Summer Institute in Radiochemistry and Radiation Chemistry at the University of California at Irvine from July 5 to August 13, 1971, and in the course Isotopic Neutron Sources at Oak Ridge Associated Universities (ORAU) from July 19 to August 13, 1971. Other summer institutes are also considering the use of californium-252 in their courses.

Summer Institute in Radiochemistry and Radiation Chemistry

In the Summer Institute in Radiochemistry and Radiation Chemistry, chemistry professors will use five 1-microgram californium-252 sources for foil activation and activation analysis experiments. The objectives of the Summer Institute are: 1) to provide college and university chemistry professors with a thorough exposure, through lectures and laboratory work, to the theory, techniques, and important applications of radiochemistry and radiation chemistry, and 2) to give them practical guidelines for incorporating this material in courses that they teach, or may teach in their own institutions.

The following general types of experiments will be conducted with the californium-252 sources placed in a circular array on a paraffin wax and water moderator assembly to provide a homogeneous flux in a central sample position:

- Activation of large samples to analyze for elements with short half-lives and large cross sections.
- Activation of metal foils, followed by determination of half-lives, beta decay energies, and back-calculation of neutron flux levels.
- Investigation of neutron-counting techniques and cross-calibration of neutron counters.
- Investigation of the properties of various materials as neutron absorbers and moderators.

A single source may be removed and used on its own.

Isotopic Neutron Sources

In the Isotopic Neutron Sources course, chemistry, physics, biology, and nuclear engineering professors will use californium-252, plutonium-beryllium, and americium-beryllium neutron sources in physics and chemistry experiments. The course is designed to

acquaint the professors with various isotopic neutron sources, experiments that can be done with these sources, and the latest techniques in nuclear measurements.

CALIFORNIUM-252 FEATURED AT GENEVA CONFERENCE

The Fourth United Nations International Conference on the Peaceful Uses of Atomic Energy will be held September 6-16, 1971, in Geneva, Switzerland. Attending will be more than 5,000 delegates and official observers, including top scientists, engineers, and administrators from more than 70 nations. Approximately 500 scientific papers will be presented, including 59 by prominent United States scientists.

An exposition of nuclear science and technology will be held in conjunction with the conference, and 21 nations have already indicated their intention to exhibit. The United States will participate with a 20,000-square-foot exhibit entitled *Nuclear USA*.

One of the feature exhibits of the United States in the area of industrial and engineering application of radiation and radioisotopes will be a demonstration of neutron activation analysis techniques using californium-252 as the neutron source. As a part of the exhibit, a small californium-252 source and sodium iodide detector will be used to emphasize the rapidity and reliability with which sample analyses can be made (see *Californium-252 Progress*, No. 5, 6 (1970)). Another part, based on the work of J. I. Trombka, Goddard Space Flight Center, and F. E. Senftle, United States Geological Survey, will use californium-252 as a neutron activation source for the analysis of complex materials. In this analysis, a complex sample is irradiated and counted, the spectrum is telemetered by satellite to the Goddard Space Flight Center for computer analysis, and the result is sent back within a few minutes.

Two exhibits highlighting the use of californium-252 in safeguards applications based upon the work of Gulf Energy and Environmental Systems, Inc. and Los Alamos Scientific Laboratory (see pages 26 and 44, respectively) will also be on display.

Other exhibits of *Nuclear USA* will feature the production of californium-252 and a graphic display of californium-252 medical sources which are being considered for possible use in cancer radiotherapy.

CALIFORNIUM-252 SHIPPING CASKS AVAILABLE

Each participant in the AEC's californium-252 market evaluation program is required to provide a cask to transfer californium-252 sources to and from the Savannah River Plant. These casks may be designed and fabricated by each participating organization, or they may be purchased or leased from commercial vendors.

As an aid to prospective californium-252 evaluators, a list of commercial companies having californium-252 shipping casks available for sale or rent and those expressing an interest in fabricating casks to custom design was published in *Californium-252 Progress*, No. 5, 8 (1970). The list is updated as the AEC's Savannah River Operations Office continues a survey to determine the availability of commercial neutron source shipping casks suitable for containing californium-252.

Companies having shipping casks for californium-252 available for sale or rent (and their contacting officials) are:

Monsanto Research Corporation
Dayton Laboratory
1515 Nicholas Road
Dayton, Ohio 45407
Attention: Mr. D. R. Adams

ATCOR, Inc.
5 Westchester Plaza
Elmsford, New York 10523
Attention: Mr. J. C. Darrin

O. G. Kelley & Company, Inc.
Neponset
Boston, Massachusetts 02122
Attention: Mr. John Bready

Protective Packaging Inc.
Post Office Box 1192
Tacoma, Washington 98401
Attention: Mr. Ken Gablin

J. L. Shepherd and Associates
703 South Pacific Avenue
Glendale, California 91204
Attention: J. L. Shepherd

In addition, the following companies have expressed

an interest in fabricating shipping casks for californium-252 to customers specifications:

Optics for Industry
Nuclear Products Division
1929 N. Buffum Street
Milwaukee, Wisconsin 53212
Attention: Mr. R. H. Eder

Ionics, Incorporated
Post Office Box 99
Bridgeville, Pennsylvania 15017
Attention: Mr. H. A. Zenk, Jr.

Health Techna Corporation
Penberthy Division
6701 Maynard Avenue, South
Seattle, Washington 98108
Attention: Mr. H. W. Sankey

Prospective californium-252 evaluators should contact these organizations for further information on cask availability and cask design specifications.

FOUR NEW PARTICIPANTS IN UNIVERSITY LOAN PROGRAM

Four more universities — Carnegie-Mellon University, Pittsburgh; University of Tennessee, Memphis; University of Texas, Austin; and Ohio University, Athens — will receive californium-252 sources under the university loan program. The addition of these universities to the program brings the number of participants to 15 (see page 51). Proposals from six additional universities have been approved, and agreements are being executed.

Under the university loan program, the AEC will lend sources containing less than one microgram of californium-252 to universities for classroom and laboratory demonstrations, but not for radiotherapy. These sources are loaned free of charge and are shipped in paraffin-filled containers, which can be retained by the university with the source. The only expense incurred by the borrower is the shipping charge from the Savannah River Laboratory near Aiken, South Carolina.

The procedure for submitting proposals can be found in *Californium-252 Progress*, No. 4, or may be obtained by writing to the address on page 50.

CALIFORNIUM-252 Questions and Answers

What types of survey meters are required to monitor radiation from californium-252?

Californium-252 emits two types of penetrating radiation which necessitate measurement — neutrons and gamma rays. Neutron radiation levels can be monitored with commercially available moderating sphere neutron and neutron rem counters that closely match the response of the human body. Gamma radiation can be monitored with commercially available ionization chamber or Geiger-Müller instruments. [For more detailed information on survey techniques for californium-252 see *Guide for Fabricating and Handling ²⁵²Cf Sources* (see page 5) USAEC Report SRO-153, Savannah River Operations Office, U. S. Atomic Energy Commission, Aiken, South Carolina (1971).]

In removing a californium-252 source from a shipping cask, what personnel dose rates would be received at distances of 5, 10, and 15 feet from 1-, 5-, and 10-milligram sources?

The total radiation dose rate from californium-252 is a combination of neutron and gamma dose rates. Approximate dose rates, in mrem per hour, based upon Savannah River Laboratory experience for the above conditions are:

	5 Feet			10 Feet			15 Feet		
	γ	n	Total	γ	n	Total	γ	n	Total
1 mg	68	965	1033	17	241	258	8	107	115
5 mg	340	4830	5170	85	1205	1290	40	535	575
10 mg	680	9650	10330	170	2410	2580	80	1070	1150

Is the \$1600 handling charge on shipments of 1-milligram or more of unencapsulated californium-252 applicable to encapsulated sources sold by the AEC?

No. Encapsulated sources sold by the AEC do not carry a handling charge, but do carry an encapsulation charge. The AEC will only provide encapsulation services if commercial services are not available at reasonable terms and conditions. The encapsulation charge will be determined for each source at the time of encapsulation. The charge for each source sold by the AEC will be published in *Californium-252 Progress* (see page 4).

Where can suitable containers for shipping californium-252 sources be obtained?

Commercial companies having californium-252 shipping casks available for sale or rent and those expressing an interest in fabricating casks to customer design are listed on page 7.

What are water-extended polyester resins?

Water-extended polyester resins (WEP[®]) are special polyesters which form an emulsion with water to extend the polyester and improve physical properties. WEP can be used as a neutron shielding material (see page 40) because it has a high water content, and because materials such as lithium, boron, and cadmium can be added.

Even though californium-252 is now for sale, the AEC expects to continue its market evaluation and other loan programs for californium-252. Information concerning any of the californium-252 programs can be obtained by writing to the address on page 50 or by telephoning 803-824-6331, ext. C252.

Questions concerning californium-252 may be directed to:

Manager, SROO
Attn: *Californium-252 Progress*
Savannah River Operations Office
U. S. Atomic Energy Commission
Post Office Box A
Aiken, South Carolina 29801

CALIFORNIUM-252 TECHNOLOGY DEVELOPMENT

Applications Development Under Market Evaluation Program

BROOKHAVEN NATIONAL LABORATORY

At Brookhaven National Laboratory, the continuing evaluation of californium-252 in radiation therapy has included cell culture studies on Chinese hamster cells to determine the dose rate effect, the relative biological effectiveness (RBE), and the oxygen enhancement ratio (OER) of californium-252. Studies of the skin tolerance for californium-252 irradiations are also continuing.

Cell Culture Studies

Cell survival curves obtained with Chinese hamster cells at californium-252 dose rates of 18.6 (D_0 , 160 rads) and 13.5 (D_0 , 210 rads) rads per hour (Figure 1) and at cesium-137 dose rates of 37.3 and 16 rads per hour (Figure 2) show that cell survival is a function of the dose rate mainly because the cellular multiplicity* changes significantly during irradiation.

When the survival curves for californium-252 were corrected for cellular multiplicity, the resulting single-cell survival curves fell on a straight line with a D_0 of 105 rads. However, because of the rapidly changing multiplicities, the single-cell survival curves are at best only rough approximations.

* Average number of cells per colony-forming unit at time of irradiation.

FIG. 1 Effect of Californium-252 Radiation and Irradiation Time on Cellular Multiplicity of Chinese Hamster Cells

This rapid change also makes it difficult to evaluate OER's, particularly since cellular proliferation is inhibited under anoxia (Figure 2). Thus, the radio-resistivity caused by anoxia may be marked by the increased survival of rapidly dividing oxygenated cells. Therefore, OER measurements are planned at higher dose rates.

With an 85% survival of control cells under anoxia, an OER of 2.6 was measured with cesium-137 (37.3 rads per hour) at a dose level of 1000 rads (Figure 3).

Skin Tolerance Tests

In continuing tests to determine the influence, or lack of influence, of dose rate of californium-252 on skin tolerance and to determine a more appropriate RBE with respect to gamma sources (radium or cesium-137), pigs are being irradiated with californium-252 at 12.5, 15, and 23 rads per hour and with gamma sources at 35, 67.8, and 192 rads per hour. Californium-252 irradiations at 60 rads per hour are planned.

FIG. 2 Effect of Cesium-137 Radiation and Irradiation Time on Cellular Multiplicity of Chinese Hamster Cells

FIG. 3 Survival Curves for Chinese Hamster Cells which Received Protracted Irradiation (37.3 rads per hour) from Cesium-137

When the total doses required to produce a moist reaction involving more than 50% of the irradiated field were plotted against exposure hours, the resulting line for gamma sources had a slope of 0.29 between about 20 and 240 hours (Figure 4). This slope is consistent with previously reported clinical studies in man. The line for californium-252 irradiations is nearly level, which indicates that any increase in dose rate will have little effect on the tolerance level. (Cellular recovery and repopulation appeared to be minimal during irradiation with californium-252.) The RBE of californium-252 with respect to radium or cesium-137 depends on the overall period of irradiation required to achieve a specific dose level as shown in Figure 5.

FIG. 4 Total Dose Required to Produce the Same Skin Reaction for Californium-252 and Radium-226 or Cesium-137 Irradiation at Different Dose Rates (X indicates single acute exposure of 250 KVP X-rays, corrected for RBE relative to high-energy gamma rays. Californium-252 irradiation at 23 rads per hour not included, but point falls at or above the line.)

FIG. 5 Effect of Dose Rate on the RBE of Californium-252 with Respect to Radium-226 or Cesium-137

UNIVERSITY OF CINCINNATI

The University of Cincinnati Departments of Radiology and Nuclear Engineering are preparing to use a 20-microgram californium-252 source to measure the neutron energy spectrum and dose with threshold damage track detectors in tissue-equivalent phantoms. A computer code based on integral transport theory is being written to calculate three-dimensional doses near the surfaces of cylindrical phantoms. Experimental results will be compared with the calculations to evaluate the biological effect of neutrons produced by implanted sources. The evaluations of californium-252 at the University of Cincinnati are under the direction of J. G. Kereiakes.

M. D. ANDERSON HOSPITAL AND TUMOR INSTITUTE

The University of Texas M. D. Anderson Hospital and Tumor Institute is continuing radiobiology and dosimetry studies with californium-252. An additional new study, diagnostic radiology with californium-252, is being considered. Investigations being considered are summarized below.

Diagnostic Radiology

The technique of prompt capture gamma ray analysis is being considered for determining the elemental metamorphosis of bone *in vivo*. With this technique, neutron-induced capture gamma rays could be the basis for qualitative and quantitative analyses. The technique may be useful for:

- Differentiating between lesions that are indistinguishable radiographically, such as fibrous versus cartilaginous lesions, osteosarcoma versus Ewing's sarcoma, or even inflammatory versus neoplastic lesions.
- Investigating metabolic diseases with gross calcium changes, such as hyperparathyroidism, osteomalacia, and osteoporosis.
- Evaluating the response of known bone lesions to therapy (for example, myeloma treated with melphalan).
- Determining the effects of various chemotherapeutic agents on normal bones (for example, methotrexate or steroids).

Prompt capture gamma analysis depends on the capture of a thermal neutron by a nucleus that causes the formation of an excited state and the immediate ($<10^{-12}$ second) emission of one or more prompt photons. The energies and relative intensities of these photons create characteristic spectra of the isotope emitting them; thus, each isotope has a unique signature. The energy of the neutron capture gamma radiation ranges from 0.07 to 10 Mev, and the spectra of most isotopes are complex. With high resolution energy spectroscopy, the isotopic components of a specimen can be determined by comparing the specimen to pure elemental samples or to known sample mixtures. If properly applied, this prompt gamma ray technique may be used to compare similar anatomical bone sites, to assay long bones or chains of bones, or to determine a temporal series of a specific bone.

Prompt capture gamma analysis may be useful in studying bone compositional changes because no other method can selectively assay a multielement substance. Two available techniques for analyzing bone *in vivo*, radiographic comparison and photon relative transmission, determine total mineral content only. The transmission method is more accurate, but is limited to a scan of only the thin bone sections. With the radiographic technique, thick sections of bone can be analyzed, but only gross mineral changes (~ 30%) can be detected.

The neutron source for the experiments will be 200 micrograms of californium-252. A convergent collimator will be used to extract a neutron beam from the biological shield. Nonuniformity of the beam across the collimator exit area is not as important as in neutron radiography; uniformity can be achieved only at the sacrifice of the flux intensity. Because the maximum thermal neutron flux from californium-252 is 1 to 2 centimeters from the source, which in many cases corresponds to the midline of the bone,* homogeneous activation can be achieved to a depth of about 4 centimeters. A gamma ray spectrometer placed near the sample irradiated can detect any prompt capture gamma ray emitted from the thermal capture *in vivo*.

The evaluation of prompt capture gamma analysis for analysis of bone will include the following experiments in the order listed.

- Determination of the characteristic prompt gamma ray energy spectra of elements found in the body by irradiating these elements in their simplest form (elemental, hydroxyl, or simplest compound). These spectra will serve as reference spectra in subsequent tests.
- Test analysis of simple composite samples, such as calcium nitrate and sodium chloride, and then of complex composite samples, such as calcium hydroxyapatite. Also, composite samples varying only in quantitative makeup will be analyzed to evaluate sensitivity factors.
- Analysis of pathologically catalogued bones of sacrificed marmosets that have undergone specific

diet and vitamin intake changes designed to effect skeletal changes. Routine examinations on these bones, such as radiographic, serum, and bone densitometry studies (gamma ray transmission method), will be compared with the prompt capture gamma ray analyses.

- Examination *in vitro* of human bones that have known disease states, if the above tests are successful.
- Application of prompt capture gamma analysis *in vivo*.

THE HOSPITAL OF THE UNIVERSITY OF PENNSYLVANIA

The Hospital of the University of Pennsylvania is evaluating californium-252 as a neutron source for clinical use in the treatment of surface and interstitial lesions. The first patient treated with californium-252 fission sources had a right cervical lymph node metastasis from a squamous cell carcinoma of the tongue. A planar interstitial implant was made with the afterloading tube technique conventionally used with iridium implants. A total of 14 afterloading cells (ALC type), each containing 1.85 micrograms of californium, was inserted. For calculation of the dose, all of the afterloading cells were assumed to be parallel, in the same plane, and spaced 1-centimeter apart. The californium was left in place for 67-1/2 hours. The calculated dose 0.5 centimeter above the central plane was 1544 rads (966 neutron and 578 gamma). In addition to the californium-252 implant, a submental metastasis was implanted with iridium, and a total dose of 8000 rads was delivered to this area. After two months, both areas had no evidence of persistent tumor.

* D. W. Magnuson. "Thermal Neutron Flux Distribution from a Cf-252 Source." *Californium-252, Proceedings of a Symposium Sponsored by the New York Metropolitan Section of the American Nuclear Society, October 22, 1968, New York*. J. J. Barker, Ed. USAEC Report CONF-681032 (1962).

NEUTRON RADIOGRAPHY

UNIVERSITY OF TEXAS AT AUSTIN

The University of Texas at Austin is evaluating a 1-milligram californium-252 source for nuclear safeguards analysis, forensic analysis, neutron radiography, and prompt capture gamma analysis.

A single container has served both as the shipping container and as the experimental irradiator. The details of the design and construction of this container have been described previously.*

Neutron Activation Analysis System

A californium-252 neutron activation analysis system has been developed for both the nuclear safeguards studies and forensic investigations. This system includes the californium-252 source, a combination shield and experimental assembly, a pneumatic transfer facility, a lithium-drifted germanium detector, and a dual-crystal sodium iodide spectrometer. The system accepts samples inserted by hand or transferred by a pneumatic transfer system.

The study of short-lived radioisotopes requires the use of rapid sample transfer, accomplished by a pneumatic transfer system. This system (Figure 6) consists of the pneumatic control subsystem, the pneumatic tubing, and transfer container (rabbit). The pneumatic control subsystem controls the irradiation time, transfer time, and counting time. The pneumatic tubing is fabricated from 1-inch-outer-diameter stainless steel tubing. The sample to be irradiated is contained in a transfer rabbit of low-density polyethylene or stainless steel, approximately 1-3/4 inches in length and 7/8 inch in diameter; 3/32-inch flanges

FIG. 6 Pneumatic Transfer System

hold the rabbit in position in the tube and also allow it to travel around turns without binding. The stainless steel rabbits are used for safeguards analysis to avoid thermalization of neutrons. The rabbits accommodate a one-dram polyethylene sample vial. The hermetically sealed vials are placed in the rabbit and held in position by a threaded end plug. The beam port plug for the pneumatic transfer system is filled with borated water-extended polyester. A mixture of 18 vol % sodium borate in water-extended polyester after curing was less prone to crumbling under pressure than previous borated mixtures.

The lithium-drifted germanium detection system is used primarily in safeguards and prompt capture gamma experiments where good resolution is necessary. The detection system consists of a lithium-drifted germanium detector coupled to a preamplifier, amplifier, and then to a Nuclear Data 2200 multi-channel analyzer. With this system and the pneumatic transfer system, a sample may be activated in cyclic fashion, i.e., irradiated, sent to the detector, counted, and then sent back to the source for further irradiations.

The dual-crystal sodium iodide spectrometer is a high efficiency counting system used primarily in forensic analysis, where high efficiency is required because of small samples. The spectrometer consists of two matched 3 x 3-inch sodium iodide detectors, preamplifiers and amplifiers for each, a summing amplifier, and a Nuclear Data 2200 multichannel analyzer.

Nuclear Materials Safeguards

For the on-line assay of the fissile content of nuclear fuels, samples will be pneumatically transferred into the shield assembly through a beam port,

* S. J. Gage, E. L. Draper, Jr., G. D. Bouchev, and R. R. Day. "Design and Construction of a Versatile Cf-252 Neutron Source Shield and Experimental Facility." *Neutron Sources and Applications: Proceedings of the American Nuclear Society National Topical Meeting, April 19-21, 1971, Augusta, Georgia.* USAEC Report CONF-710402 (1971).

irradiated with californium-252 neutrons, rapidly transferred to the high-resolution lithium-drifted germanium detector, and then returned to the source for further irradiations. Cyclic irradiation increases the number of counts accumulated in a given period, particularly when short half-life isotopes are to be measured.

Preliminary experiments involving cyclic activation of thorium and uranium dioxide samples are in progress to evaluate experimental procedures and to test the irradiation and counting equipment.

Forensic Analysis

For forensic activation analysis, samples are pneumatically transferred to the shield assembly, irradiated

with californium, and then transferred to the dual-crystal sodium iodide detector.

Neutron Radiography

The californium-252 source is being used to study the effect of neutron energy spectrum, converter screen, and film type on the quality of neutron radiographs of a multielement test object. Results for four beam configurations with four different converter screens have been reported,* and are given in Table I.

* G. D. Bouchey, E. L. Draper, Jr., and S. J. Gage. "Neutron Radiography with Cf-252: The Effect of Tailoring Neutron Energy Spectra on Photographic Images." *Neutron Sources and Applications: Proceedings of the American Nuclear Society National Topical Meeting, April 19-21, 1971, Augusta, Georgia*. USAEC Report CONF-710402 (1971).

TABLE I
Characterization of Radiographic Results

Screen	Beam 1	Beam 2	Beam 3	Beam 4 ^a
LiF-ZnS	Excellent Resolution Good Thermal Neutron Imaging No Fast Neutron Imaging No Gamma Contribution	Excellent Resolution Good Thermal Neutron Imaging No Fast Neutron Imaging No Gamma Contribution	No Image at All	Not Used
Gadolinium	Fair Resolution Substantial Thermal Neutron Imaging Some Fast Neutron Imaging Strong Gamma Contribution	Fair Resolution Substantial Thermal Neutron Imaging Some Fast Neutron Imaging Strong Gamma Contribution	Good Resolution Some Thermal Imaging Substantial Fast Neutron Imaging Strong Gamma Contribution	Not Used
Indium (Direct)	Poor Resolution Substantial Thermal Neutron Imaging Substantial Fast Neutron Imaging Strong Gamma Contribution	Poor Resolution Substantial Thermal Neutron Imaging Substantial Fast Neutron Imaging Strong Gamma Contribution	Fair Resolution Some Thermal Neutron Imaging Substantial Fast Neutron Imaging Strong Gamma Contribution	Not Used
Indium (Transfer)	Not Used	Not Used	Not Used	Poor Resolution No Thermal Contribution Excellent Indium Self-imaging No Gamma Contribution
Organic Scintillator	Low Quality Gamma Radiograph	Low Quality Gamma Radiograph	Low Quality Gamma Radiograph	Low Quality Gamma Radiograph

Excellent - All holes and spacings between holes could be distinguished on the cadmium test object.

Good - The 0.020-inch holes could be distinguished, but smaller hole spacings not visible.

Fair - The 0.0625-inch hole distinguished, but 0.020-inch holes not visible.

Poor - Only the 0.250-inch hole was distinguishable.

^a Scattered thermal neutrons were eliminated in this beam by encasing the object, screen, and film in cadmium.

Four beam configurations were used: a thermal neutron beam, an epithermal neutron beam, a fast neutron beam, and a high-intensity fast neutron beam. The thermal neutron beam (Beam 1) was achieved by placing a water-filled polyethylene container with a penetration for a 4-inch-diameter collimator into the central cavity of the shield assembly and locating the source approximately 1 inch below the collimator entrance. Offsetting the source should improve the image quality and the neutron-to-gamma ratio. The epithermal neutron beam (Beam 2) was achieved by inserting a 4-inch-diameter by 1-inch-thick lead disk between the source and collimator entrance in the above configuration. A fast neutron beam (Beam 3) was achieved by lining the central cavity with 2 inches of sodium borate and placing the source directly in the cadmium-lined collimator entrance. For a very high-intensity fast neutron beam (Beam 4), the object, screen, and film were encased in cadmium and placed in the 10-inch beam port 8 inches from the source. No collimator was used.

For thermal neutron radiography, the $^6\text{LiF-ZnS}$ scintillator with light sensitive Type 51 and 57 Polaroid film is best (Figure 7) because (1) it eliminates gamma fogging of the film, and (2) it is an efficient neutron converter-film combination, which allows shorter exposure times.

FIG. 7 Thermal Neutron Radiograph with LiF-ZnS Screen and Type 57 Polaroid Film

For fast neutron radiography, gadolinium and indium screens gave the most satisfactory results for imaging epithermal neutrons (Figure 8); however, with the direct exposure method, the resolution is poor because of gamma interference from neutron capture in the shielding material.

FIG. 8 Fast Neutron Radiograph of University of Texas Test Object with Gadolinium Screen, "Kodak" Royal Blue Film, and Beam 3

The high-intensity fast beam was used to demonstrate a technique called "self-imaging", whereby the relative contrast of strong resonance materials in a radiograph is enhanced by using that same material as a screen (Figure 9). The main difficulties in this method are eliminating excessive gamma exposure and neutron scatter in thick objects. Activation-transfer techniques can be used to reduce gamma exposure; anticatter grids will decrease neutron scatter.

In future work, the homogeneous uranium-polyethylene assembly and a subcritical heterogeneous array of TRIGA reactor fuel elements will be investigated to assess the flux enhancement of californium-252 neutrons and conventional radioisotopic source neutrons.

FIG. 9 Fast Neutron Radiograph by Indium Self-Imaging Technique Using Indium Screen (Transfer Exposure) and "Kodak" Royal Blue Film

Prompt Capture Gamma Analysis

Californium-252 will also be evaluated as a neutron source for prompt capture gamma and prompt X-ray analysis techniques.

For these analyses, a sample is irradiated in an extracted beam from the shield assembly, and the prompt gammas are counted by a detector directly below the sample. Because there is a large gamma ray background at the sample (primarily 2.223-Mev hydrogen capture gamma rays), a detector shield has been designed and fabricated. The shield consists of contoured lead bricks and a section of poured lead. The lead has a minimum thickness of 4 inches and is lined with sheets of copper and tin to minimize the number of X-rays reaching the detector. The shield is on a steel table on rollers which can be positioned below the sample to be analyzed. The liquid nitrogen Dewar and the lithium-drifted germanium and lithium-drifted silicon detectors are on a shelf near the bottom of the table. The detectors are surrounded radially by the shield. A slot in the shelf supporting the lead brick allows the Dewar and detector to be removed from the shield without disassembling the apparatus.

PICATINNY ARSENAL

The Picatinny Arsenal of the U. S. Department of the Army, at Dover, New Jersey, is evaluating the use of californium-252 in neutron radiography, activation analysis, moisture measurements, and charge inspections of munitions. Picatinny has received a 4.22-microgram californium-252 source for the evaluations and is constructing a shipping-storage container for a 10-milligram source.

Preliminary experiments with the 4.22-microgram source indicate a californium-252 source is better than a plutonium-238-beryllium source for low-level moisture measurements.

The shipping-storage container for the 10-milligram source will be a five-foot-diameter steel sphere fabricated by welding together two hemispherical segments. Facilities for loading artillery shells with high explosives will be used to fill the spherical cavity with a molten lithium-wax mixture.

BATTELLE MEMORIAL INSTITUTE

A 1-milligram californium-252 source is being evaluated for neutron radiography and neutron activation analysis at Battelle Memorial Institute, Columbus, Ohio.

The californium source is stored and neutron radiographs are made in the pool shielding facility at the Battelle Research Reactor. The peak thermal flux was measured to be about 0.5 inch from the surface of the source. The flux distributions agreed with those determined by Barton* and Magnuson** except near the source. This disagreement was thought to be caused by the source encapsulation material, a study of which is reported below.

Preliminary measurements indicated the neutron-to-gamma ratio in the radiographic beam was not sufficient for neutron radiography with gadolinium foil and X-ray film. To increase the ratio, an air gap was added to the collimator inlet to minimize flux perturbation effect of the cadmium-lined collimator and to move the source away from the collimator inlet to prevent gamma streaming.† Continuing studies of the effect of the source-collimator geometry on the thermal neutron flux and on the neutron-to-gamma ratio are reported also.

Evaluation of Encapsulation Material

The effect of californium-252 source encapsulation material on the peak thermal neutron flux in a moderator region surrounding the source was analyzed for a stainless steel-clad californium-252 source (standard SR-Cf-100 series source) from the Savannah River Laboratory and an aluminum-clad source from Oak Ridge National Laboratory. The thermal neutron flux distributions measured at Battelle for the stainless steel-clad source and at Oak Ridge** for the aluminum-clad source are shown in Figure 10. The fluxes were normalized to a source strength of 1 neutron per second. For the stainless steel-clad source the

* *Californium-252 Progress*, No. 2, 20 (1969).

** D. W. Magnuson. "Thermal Neutron Flux Distribution from a Cf-252 Source." *Californium-252, Proceedings of a Symposium Sponsored by the New York Metropolitan Section of the American Nuclear Society, October 22, 1968, New York*. J. J. Barker (Ed.). USAEC Report CONF-681032 (1969).

† *Californium-252 Progress*, No. 6, 23 (1971).

FIG. 10 Thermal Neutron Flux Distribution Measured from Aluminum-Clad and Stainless Steel-Clad Californium-252 Sources

peak normalized flux is 1.06×10^{-2} neutrons/(cm²-source neutron), which corresponds to an absolute flux of 2.53×10^{-7} neutrons/(cm²-sec) at 0.75 inch from the center of the source. For the aluminum-clad source the peak normalized flux is 1.37×10^{-2} neutrons/(cm²-source neutron), which is 25% greater than the peak obtained with the stainless steel-clad source. The fluxes converge as the distance from the source increases and are nearly the same at a distance of 3 inches or greater.

The primary difference between the two sources is the encapsulation materials. The absorption cross sections for thermal neutrons are greater for the stainless steel, platinum, and rhodium in the SR-Cf-100 series source than for the aluminum in the ORNL source. Therefore, the thermal flux near the source is perturbed by the materials used for the californium-252 encapsulation.

The DOT-II-W code* was used to calculate the thermal neutron flux in a water moderator surrounding the two types of californium sources (Figure 11) in order to corroborate the above experimental conclusion. The magnitude and shape of the calculated distributions are the same as for the experimental data up to about 1-1/2 inches from the source. At greater distances, the calculated fluxes decrease more rapidly than the experimental fluxes; however, the calculated fluxes for both sources become identical beyond 3 inches, as did the experimental fluxes. The calculated thermal flux appears to decrease more rapidly 1-1/2 inches from the source than the experimental fluxes because the mesh spacing in the calculation was increased at these distances. The mesh

FIG. 11 Thermal Neutron Flux Distributions Calculated for Aluminum-Clad and Stainless Steel-Clad Californium-252 Sources

spacing was increased because of computer size limitations and the desire to obtain an extremely fine mesh near the source, which was the area of prime interest. This rapid decrease in flux has very little effect on the accuracy of the solution near the source because the distribution is calculated from the source outward.

The experimental and calculated thermal flux measurements show that the peak thermal flux for a californium-252 source depends on the encapsulation material. A high-strength material with low neutron cross section, such as zirconium (in which californium-252 sources are being encapsulated**), should also be considered as an encapsulation material for californium-252 sources used for neutron radiography.

Beam Extraction Geometries

The effect of source-collimator geometry on the thermal neutron flux and the ratio of thermal neutron flux to gamma dose rate at the collimator outlet was evaluated with two collimators having the same effective length-to-diameter ratio. One collimator was 7.5 inches long with a 0.5 x 0.5-inch entrance aperture and a 4 x 5-inch exit area. The second collimator was 15 inches long with a 1 x 1-inch entrance aperture

* R. G. Soltesz and R. K. Disney. *User's Manual for the DOT-II-W Discrete Ordinates Transport Computer Code*. USAEC Report WANL-TME-1982, Westinghouse Electric Corp., Astronuclear Laboratory, Pittsburgh, Pa. (1969).

** *Californium-252 Progress*, No. 6, 26 (1971).

and an 8 x 10-inch exit area. Both collimators were air-filled aluminum pyramids lined with 0.021-inch-thick cadmium.

Thermal neutron fluxes and neutron-to-gamma ratios were measured (Table II) at the collimator outlet by moving the source both vertically and horizontally with respect to the collimator inlet and with respect to the inlet of an air gap between the source and collimator. In the vertical configuration, measurements were made with the source aligned with the central axis of the collimator and with different air gaps. In the horizontal configuration, measurements were made with the midpoint of the source coplanar with the inlet aperture of the collimator and with different air gaps. The air gaps were of the same cross sectional area as the collimator inlet aperture.

The best combination of thermal neutron flux and neutron-to-gamma ratio for the 7.5-inch-long collimator is at the 0.5-inch vertical position with a 2-inch air gap. This air gap with the vertical configuration produces a 25% increase in the thermal neutron flux and a threefold increase in the neutron-to-gamma ratio.

The best combination of thermal neutron flux and neutron-to-gamma ratio for the 15-inch-long collimator is at the 1.0- and 1.5-inch horizontal positions with a 2-inch air gap. A 2-inch air gap produces a 50% increase in the thermal neutron flux and nearly a two-fold increase in the neutron-to-gamma ratio.

The data in Table II indicate that the longer collimator has a substantially lower exit flux, which is unusual because the effective length-to-diameter ratio is the same for both collimators. This difference probably results from the source not being effectively infinite in size with respect to the dimensions of the collimator inlet aperture.

TABLE II

Thermal Neutron Flux and Neutron-to-Gamma Ratios at Collimator Outlet

Air Gap Length, in.	Source-to-Collimator ^a Separation Distance, in.	Thermal Neutron Flux, $n/(cm^2 \cdot sec)$	Neutron-to-Gamma Ratio, $n/(cm^2 \cdot mR)$
<u>7.5-in.-Long Collimator, Vertical Traverse</u>			
0	1.0	1.07×10^4	0.6×10^4
1.0	1.0	1.31×10^4	1.2×10^4
2.0	0	1.13×10^4	0.6×10^4
	0.5	1.35×10^4	2.0×10^4
	1.0	0.98×10^4	1.8×10^4
3.0	1.5	0.65×10^4	1.2×10^4
	1.0	0.96×10^4	
<u>7.5-in.-Long Collimator, Horizontal Traverse</u>			
0	1.0	2.20×10^4	0.9×10^4
1.0	1.0	1.93×10^4	1.3×10^4
2.0	0.75	1.37×10^4	1.1×10^4
	1.0	1.29×10^4	1.1×10^4
	1.5	1.40×10^4	1.1×10^4
3.0	1.0	0.80×10^4	0.9×10^4
<u>15-in.-Long Collimator, Vertical Traverse</u>			
0	1.0	0.49×10^4	0.6×10^4
1.0	1.0	0.70×10^4	1.1×10^4
2.0	0	0.61×10^4	0.9×10^4
	0.5	0.64×10^4	1.0×10^4
	1.0	0.65×10^4	1.2×10^4
	1.5	0.65×10^4	1.2×10^4
<u>15-in.-Long Collimator, Horizontal Traverse</u>			
0	1.0	0.52×10^4	0.7×10^4
1.0	1.0	0.70×10^4	1.1×10^4
2.0	1.0	0.75×10^4	1.2×10^4
	1.5	0.75×10^4	1.2×10^4
	2.0	0.22×10^4	0.4×10^4
3.0	1.0	0.57×10^4	1.5×10^4

^a With air gaps, this is the source to air gap distance.

NATURAL RESOURCES EXPLORATION

UNITED STATES GEOLOGICAL SURVEY

The United States Geological Survey is evaluating mineral exploration with a californium-252 source by *in situ* neutron activation analysis.

Borehole Activation Studies

A 2-1/4-inch logging sonde containing a californium-252 source and a 1-1/2 x 2-inch-long sodium iodide detector has been evaluated in a simulated borehole of fireclay, which contains Al_2O_3 , to determine how borehole diameter, water in the borehole, and sonde misalignment affect the area of a spectral peak.

When the diameter of a dry borehole was increased from 3 to 6 inches, the 1.78-Mev aluminum-28 peak was diminished by one-half, but the decrease did not appear to be linear with change in the diameter. When the borehole was filled with water, the peak increased from ~60% (6-inch diameter) to ~100% (3-inch diameter) as compared to dry boreholes of the same diameter. When the sonde was placed off-center, adjacent to the borehole wall, and the borehole diameter was about three times larger than the crystal diameter, the peak area increased appreciably. Consequently, when a large diameter borehole is logged with a small diameter sonde, a centering device should be used.

A logging sonde containing a californium-252 source and a lithium-drifted germanium detector has been constructed* and is being evaluated. The sonde is 2-7/8 inches in diameter by about 7-feet long, and weighs somewhat over 35 pounds. The detector is cooled by a sealed cryostat containing melting propane. Therefore, the sonde does not require venting or power for cooling during operation, and may be used in liquid-filled boreholes. The cryostat will hold at the temperature of melting propane (8°C above boiling nitrogen at atmospheric pressure) for about 10 hours. The propane is refrozen by conduction through a cold finger in contact with a liquid-nitrogen reservoir, which is attached when the sonde is out of the borehole.

Detector Calibration

The NASA Langley Research Center, Hampton, Virginia, used a 3.25×10^8 neutron per second californium-252 source at the U. S. Geological Survey to calibrate detectors used to measure absolute neutron fluences from a dense plasma focus experiment. The plasma focus apparatus produces a dense deuterium plasma at thermonuclear temperature over a volume of about 0.001 cubic centimeter. A burst of up to 10^{10} neutrons at 2.45 Mev is emitted during a 100-nanosecond period. The spatial distributions of neutron and X-ray fluence and energy are being measured to determine the deuterium velocity distribution of the plasma focus. The neutron fluence is determined by measuring the intensity of the 656-keV gamma peak from a 230-gram silver target activated by the neutron burst from the plasma focus. The small physical size, the energy spectrum centered near 2.45 Mev, and the high intensities available ($>10^8$ neutrons per second) make the californium-252 source an ideal choice for detector calibration in this particular application.

Process Stream Analysis

Simulated laboratory experiments have been made to test the feasibility of using a radiative capture gamma method for industrial process stream analysis and control.** 500- to 1000-gram samples of taconite, magnetite, copper, and nickel ores were irradiated with a 125-microgram californium-252 neutron source for a combined irradiation-counting time of 100 minutes. Because the larger sample sizes encountered in typical industrial process streams would require considerably shorter counting times, this method

* A. B. Tanner, R. M. Moxham, F. E. Senftle, and J. A. Baicker. "A Borehole Sonde Using a ^{252}Cf Source and a Ge(Li) Detector Cooled by a Melting Cryogen." *Neutron Sources and Applications: Proceedings of the American Nuclear Society National Topical Meeting, April 19-21, 1971, Augusta, Georgia.* USAEC Report CONF-710402 (1971).

** D. Duffey, P. F. Wiggins, and F. E. Senftle. "Application of Neutron Capture Gamma Rays Using a ^{252}Cf Neutron Source to Industrial Process Stream Analysis." *Neutron Sources and Applications: Proceedings of the American Nuclear Society National Topical Meeting, April 19-21, 1971, Augusta, Georgia.* USAEC Report CONF-710402 (1971).

appears feasible for on-line analysis and control. The capture gamma method is particularly effective for detecting iron, copper, and nickel, which are difficult to detect by delayed gamma techniques under similar conditions.

TEXACO INC.

In a continuing evaluation of californium-252 in well logging for petroleum, Texaco Inc. is investigating both dual-spaced detector porosity logging and neutron activation logging with a 700-microgram source.

Laboratory investigations have shown that dual-spaced detector porosity logging can produce a porosity log in cased or uncased wells.* Porosity logs made in cased test formations were only slightly affected by variations in the borehole size and in the saltwater or boron content of the formation, but were quite sensitive to the formation matrix composition. Comparative porosity logs made in uncased test formations showed that the logs are almost insensitive to casing in the borehole and to variations in the borehole salinity and are only affected slightly by the position of the logging instrument in the borehole.

Activation logs (aluminum logs) of test formations of mixtures of Al_2O_3 and SiO_2 have shown that the lower average energy of californium-252 neutrons produces much less interference from silicon than from the higher energy plutonium-beryllium neutrons. Also, the higher neutron output of californium-252 makes it possible to log a well at an economical speed.

Dual-Spaced Neutron Detector Logging for Porosity

Porosity logs of two Texaco wells were made with the dual-spaced neutron detector logging sonde containing the helium-3 detectors at 60 and 90 centimeters from the 700-microgram californium-252 source (Table III).

In Well 1, porosity logs were run with the detectors at different combinations of spacing from the neutron source because the porosities in this well varied over a wide range. The shorter spacings were for porosities from 10 to 30%; the longer spacings for 0 to 10% porosities. The log analysis was concentrated on the uncased section below a depth of 4612 feet. The logging sonde was not fitted with a decentralizer spring because parts of the borehole were closing in and a

spring would have increased the possibility of the sonde becoming stuck in the well. Consequently, the sonde was allowed to find its own position in the borehole. Because logs were run as the sonde was pulled out of the well, the sonde dragged against the borehole wall most of the time.

The lithology and porosity of the formation surrounding Well 1 are not known accurately because core samples were not taken during drilling of the well. However, a lithology log was obtained from drill cuttings collected from the mud stream at the surface. Also, a commercial logging company made a Borehole Compensated Sonic Log and a natural gamma ray log of the well. Although the sonic log gives some information on porosity, the porosity may be better defined if sidewall cores are taken of the well.

TABLE III
Californium-252 Porosity Logs for Well 1 and Well 2

Log Type	Neutron Source	Detector Positions, cm	Logging Speed, ft/min	Interval Logged, ft
Dual Neutron of Well 1	700 μg ^{252}Cf	72 and 102 60 and 90	20 30	6200 - 4300 6200 - 4300
Dual Neutron of Well 2	700 μg ^{252}Cf	60 and 90	20	5090 - 3600

Well 1 Description

Location:	West Texas
Casing:	0 - 4612 ft, steel casing 9-5/8 in. ID 4612 - 6300 ft, no casing
Borehole fluid:	Saturated saltwater
Borehole diameter:	9 1/4 to 12 in. in uncased section
Lithology control:	Log of drill cuttings
Porosity control:	Borehole Compensated Sonic Log No core porosities available

Well 2 Description

Location:	Texas Gulf Coast
Casing:	5106 to 5057 ft, steel casing 5 in. ID 5120 to 0 ft, steel casing 7 in. ID
Borehole fluid:	Saltwater
Borehole diameter:	9-7/8 in.
Lithology control:	None available
Porosity control:	None available

Logs of Well 1 are compared in Figure 12 for the uncased interval from 4700 to 5350 feet. The sonic log and the neutron log are similar. However, the neutron log is much more sensitive in the lower porosity zones such as the interval from 4754 to 4810 feet.

Porosities in Well 1 were determined from the ratio of the count rates in the two detectors using the known lithology and the calibration data presented in *Californium-252 Progress*, No. 6, 26. In deriving porosity from

* *Californium-252 Progress*, No. 6, 27 (1971).

FIG. 12 Logs of Well 1

the sonic log, the lithology and the sonic velocity in the formation matrix and in the formation fluid must be known. The matrix velocity in limestone was assumed to be 23,000 feet per second, in sandstone 21,000 feet per second, and in the formation fluid 5300 feet per second. Some judgment is required in choosing these velocities because experience has shown that the best values vary with the location of the well and the formations traversed.

Porosities derived from the sonic log and the neutron log of Well 1 are compared in Table IV. The porosities are in general agreement for 10 to 20% porosity. Below 10% porosity, the neutron log generally gives a lower value than the sonic log. It is not known which log gives the correct porosity, because no cores have been taken from the well to accurately measure the porosity.

In interpreting dual-spaced detector porosity logs the following factors must be considered:

- Because the californium-252 dual-spaced detector porosity logs have detectors spaced at large distances from the source, formation beds with thick-

TABLE IV

Porosities Calculated from Sonic and Neutron Logs for Well 1

Depth, ft	Sonic Log		Dual Neutron Log	
	Transit Time, μ sec	Porosity, %	72/102 cm Porosity, %	60/90 cm Porosity, %
Limestone				
4764	51.5	5.5	0.3	0.2
4778	52.5	6.2	0.5	0.9
4802 - 06	53	6.5	0.9	1.1
4824 - 28	59.5	11.0	14	11
4900 - 05	64.5	14.5	19	15
4943 - 50	61.5	12.5	16	15
4960 - 70	60	11.3	13	11
4999 - 5002	54.5	7.5	8.0	6.6
5300	55.5	8.2	4.8	3.5
Sandstone				
5100	61.5	10.0	11.5	11
5150 - 12	65.5	13.0	16	14
5207 - 12	71	16.8	20	19
5238 - 54	71	16.8	18	16
5700 - 20	71.5	17.1	19	20

^a Calculated assuming $V_{LS} = 23,000$ ft/sec, $V_{SS} = 21,000$ ft/sec; $V_{fluid} = 5300$ ft/sec

nesses less than the source-to-detector spacings cannot be resolved. For typical source-to-detector spacings of 60 and 90 centimeters the minimum bed resolution is about 2 and 3 feet, respectively. In some circumstances, this limitation could be a disadvantage.

- In processing the data, the logs must be carefully aligned.
- The two detectors measure neutrons which have traversed different lengths of formations. This can cause some problems if the porosity changes rapidly with depth.

The porosity log of Well 2 was made to assist in the interpretation of other californium-252 logs measured in this well and has limited usefulness in the porosity log evaluation program because no accurate lithology or porosity data are available for this well.

Continuous Activation Logging

An activation log was made in Well 1 with the 700-microgram californium-252 neutron source. The log recorded four gamma ray energy ranges (0.75 to 1.0 Mev, 1.0 to 2.0 Mev, ≥ 2.0 Mev, and the total gamma ray count rate), and was made at a logging speed of 20 feet per minute. The activation log was recorded with the source preceding the sodium iodide detector by 86 inches. A background log was recorded at the same speed when the sonde was pulled out of the well after a 10-minute delay at the bottom of the well to allow the aluminum-28 activation to decay.

The activation log for the 1.0 to 2.0-Mev range is shown in Figure 12. A low count rate was recorded in the limestone section down to about 5000 feet. On entering the shaley sandstone section the increase in count rate necessitated a factor of 2 scale change on the recorder. In general, the character of the activation log is similar to that of the natural gamma ray log. This similarity was first shown by Keys and Boulogne,* who noted that sediment beds are more sharply delineated in a californium-252 activation log than in a natural gamma ray log. The present data confirm their observation. The activation log in Figure 12 is being studied to determine the effect of porosity changes on the activation log.

Californium-252 activation logs were made in Well 2 at speeds of 30, 20, 10, and 3 feet per minute to determine the optimum logging speed. Results of this work will be reported later.

SCHLUMBERGER TECHNOLOGY CORP.

Schlumberger Technology Corporation is evaluating californium-252 in well logging for petroleum and natural gas.

Porosity Logging

Porosity logs are being made to compare californium-252 sources with plutonium-beryllium sources.

In tests comparing a 10^8 neutrons per second californium-252 source with a 10^7 neutrons per second plutonium-beryllium source in a sidewall epithermal neutron porosity sonde in a simulated borehole, the californium-252 source had higher porosity resolution and a higher count rate, but an increased mud cake effect.

In tests comparing a 10^8 neutrons per second californium-252 source with a 10^8 neutrons per second plutonium-beryllium source in an experimental compensated neutron porosity sonde in a simulated borehole, the californium-252 source had a higher porosity resolution, about the same mud cake and cement effects, but a lower count rate. Comparative logs were also run in a cased and cemented test well in the Gulf Coast area. Results were inconclusive because of low count rates and poor statistics. The sonde is being redesigned to increase the count rate.

Neutron Capture Gamma Logging

A californium-252 source is also being evaluated for neutron capture gamma ray measurements for chemical analysis of borehole formations. Comparisons are being made with a 14-Mev pulsed neutron source and a plutonium-beryllium source. Laboratory measurements have confirmed that the californium-252 source has the advantages of a lower production of inelastic gamma rays and less shielding needed to prevent source neutrons from entering the detector.

THE UNIVERSITY OF GEORGIA

The University of Georgia is continuing the development of an undersea neutron activation analysis system with californium-252 for *in situ* elemental analysis of undisturbed mineral deposits on the sea floor. Three modes of undersea activation analysis have been evaluated: an undersea submersible for deep water capabilities, a towed sled for intermediate depths, and scuba divers for shallow water operations.** Simulated terrestrial ore-grade samples, a nominal 200-microgram californium-252 source, and a sodium iodide detector were used.

Ore samples were prepared for the 14 elements marked with an asterisk in Table V. These elements were selected because of their favorable nuclear properties (see Table I in reference below) for rapid neutron activation analysis and their probable occurrence in the marine environment.

In all three modes of analysis, the samples were irradiated with a flux of 1×10^6 neutrons/(cm²-sec) for 4 minutes, allowed to decay for 30 seconds, and then counted for 4 minutes. In some samples, the radioactivity was sufficiently high to register a 15 to 40% dead time in the analyzers. Sea water enhanced

* W. S. Keys and A. R. Boulogne. "Well Logging with Californium-252." *Transactions of the SPWLA Tenth Annual Logging Symposium, The Society of Professional Well Log Analysts, Houston, Texas, P (1969).*

** J. E. Noakes, G. A. Smithwick, J. L. Harding, and A. Kirst, Jr. "Undersea Mineral Analysis with Californium-252." *Neutron Sources and Applications: Proceedings of the American Nuclear Society National Topical Meeting, April 19-21, 1971, Augusta, Georgia. USAEC Report CONF-710402 (1971).*

TABLE V

Ore Grade of Currently Exploited Terrestrial Minerals

<u>Element</u>	<u>Ore Grade, %</u>	<u>Element Content, %</u>
U	0.1 - 0.2 U_3O_8	0.175
Fe	20 - 70 Fe	20 - 70.0
*Mn	15 - 55 Mn	15 - 55.0
*Cr	15 - 68 Cr_2O_3	46.0
Ni	0.9 Ni	0.9
*Mo	0.2 Mo	0.2
*V	1.0 - 2.0 V_2O_5	1.1
*W	0.3 - 1.0 WO_3	0.8
*Co	0.2 - 1.5 Co	0.2 - 1.5
*Cu	0.4 Cu	0.5
*Pb	3.5 - 5.0 Pb	3.5 - 5.0
Zn	5.0 Zn	5.0
*Sn	0.5 Sn	0.5
*Al	50 Al_2O_3	27.0
Au	~5 oz/ton Au	~0.0001
*Ag	~10 oz/ton Ag	~0.03
Be	10 - 12 BeO	4.3
*Hg	~ 5 lbs/ton Hg	.25
*Ti	20 - 60 TiO_2	36.0
*Mg	20 - 69 MgO	41.0
S	15 - 40 S	15 - 40.0
K	14 K_2O	13.0
P	15 P_2O_5	6.6

TABLE VI

Undersea Activation Analysis of Synthetic Ore Samples with Cf-252

<u>Element</u>	<u>Terrestrial Ore Element Content, %</u>	<u>Detected Simulated Ore Element Content, %</u>	<u>Estimated (Lower Range) Simulated Ore Element Content, %</u>
Mn	15 - 55.0	10.0	0.1
V	1.1	1.0	0.5
Cu	0.5	10.0	2.0
Al	27.0	10.0	1.0
Sn	0.5	15.0	3.0
Ag	~0.03	5.0	0.3
Hg	0.25	20.0	10.0
Ti	36.0	20.0	4.0

sample activation by thermalizing the higher energy californium-252 neutrons and also served as a neutron shield for personnel in the water.

These tests showed that undisturbed sea floor mineral deposits can be analyzed for element concentration levels of commercial interest (Table VI). Of the three modes of activation, a scuba diver team was excellent for investigating ocean bottom depths to 60 feet. The Westinghouse DEEPSTAR 2000 submersible, with its self-contained power and life support system, was superior for depths greater than 60 feet. Analysis can be made with the underwater sled, but a more efficient analysis would be obtained if the sled were self-propelled instead of towed by a surface ship and if a diver rode the sled and positioned the source and detector.

The health physics study of the total marine exploration study showed that a californium-252 source could be used in all phases of marine work without undue radiation hazards to participating personnel.

KERR-McGEE CORPORATION

Kerr-McGee Corporation of Oklahoma City has received a 1.0-milligram californium-252 source to evaluate uranium exploration by borehole logging. The source was transported in a shielded cask to Oklahoma City, where it was transferred to a storage location 23 feet underground.

The source will be used to determine the extent and uranium content of deep-lying ore deposits. To date, all efforts have been directed toward developing equipment sufficiently rugged and reliable to withstand the roughest field use. Figure 13 shows a californium-source trailer attached to the logging truck. The truck is equipped with data processing equipment. The source shield is integral with the heavy duty trailer, which carries a probe washing system and is equipped with a red warning light that flashes automatically when the entrance lid to the source is open.

FIG. 13 Californium-252 Shield for 1-milligram Source

The shield (Figure 14) is filled with an 18% solution of boric acid in glycerin. Since boric acid forms a true solution in glycerin, rather than a suspension, uniform distribution is assured, and the concentrations obtainable in glycerin are higher than those ordinarily attempted in paraffin. Boric acid, rather than borax, is used, to avoid producing radioactive sodium-24, which emits high energy gamma radiation. Calculations indicate that this shield can hold up to 10 milligrams of californium-252 and still meet DOT external radiation requirements.

The borehole probe carries the source holder (Figure 15) as well as three helium-3 neutron detectors, their high-voltage power supply, and a shield to prevent direct exposure of the detectors to the source.

A remotely operated hydraulic manipulator enables the operator to remove the source from the shield after the probe has been manually attached to the source holder, to lift up the source, to rotate the probe, and then to swing it into the borehole (Figure 16).

Handling will be practiced with a dummy source holder until all operations are smoothly executed. Preliminary work with the actual source will simulate field conditions: tests will be carried out in five 22-foot-deep circular pits filled with uranium-free and uranium-bearing strata of known thicknesses.

FIG. 14 Californium-252 Shield for 1-milligram Source

FIG. 15 Californium-252 Source Holder

FIG. 16 Logging Probe and Source Being Removed from Shield by a Remote Hydraulic System which Allows Operator to be 50 feet Away

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

The National Aeronautics and Space Administration will use three 1-microgram californium-252 sources at the Goddard Space Flight Center, Greenbelt, Maryland, to:

- Evaluate californium-252 as a neutron source in a neutron radiative-capture method to perform elemental analysis of the Cytherean (Venus) surface.

- Determine whether instruments and data analysis systems developed for space exploration are applicable for determining water contaminants at remote stations.
- Calibrate the instrumentation systems planned for a gamma ray survey of the lunar surface.

Under a letter agreement signed with the USAEC, the program will be under the overall direction of J. I. Trombka.

NUCLEAR SAFEGUARDS

GULF ENERGY AND ENVIRONMENTAL SYSTEMS, INC.

Gulf Radiation Technology, a Division of Gulf Energy and Environmental Systems, Inc., is evaluating californium-252 as a neutron source in a nondestructive assay device to determine the fissile and fertile content of various nuclear materials. In a new evaluation effort, Gulf is considering the development of a system for analyzing pollutants in water.

Isotopic Source Assay System

An Isotopic Source Assay System (ISAS) has been built to nondestructively assay the fissile and fertile material content of nuclear fuel materials. The ISAS

(described in detail in *Californium-252 Progress, No. 6*) consists of a source collimator assembly and a fission multiplicity detector with associated electronics. Tests with the ISAS are continuing.

Water Pollution Monitor

It is anticipated that a source strength of up to 10 milligrams of californium-252 will be required to develop a system for analyzing pollutants in water. The proposed system will use californium-252 sources for neutron activation analysis, prompt capture, and inelastic neutron scattering techniques. This system will be designed to analyze samples brought to the laboratory from various locations as well as to monitor water from a river or stream on a continuous basis.

BUREAU OF MINES

The Bureau of Mines, U. S. Department of the Interior, is evaluating californium-252 as a neutron source for process control in the coal industry. Californium-252 is being tested for use in a meter that monitors tonnage flows of coal for sulfur content. In this meter, 5.4-Mev sulfur gamma rays from prompt neutron capture reaction must be measured rapidly to provide process control in coal cleaning plants.

Tests have shown that with a californium-252 neutron source the signal-to-noise ratios are higher and the interference is less than with americium and plutonium neutron sources.

For precision measurements, the gamma spectra must be corrected as the iron content of the coal changes. Therefore, 20-pound increments of pyrite (each increment corresponding to a 0.26% sulfur increase in the coal) were added to the 50-ton per hour coal system, and the 5.4-Mev sulfur gamma rays were measured (Figure 17). The increased count rate in the sulfur peak is a linear function of pyrite addition. This measurement gives a precision of 0.05% sulfur.

Additional correction factors are being determined for changes in moisture content and bulk density during monitoring of sulfur, moisture, and bulk density with a single californium-252 source.

FIG. 17 Measurement of Sulfur in Coal

REPUBLIC STEEL CORPORATION

Republic Steel Corporation is evaluating californium-252 for determining important elements in typical steelmaking materials and products. Initial laboratory studies of the detectability of manganese in an iron matrix have been made by neutron inelastic scattering, neutron radiative capture, and neutron activation analysis techniques.* Results of recent tests are reported below.

Neutron Activation Analysis

A 1120.9-microgram source of californium-252 was used for neutron activation analysis of manganese in steels.** Test analyses show that the neutron activation method is a fast and accurate process measurement tool: the accuracy equals that of conventional methods, yet the neutron activation analysis takes less than half the time.

Interference from activation of iron-56 is smaller with californium-252 neutron sources than with other conventional isotopic sources because californium-252 has a lower average neutron energy.

Matrix effect errors can be minimized by a suitable choice of sample irradiation position. Two sets of specially made steels were irradiated at two positions to develop curves to define the limits of matrix effect errors inherent in the analyses. Standards were activated simultaneously with the samples to be used as time bases for performing time-independent activation analysis. All samples were right circular cylinders with a diameter of 2 inches and a length of 1 inch.

* *Californium-252 Progress*, No. 6, 34 (1971).

** S. J. Aron, Jr. "Fast Process Measurement for Manganese in Steels by Neutron Activation Analysis." *Neutron Sources and Applications: Proceedings of the American Nuclear Society National Topical Meeting*, April 19-21, 1971, Augusta, Georgia. USAEC Report CONF-710402 (1971).

Because the composition of most steels falls between the low and high alloy content of samples used for the analyses, the maximum matrix error is specified by the curves in Figures 18 and 19. Figure 18 shows that the matrix errors present in the activation analysis of high and low alloy steels at 10 inches from the californium-252 source are $\pm 8\%$ of the manganese concentration. At 1 inch from the source, the matrix errors are reduced to $\pm 3\%$, as shown in Figure 19.

FIG. 18 Neutron Activation of Steels at 10 inches from the Californium-252 Source

FIG. 19 Neutron Activation of Steels at 1 inch from the Californium-252 Source

In samples of unknown alloy content, manganese can be determined to an accuracy of $\pm 3\%$ of the concentration. Neutron activation analyses of low alloy bar stock and basic oxygen furnace steels agreed to within $\pm 0.02\%$ manganese when compared with analyses by conventional methods (wet chemistry, emission spectroscopy, or X-ray fluorescence).

COLUMBIA SCIENTIFIC INDUSTRIES

The application of californium-252 to discrete sample and on-stream neutron activation analysis is being evaluated by the Columbia Scientific Industries of Austin, Texas. Preliminary data have been obtained on the sensitivity and potential interferences for analyses having applications in industrial process control. Analyses studied include cobalt in process stream solutions; copper, molybdenum, silver, and gold in ore slurries; and calcium in cement raw mix.

The slurry loop used for on-stream neutron activation analysis was described in *Californium-252 Progress*, No. 5, 29, (1970), except that a bypass to the activation cell was installed to allow recirculation and counting of samples without simultaneous irradiation. All slurries contained 12.5% by weight of solids (1 kilogram of solids plus 7 liters of water) because only limited quantities of solids were available; full capacity of the slurry loop is over 50% solids. Because actual analyzed samples did not include a sufficient concentration range of each element to be determined, synthetic samples were prepared.

The activation cell contained 400 micrograms of californium-252 during all measurements.

The sensitivity limits of neutron activation analyses are determined by many analysis conditions, such as sample size, irradiation time, counting time, source strength, and detector efficiency and geometry. These factors must be adjusted for maximum sensitivity under each analysis situation. The limits reported below were observed under the specific conditions described; these limits might be improved under actual analysis situations if the proper conditions are selected.

Determination of Cobalt in Process Streams

Cobalt is a catalyst (from 10 to 10^4 ppm) in some organic and aqueous process streams. The preferred nuclear reaction for activation of cobalt by thermal

neutrons is $^{59}\text{Co}(n,\gamma)^{60\text{m}}\text{Co}$ (gamma energy, 1.33 Mev; half-life, 10.5 min).

Aqueous cobalt nitrate standards containing 40 to 800 ppm cobalt were recirculated for 30 minutes, and 5-minute counts were taken after recirculation for 1 and 20 minutes. The resulting calibration curves are summarized in Figure 20. Background under the 1.33-Mev peak was estimated by counting windows on each side of the peak. The slope of the calibration curve obtained after 20-minute irradiations was 7.8 net counts per ppm cobalt. The standard deviation of the net count was 200 in 5 minutes, equivalent to a detection limit (1 standard deviation) of 25 ppm cobalt. For the measurements one minute after irradiation started, the corresponding detection limit is 40 ppm cobalt.

FIG. 20 Calibration Curves for Cobalt in Aqueous Solution

The sensitivity is suitable for on-stream cobalt determination in solutions containing more than 250 ppm. The sensitivity would be increased in proportion to increases in the source strength provided the background does not increase. The sensitivity could only be doubled by increasing the counting time because of the 10.5-minute half-life of cobalt-60m.

Ore Slurries

An ore matrix simulating the average elemental composition of sedimentary or igneous rocks was prepared by mixing powdered chemicals as follows: SiO_2 , 58.0%; Al_2O_3 , 14.0%; CaCO_3 , 10%; Fe_2O_3 , 5.5%; Na_2CO_3 , 4.5%; K_2CO_3 , 4.2%; MgO , 3.0%; and TiO_2 , 0.8%. Activation of this matrix indicated that the main source of background is from the 1.78-Mev gamma rays from $^{27}\text{Al}(n,\gamma)^{28}\text{Al}$ (and, to a lesser extent, from $^{28}\text{Si}(n,p)^{28}\text{Al}$).

Figure 21 shows the spectrum from a natural ore sample counted for the last 5 minutes of a 7-minute recirculating activation. The ore sample contained 12.7% Cu, 0.1% Mo, 3.1 oz/ton Ag (90 ppm), and 0.03 oz/ton Au (0.9 ppm). Peaks from activation of copper and silver are present, but none from activation of gold or molybdenum. The aluminum-28 peak is prominent; its Compton continuum is a major source of background at lower energy. The aluminum-28 peak in the simulated ore matrix was about twice the intensity of that of the ore in Figure 21, indicating that the sensitivities reported below are conservative. The manganese-56 peak is much more prominent than that from the simulated ore matrix. This may be attributed to a (n, γ) contribution from a small amount of manganese in addition to the (n,p) contribution from the large amount of iron present in the natural ore.

FIG. 21 Gamma Ray Spectrum from Copper-Silver Ore Stream Activated with Californium-252

Determination of Silver. The sensitivity for determination of silver in ores was estimated by recirculating the simulated ore matrix spiked with known quantities of Ag_2O . For the most-sensitive analysis, the activation reaction is $^{109}\text{Ag}(n,\gamma)^{110}\text{Ag}$ (gamma energy, 0.66 Mev; half-life, 24.5 seconds). Recirculating activation time was 2 minutes with a 50-second count started 50 seconds after commencing the activation.

Net counts (after subtracting background) in the 0.66-Mev gamma peak for various Ag_2O contents are shown in Figure 22. The curve slope indicates 5.8 net counts per ppm Ag_2O . The standard deviation on the net count between 0 and 1000 ppm Ag_2O was about 170 counts, equivalent to 30 ppm Ag_2O , or 1 ounce per ton.

FIG. 22 Calibration Curve for Silver in Ore Slurry

The tests indicate that the method is sufficiently sensitive to monitor silver ore.

Determination of Copper and Molybdenum. Copper is activated by $^{65}\text{Cu}(n, \gamma)^{66}\text{Cu}$ (gamma energy, 1.04 Mev; half-life, 5.1 minutes). For molybdenum, the selected reaction is $^{100}\text{Mo}(n, \gamma)^{101}\text{Mo}$; ^{101}Mo decays by emission of 0.19-Mev gamma rays (half-life, 14.6 minutes) to tellurium-101m (gamma energy, 0.31 Mev; half-life, 14 minutes).

Simulated ore matrix samples spiked with CuO and MoO_3 were activated and counted as follows. Ten minutes of recirculating activation was followed by 20 minutes for recirculating decay (using the activation cell bypass). Five-minute counts were taken beginning 1 minute after activation began, and 5 and 14 minutes after commencing the decay cycles. Net counts were obtained for the 1.04-Mev copper peak and the 0.31-Mev molybdenum peak.

Figure 23 shows results for copper. The relative slopes and standard deviations are tabulated below.

<i>Time after Commencing Activation, min</i>	<i>Net Counts per % CuO</i>	<i>Std Dev, % CuO</i>
1	3350	0.13
15	2800	0.09
24	1100	0.20

The earliest count shows a relatively high background due to aluminum-28 activity (note that net counts are plotted in Figure 23). The count begun after 5 minutes delay represents near optimum sensitivity because of the more rapid decay of aluminum-

FIG. 23 Calibration Curves for Copper in Ore Slurry

28, but the count begun after 14-minutes delay was less sensitive because the copper-66 had also decayed.

These results show that the method is sufficiently sensitive to determine copper in ore processing, and the sensitivity could be readily increased.

Results for molybdenum are shown in Figure 24. Only the counts after 5- and 14-minutes delay were reasonable. Background could not be accurately subtracted because the lower background window included part of the 0.19-Mev molybdenum-101 peak. The results would have been better if the spectrometer

FIG. 24 Calibration Curves for Molybdenum in Ore Slurry

gain and bias settings had been changed to count both the 0.19- and 0.31-Mev peaks as signal, and background windows had been accessible both above and below the pair of peaks. Although these conditions could not be obtained conveniently in the present tests, the results do allow evaluation of the sensitivity for molybdenum determination.

The slopes of the calibrations, and standard deviations, are tabulated below.

<i>Time after Commencing Activation, min</i>	<i>Net Counts per % MoO₃</i>	<i>Std Dev, % MoO₃</i>
15	1700	0.17
24	1550	0.14

The sensitivity observed was inadequate for monitoring molybdenum in ore processing streams, where the detection limit must be <0.002% molybdenum.

Determination of Gold. The selected reaction for gold determination is $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ (gamma energy, 0.41 Mev; half-life, 2.7 days). The long half-life permits long activation, decay, and counting times, all of which can increase sensitivity. However, an analysis must take less than one day, or classical methods would be more practical.

Artificial ore slurries with up to 5500 ppm gold added were recirculated for just over 20 minutes each, and a 10-minute count was started after 10 minutes. Measurement of the 0.41-Mev gamma ray peak produced a linear calibration with a standard deviation of 400 ppm gold.

Additional studies are needed to determine the conditions for maximum sensitivity for gold determination. The sensitivity might be increased to 10 ppm in a reasonable measurement time. Although this sensitivity is inadequate for analysis of gold ore, it may be adequate for analysis of process streams and plating bath solutions.

Determination of Calcium in Cement Raw Mix

One of the most important potential applications of on-stream neutron activation analysis is to the determination of "basicity ratio" in iron ore processing and cement production. This involves deter-

mination of calcium and silicon, and possibly of iron, aluminum, and magnesium.

The key analysis is the calcium determination. Despite interference from the light pulser peak (equivalent energy, 2.8 Mev), a preliminary study was begun of the determination of calcium using the reaction $^{48}\text{Ca}(n,\gamma)^{49}\text{Ca}$ (gamma energy, 3.1 Mev; half-life, 8.8 minutes).

Using a simulated cement raw mix slurry (12.5% solids composed of 80.5% CaCO_3 , 14.0% SiO_2 , 4% Al_2O_3 , and 1.5% Fe_2O_3), the statistical accuracy for counting the 3.1-Mev peak was estimated to be 1.0% CaCO_3 (in the absence of the pulser peak) in a 5-minute count after a 24-minute recirculating activation. Investigations will continue after the detector is modified to separate the pulser peak from the calcium-49 peak.

NATIONAL BUREAU OF STANDARDS

The National Bureau of Standards of the U. S. Department of Commerce is using eight 100-microgram californium-252 sources to investigate neutron activation analysis and its use in on-stream process analysis.

Storage and Irradiation Container

The californium-252 sources arranged in an octagon are stored in a 3 x 3 x 7-foot aluminum tank filled with demineralized water (Figure 25). The tank is surrounded by approximately 12 inches of paraffin, 4 inches of lead, and 8 inches of high-density concrete. The octagonal source array produces a homogeneous flux (Figure 26) for irradiation with a net neutron loss of approximately 10% as opposed to a single source containing 800 micrograms of californium-252. The loss is probably due to the capture of reflected neutrons in the encapsulation material of neighboring sources because a four-source configuration gave negligible neutron losses.

The sample in a 2-dram polyethylene vial, 14 millimeters in diameter and 48 millimeters in height, is propelled into the irradiation position using nitrogen, is irradiated, and then is returned and counted between two matched 3 x 4-inch sodium iodide detectors. Transfer time is less than 1 second.

FIG. 25 Californium-252 Storage and Irradiation Container

FIG. 26 Vertical Flux Distribution in Californium-252 Facility Measured with Copper Foils and with Source at Various Distances from the Sample Holder

Reproducibility of the system is excellent. A series of 10 analyses on a sample of bis(1-phenyl-1, 3 butanedione) oxovanadium (IV) (NBS Standard Reference Material 1052a) showed a standard deviation of less than 0.1%.

Fluorine Analysis

Fast neutron activation analysis was used to determine fluorine in many materials by measuring the short half-life isotope nitrogen-16 produced by the $^{19}\text{F}(n, \alpha)^{16}\text{N}$ reaction.

A weighed sample was irradiated for 30 seconds; after a 2-second delay, the nitrogen-16 gamma rays were counted for 60 seconds. One gram of fluorine

gave about 16,000 counts of gamma activity. The samples were irradiated and counted ten times with a standard deviation of about 0.1% for fluorine determination (Table VII).

TABLE VII

Specific Activities of Fluorine Using Californium-252

Sample	Sample Size, g	Calculated Fluorine Content, g	^{16}N Total Counts	Specific Activity, counts/mg F
p-Fluorobenzoic Acid	1.3142	0.1782	28,580	160.4
NaF^a	1.9450	0.8801	144,865	164.4
LiF^b	1.2110	0.8870	146,532	165.2
LiF^b	1.2110	0.8870	145,853	164.4
Decafluorobiphenyl	2.0082	1.1419	185,617	161.7

^a Certified A.C.S. (Fisher)

^b Fisher, Certified Reagent (Fisher)

AMERICAN SCIENCE AND ENGINEERING, INC.

A 12-microgram californium-252 source is being evaluated for measuring the sulfur content of metallurgical-grade coal by prompt gamma activation analysis. Prompt gamma spectra of metallurgical-grade coal have been obtained with the coal analysis system, the californium-252 source, and a lithium-drifted germanium detector.

Coal of very low sulfur content is being analyzed to determine the feasibility of measuring the ash content with californium-252. The sulfur content is low enough to be ignored, but the large and variable amounts of ash have caused problems with customers. Initial measurements disclosed no chlorine in the coal (chlorine masks some of the ash peaks). Additional studies are determining whether the ash composition is sufficiently uniform to permit assumptions to be made about the silicon content. The uniformity of the ash constituents also affects the data analysis procedures.

KANSAS STATE UNIVERSITY

Kansas State University will evaluate the application of californium-252 in the following:

- Protein analysis of cereal grains by neutron activation.
- A study of electronic materials and components in nuclear radiation environments.

- Quantitative analysis for mercury and other selected toxic materials in commercially processed foods for humans. These evaluations will be made with 5 californium-252 sources totaling 50 milligrams.

Facilities have been built for storing 50 milligrams of californium-252 and for irradiating samples. These facilities include a storage tank, traversing mechanism, and sample entry mechanism.

Storage Tank

The sources will be used and stored at the bottom of an 8-foot-long, 5-inch-diameter stainless steel pipe. The pipe is attached to the bottom of a 4-foot-diameter, 5-foot-high stainless steel tank (Figure 27). The tank is filled with water and has an attached circulating system to deionize the water. The tank was tested for leaks and then positioned in its permanent location (Figure 28).

FIG. 27 Californium-252 Source Storage Tank

FIG. 28 Californium-252 Storage Pit

Source Thimble and Traversing Mechanism

The central thimble is used for sample entry to the sources (Figure 29). The bottom 6-foot section of the thimble is made of acrylonitrile butadiene styrene (1-1/4-inch plastic pipe); the top 6-foot section is made of polyvinyl chloride. The source holder (1-inch polyethylene plate) is attached to the central thimble. The top of the thimble is guided and clamped into place by a three-pronged laboratory clamp. The fitting at the 6-foot level prevents movement of the central thimble past this clamp.

The thimble will be clamped to the traversing mechanism (Figure 30) on the vertical movement bar, which will traverse 3 feet. The clamps will have to be reset before further movement is possible. The mechanism shuts off automatically if the 3-foot limit is exceeded. Two traverses will raise the sources to the 6-foot level.

FIG. 29 Central Source Thimble in Storage Tank

FIG. 30 Traversing Mechanism on Storage Tank

Sample Handling Mechanism

Samples will be injected into the system in 1-1/8-inch plastic vials. A remote control device is being constructed for sample entry and removal. The sources can be raised to the bottom of the tank if the size of the sample prevents the use of the central thimble. The thimble is removable to allow changes from wet to dry sample entry condition.

UNIVERSITY OF UTAH

The University of Utah, Salt Lake City, will use three 5-milligram californium-252 sources to evaluate the application of californium-252 neutron sources in exploration and process control in mining, milling, and refining of copper products and byproducts; for activation analysis and neutron radiography in a university-related program; and as a laboratory teaching device. The evaluation will be directed by G. M. Sandquist, Director of the Nuclear Engineering Program.

The copper program, a cooperative effort with the Kennecott Copper Research Center, Salt Lake City, will include determination of copper and iron concentration profiles in cased boreholes in Kennecott overburden dumps, and the evaluation of californium-252 as a neutron source for activation analysis in development of a method for rapid on-line process control of copper ore feed composition to the mill.

The activation analysis and neutron radiography program will include development of techniques suitable for use in a center to serve the general research and industrial community and evaluation of californium-252 for neutron activation analysis for trace element survey in geological and biomedical fields.

Evaluations of californium-252 as a teaching device will include laboratory studies in nuclear engineering and related fields and comparison of the source with the low-power nuclear reactor at the University of Utah.

Other Application Developments

DOSIMETRY

SAVANNAH RIVER LABORATORY

The radiation dose rate from a californium-252 needle designed for radiotherapy has been calculated using the ANISN code (discrete ordinates method of approximating the solution of the energy-dependent linear Boltzmann transport equation with anisotropic scattering).* Earlier published dose rates** were hand-calculated. The source was assumed to be a 1-microgram californium-252 needle with a diameter of 0.165 centimeter and an active length of 2 centimeters, embedded in a 20-centimeter-diameter sphere of tissue.

The neutron and gamma ray energies from the californium-252 source were divided into 22 energy groups. The flux density for each group was calculated as a function of tissue depth (Figures 31-33).

FIG. 31 Flux Density per Microgram from a 20-mm Needle as a Function of Tissue Depth for Neutron Energy Groups 1 through 8

The absorbed dose rate was then obtained by multiplying the flux density times the Kerma rate factor. In Figures 34 and 35, the calculated dose rates are compared with measured dose rates from a californium-252 needle immersed in a tissue-equivalent solution.

FIG. 32 Flux Density per Microgram from a 20-mm Needle as a Function of Tissue Depth for Neutron Energy Groups 9 through 13

* D. H. Stoddard. "Calculated Dose Rate from ²⁵²Cf Radiotherapy Source." Proposed for publication.

** C. S. Schlea and D. H. Stoddard. "Californium Isotopes Proposed for Intracavity and Interstitial Radiation Therapy with Neutrons." *Nature* 206, 1058 (1965).

FIG. 33 Flux Density per Microgram from a 20-mm Needle as a Function of Tissue Depth for Neutron Energy Groups 14 through 22

FIG. 34 Distribution of Fast Neutron Dose Rate from Californium-252 as a Function of Tissue Depth

FIG. 35 Distribution of Gamma Dose Rate from Californium-252 as a Function of Tissue Depth

NEUTRON RADIOGRAPHY

BATTELLE-NORTHWEST

Under a program sponsored by the USAEC Division of Isotopes Development, Battelle-Northwest is studying the use of californium-252 for thermal neutron radiography. A portable storage shield and radiographic device* has been designed and constructed. The device, exclusive of its stand, weighs about 350 pounds, which is light compared to the weight of a conventional neutron source shield. Calculations to determine the optimum moderator and imaging materials were described in *Californium-252 Progress*, No. 3, 25 (1970).

Radiographic Device

The storage and radiographic device is shown in Figure 36. The outer, egg-shaped shell (26 inches on the major axis and 22 inches on the minor axis) is a two-piece container of laminated fiberglass with a minimum wall thickness of 0.500 inch. The halves are connected by 1-3/4-inch-wide flanges. A 5-inch-diameter beam port in the small end accepts a moderator and collimator holder.

Laminated fiberglass (Fabmat-2415) was used because of its light weight, good physical strength, self-extinguishing fire characteristics, ease of fabrication, cheapness, and durability. A comparable thickness of stainless steel would be much more expensive, heavier, and more difficult to fabricate in the chosen design.

The device (Figure 37) can be used for both storage and radiography by remotely shifting the position of the californium-252 source within the container. The source is located on the periphery of a drum within the outer shield. By rotating the drum through a 180° arc on a vertical axis, the source is positioned either near the beam port so neutrons can be extracted and selectively moderated for radiography, or near the middle of the major axis of shielding for storage.

The source drum is rotated with a remotely controlled reversible motor. An electromagnetic brake and limit switches control the positioning of the source drum. The control panel indicates the position

FIG. 36 Storage and Radiographic Device

FIG. 37 Storage and Radiographic Device Showing Californium-252 Source in Radiographic Position

* J. L. Cason. "Design and Fabrication of a Portable Device for Neutron Radiography." USAEC Report BNWL-1329, Battelle-Memorial Institute, Pacific Northwest Laboratories, Richland, Washington (1971).

of the source and the relative dose rate near the container, and has provisions for interlocks and warning lights. When power is not available, the source drum can be moved into its exposure position with a manual crank.

Shielding Design

In determining the optimum geometry and materials for shielding californium-252, six neutron shielding materials were evaluated: zirconium hydride, titanium hydride, paraffin, gadolinium oxide, cadmium, and boron carbide-epoxy resin compound (approximately 4 parts boron carbide to 1 part resin by weight). The boron carbide-epoxy resin (B-E) compound was more effective for fast neutrons and more versatile than paraffin, water, or any of the metal hydrides evaluated. The hydrogen in the epoxy resin moderated the fast neutrons (half-value layer of 1 inch for fast neutrons), and the boron carbide captured the thermal neutrons. The neutron dose rate versus shielding thickness for the B-E compound is given in Figure 38. The B-E compound was easy to mold or cast in any shape or size without special equipment.

Gamma shielding was determined by considering: 1) the inverse square principle in determining the dimensions; 2) the maximum gamma shielding obtainable from the neutron shielding and moderator materials; and 3) the use of a small volume of depleted uranium rather than a larger volume of lead.

Table VIII gives the approximate thickness of some of the materials in the shield.

With a 268-microgram californium-252 source in the storage position, the neutron dose rate at the surface is 200 mrem/hr and the gamma dose rate is 50 mR/hr. At three feet from the surface, the dose rate drops to about 50 mrem/hr total. In the radiographic position, a collimated thermal neutron beam of approximately 1×10^4 neutrons/(cm²-sec) is obtained.

FIG. 38 Shielding Effectiveness of B-E Compound for Californium-252

TABLE VIII

Shielding Material Thicknesses in Radiographic Facility

<u>Material</u>	<u>Effective Thickness, cm</u>	<u>Radiation Shielded</u>
ZrH ₂	0.82	Fast Neutron & Gamma (Neutron Moderator & Reflector)
Be	1.91	Thermal Neutrons (Neutron Moderator & Reflector)
UO ₂	0.64	Gamma
B-E	7.6	Fast & Thermal Neutrons
CH ₂ (paraffin)	15.0	Neutrons
Fiberglass	1.27	Neutrons

IDAHO NUCLEAR CORPORATION

Idaho Nuclear Corporation designed and built a multipurpose storage container (Figure 39) for Sandia Corporation's 2-milligram californium-252 source. This container not only stores the source, but also can function as a radiation facility without the source being removed. For radiography, a neutron beam can be extracted through a large beam hole; for activation analysis, samples can be exposed by inserting them near the source via either of two small beam holes.

The 5-foot-diameter, 5-foot-long cylindrical container consists of a large outer tank filled with a saturated aqueous solution of boric acid, and a small inner tank containing unborated water (Figure 40). When filled, the storage container weighs about 7000 pounds.

The source, attached to the end of a "Lucite"* rod which in turn is mounted on the end of a borated-polyethylene plug, is centrally positioned within the storage container. Radiation dose rates from a 2-milligram source were measured with conventional health physics instruments at the outer surface of the container. The gamma dose rate (measured with an Eberline Model E510 Geiger-Müller counter) plus the fast neutron dose rate (measured with a Fairport Model 420 neutron dosimeter counter) totaled less than 25 mrem/hour. A thermal neutron flux of only 30 neutrons/(cm²-sec), equivalent to a dose rate of about 0.1 mrem/hour, was measured with a PAC-4S slow neutron counter.

The main beam hole, constructed of stepped aluminum pipe, allows insertion of a divergent collimator. The collimator is made of polyethylene containing 1 to 2 weight % boron and is lined with 0.020 inch of cadmium. A removable piece of unborated polyethylene is located at the inlet of the collimator, adjacent to the source. Two smaller beam holes are also available. All beam holes are plugged with borated polyethylene when not in use.

Thermal neutron radiographs can be made by replacing the large beam hole plug of borated polyethylene with the divergent collimator, and aligning the object and detector foil in front of the beam

FIG. 39 Californium-252 Storage and Radiographic Container (The open sliding door by the collimator exit and one of the smaller beam ports (uncovered) are shown. The fill pipes for the inner and outer tanks are visible at the top of the container.)

FIG. 40 Californium-252 Storage and Radiographic Container

port. Objects and detector foils can be changed while most of the neutron beam has been cut off by sliding the 1/4-inch boral door over the collimator exit.

Fast neutron radiographs can be made by removing the unborated polyethylene moderator from in front of the collimator inlet and draining the unborated water from the inner tank.

* Trademark of Du Pont

Samples for neutron activation analysis can be exposed to maximum neutron fluxes by inserting them close to the source through either of the smaller beam holes. These smaller ports can also be used for neutron beam studies.

SAVANNAH RIVER LABORATORY

The shielding capabilities of water-extended polyester (WEP®) and glycerin for californium-252 neutron and gamma radiation have been calculated with the ANISN code.*

WEP shields have many advantages: ease in preparation, low cost, solid form, strength, high water content, and resistance to fire. WEP resins are already in use at the M. D. Anderson Hospital and Tumor Institute for a californium-252 source transport shield and a bedside shield to protect attendants and visitors of patients with californium-252 implants.** The University of Texas at Austin used a WEP resin as the shielding material in their californium-252 storage and experimental facility.† Two shipping casks of WEP-60 (60% water) are being fabricated at the Savannah River Laboratory.

The calculated attenuation of neutron and gamma radiation provided by spherical shields of WEP-65 (65% H₂O) is shown in Figure 41. Measured neutron transmission curves for WEP resins have already been published.‡

Glycerin (C₃H₈O₃) makes an even better californium-252 shield than WEP resins, according to the calculated attenuation properties shown in Figure 42. However, because of low densities, both WEP-65 (1.1 gram/cm³) and glycerin (1.26 gram/cm³) may require additional shielding by a more-dense material to reduce the gamma radiation dose rate from larger sources.

Boron Addition

Interaction of neutrons with hydrogen atoms in any hydrogenous shield releases 2.2-Mev secondary gamma rays, which may in some cases be the predominant contributor to the penetrating dose. Production of this energetic gamma radiation by the H(n,γ)D reaction can be partially suppressed by adding boron to the hydrogenous shield; the com-

FIG. 41 Dose Rates in an Infinite Medium of Water-Extended Polyester (65% H₂O) from a Point Source of Californium-252

® Trademark of Ashland Chemical Company

* D. H. Stoddard and H. E. Hootman. *²⁵²Cf Shielding Guide*. USAEC Report DP-1246, E. I. du Pont de Nemours and Company, Savannah River Laboratory, Aiken, South Carolina (1971).

** *Californium-252 Progress*, No. 1, 19 (1969) and No. 5, 13 (1970).

† S. J. Gage, E. L. Draper, Jr., G. D. Bouchev, and R. R. Day. "Design and Construction of a Versatile Cf-252 Neutron Source Shield and Experimental Facility." *Neutron Sources and Applications: Proceedings of the American Nuclear Society National Topical Meeting, April 19-21, 1971, Augusta, Georgia*. USAEC Report CONF-710402 (1971).

‡ G. D. Oliver, Jr. and E. B. Moore. "The Neutron Shielding Qualities of Water-Extended-Polyesters." *Health Physics* 19, 578 (1970).

FIG. 42 Dose Rates in Glycerin from a Point Source of Californium-252

peting reaction, $^{10}\text{B}(n, \alpha)^7\text{Li}$, is accompanied by more easily attenuated 0.5-Mev gamma rays. For this purpose, boron compounds such as boric acid can be readily dissolved and uniformly distributed in WEP resins and in glycerin. One evaluator, the Kerr-McGee Corporation, uses an 18% solution of boric acid in glycerin (page 24) to shield a 1-milligram californium-252 source.

Dose Rate Calculation

The total dose rate at the surface of a spherical shield of WEP-65 or glycerin, without any dissolved boron and without any supplementary iron or lead shields, can be determined from Figure 41 or Figure 42 as follows:

- Add the fast neutron and the thermal neutron dose rates (shown in the figures) corresponding to the shield radius $r(\text{cm})$ to obtain the total neutron dose rate D_n .
- Add the prompt gamma and the capture gamma dose rates at this same shield radius to obtain the total gamma dose rate D_γ .
- Add D_n to D_γ , and multiply by the actual source weight (milligrams of californium-252) and by 2.3×10^9 neutrons/(second-milligram).
- Divide by $4\pi r^2$ to obtain the total dose rate in mrem/hour.

The reduction in the capture-gamma dose rate brought about by adding boron to a hydrogenous shield and the effect of adding iron or lead shields to the inside or outside of a hydrogenous shield can be readily estimated.*

* H. E. Hootman. *Estimation of ^{252}Cf Shielding Requirements*. USAEC Report DP-1232. E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, South Carolina (1970); *Californium-252 Progress*, No. 2, 36 (1970); No. 4, 28 (1970); and No. 5, 39 (1970).

LOS ALAMOS SCIENTIFIC LABORATORY

Six common moderating materials have been compared in studies to optimize a californium-252 moderator system (see page 44) for the assay of low-enrichment reactor fuel rods.* Previously, calculations and measurements** have shown that this system could measure the fissile material content in reactor fuel rods (e.g., UHTREX rods†).

The six moderating materials are CH_2 , ZrH_2 , and H_2O , which have large slowing down powers ($\xi\Sigma_s$), and Be, C, and D_2O , which have large moderating ratios ($\xi\Sigma_s/\Sigma_a$). The slowing down power is a measure of how well a moderator slows down neutrons, while the moderating ratio represents the ability of a moderator to slow down neutrons without capturing them.

The thermal neutron flux versus radius for a californium-252 source in 30-centimeter-radius spheres of five moderators (Figure 43) was calculated with the DTF-IV code and 25-group cross sections. The CH_2 and H_2O produce very similar thermal flux spectra because of their similar H_2 densities. The neutron flux peaks at the center and drops off rapidly with increasing radius resulting in a rather large flux gradient at interior positions for sample irradiations. ZrH_2 yields a similar curve for thermal flux versus radius. The moderators with the largest moderating

FIG. 43 Calculated Thermal Neutron Flux from a Californium-252 Source in a One-Region 30-cm-radius Spherical Assembly for Five Moderators

ratios yield much flatter thermal fluxes as a function of radius because of less thermal absorption. Beryllium yields a larger flux than D_2O or C because of its larger macroscopic thermal scattering cross section. Figure 43 shows that both Be and D_2O are more suitable than the other moderators for thermal neutron irradiations involving energy-biased detectors which must be suitably displaced from the high energy ($\bar{E}_n \sim 2$ Mev) californium-252 source neutrons. This is illustrated in Figure 44 which shows calculated $^{235}\text{U}/^{238}\text{U}$ ratios, assuming equal masses, versus radius for the various moderators. These large

FIG. 44 Calculated $^{235}\text{U}/^{238}\text{U}$ Fission Ratio for a Californium-252 Source in a One-Region 30-cm-radius Spherical Assembly for Five Moderators (Equal masses of both isotopes)

* R. A. Forster and H. O. Menlove. "Moderators for ^{252}Cf Assay Systems." In *Nuclear Safeguards Research and Development, Program Status Report, September-December, 1970*. G. R. Keepin (Ed.) USAEC Report LA-4605-MS, Los Alamos Scientific Laboratory, Los Alamos, New Mexico, 8 (1971).

** R. A. Forster and H. O. Menlove. "Neutron Source Moderator-Shield Assemblies." In *Nuclear Safeguards Research and Development, Program Status Report, May-August, 1970*. G. R. Keepin (Ed.) USAEC Report LA-4523-MS, Los Alamos Scientific Laboratory, Los Alamos, New Mexico, 11-13 (1970).

† H. O. Menlove, J. L. Parker, and H. A. Walter. "Assay of Hot UHTREX Fuel Rods Using ^{252}Cf ." In *Nuclear Safeguards Research and Development, Program Status Report, May-August, 1970*. G. R. Keepin (Ed.) USAEC Report LA-4523-MS, Los Alamos Scientific Laboratory, Los Alamos, New Mexico, 14-15 (1970).

FIG. 45 Calculated Thermal Neutron Fluxes from a Californium-252 Source in a Composite Moderator Assembly with Various Moderators in the Middle Region

$^{235}\text{U}/^{238}\text{U}$ ratios ($>60,000$) show that the fissile component (^{235}U) can be preferentially interrogated in low enrichment or natural uranium. Also, the $^{235}\text{U}/^{238}\text{U}$ ratio is proportional to the signal-to-background ratio for a detector biased at the ^{238}U fission threshold. At radii greater than 12 centimeters, Be and D_2O yield $^{235}\text{U}/^{238}\text{U}$ ratios greater than the hydrogenous moderators. Carbon does poorly because neutrons do not lose much energy per collision.

One method to increase both the thermal flux and $^{235}\text{U}/^{238}\text{U}$ ratios in the interior of an assembly of a given volume is to surround the californium-252 source with a small sphere of material with a high slowing down power such as CH_2 . This hydrogenous moderator softens the neutron spectrum more than would the same volume of high moderating ratio material; however, because of the small volume fraction of the hydrogenous core, the thermal neutron capture in the hydrogen is minimal. In addition, surrounding the entire moderating assembly with a layer of CH_2 increases the thermal flux throughout the assembly as well as provides exterior neutron shielding.

Figures 45 and 46 show calculations of the thermal flux and $^{235}\text{U}/^{238}\text{U}$ ratios versus radius for some moderator systems composed of CH_2 in the center and periphery, and either D_2O , C, or Be in the intermediate region. The outer radii of the three regions are 5, 20, and 30 centimeters, respectively. The radius of the inner sphere was chosen as 5 centimeters to maximize the thermal flux in the outer regions of the moderator. The thermal fluxes in Figure 45 for both C and D_2O in the range 5 to 20 centimeters are greatly enhanced by this technique, while the flux in Be remains essentially the same. At a radial position of 20 centimeters, the thermal flux in the CH_2 - D_2O system is slightly greater than the flux at the same position in the pure Be assembly. Figure 46 shows that the largest change caused by the composite moderators is in the $^{235}\text{U}/^{238}\text{U}$ ratio. Both C and D_2O yield much larger ratios for the composite systems, while the ratio for Be remains essentially unchanged. Therefore, such composite assemblies represent a significant improvement over a one-region moderator for certain assay applications.

FIG. 46 Calculated $^{235}\text{U}/^{238}\text{U}$ Fission Ratios for a Californium-252 Source in a Composite Moderator Assembly with Various Moderators in the Middle Region (Equal masses of both isotopes)

LOS ALAMOS SCIENTIFIC LABORATORY

A reactor fuel rod assay system using californium-252 has been designed* from moderator calculations and measurements discussed on page 42. The fissile content of low enrichment fuel rods for power reactors is measured by irradiating the fuel with moderated neutrons from the californium-252 source, counting the prompt neutrons from the induced fission reactions with energy-biased fast-neutron detectors, and comparing the results with standard measurements. The system is limited to material with low fissile content density because of the self-absorption of thermal neutrons in the fuel.

The cylindrical assay device (Figure 47) has the californium-252 source located at the center and ^4He -filled fast-neutron detectors located near the sample holes. The californium-252 source is surrounded by a 2-centimeter-thick tungsten** shell to take advantage of the large cross section of W for inelastic neutron scattering and also to shield the gamma rays emitted in the spontaneous fission of the californium-252. The adjacent layer of ZrH_2 (4 centimeters thick) further moderates the source neutrons by virtue of its large slowing down power ($\xi\Sigma_s$) (i.e., the ZrH_2 performs the same moderator function as the CH_2 core in Figure 45). In addition, the Zr helps reduce the fast neutron flux by inelastic scattering and shield gamma rays. The central hydrogenous core (ZrH_2 plus $\sim 20\% \text{CH}_2$) was held to a 6-centimeter-outer radius because the capture of thermal neutrons by the hydrogen becomes excessive if more hydrogen is added to the central region. The surrounding shell of D_2O (14 centimeters thick) further moderates the source neutrons without significant capture of the thermalized neutrons. The outer layer of CH_2 serves primarily as a neutron reflector and also provides additional neutron shielding at the exterior of the system. The Li added to the thin layer of polyvinyl chloride (PVC) captures a large fraction ($>80\%$) of the thermal neutrons prior to their leakage into the outer shield. This reduces the exterior gamma ray dose because, unlike neutron capture in B, the capture of neutrons in Li does not produce secondary gamma rays.

FIG. 47 Cylindrical Composite Moderating Assembly for Assaying Power Reactor Fuel Rods

Reentrant holes to accommodate the samples to be assayed (i.e., fuel pins or small samples) are located in the outer cylindrical layer of CH_2 at a radius of 22 centimeters (Figure 47), which is roughly the position where the signal-to-background ratio reaches its maximum, as indicated in Figure 46.

* H. O. Menlove and R. A. Forster. "Design of a Reactor Fuel Rod Assay System." In *Nuclear Safeguards Research and Development, Program Status Report, September-December, 1970*. G. R. Keepin (Ed.) USAEC Report LA-4605-MS, Los Alamos Scientific Laboratory, Los Alamos, New Mexico, 10 (1971).

** The tungsten was in the form of an alloy containing 90% W, 6% Ni, and 4% Cu.

Helium-4 gas tubes* were selected as the fast-neutron detectors because of: (1) favorable efficiency for detecting fission spectrum neutrons, (2) low sensitivity to gamma rays, (3) electronic stability and simplicity, and (4) close geometrical coupling with fuel rods. The results of the signal-to-background ratio at the sample position as a function of bias energy of the ^4He are shown in Figure 48. Several different core configurations are shown for comparison. The signal-to-background is increased $\sim 10\%$ and 20% by using ZrH_2 in place of CH_2 and D_2O , respectively, in the core. Note that the signal-to-background ratio can be further increased by enlarging the outer radius of the D_2O layer (thus increasing the volume of the assay device). However, the cost of the additional D_2O and other materials increases at a much faster rate than does the signal-to-background ratio.

In the design of the fuel rod assay system, six sample holes are included (Figure 47) to increase the assay capacity of the device. All sample holes can be used simultaneously with negligible crosstalk ($<1\%$) between adjacent rods because of the shielding effect of the intervening moderator and the energy bias in the ^4He detectors. The detectors are biased such that the signal-to-background ratio is maximized; this corresponds to a ^4He recoil energy of ~ 0.25 Mev or an incident neutron energy of ~ 0.4 Mev as can be seen in Figure 48. The counting rate for a typical 4% enriched fuel rod is calculated to be ~ 1000 cps for a californium-252 source of about 200 micrograms. Thus, it should be possible to measure the total ^{235}U content of six fuel rods in less than one minute to a counting statistical accuracy of better than 1%.

In addition to the assay of reactor fuel rods and pellets, the californium-252 moderator system can be used to assay small uranium or plutonium samples for which the fissile density is not too large for adequate penetration by thermal neutrons. To demonstrate this capability, small uranium-235-thorium carbide inventory samples were assayed for the ^{235}U content. The samples, packaged in 0.5-inch-diameter vials, contained a large fraction of carbon and thorium relative to the uranium (93% enriched in ^{235}U) so that thermal neutron absorption was negligible for all but the heaviest sample (Sample E).

The californium-252 assay results are listed in Table IX together with the results using other non-destructive assay techniques. The californium-252 data agree very well with the more accurate delayed neutron assay results using Van de Graaff neutron interrogation.**

FIG. 48 Signal-to-Background Ratios at the Sample Position Calculated for Helium-4 Detector Bias Energies and Various Moderators in the Inner 5-cm Region Surrounding the Californium-252 Source (The uranium-235 sample was in the 10-cm-thick CH_2 shell at a radius of 22 cm with D_2O in the region from 5 to 20 cm)

The fuel rod assay system described has been constructed at Los Alamos Scientific Laboratory and will be evaluated in the plant production lines of two major U. S. manufacturers of power reactor fuel elements.

TABLE IX
Nondestructive Assay of ^{235}U -Thorium Carbide Samples

Sample	Delayed Neutron Assay ^a	^{252}Cf Assay	Gamma Ray Assay ^b
A	0.148 $\pm$ 0.003	0.147 $\pm$ 0.006	0.131 $\pm$ 0.009
B	0.441 $\pm$ 0.005	0.438 $\pm$ 0.018	0.415 $\pm$ 0.026
C	0.212 $\pm$ 0.003	0.202 $\pm$ 0.008	0.188 $\pm$ 0.011
D	0.383 $\pm$ 0.004	0.406 $\pm$ 0.016	0.368 $\pm$ 0.028
E	0.905 $\pm$ 0.010	...	0.887 $\pm$ 0.098
F	0.328 $\pm$ 0.004	0.333 $\pm$ 0.013	0.331 $\pm$ 0.025

^a Delayed-neutron assay with Van de Graaff using subthreshold neutron interrogation.
^b 185-keV ^{235}U gamma ray assay.

* H. O. Menlove, R. A. Forster, R. H. Augustson, A. E. Evans, and R. B. Walton. "Characteristics of ^4He -Gas Tubes for Fast-Neutron Detection." In *Nuclear Safeguards Research and Development, Program Status Report, September-December, 1970*. G. R. Keepin (Ed.) USAEC Report LA-4605-MS, Los Alamos Scientific Laboratory, Los Alamos, New Mexico, 13 (1971).

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TECHNICAL UNIVERSITY HANNOVER

In cooperation with the Nuclear Research Center, Karlsruhe, Germany, a program has been started to evaluate the use of californium-252 as a randomly pulsed neutron source for reactor-noise investigations. The main goal of this program is to design and to develop an on-line reactivity meter as an experimental tool in determining shutdown reactivity of fast reactors.*

The measuring system consists primarily of two ionization chambers in the current mode of operation which are located in the reactor system under investigation. The first chamber contains a layer of californium-252 and a neutron insensitive filling gas (e.g., argon). Each spontaneous fission of the californium-252 produces an ionization pulse by the fission products in the gas and establishes the time at which neutrons are injected into the assembly where they initiate fission chains. The second neutron sensitive chamber contains helium-3 as the target gas and monitors the neutronic response of the reactor system.

By working out the theory it can be shown* that the cross-power spectral density (CPSD) between the two fluctuating chamber output signals in the frequency range $\lambda_i \ll \omega \ll a$ (plateau region of the source transfer function) can be given by the simple relationship

$$\text{CPSD}(\lambda_i \ll \omega \ll a) = \frac{\text{const.}}{1 - \$}$$

where

- λ_i = decay constant of the i^{th} precursor group
- a = $a_c (1 - \$)$ = prompt neutron decay constant, sec^{-1}
- a_c = β / l = prompt neutron decay constant at delayed critical, sec^{-1}
- β, l = fraction of delayed neutrons and prompt neutron lifetime, respectively
- $\$ \leq 0$ = reactivity of the reactor system, dollars

The CPSD will be measured using two ac-amplifiers including bandfilters, an analog multiplier, and an active RC-smoothing network. The whole equipment must be calibrated only once at delayed critical ($\$ = 0$) to determine the constant in the previous equation. Thus, the reactivity of the reactor system under investigation can be directly monitored on an analog voltmeter scaled in dollars.

* W. Seifritz. "Ein On-line Reaktivitätsmeter für schnelle Reaktoren mit Hilfe von Cf^{252} -Neutronen." Paper to be presented at the Reaktortagung des deutschen Atomforums am 30. März - 2. April 1971, Bonn, Germany.

The specific problems concerning the signal-to-noise ratios in the two channels which affect the statistical precision of the reactivity measurement have been solved satisfactorily: the uncorrelated alpha particles from the alpha decay in the californium chamber affect adversely the signal-to-noise ratio in the first channel. However, it could be shown that due to the Campbell effect and due to the geometrical discrimination by a specially designed geometrical shape of the chamber, the contribution from the alpha particles to the current fluctuations are suppressed sufficiently. The signal-to-noise ratio in the second channel is determined mainly by the detector efficiency of the neutron sensitive chamber (given in counts per fission in the reactor), the reactivity of the reactor system, and the ratio of the (importance weighted) californium source strength to the effective source strength of the inherent neutron sources of the reactor (e.g., caused by spontaneous fissions of plutonium-240 and by (a,n) reactions in a plutonium-oxide-fueled system).

For a typical plutonium-oxide-fueled fast breeder reactor ($a_c = 10^4 \text{ sec}^{-1}$) possessing an effective inherent neutron source strength of about 10^8 n/sec , about 200 micrograms of californium-252 in the californium chamber is a sufficient amount of this material resulting in a neutron yield of $\approx 5 \times 10^8$ prompt californium neutrons per second. Under the realistic assumption of the detector efficiency of the second chamber of $w = 5 \times 10^{-5}$ counts per fission a shutdown reactivity of -5 dollars can be determined with an accuracy already of $\approx 3\%$ within a measuring time of 10 seconds.

This example indicates that the proposed reactor noise method using californium neutrons as a randomly pulsed neutron source provides the reactivity of a fast reactor much more efficiently than the familiar two-detector cross correlation technique using only two neutron sensitive detectors.

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Related information on this application may be found in:

R. A. Goldstein and E. Melkonian. "New Neutron Pulse Technique to Measure Space-Time-Dependent Important Functions." *Trans. Amer. Nucl. Soc.* 11, 228 (1968).

J. T. Mihalcz. "Use of Californium-252 as a Randomly Pulsed Neutron Source for Prompt Neutron Decay Measurements." *Nucl. Sci. Eng.* 41, 296 (1970).

ERRATA

In *Californium-252 Progress*, No. 6, January 1971, your copy may have an error in Figure 7 (page 15), Figure 8 (page 16), in the text on page 16, and in the text on page 17. To correct your copy, paste the corrected figures or text from this page and the following page in the appropriate place.

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FIG. 7 LET Distribution for Californium-252

depth, peaks are lowered and broadened, and the LET is decreased, as shown in Table II.

Table III shows measured physical depth dose rate from neutrons emitted by a californium-252 medical afterloading tube (AT Type) in TES as a function of depth. These measured values agree with other published data on tubes. Figure 9 shows measured depth dose curves for an ALC Type source. The only noted difference in these curves and those from an AT Type source is in the gamma dose rates.

Figures 10 and 11 show measured isodose contours for gamma rays and for neutrons emitted from a californium-252 needle in TES. The gamma dose is depressed at the ends of the needle by the internal sealing plugs; the fast neutron dose rate distribution exhibits no such end effect.

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FIG. 8 Effect of Depth on Californium-252 LET Distribution

SLOAN-KETTERING INSTITUTE

Californium-252 is being evaluated in a medical research and development program at the Sloan-Kettering Institute for Cancer Research and the Memorial Hospital for Cancer and Allied Diseases. Under the direction of J. S. Laughlin and G. D'Angio, 32 californium-252 sources totaling 240 micrograms are being used in radiodosimetry, radiobiology, and human radiobiology studies.

Radiodosimetry

The following dosimetry measurements will be made on californium-252 needles:

- Fast neutron flux density
- Total kerma rate for tissue
- Gamma ray dose rate
- Fast neutron flux, thermal neutron flux, total tissue dose, and gamma ray dose distribution around the sources in a tissue-equivalent medium
- LET spectral distribution in a tissue-equivalent medium
- Source standardization
- Neutron-to-gamma dose ratio

The dose and flux distribution data will be used to modify existing computer programs to permit the calculation of gamma ray and neutron absorbed dose distributions for all possible clinical applications.

Radiobiology

The RBE and OER of californium-252 relative to cobalt-60 gamma rays will be determined with HeLa tumor cells by examining cell survival and clone size.

Biochemical studies on repair mechanisms will be made. The effect of dose rate on RBE will also be studied.

Experiments characterizing the effects on spermatogenesis in rabbits will be extended from cobalt-60 and radium to include RBE comparisons with californium-252.

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ADDITIONAL INFORMATION AVAILABLE

LITERATURE

Californium-252, Its Use and Market Potential is a brochure that describes the characteristics, availability, applications, production, and properties of californium-252. The brochure also describes the market evaluation program by which industry is invited to help in the evaluation of the usefulness of californium-252.

Californium-252 Progress is issued quarterly to supplement the information in the brochure, and consists of two main sections:

Californium-252 News. Reports on participants in the program, cost and availability of californium-252, market projections, announcements of meetings and publications, and other topics related to the development of californium-252 applications, which are expected to lead, eventually, to a large market for californium-252.

Californium-252 Technology Development. Reviews of applications technology developed under the market evaluation program, californium production technology, and other technical accomplishments related to californium-252 utilization.

Guide for Fabricating and Handling Californium-252 Sources is a comprehensive review of:

- Radiation and decay properties of ^{252}Cf .
- Permissible contamination, posting of areas, labeling of containers, survey techniques, personnel monitoring.
- Routine handling techniques for sealed sources and unencapsulated californium.
- Permissible radiation doses, shielding requirements, shield design.
- Containment and ventilation.
- Packaging and labeling of shipping containers.
- Inventory and control requirements.
- Purification techniques and product description.
- AEC facilities and techniques for fabricating and testing encapsulated sources.
- Radiation units.

Copies of the guide (Document Number SRO-153) may be purchased for \$3.00 each from National Technical Information Service, U. S. Department of Commerce, Springfield, Va. 22151.

Californium-252 Buyers' Guide presents the AEC's californium-252 sales program. Information is furnished about: price and availability; product specifications and packaging; shipping and licensing requirements; and ordering procedure. An order form and a brief summary of safety considerations are included.

INFORMATION CENTER

A californium-252 information center has been established at the AEC's Savannah River Plant. The growing number of inquiries about the californium-252 market evaluation program, as well as technical questions about californium-252, have made it necessary to centralize and make readily available such information. A current bibliography of published literature on californium-252 is maintained. Questions are directed to appropriate groups for answers.

Information regarding the AEC's various evaluation programs and the AEC sales program, copies of the brochure, progress reports, buyers' guide, order forms, and information and answers to questions about californium-252 may be obtained by telephoning 803-824-6331, ext. C-252, or by writing to

**Manager
Savannah River Operations Office
U. S. Atomic Energy Commission
Post Office Box A
Aiken, South Carolina 29801**

PARTICIPANTS IN UNIVERSITY LOAN PROGRAM

University of California, Santa Barbara, California

University of Missouri, Rolla, Missouri

University of Minnesota, Minneapolis, Minnesota

Tennessee Technological University, Cookeville, Tennessee

Ohio State University, Columbus, Ohio

Eastern New Mexico University, Portales, New Mexico

Washington State University, Pullman, Washington

Wayne State University, Detroit, Michigan

Mississippi State University, State College, Mississippi

University of Washington, Seattle, Washington

Sam Houston State University, Huntsville, Texas

Carnegie-Mellon University, Pittsburgh, Pennsylvania

University of Tennessee, Memphis, Tennessee

University of Texas, Austin, Texas

Ohio University, Athens, Ohio

**INVESTIGATORS OUTSIDE MARKET EVALUATION
PROGRAM USING CALIFORNIUM-252 SUPPLIED BY THE AEC**

Organization	Application
National Bureau of Standards* Washington, D. C.	Neutron spectrometry and Calibration
Idaho Nuclear Corporation† Idaho Falls, Idaho	Reactor physics experiments, Neutron radiography, and Neutron activation analysis
Battelle-Northwest† Richland, Washington	Neutron radiography and Mineral exploration
Oak Ridge National Laboratory† Health Physics Division Oak Ridge, Tennessee	Health physics studies
Sandia Laboratories† Albuquerque, New Mexico	Development of neutron detector techniques
Oak Ridge Associated Universities† Special Training Division Oak Ridge, Tennessee	Training activities involving: Neutron activation, Nuclear physics, and Health physics
Los Alamos Scientific Laboratory† Los Alamos, New Mexico	Research and development of non- destructive assay methods for safeguarding nuclear materials
Lawrence Radiation Laboratory† Livermore, California	Instrument calibration
Oak Ridge National Laboratory† Analytical Chemistry Oak Ridge, Tennessee	Activation analysis studies in analytical chemistry
Argonne National Laboratory† Argonne, Illinois	Fast and thermal neutron radiography
National Aeronautics and Space Administration* Jet Propulsion Laboratory Pasadena, California	Simulate spontaneous neutron and gamma emission from radioisotope power supplies
Savannah River Laboratory* Aiken, South Carolina	Neutron detection, Shielding, Water content and sediment, Density measurements, and Neutron radiography
Health and Safety Laboratory† USAEC New York Operations Office Brookhaven Office, New York (Upton)	Calibration of detectors
National Aeronautics and Space Administration* Goddard Space Flight Center Greenbelt, Maryland	Instrument calibration
United Nuclear Corporation* Wood River Junction, Rhode Island and Hematite, Missouri (Sponsored by USAEC, Office of Safeguards and Materials Management)	Research and development of non- destructive assay methods for safeguarding nuclear materials

* ²⁵²Cf source obtained from Savannah River Laboratory.

† ²⁵²Cf source obtained from Oak Ridge National Laboratory

PARTICIPANTS IN MARKET EVALUATION PROGRAM

Organization	Principal Investigator	Application	Effective Date*	Date First ²⁵² Cf Shipped	Number of ²⁵² Cf Sources	Total ²⁵² Cf, µg
Brookhaven National Laboratory Upton, N. Y.	H. E. Atkins	Radiotherapy	Sept. 13, 1967	June 1967	118	420
U. S. Geological Survey Washington, D. C.	F. F. Suttle	Mineral Exploration	Feb. 2, 1968	March 1969	5	273
M. D. Anderson Hospital and Tumor Institute Houston, Texas	R. E. Shalek G. H. Fletcher	Radiotherapy	Oct. 15, 1968	Nov. 1968	76	259
Bureau of Mines, Morgantown Research Center Morgantown, W. Va.	R. E. Stewart	Air Pollution Control	Dec. 19, 1968	Feb. 1969	5	115
Argonne National Laboratory Argonne, Ill.	J. P. Barton	Neutron Radiography	March 4, 1969	March 1969	1	141
Texaco, Inc. Beaumont, Texas	H. D. Scott	Well Logging	May 15, 1969	Aug. 1969	2	761
Schlumberger Technology Corporation Ridgely, Conn.	J. Tittman	Well Logging	May 29, 1969	Dec. 1969	3	473
Republic Steel Corporation Cleveland, Ohio	E. R. Stone	Process Control	Oct. 15, 1969	March 1970	2	1221
GEOSENSORS, Inc. Dallas, Texas	N. B. Humphrey	Mineral Exploration	Nov. 5, 1969	April 1970	1 1 [†]	50 500 [†]
General Dynamics Corporation Fort Worth, Texas	W. E. Dungan	Neutron Radiography	Nov. 21, 1969	Feb. 1970	1	920
Columbia Scientific Industries Austin, Texas	E. R. Rhodes	Process Control	Nov. 26, 1969	Feb. 1970	5	400
Hospital of the University of Pennsylvania Philadelphia, Pa.	E. W. Davis	Radiotherapy	Dec. 4, 1969	Feb. 1970	52	249
Georgia Institute of Technology Atlanta, Ga.	C. E. Robert	Education	Dec. 16, 1969	Dec. 1969	1	129
Goit Research & Development Company Pittsburgh, Pa.	J. G. Larson	Process Control	Jan. 26, 1970	May 1970	1	500
University of Georgia Athens, Ga.	J. E. Snokes	Mineral Exploration	March 20, 1970	May 1970	1	200
University of Texas at Austin Austin, Texas	S. J. O'Leary C. E. Dwyer	Nuclear Safeguards Instrument Analysis Neutron Radiography Process Control	May 8, 1970	July 1970	1 1 [†]	1000 200 [†]
Goit Energy & Environmental Systems, Inc. San Diego, California	E. Goetz D. Goetz	Nuclear Safeguards Pollution Control	June 10, 1970	July 1970	1 1 [†]	20 10,000 [†]
Battelle Memorial Institute Columbus, Ohio	J. W. Ray	Neutron Radiography Environmental Pollution Control	June 24, 1970	Aug. 1970	1	946
Plasma, Arsenal Department of the Army Dover, New Jersey	S. Hell E. G. Barnes	Neutron Radiography Quality Control	July 2, 1970	Nov. 1970	1 1 [†]	5 5000-10,000 [†]
University of Hawaii Honolulu, Hawaii	E. L. Peterson	Well Logging	July 6, 1970		1 [†]	200 [†]
National Bureau of Standards Gaithersburg, Maryland	P. D. LaBelle	Activation Analysis	Aug. 5, 1970	Sept. 1970	8	800
American Science and Engineering, Inc. Cambridge, Mass.	G. Davidson	Air Pollution Control Process Control	Aug. 12, 1970	Aug. 1970	1 1 [†]	10 300 [†]
Kerr McGee Corporation Oklahoma City, Oklahoma	H. C. Eberline J. D. Shreve	Mineral Exploration	Aug. 14, 1970		1	1000
U. S. Department of Agriculture Oxford, Miss.	J. R. McHenry	Pollution Control	Sept. 29, 1970	Oct. 1970	1	50
Kansas State University Manhattan, Kansas	C. G. Chezem	Quality Control Process Control	Oct. 7, 1970		5	50,000
University of Cincinnati Cincinnati, Ohio	J. G. Kenenak	Radiobiology	Oct. 7, 1970	Nov. 1970	1	20
International Systems, Inc. Los Altos, California	K. J. Bialy	Pollution Control	Nov. 12, 1970		4 [†]	5000 [†]
Sloan-Kettering Institute New York, N. Y.	J. S. Laughlin G. D. Angelo	Radiotherapy	Dec. 17, 1970	Feb. 1971	32	240
University of Utah Salt Lake City, Utah	G. M. Sandquist	Process Control Mineral Exploration Neutron Radiography Education	Jan. 11, 1971		1 [†]	15,000 [†]
National Aeronautics and Space Administration Goddard Space Flight Center Greenbelt, Maryland	J. E. Trumbka	Activation Analysis Instrument Calibration	Feb. 5, 1971	April 1971	1	3
Army Materials and Mechanics Research Center Department of the Army Watertown, Mass.	J. J. Antal	Neutron Radiography	March 1, 1971		1 [†]	5000 [†]
EURATOM Joint Research Centre Ispra, Italy	M. Y. R. Cuypers F. Garardi	Nuclear Safeguards	March 19, 1971		1 [†]	25 [†]

* Date of contract with private organizations or agreement with other government agencies
† To be shipped

**DATE
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