Isotopes Development Programs

Research
and
Development
1971

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ISOTOPES DEVELOPMENT PROGRAMS RESEARCH AND DEVELOPMENT: 1971

Progress Reports on Sponsored Work

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Issued: February 1972

Division of Applied Technology
United States Atomic Energy Commission
Washington, D. C. 20545

Isotopes Development Programs

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FOREWORD

Isotopes are contributing to the advancement of science and technology and human betterment to an extent equaled by few discoveries or inventions in history. Their use spans the whole of our scientific disciplines, and many of them are being brought to bear on urgent human problems and national requirements. In some areas, such as medicine and industry, this new technology is now well established and its benefits are very significant. In others, such as pollution control and forensic analysis, its potential is just beginning to be realized. Through its Isotopes Development Program, the Atomic Energy Commission is endeavoring to establish the basic technology necessary for realization of these values; the specific objectives of this program are to

Bring about productive utilization of radioisotopes and radiation, important to the national economy and welfare through research and development and demonstration programs and to assist in the introduction of this technology into wide scale use

Develop technology for the production, separation, purification, and encapsulation of radioisotopes utilizing resources of both the Atomic Energy Commission and industry

Ensure that national requirements for the production and distribution of radioisotopes are satisfied by the AEC or by private industry and that current AEC activities in this area are transferred to private industry in keeping with the best interest of the public

To achieve these objectives, the Atomic Energy Commission sponsors research and development at its laboratories and plants such as Oak Ridge National Laboratory and Brookhaven National Laboratory and at various industrial and academic facilities. Major technical program areas and goals are

Radioisotope Production and Materials to improve methods for the production of radionuclides

Thermal Applications to develop applications using the thermal energy generated by radioisotope decay as a heat source or for conversion to mechanical power

Environmental and Ocean Sciences to develop radioisotope techniques to aid in environmental problems such as pollution and water management and to assist in the exploration and economic development of the oceans

Radiation Analysis and Control to develop basic technology and instrumentation for detection and measurement of radioisotopes and to apply the technology to current problems

Process Radiation to develop methods for the production of chemicals and other materials using large sources of radiation and to develop efficient, safe radiation sources for process radiation

Radiation Preservation of Foods to develop a low dose radiation processing method to extend marketing times and reduce spoilage of selected foods

Technology Analysis and Utilization to evaluate Program requirements, objectives, and outputs and to expedite the wide scale use of technology developed under the Program

This report is the fifth edition of the Isotopes Development Program Project Summaries. It presents a short summary of objectives, results, and future plans for each research or development project within the program areas described above.

Assistant Director
for Isotopes Development
Division of Applied Technology
U.S. Atomic Energy Commission

The project summaries in this report were prepared by the contractors who are performing the work and were collected and edited for technical content by members of the technical staff of the Isotopes Development Branch*, Division of Applied Technology, USAEC. Final editing and preparation for publication was done by the staff of the Isotopes Information Center, Oak Ridge National Laboratory.

*Formerly Division of Isotopes Development. Since the reorganization of the AEC occurred subsequent to the editing of the draft of this report, changes could not be made in the text to reflect the new organizational structure. Hence the divisions referred to are those as they existed prior to December 7, 1971.

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TECHNOLOGY UTILIZATION, ISOTOPES INFORMATION CENTER*

Objective of Work

The project objective is to collect, evaluate, and disseminate information as reported in world literature — primarily on the production and uses of radioisotopes in industry, medicine, and research. The Center also maintains a current awareness of uses of isotopes in biology, agriculture, education, forensics, and the military.

Progress and Results

During the current period, the first of a number of "Radioisotope Briefings for Industry" was completed. These consist of annotated bibliographies covering specific areas of isotope applications — e.g., "Non-ferrous Metals." These are to be available as handouts to interested users and will be submitted to appropriate trade journals for publicity.

The Center's total of indexed documents is now nearly 23 thousand. The information contained in these documents was used by members of the Center for the following purposes.

To write four issues of the quarterly technical progress review, *Isotopes and Radiation Technology*, for publication by the Division of Technical Information (This review is available from Superintendent of Documents, U. S. Government Printing Office, Washington, DC 20402, subscription \$2.50 per year, \$0.70 for individual issues, non-U.S. subscriptions \$3.25 per year, \$0.90 for individual issues.)

*P. S. Baker, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, TN 37830, Contract W-7405-eng-26.

To answer about 350 requests for specific information and to fill over 850 individual requests for about 2500 copies of IIC publications. The information requests came from industry (30%), Government (12%), educational institutions and research institutes (30%), individuals, no affiliation indicated (13%), and miscellaneous sources (15%).

To provide information — chiefly in the form of bibliographies — needed by personnel of DID to evaluate research proposals.

To provide information for DID personnel for inclusion in reports and for presentation at meetings and to help prepare the Annual Summaries of DID's *Research and Development Projects*.

To prepare a number of summary review articles and other special reports and bibliographies. These reports are listed under *Reports Published*.

The previously reported (1969) *Isotope User's Guide* continued to be a very popular item and a reprinting was necessary.

More than one hundred persons visited the Center during the year.

Plan for Next Period

The continuing program of the Center comprises all of the above activities, with increased emphasis on providing for the preparation of more summary review articles and on extending and promoting the interplay between the IIC and industry so as to improve the technology utilization aspects of DID's programs.

Reports Published

P. S. Baker, A. F. Rupp, and Associates, *Isotopes and Radiation Technology*, Volumes 8(3), 8(4), 9(1), and 9(2).

Ruth Curl, Comp , *List of AEC Radioisotope Customers with Summary of Radioisotope Shipments, FY 1970*, USAEC Report ORNL-IIC-32, February 1971

R E Greene, H S Warren, and P S Baker, *Patent Literature on Process Radiation and Irradiator Design Part 1 U S Patents 1950 Through 1968*, USAEC Report ORNL-IIC-34, June 1971

R Shor, R H Lafferty, and P S Baker, Comps , *Strontium-90 Heat Sources*, USAEC Report ORNL-IIC-36, May 1971

Martha Gerrard and P S Baker, *Selected Abstracts of World Literature on Production and Industrial Uses of Radioisotopes*, USAEC Reports ORNL-IIC-30 (Part 3) December 1970, ORNL-IIC-30 (Part 4) April 1971, ORNL-IIC-37 (Part 1) in press

IIC Staff, Eds , *Division of Isotopes Development Research and Development Projects, 1970*, USAEC Report TID-4066, February 1971

Estimated Date of Final Report

Continuing project

RADIOISOTOPE PRODUCTION AND MATERIALS

BLIP target.

HAPO I cask arriving at
ORNL's FPDL from Hanford.

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Radioisotope Production and Materials

The radioisotope production and materials development work has been concerned with meeting isotope requirements in four principal areas: (a) diagnostic and therapeutic medicine, (b) analysis and controls, (c) process radiation, and (d) thermal energy. As a result of the AEC's program, radioisotopes are now firmly established as essential materials in medicine and in analysis and controls. In addition, considerable progress has been made toward establishing the opportunity for economical use of large quantities of the radioactive nuclides for process radiation and thermal power applications. This effort also includes the pilot production of radioisotope products, unique to AEC capabilities, as part of the program to develop radioisotope applications for the national benefit.

The *Fission Product Studies* represent continuing efforts at Oak Ridge National Laboratory (ORNL) and Pacific Northwest Laboratory (PNL) to develop production technology and to characterize products. Also supported is a small effort at Atlantic Richfield Hanford Company on fission product availability from Hanford reactor wastes. Fission products, particularly from power reactors, represent a potentially valuable source of long-lived radioisotopes including Sr-90, Cs-137, Kr-85 and Pm-147. Work at ORNL is principally on enrichment of Kr-85, preparation of Cs-137 sources for optimized process radiation applications, and development of encapsulation technology for Sr-90 sources. In addition, pilot plant operations at ORNL supply Sr-90, Cs-137, and Kr-85 along with a broad spectrum of shorter-lived fission products for applications development. The PNL studies include Pm-147,

Sr-90, and Cs-137 and the stable fission products rhodium, palladium, and xenon. In addition, studies are made of projected isotope availability based on power reactor operations. Pilot production operations provide Pm-147 for applications development.

The *Reactor Product Studies* involve work at ORNL and the Savannah River Laboratory. The studies at ORNL employ two high flux reactors in the development of new products—primarily for medical applications. Specific products investigated include P-33, Cl-36, K-43, Ru-97, La-137, Ba-135m, Gd-153, Tm-170, Pt-195m, and Rh-105.

The *Accelerator Product Studies* at Brookhaven National Laboratory (BNL) and ORNL are continuing, with emphasis on research and medical products. In these studies, the 86-inch cyclotron at Oak Ridge along with the 60-inch cyclotron and Van de Graaff machines at Brookhaven are used. In addition, facilities are being established to use the Linear Accelerator Injector of the Brookhaven Alternating Gradient Synchrotron in 1972 for spallation reaction studies. Specific product studies include Ga-67, In-111, Rb-83, Nd-140, F-18, Cs-129, Ba-133m, Tm-167, and Pb-203. General technology studies include radionuclide recoil labeling.

The *Applications and Technology Support Studies* include work principally at BNL, ORNL, and PNL. Both Brookhaven and Oak Ridge have major supporting activities in the radiopharmaceutical area. Studies involving radiation sources, standards, and regulatory questions are carried out at ORNL. At PNL research on nondestructive testing is in progress.

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ORNL FISSION PRODUCT DEVELOPMENT PROGRAM*

Objective of Work

The objective of the ORNL fission product development program is to support development activities in the areas of thermal and radiation applications. Radioisotopes that result from the fission process are the most abundant sources of unique energy forms. In addition, technical support is provided for the Fission Products Development Laboratory (FPDL) and the short lived fission product processing unit. Specific areas of application include (1) materials sterilization, (2) food preservation, (3) radiation stimulated chemical processing, (4) pollution abatement, (5) extraterrestrial, terrestrial and oceanographic thermal control and (6) medical research.

Progress and Results

The usefulness of Kr 85 can be increased by removing from it the nonradioactive krypton isotopes thus increasing the specific activity of the product. The technique of thermal diffusion enrichment is being applied to isotopic enrichment of Kr-85. Progress in the development of this technique is reported in a separate summary. Cesium-137 chloride process improvement and source characterization studies were continued with emphasis on (1) chemical process improvement and (2) the effect of KCl on the physical properties of Cs(137)Cl sources. Both items are reported in individual summaries. Alternative Cs-137 compounds to Cs(137)Cl were prepared and evaluated on the basis of technical feasibility and economic potential. Progress in the examination of niobates, tantalates, and silicates is reported in a separate summary.

The preparation of fission-product xenon highly purified and decontaminated from Kr-85 was continued in order to supply Battelle Northwest Laboratory with material for evaluation of applications in medicine. A separate technical summary describes this work.

Plan for Next Period

See summaries of individual projects.

Reports Published

See summaries of individual projects.

Estimated Date of Final Report

Continuing Project

KRYPTON-85 ENRICHMENT*

Objective of Work

The objective of the Kr 85 enrichment program is to provide krypton gas containing a higher concentration of radioactive mass 85 isotope than the approximately 5% that is found in normal fission product krypton. The minimal biological hazard and ease of handling of the inert gas krypton are desirable features of this radioisotope. Fission krypton has been used in such applications as beta thickness gages, wireless emergency exit and highway signs, and kryptonated metal parts for wear and temperature measurements. The availability of krypton with an increased specific activity has improved the effectiveness of most of these applications and may open new uses, such as in isotopic power sources.

Progress and Results

The thermal diffusion pilot plant constructed to provide operational experience, is routinely producing approximately 1200 Ci/year of material containing greater than 15% Kr 85 (214 Ci/liter) and has produced material containing as much as 43.8% (625 Ci/liter) Kr-85, representing an enrichment factor of nearly 12 times the specific activity of the starting feed gas. The present system has been in operation for more than two years with remarkably few operational difficulties or maintenance problems. The only major repairs needed during this period involved replacing several manual valves in one cell containing three thermal diffusion units.

One of the six column bundles has been used to demonstrate a method for decontaminating fission xenon to a low Kr-85 level so that the xenon could be used in unregulated applications, such as in light bulbs or as an anesthetic. Fission xenon which had been decontaminated to a Kr-85 activity level of 0.4 μ Ci/ml by chromatographic methods was further decontaminated to an activity level of 0.002 μ Ci/ml by thermal diffusion. Twenty days of column operating time was required to reach this activity level.

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Plan for Next Period

Operation of the pilot plant will continue, with emphasis on producing material enriched to greater than 43.8% of Kr-85.

Reports Published

None.

Estimated Date of Final Report

Continuing project.

CESIUM-137 CHLORIDE PROCESS IMPROVEMENT AND SOURCE CHARACTERIZATION – CHEMICAL PROCESS IMPROVEMENT*

Objective of Work

The objectives of this project are (1) to develop an improved process to replace the one in use at the Fission Products Development Laboratory (FPDL) for conversion of cesium eluted from the inorganic ion exchanger shipping cask to Cs(137)Cl and (2) to carry out experimental studies on technical feasibility of the selected processes

Progress and Results

A preliminary evaluation of fixed capital costs (relative figures) based on a generalized semi-empirical equation has been carried out for five processes under consideration for purification and conversion of the Cs-137 feed from an inorganic ion exchanger into Cs(137)Cl. Conceptual process flow diagrams and estimated values of various parameters were used in the computation. The result, summarized in the table, indicates that Process 1 (NH₄Cl eluant) is economically the most attractive from the viewpoint of the fixed capital costs. Process 4 (HCl eluant) requires the second lowest fixed capital costs, but experimental results revealed that the elution efficiency of the acid-type eluant is appreciably lower than that of the ammonium salt type. A similar statement is applicable to Process 5 (HNO₃ eluant). For these reasons, Processes 1, 2, and 3 are preferred to Processes 4 and 5

In progress is the study on the corrosion of materials for construction of processing vessels (several grades of stainless steel) by NH₄Cl and (NH₄)₂CO₃–NH₄OH solutions. The initial examination of the test specimens

Comparison of Fixed Capital Costs for Various
Conceptual Cs(137)Cl Processes

Eluant	Fixed Capital Costs ^a (Relative Figure)
NH ₄ Cl	14.9 K
(NH ₄) ₂ CO ₃ –NH ₄ OH	26.2 K
(NH ₄) ₂ SO ₄	33.4 K
HCl	23.7 K
HNO ₃	30.9 K

^aK = CPⁿ(I/Ī), where C is a constant, P is the production capacity (Ci/yr), n is a factor derived from a statistical correlation between F and P, and I and Ī are construction-cost indices. K is assumed to be common to all the processes.

exposed to the solutions circulating at ~75°C under Cs(137)Cl radiation showed almost negligible attack in all cases after about two months. Further studies are required before any interpretation and conclusion can be made in regard to the compatibility of the eluant solutions with the structural materials.

Plan for Next Period

Experimental studies will be continued to confirm technical feasibility of the selected Cs(137)Cl processes. The experimental data obtained will be used as the basis for the preliminary process design and economic evaluation of commercial-scale plants.

Reports Published

None.

Estimated Date of Final Report

Continuing project.

CESIUM-137 CHLORIDE PROCESS IMPROVEMENT AND SOURCE CHARACTERIZATION – EFFECT OF KCl ON PHYSICAL PROPERTIES OF Cs(137)Cl SOURCE*

Objective of Work

The objectives of this project are (1) to investigate how presence of the KCl impurity in Cs(137)Cl gamma sources influences dimensional stability of the source compound and (2) to determine the limits of application conditions based on the study.

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Progress and Results

Further studies on the effect of KCl on the dimensional stability of CsCl have been carried out using dilatometry and high-temperature x-ray diffractometry. Emphasis was placed on elucidating the solid phase transformations as related to the thermal expansion as a function of several variables. Both equilibrium and kinetic effect were investigated.

The data obtained from previously reported differential thermal analysis (DTA) studies were combined with the dilatometry and x-ray diffractometry data to establish a clear understanding of the equilibrium phase diagram for the solid phase region. Previous DTA data were inadequate for this purpose due to small energies of the transformations. The DTA data did indicate, however, that kinetic effects are very significant factors. The addition of KCl (up to 2 mole%, 0.9 wt%) does not significantly alter the phases present at equilibrium. The addition of 2 mole% KCl does significantly alter the kinetics of the solid-solid phase transformation. For example (see the figure)

- 1 The solid-solid phase change from β to α at approximately 460°C takes place very slowly in 2 mole% KCl-CsCl mixture, whereas in pure CsCl the transformation at this temperature is rapid both on heating and on cooling.
- 2 The linear coefficient of thermal expansion of the high-temperature form (β phase) is much less than for pure CsCl. The linear coefficient of thermal expansion of pure CsCl in the high-temperature form (above 465°C) is 22 to 44 times that of the low-temperature form (below 465°C). When 2 mole% KCl is added to CsCl the relative expansion is reduced by a factor of about 2.
- 3 The residual expansion of pure CsCl when repeatedly cycled about the solid-solid transformation point is about 10%. When 2 mole% KCl is present, the residual expansion is about 6.5%. The addition of an amount of KCl greater than the 2 mole% alters significantly the phases present as a function of temperature. In addition severe kinetic effects are encountered, especially in samples not exposed to ambient air.

Phase diagram for CsCl-KCl system.

Based on the findings described above, the following recommendations are made in regard to the limits of application conditions

- 1 For a conservative design of the source capsule, a sufficient volume should be allowed for thermal expansion based on pure CsCl, although the presence of a small amount of KCl tends to reduce the extent of such expansion
- 2 Thermal cycling through the solid-phase transformation temperature (depends on KCl content) should be minimized because it tends to increase the amount of residual expansion of the salt

Plan for Next Period

This phase of the project has been completed

Reports Published

None

Estimated Date for Final Report

Second quarter of 1972

ISOTOPE PRODUCTION EVALUATIONS*

Objective of Work

The objective of this project is to evaluate major aspects of isotope production, including (1) economic evaluations of isotope production by target irradiation or recovery from fuel reprocessing waste, (2) preparation of isotope availability projections, and (3) development of a computerized isotope data system

Progress and Results

The study to define the time and circumstances as to when recovery of fission product cesium and strontium might be attractive to the nuclear fuel reprocessing industry was completed. Initial results are described in the 1970 report (TID-4066)

An important aspect of this study was determination of the value of cesium recovered from power reactors and from Hanford Waste Management Program (HWMP) relative to Co-60. The value of power reactor cesium as a gamma radiation source is enhanced by the larger gamma energy contribution of the contained Cs-134 (~2.5 wt%), while the value of HWMP cesium is reduced by the large self-absorption effect in the 2.6-in

dia capsules. The values relative to Co-60 at \$0.40/Ci and based on a discounted cash flow analysis were found to be in the range of \$0.25 to \$0.30/Ci Cs-137 for power reactor cesium and \$0.07 to \$0.09/Ci Cs-137 for the HWMP cesium.

The computer program mentioned in TID-4066 was designed to calculate price requirements for a profitable venture. This computer program was modified to include more realistic market projection options as well as market sharing options. This permits examination of the gamma-radiation source market as a whole with a suitable apportionment of the market between Co-60, power reactor cesium, and HWMP cesium. For different source materials, the different initial and replenishment requirements result in quite different annual curie requirements for the same basic market.

Using this computer program, a series of cases was prepared which simulated the development of a Cs-137 and Sr-90 recovery industry. The gradually developing market is first supplied from HWMP cesium and strontium resources, and when the market has grown large enough to justify the large investment required, a private reprocessor begins operations. The results of these calculations indicated that this kind of two-phase market evolution could result in feasible private recovery operations in the late 1970's or early 1980's. Results for the base-case assumptions are shown in the first table.

Target irradiation capabilities of a pressurized water reactor (1975-startup design) were calculated using the 2DB and ALTHAEA computer codes. It was assumed that targets could be inserted in unused rod cluster control assembly locations. Results are summarized in the second table for 11-month irradiations (one refueling cycle).

Revised projections of isotope availability from power reactor spent fuel processing were developed following the procedures outlined in BNWL-716 (July 1968). A slower rate of nuclear plant startups and lower capacity factors have reduced projected quantities relative to those in BNWL-716. The difference was large in the early 1970's but amounted to only a one-year offset in the 1980's.

A computerized isotope data system was developed and tested on a sample data file of 50 nuclides. The system was designed for remote accessing via a teletype terminal with queries in the English language. All design options were tested and found operable.

The system permits searching the data file with up to 50 constraints imposed on items to be reported. For example, maximum or minimum constraints can be specified for half-life, gamma energies or abundances,

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Price Requirements for Base Case Power Reactor-Cesium Recovery
(\$/Ci Cs-137)

Note Feasible prices enclosed in rectangles

Target Irradiation Capabilities of a Power Reactor (PWR)

Target	Reaction	Product	Target conc g/cm ³	Enrichment required in assembly, % U-235*	Product Yields (no loss basis)				Product activity, Ci per gram of element
					g/g	kg/assembly	Maximum kW per 1000 MWe per year	Maximum kCi per 1000 MWe per year	
Np-237 (oxide)	n,γ	Pu-238	0.5	4.09	0.193	0.05	32.1		
Am (50/50) 241 & 243 (oxide)	n,γ	Cm-242 Pu-238 Cm-244	0.25	4.24	0.115 0.106 0.113	0.150 0.138 0.147	2050 8.8 47		
Cobalt	n,γ	Co-60	(20 g/ft)	3.99	0.028	0.134		17,300	31.5
Thulium (Tm ₂ O ₃)	n,γ	Tm-170	0.5	3.80	0.108	0.280	560	194,000	650
Bismuth	n,γ	Po-210	9.8	3.30	1.6×10^{-5}	7.2×10^{-4}	12	370	4490
Nitrogen (Be ₃ N ₂)	n,p	C-14	0.8	3.43	1.3×10^{-3}	5.5×10^{-3}	0.9×10^{-3}	2.8	4.5
Lithium (Li ₂ O)	n,α	H-3	0.1	3.87	0.262	0.136	5.6	150,000	9690
Phosphorus (P ₂ O ₅)	n,γ	P-32	1.0	3.40	1.6×10^{-3}	8.0×10^{-3}	875	260,000	440
Sulfur	n,γ	S-35	1.9	3.41	1.6×10^{-4}	1.6×10^{-3}	2.5	7,600	6.7
Tellurium	n,γ	I-131 I-127 I-129	5.7	4.14	2.6×10^{-6} 1.6×10^{-4} 1.4×10^{-4}	7.6×10^{-5}	3.6	1,100	1040

*Based on enrichment without target = 3.3% U-235

beta energies, cross sections, etc., and only data on isotopes satisfying these constraints will be reported. Results can be reported in tabular form in one to ten evenly spaced columns, with items sorted in either increasing or decreasing order.

The Nuclide Data File (2000 nuclides) was completely revised and updated and the file is ready for loading into the data system.

Plan for Next Period

No further effort is anticipated.

Reports Published

None

Estimated Date of Final Report

None planned

LOW-COST CESIUM AND STRONTIUM SOURCES*

Objective of Work

The objective of this program is to provide technology for the production of inexpensive cesium and strontium sources by using the purified compounds generated in the Hanford Waste Management Program (HWMP). Large scale use of these radioisotopes will require an assessment of the capabilities of HWMP for producing suitable radiation and heat sources, acceptance of the HWMP product by the customer, licensing of the sources, and market development activities.

Progress and Results

Construction has been started by Atlantic Richfield Hanford Company (ARHCO) on the encapsulation portion of the Waste Management Plant. The facility is expected to be ready for cold testing in 1973, and for full hot operation in January 1974. Ion-exchange purification equipment for cesium is being installed, and final purification of cesium is expected to start in FY-1972. Purified strontium is currently being accumulated. One of the strontium product tanks contains about 12 MCi of strontium of 95% chemical purity. The Sr-90 content is 53.7 wt% based on total cation content.

At the present processing rate, the waste storage tanks will have been completely processed in about 1976. A few additional years will be required for conversion to solids and encapsulation.

Container sizes for both cesium and strontium were recently changed by HWMP to provide easier handling, welding, and nondestructive testing (NDT), however, the amount of material per container was not changed. In each case new dimensions are 2.625-in. o.d. for the outer capsule and 2.500-in. o.d. for the inner capsule. The total wall thickness for the cesium capsule was reduced slightly to 0.179 in. The utility of each type of capsule for radioisotope applications is not affected appreciably.

The work described above was funded by the Division of Production, but the status is described here because of the strong interaction with the Division of Isotopes Development program described below.

The process for encapsulation of Cs-137 in the HWMP involves casting of molten CsCl into a series of nine containers. Optimization of radiation sources will require the ability to transfer the molten CsCl through

small diameter lines into a series of 1-in. o.d. capsules a size that would be attractive as a radiation source.

A device for loading a series of ten 1-in. o.d. by 18-in. long capsules with 0.035-in. wall was fabricated for testing. Each capsule was mated and sealed to a transfer head that contained 1/4-in. lines for conducting the molten salt to and from each capsule in series. Molten CsCl was transferred into the series of ten capsules without difficulty. The density of the CsCl was 2.68 g/cm³ (~78% theoretical density). A uniform cylinder of CsCl was produced with only slight (~1/2 in.) depression of the top surface caused by cooling. This demonstrates that HWMP could load the small diameter capsules without adjusting the batch size normally used in the Waste Management Plant.

A study of technical feasibility and cost has shown that multimegacurie amounts of strontium as the fluoride, titanate, or oxide can be encapsulated by HWMP. Low costs are possible due to low waste disposal costs, a large amount of available hot-cell space, large-scale equipment, and a large enough workload to permit optimum assignment of personnel.

Analyses of selected samples of the strontium product showed about 10 mole% calcium and less than 1 mole% of any other single cation. The flowsheet for isolation of the strontium from Purex high level waste uses several cycles of solvent extraction to concentrate the product from very large volumes of waste and effect a gross separation from the major contaminants. The product from solvent extraction is purified from calcium using a sulfate purification step. Except for the rare earth and lead, this material meets the requirements for encapsulation. Adjusting to a pH of 10 and filtering should remove these elements from solution.

Compatibility of strontium fluoride with container materials was reviewed. The strontium purity being achieved by HWMP is adequate for storage, but may not be adequate for use in heat sources. The presence of trace impurities such as sulfate, phosphate, lead, manganese, and sodium may have a deleterious effect on the high temperature compatibility of the fluoride with materials of containment. This, in turn, can endanger the long-term integrity of the strontium fluoride package. Elimination of traces of water in the final encapsulated package could require modifications to the proposed installation. Additional data on strontium fluoride compatibility are necessary to determine the required purity for heat source uses.

Phase diagrams including strontium fluoride as one of the components were reviewed. Sodium fluoride and strontium fluoride do not form a solid solution under any conditions, but they do form an equimolar eutectic

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with a melting point of about 850°C. Thus, the thermal equilibrium composition expected in HWMP strontium product is a bulk phase consisting of alkaline earth fluorides and a small amount of a second phase containing alkali-alkaline earth fluoride eutectic. The properties of the bulk phase can be evaluated with sufficient accuracy from existing data. The properties of the impurity phase cannot be evaluated adequately from existing data.

As decay of the strontium proceeds, ZrF_2 is created in the lattice. This rather unstable molecule has been reported and its properties are said to be those of group IV-A difluorides. Disproportionation would be expected to occur when a sample of old encapsulated SrF_2 is brought to 800°C for the first time. This would probably mean a decrease in melting point as the ZrF_4 is formed and makes eutectic mixtures with other fluorides.

Plan for Next Period

Progress of the HWMP will be followed by close coordination with ARHCO and Division of Production. Changes in process and encapsulation equipment will be recommended when necessary to permit commercial utilization of the HWMP products.

Equipment similar to that used for producing non-radioactive cesium sources will be acquired and used for fabrication of fully radioactive sources. Prototype sources will be evaluated for licensing and suitability for commercial use.

Process technology for preparing strontium compounds other than the fluoride in HWMP will be developed. Accumulation of currently available data for licensing strontium fluoride will be completed and additional data will be developed.

Reports Published

None

Estimated Date of Final Report

Continuing project

HANFORD FISSION PRODUCT PRODUCTION CAPABILITIES*

Objective of Work

The objective of this project is to evaluate the production of heat and radiation sources to customer

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specification, using the fission products separated as part of the Hanford Waste Management Program (HWMP). Construction of a plant for the encapsulation of CsCl and of SrF_2 is in progress and startup is scheduled for 1974.

Progress and Results

Three separate studies were undertaken to evaluate the production of special Sr-90 and Cs-137 sources. In the first study, the capability of the encapsulation plant to produce up to 5 megacuries annually as doubly clad Sr-90 sources (as SrF_2 , SrO , or SrTiO_3) in capsules ranging from 1 to 4 in. in dia and having thermal output of from 20 to 2000 W each was evaluated. Encapsulation plant equipment additions, modifications, and development work costs were estimated.

In the second study, information regarding the production of special Cs-137 radiation sources and encapsulation plant operating cost was developed.

Production of 10 and 100 CsCl sources similar in design to the HWMP capsule but containing inert partial inserts was found feasible. This work could be performed without modification to the existing equipment and with a minimal capital expenditure. The number of capsules per year and the insert diameters are listed below for production of this type of source.

Number of Capsules per Year	Insert Diameter
10	1 in
10	1½ in
100	1 in
100	1½ in

Evaluation of other capsule sizes indicated that production of forty 1-in. dia sources without the inserts is feasible with the work force presently planned. If 400 of these capsules were to be produced annually, the staff would have to be increased by ~30%.

Plan for Next Period

A report will be issued annually, giving a current assessment of Hanford's fission product production capabilities. Included will be information on plant flow sheets, operating costs, production rates and schedules, existing plant facilities, and isotope supplies and purities.

Reports Published

None

Estimated Date of Final Report

July 1, 1972 (Annual Report)

**FISSION PRODUCT RHODIUM,
PALLADIUM, RUTHENIUM, AND
TECHNETIUM RESEARCH*****Objective of Work**

The initial objective of this program was to measure the radiation characteristics of the fission product rhodium and palladium produced in power reactors, and to study ways of recovery of these two elements from the fuels reprocessing wastes. The program has been expanded to include ruthenium and technetium, and the emphasis of the goals has been placed on the practical aspects of the safe use of these four elements.

The expanded program has five tasks:

- 1 Evaluation of rhodium and palladium produced in power reactors
- 2 Technology for recovery of Rh, Pd, Tc, and Ru

*F. P. Roberts, J. C. Sheppard, and R. S. Kemper, Battelle, Pacific Northwest Laboratory, Richland, WA 99352, Contract AT(45-1)-1830

- 3 Market analysis of Rh, Pd, Tc, and Ru
- 4 Economic analysis of Rh, Pd, Tc, and Ru production from nuclear wastes
- 5 Fabrication process development for nuclear source Rh, Pd, Tc, and Ru

The overall program is designed to establish whether or not any of these fission products are candidates for industrial application. The desirability of use of each of these elements must be based on future requirements, economic feasibility, and the degree of the radiological hazards inherent in their use.

Progress and Results

The principal radiation characteristics of the rhodium and palladium formed in power reactors have been established. Palladium-103 and Rh-102m are the radioactive isotopes which will be important in defining restrictions placed on the use of these two fission products. Ruthenium has two radioactive isotopes with significantly long half-lives, Ru-103 and Ru-106. The Ru-106 will be the limiting radioactivity. Technetium from fission is essentially 100% Tc-99. The radiation characteristics of these fission products are given in the table.

Radiation Characteristics of Rh, Pd, Ru, and Tc Isotopes

Isotope	Half-life	Decay mode	Maximum beta energy, MeV	Gamma energies, MeV	Specific activity, ^a $\mu\text{Ci/g}$
Rh-102	206 d	Beta Positron E.C.	1.29	0.475(57%) 0.511(25%) 0.632(54%) 0.698(41%) 1.110(22%)	1900
Rh-102m	3.38 y	E.C.	none	0.418(13%) 0.475(95%) 0.632(54%) 0.768(30%) 1.050(41%) 1.110(22%)	290
Pd-107	7×10^6 y	Beta	0.04	none	90
Ru-103	39.6 d	Beta	0.70	0.497(88%) 0.610(6%)	5×10^9
Ru-106	368 d	Beta	3.54^a	0.512(21%) ^b 0.622(11%) 1.050(1.5%) 1.130(0.5%) 1.550(0.2%)	5×10^8
Tc-99	2.12×10^5 y	Beta	0.292	none	1.71×10^4

^aThe specific activities are for the fission product mixture. The values for ruthenium are the "worst case" and are probably too high.

^bThese include the daughter radiations from Ru-106.

The half-life of Rh-102m has been determined. Its value is 3.38 ± 0.05 yr. The precise value of this half-life is important in projecting the specific activity of a fission product at various cooling times. The specific activity of rhodium from a typical power reactor was found to be $290 \mu\text{Ci/g}$ at discharge.

The dose rate from fission product palladium has been measured. Surface dose rates from unshielded slabs is 0.83 rad/hr , but with 7 mg/cm^2 shielding (this is the thickness of the human skin) the dose rate is essentially zero.

An examination of the supply-demand relationships for Rh, Ru, Pd, and Tc was made. Recent geological data indicate that the world's platinum metal reserves are well established and that major discoveries are unlikely. Bureau of Mines data and calculations using a logistical growth model indicate expanded use of the platinum metals and a resource deficit after 1980. Breeder reactors, such as the LMFBR, may be a significant source of light platinum metals if these reactors are built in projected numbers.

The future use of these fission-produced elements is contingent on establishment of their radiological hazards. The control potential of these radioactive platinum metals was investigated in terms of environmental dispersion and mixing with nonradioactive platinum metals. Vaporization appears to be the primary dispersion mode for palladium, largely used for switching contacts, and rhodium, used in the catalytic production of nitric acid. There are sufficient thermodynamic and experimental data available to make qualitative estimates of these losses.

Analysis of potential radiological hazards of fission-produced palladium suggests that internal doses are very small and that the skin-contact dose may be biologically insignificant. More work should be done to confirm these conclusions before radio-palladium is used in large amounts.

Rhodium and ruthenium produced by nuclear reactors are radioactive enough to represent a present hazard and should be stored 20 to 40 yr before use as substitutes for nonradioactive material. When considered in terms of future resource scarcity, there may be justification for the separation and storage of these fission-produced platinum metals.

Plan for Next Period

The radiation characteristics of the fission products Rh, Pd, Ru, and Tc are now sufficiently well defined to permit analyses of the hazards associated with their use. No further studies are planned at this time. A report

summarizing all of the data obtained will be issued during the coming period.

Further studies on the technology for recovery of the fission products Rh, Pd, Ru, and Tc from nuclear fuels reprocessing wastes will be carried out. The objective is to find a simple economical process for isolating Rh, Pd, Ru, and Tc from the other fission products.

The market and safety analysis will be completed and a report will be issued.

The economic analysis to determine the feasibility of recovery and use of the fission products Rh, Pd, Ru, and Tc will be completed. Efforts will be initiated to develop fabrication processes for the fission products Rh, Pd, Ru, and Tc.

Reports Published

- R. G. Nelson and D. P. O'Keefe, *A Study of Tungsten-Technetium Alloys - Concluding Progress Report October 1, 1966 - August 1, 1968*, USAEC Report BNWL-865, Battelle-Northwest, 1968.
- C. A. Calvin, *Recovery of Palladium from Nuclear Waste Solutions Using a Packed Column of Tri-n-propyl Monomethyl Ammonium Chloride on an Inert Support*, USAEC Report ARH-1346, 1969.
- J. V. Panesko, *Recovery of Rhodium, Palladium, and Technetium on Strongly Basic Anion Exchange Resin*, USAEC Report ARH-1279, 1969.
- J. V. Panesko, *Process for Recovery of Rhodium, Palladium, and Technetium from Aged Reprocessing Wastes at Hanford*, USAEC Report ARH-733, 1968.
- F. P. Roberts, *Fission Product Rhodium and Palladium*, USAEC Report BNWL-CC-2239, Battelle-Northwest, 1969.

Estimated Date of Final Report

The next major report will be issued in January 1972.

EVALUATION OF LARGE-SCALE USES FOR PURIFIED FISSION PRODUCT XENON*

Objective of Work

The objectives of this work are to reprocess existing stocks of crude fission product xenon to such a purity (low Kr-85 content) that specimens can be offered to interested large scale users in exchange for the results of

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their evaluation of the product in their various applications

Progress and Results

Purification of the first volume (approximately 900 liters, STP) of fission product xenon was completed at ORNL. The composition and quality of this material was as follows

Xenon: 99.93%

Kr-85 less than 1 (one) microcurie per liter (STP) xenon

Isotopic Composition	F P Xe	Atmospheric Xe
Xe-124		0.096
126		0.090
128	0.01%	1.92
129	0.08	26.44
130	0.04	4.08
131	10.34	21.18
132	16.61	26.89
134	29.21	10.44
136	43.70	8.87

Other Gases

CO	None	H ₂	Less than 200 ppm (0.02%)
CH ₄	None	N ₂	Less than 300 ppm
Hydrocarbons	None	O ₂	Less than 200 ppm
H ₂ O	None	CO ₂	Less than 200 ppm
		Ar	Less than 200 ppm

These figures for other gases are the limits of detection by the ORNL methods used. From the xenon purity figure it is concluded that the actual concentration of these other gases is far below the indicated numbers. It is also concluded that the sum of the concentrations of all such contaminant gases could not reach 700 ppm.

Contacts with prospective evaluators or users of this material were continued. The progress of the project was also reviewed with visitors.

Plan for Next Period

Formal announcement of the quality and availability of the existing stock of purified gas will be made to a variety of producers, users, and nuclear fuel processors with the expectation that some requests for the material will be received. Purification of the remaining stock of available crude fission product xenon will be completed to yield a total of about double the present volume of pure material. Progress being made by those who will be provided with material for test or evaluation will be followed.

Reports Published

C. A. Rohrmann, Fission Product Xenon and Krypton - An Opportunity for Large Scale Utilization, *Isotop Radiat Technol*, 8(3) 253-260 (Spring 1971)

Estimated Date of Final Report

The next major report on this program will probably be the Annual Report for FY-1972, which will be issued early in FY-1973.

PROMETHIUM-146 REDUCTION*

Objective of Work

The objective of the Pm-146 burnout program is to demonstrate the technical feasibility of reducing the Pm-146 content in power source promethium to less than 0.1 ppm by neutron capture. Such a reduction of the Pm-146 content would substantially lower the radiation level of promethium and enhance its value for biomedical applications.

Progress and Results

Previous Pm-146 burnout experiments in Hanford and Savannah River reactors did not reduce the Pm-146 level in promethium to the desired level of less than 0.1 ppm. Analysis of these experiments and those related to the production of Pu-238 heat source material containing low Pu-236 concentrations indicated the importance of the Pm-147(n,2n)Pm-146 and Pm-147(γ,n)Pm-146 back reactions. Additional analysis showed that, in addition to the high Pm-146 cross section of 3500 barns, the extent of Pm-146 burnout is determined by the following factors: (1) Structural members of reactors containing iron, chromium, and nickel are important sources of high energy gammas capable of inducing the Pm-147(γ,n)Pm-146 reaction and should be avoided; (2) Promethium target matrix and cladding materials can be important sources of the Pm-147(γ,n)Pm-146 reaction. Aluminum may be an important high energy gamma source and should be avoided. Impurity levels of iron, nickel, and chromium may be important contributors to the Pm-147(γ,n)Pm-146 back reaction. Zirconium, beryllium, tantalum, and graphite are desirable cladding and matrix materials; (3) Location of the promethium target element in a well moderated region of a reactor.

*J. C. Sheppard, Battelle, Pacific Northwest Laboratory, Richland, WA 99352, Contract AT(45-1)-1830

can significantly reduce the contribution of the $\text{Pm-147}(n,2n)\text{Pm-146}$ back reaction

Analysis has shown that the Pm-146 content in promethium can be successfully reduced to 0.1 ppm or below by neutron irradiation, provided that the contributions of the $\text{Pm-147}(\gamma,n)\text{Pm-146}$ and $\text{Pm-147}(n,2n)\text{Pm-146}$ back reactions are carefully minimized. Other data indicate that such irradiation conditions are attainable in the HFIR, ORR, Savannah River Cf-252, and Hanford N reactors.

Plan for Next Period

This program was terminated at the end of FY-1971.

Reports Published

None

Estimated Date of Final Report

Results of this program were reported in BNWL-1308-7, Pacific Northwest Laboratory, Division of Isotopes Development Programs Quarterly Report May-July 1971, August 1971.

FISSION PRODUCTS DEVELOPMENT LABORATORY*

Objective of Work

The Fission Products Development Laboratory provides Sr-90 and Cs-137 products on the megacurie level of annual production. Feed material is received from Atlantic Richfield Hanford Corporation (ARHCO) as purified Sr(90)CO_3 and crude Cs-137 sorbed on an inorganic ion exchanger. The Sr-90 is converted to strontium titanate or strontium oxide, densified by hot pressing, and encapsulated as heat sources. The Cs-137 is purified by alum crystallizations, converted to Cs(137)Cl , and encapsulated as radiation sources or shipped as powder. Pilot plant testing of chemical process and mechanical fabrication techniques and production are accomplished simultaneously.

Progress and Results

Shipments of fission product feed material from ARHCO included two shipments of Sr-90 totaling 1 million Ci and one shipment of Cs-137 containing 400 thousand Ci. An improved Cs-137 shipping cask design and improvements in the cask elution process resulted

in a ten-fold increase in Cs-137 concentration in the process equipment with corresponding savings in processing costs. Detailed cost records for one process cycle resulting in 70 thousand Ci of Cs(137)Cl product revealed a basic product cost of \$0.14 per curie. A total of 19 Sr-90 sources containing 1.065 million Ci and 41 Cs-137 sources containing 47 thousand Ci were fabricated.

One shipment of Sr-90 from ARHCO contained unusually high (3.4 mole%) stable rare-earth content that would diminish the power density of the product fuel form. A separation process was developed and used to give 63% removal of the rare earths by an oxalate precipitation technique.

Hot pressing of the strontium titanate fuel form is used to provide the high power density pellets required for isotopic power generators. Improvements in hot press operating efficiency resulted from the design improvements in vacuum-atmosphere—resistance heated and inert-atmosphere—induction-heated hot presses. Die body life was prolonged by using carbon filament reinforced strength members.

Test pressings of stable strontium titanate powder are being made to define the optimum process conditions. A problem strontium titanate composition, $\text{Sr}_3\text{Tl}_2\text{O}_7$, that results in low pellet density was defined by x-ray diffraction analysis of test pellets. A procedure utilizing calorimetric analysis of Sr(90)CO_3 followed by dry mixing with TiO_2 was developed to provide closer control of product composition.

Containment of strontium titanate fuel forms is usually provided by Hastelloy-C-276 capsules which are sealed by inert-gas, tungsten-arc welding to provide 0.055-in penetration. Small variations in alloy formulation or in fit between the cap and capsule body can cause welds of insufficient penetration. A remote plasma arc weld technique was developed to provide greater than 0.080-in penetration in capsule seal welds.

Plan for Next Period

Several of the operating cells need refurbishing to correct equipment failures which have occurred over the past several years that the equipment has been in use. Work will continue to improve the fuel forms which affect hot-pressed pellet integrity.

Reports Published

None

Estimated Date of Final Report

Continuing project

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ORNL REACTOR-PRODUCED RADIOISOTOPES PROGRAM*

Objective of Work

The principal objective of the ORNL reactor-produced radioisotopes program is the development of new and improved techniques and procedures to prepare, characterize, and support the use of radioisotopes produced by neutron irradiation. Improved or previously unavailable research materials for use in medical, biological, and physical sciences are provided.

Experimental irradiations are performed in the HFIR (see figure) and other AEC research or production reactors in order to obtain very high neutron fluxes

($>10^{15}$ n/cm²·sec) and desired neutron energy spectra for the reactions being investigated.

The program objective is accomplished by (1) examination of the principal modes of neutron interaction [(n,α), (n,p), and (n,γ)] with naturally occurring or enriched isotope targets, (2) development of appropriate chemical or physical processes to recover and purify the desired radioisotope, (3) characterization of the product purity and determination of unknown or uncertain nuclear properties such as half-life and decay scheme, and (4) establishment of cooperative programs with research groups to promote the beneficial applications of these radioisotopes.

Progress and Results

The preparation of radioisotopes for use in the development of medical diagnostic procedures and

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Plan view of High Flux Isotope Reactor (HFIR) core.

clinical applications through cooperative programs was emphasized as part of our special research materials interests. Cooperative research studies demonstrated the value of K 43 in imaging the myocardium and quantizing infarction. An appropriate increase in the ORNL pilot production effort was made in order to supply the increasing demand for K 43 for use in clinical evaluation studies. Cerium 141 in the form of high specific activity wafers of $\text{Ce}(141)\text{O}_2$ was supplied as a special research material to the Los Alamos Scientific Laboratory.

Separate project summaries describe work on K 43, P 33, Si 32, Tm 170, and Pt 195m.

Plan for Next Period

This continuing project will emphasize the use of reactor produced radioisotopes by continuing to develop special research materials for cooperative programs with publicly or privately funded research groups and to improve the quality of the radioisotope preparations.

Reports Published

See summaries of individual projects.

Estimated Date of Final Report

Continuing project

POTASSIUM-43*

Objective of Work

The objectives of this project are to prepare K 43 by the $\text{Ca-43}(n,p)\text{K-43}$ reaction, using isotopically enriched $\text{Ca}(43)\text{O}$ targets, in quantities sufficient for medical and biological experiments, to define a method for separating K 43 from the target in a purity suitable for medical use, and to establish cooperative programs with medical institutions interested in evaluating its usefulness. Potassium 43, with a half life of 22.5 hr and gamma ray emissions of 0.373 and 0.617 MeV, is potentially useful for metabolic and clinical studies of blood flow, rejection of transplanted organs, and kidney function because the dose rate would permit multiple doses.

Progress and Results

About 20 targets of enriched $\text{Ca}(43)\text{O}$ were irradiated in a fast neutron flux of $\sim 1 \times 10^{15} \text{ n/cm}^2 \text{ s}$ to deliver over 150 mCi K 43. Most of the activity was supplied for evaluation under medical cooperative programs to the University of Mississippi Medical Center, Peter Bent Brigham Hospital in Boston, the Johns Hopkins Medical Institutions in Baltimore, the University Hospitals of Cleveland and V A Hospital, Durham, NC. These cooperative research studies have demonstrated the value of K 43 in imaging the myocardium and quantizing infarction (see figures).

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Anterior and left anterior oblique gamma camera images of a patient with extensive anterior and inferior myocardial infarction

The effectiveness of the calcium recovery process and the ability of the ion-exchange procedure to produce a product acceptable for human use have been demonstrated. The recovered calcium target shows no degradation of enrichment after a year of use. Radioactivities other than K-42 in the product are usually at a level less than 10 nanocuries/ml. The K-42 is about 12% of the activity at delivery.

The physical decay properties of K-43 have been studied and evaluated.

Plan for Next Period

Efforts at process improvement will continue to shorten the calcium recovery procedure and maintain reliability in processing larger batches. Improvements in labeling and handling of the product will be effected. The study of the decay properties and production technique will be published in reports.

Reports Published

- P. J. Hurley, M. Cooper, R. C. Reba, J. K. Poggenburg, and H. N. Wagner, Jr., ^{43}KCl A New Radio-pharmaceutical for Imaging the Heart, *J Nucl Med*, 12 516 (1971)
- B. L. Holman, J. K. Poggenburg, D. F. Adams, P. Eldh, and S. J. Adelstein, Assessment of Myocardial Perfusion and Viability with K-43, *J Nucl Med*, 12 366 (1971) (Abstract)
- R. O. Smith, P. H. Lehan, H. K. Hellems, and J. K. Poggenburg, Limitation of the Bolus Isotope Infusion for Myocardial Scanning — A Possible Solution, *J Nucl Med*, 12 394 (1971) (Abstract)
- J. K. Poggenburg, K-43 The Reactor Production, Physical Properties, and Potential Availability of a Useful Radioisotope, *J Nucl Med*, 12 457 (1971) (Abstract)
- K. O. Smith, P. H. Lehan, H. K. Hellems, and J. K. Poggenburg, Scatter Problem When Counting the Lower Energies of K-43, *J Nucl Med*, 12 464 (1971) (Abstract)

Estimated Date of Final Report

1972

PHOSPHORUS-33*

Objective of Work

The purpose of this project is to develop methods of preparing hundred-millicurie quantities of carrier-free

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P-33 containing $\leq 5\%$ P-32. The two methods of producing P-33 being evaluated are based on the irradiation of highly enriched targets of S-33 ($>92\%$) or Cl-36 ($\sim 69\%$), in a fast neutron flux. Phosphorus-33 (25.2 d, 0.248-MeV β_{max}) has both a longer half life and a lower energy beta than P-32 (14.3 d, 1.709-MeV β_{max}), which makes it advantageous for autoradiography, ecological and agricultural studies, synthesis of tagged complex organophosphorus compounds, and double labeling experiments.

The HFIR is utilized for quantity production of high quality P-33 at reasonable cost for utilization by commercial vendors in providing tagged compounds and specialized preparations.

Progress and Results

The Cl-36(n, α)P-33 reaction has been determined to be the most practical method for large-scale production, but the available ($>92\%$) enriched $\text{K}_2\text{S}(33)\text{O}_4$ was also used to produce over 4 Ci of P-33 product. The recycled KCl(36) target has attained an enrichment of 69% Cl-36 with Cl-35 depleted to 3%. The P-32 content in a product produced from this target is $<1\%$ at processing within one week after reactor discharge. The capability of routinely producing curie batches of P-33 has been demonstrated. Efforts have been directed at building up an inventory of KCl(36) of intermediate enrichment for further irradiation enrichment and blending into existing target material.

Process improvements include the addition of a solvent extraction step using methyl isobutyl ketone to remove the bulk of the iron used as a phosphate carrier. This step was found to remove concomitantly the traces of antimony activity leached from the quartz target ampules. A small cation exchange column is used as a final cleanup step. A procedure was developed for removing casual Cr-51 activity from the product. The Cr-51 is usually in the CrO_4^{2-} form and is reduced to Cr^{3+} with SO_2 gas so it can be removed on a cation exchange column.

Plan for Next Period

Efforts will continue towards process and target recovery improvements. The problem of long-term buildup of Cs-134 activity by activation of cesium impurity in the targets will be studied and a method for its removal will be devised. Buildup of enriched target will continue.

Reports Published

- L. C. Brown, Determination of P-32 and P-33 in Aqueous Solution, *Anal Chem*, 43 1326 (1971)

Estimated Date of Final Report

Continuing project

SILICON-32***Objective of Work**

The objective of this project is to develop the reactor technology for the preparation of microcurie quantities of Si-32 (~650 y). Silicon-32 exhibits many desirable physical decay characteristics that make it an interesting and attractive radionuclide for a variety of uses: (1) as the only long-lived radionuclide of silicon for use in agriculture, geology, solid-state physics, etc., (2) as a parent for a Si-32–P-32 generator system which would offer a constant supply of P-32, and (3) as a long-lived counting standard for P-32 determinations.

Progress and Results

Three multiple-neutron-capture reaction paths have been considered for the production of microcurie quantities of Si-32 in the HFIR. Two of these reaction paths which should yield carrier-free Si-32 were selected for investigation: $[P-31(n,\gamma)P-32(n,p)Si-32]$ and $[S-34(n,\gamma)S-35(n,\alpha)Si-32]$. A short irradiation of a small $AlPO_4$ target has been performed to produce a tracer level sample for chemical process and separations development. Larger targets of $AlPO_4$ and $K_2^{34}SO_4$ have been fabricated and inserted for extended irradiation, i.e., ≥ 6 to 12 months, at high thermal and fast neutron fluxes for microcurie production.

Plan for Next Period

Process development will be undertaken using the tracer level sample mentioned above while larger production targets are irradiated. Production samples will be processed as the irradiations are completed. Measurements of the Si-32 half-life will be made and P-32 burnup cross section will also be determined in the course of this work.

Reports Published

None

Estimated Date of Final Report

Continuing project

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THULIUM-170***Objective of Work**

The objectives of this research are to provide sufficient data on Tm-170 to permit reasonable assessment of its potential application, to permit preliminary engineering design of power sources, and to develop data for safety analysis. It has been proposed as an isotopic power source for short-duration missions. The economics for reactor production of Tm-170 (128.5 days, 0.32 MeV β_{av}) from natural Tm-169 are favorable and the sesquioxide (Tm_2O_3) offers a promising fuel compound, having a practical specific power of approximately 2 W/g and a power density of approximately 16 W/cm³. The study includes measurements of thermal conductivity, leach rates, high-temperature compatibility with suitable containment metals, vapor pressure, and fuel-form densification.

Progress and Results

The effusion method, using two Knudsen cells of different sizes, was employed in the determination of rates of effusion and partial and total pressures of various vapor species above solid Tm_2O_3 and Yb_2O_3 (both nonradioactive). Two Tm_2O_3 – Yb_2O_3 mixtures (17 wt% and 28 wt% Yb_2O_3) have been prepared by the coprecipitation method for this work. The selection of the first Yb_2O_3 – Tm_2O_3 mixture (17 wt% Yb_2O_3) is based on the typical initial composition of the thulium(170) sesquioxide fuel source. The second mixture is chosen to represent the composition of the fuel source after a period of one half-life (128 days) which was assumed to be the maximum mission life of a thulium(170) sesquioxide source.

Values of the apparent vapor pressure of Tm_2O_3 (sum of the partial pressures of vapor species above solid Tm_2O_3) thus obtained at 1990°C ranged from 2.38×10^{-7} to 3.25×10^{-7} atm. The apparent vapor pressure of Yb_2O_3 at 1990°C is 2.22×10^{-6} atm.

In the thulium(170) sesquioxide–refractory metal compatibility studies, 16 compatibility couples have completed exposure runs at 1600°C. Of these, the exposure time for 8 couples was 2500 hr, and that for the other 8 was 5000 hr. The potential capsule materials tested included TZM, Ta–10% W, T-111, and tungsten. Two thulium(170) sesquioxide–tungsten couples were exposed at 2000°C for 500 hr. The results

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of the examination of the radioactive samples will be compared with the data from a similar study using nonradioactive Tm_2O_3 and Yb_2O_3 to determine whether there was any effect attributable to radioactivity

Plan for Next Period

Further experiments will be carried out to complete the vapor pressure data for Tm_2O_3 , Yb_2O_3 , 17 wt% Yb_2O_3 – Tm_2O_3 , and 28 wt% Yb_2O_3 – Tm_2O_3 in the temperature range from 1800 to 2300°C. The compatibility study will emphasize examination of the exposed compatibility couples and assessment of the results of examination.

A final summary report of all AEC-sponsored research on thulium oxide properties will be completed.

Report Published

None

Estimated Date of Final Report

1972

PLATINUM-195m*

Objective of Work

The objectives of this work are to develop methods for preparing platinum isotopes suitable for whole-body scanning in biological and medical experiments and to establish cooperative programs with medical research groups interested in evaluating their usefulness.

Recent medical research has shown that certain platinum compounds act as chemical therapeutic agents towards certain tumors. There is need for platinum radioisotopes as tools for investigating the therapeutic mechanism involved. Platinum-195m (4.1 d) has gamma emissions of 99 keV (12%) and 129 keV (3%) which make it suitable for whole-body scanning.

Progress and Results

Several reactor irradiations of 57% enriched Pt-194 were made and a cooperative research program was established with Michigan State University to test the radioisotope in their antitumor studies. The Pt-195m was supplied in an aqua regia solution from which both the cis- and trans- dichloro-diammine Pt(II) compound was synthesized and administered intraperitoneally to tumor-infected mice.

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The irradiations identified several problems with the Pt-195m. The best enrichment of Pt-194, the 57% target used, contains 8% Pt-196 and 0.4% Pt-198. Because of the larger capture cross sections of Pt-196 and Pt-198, the irradiated target contains much more Pt-197 (20 hr) and Au-199 (from Pt-199 β^- Au-199) activity than Pt-195m activity. The gold activity can be completely removed by two extractions with methyl isobutyl ketone. The presence of Pt-197 requires an additional correction in the whole-body counting procedure and requires the use of a silicon or germanium detector to assay both the Pt-195m and Pt-197 which have photon energies too close to be adequately resolved with NaI. The results of a 10-day irradiation suggest that the activation cross section to produce Pt-195m may be an order of magnitude less than the 0.09 barn quoted in BNL-325.

Plan for Next Period

Platinum-195m will continue to be supplied for cooperative research. Other platinum isotopes will be investigated. Experiments will be directed toward establishing the production cross sections for Pt-195m.

Reports published

None

Estimated Date of Final Report

1973

BNL ACCELERATOR PRODUCTS DEVELOPMENT*

Objective of Work

The overall objective of the BNL accelerator products development program is to increase the use of radionuclides and techniques involving their use in medicine, industry, and research. The program is designed to make maximum use of the unique accelerator capabilities available at Brookhaven, including a new high-energy proton linear accelerator presently under construction, as an injector for the Alternating Gradient Synchrotron as well as the rebuilt 60-in cyclotron and two Van de Graaff generators. The work is primarily exploratory in nature and involves determining the feasibility of radionuclide production by new techniques or improving existing methods. Goals of this

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program are the development of new radionuclides as well as increasing the quantities, improving the purity, and reducing the cost for existing nuclides. The effort also entails the development of technology required for radionuclide preparation in existing and planned medical and commercial cyclotrons and the supplying of special radioisotopes for the collaborative BNL biomedical radioisotope program and collaborative programs with other institutions. The potentialities of high- (spallation) and low-energy proton or triton reactions and the use of on-machine recoil labeling techniques are being evaluated for product development. The program is divided into three categories: (1) the BNL Linac Isotope Producer (BLIP), (2) cyclotron products, and (3) Van de Graaff generator technology.

Progress and Results

See summaries of individual projects

Plan for Next Period

See summaries of individual projects

Reports Published

See summaries of individual projects

Estimated Date of Final Report

Continuing project

THE BROOKHAVEN LINAC ISOTOPE PRODUCER (BLIP)*

Objective of Work

The objective of BLIP project is to explore the capabilities of a high-energy proton linear accelerator for the production of a large variety of radionuclides by spallation reactions as well as by low-energy proton reactions.

The construction of a 200-MeV proton Linac is presently nearing completion at Brookhaven National Laboratory to inject high-intensity pulses of protons into the Alternating Gradient Synchrotron (AGS), a machine used for high-energy physics research. Excess beam capacity of the Linac (average current of $\sim 180 \mu\text{A}$), required for beam monitoring and maintenance of equipment stability, is available for radionuclide production studies in a beam-stop facility where targets

may be placed to intercept the beam. Such a facility will have unique potential for isotope production for two reasons: (1) the high proton energy combined with a relatively high beam intensity and (2) the continuous mode of operation. High incident particle energy will allow the bombardment of multiple independently interchangeable targets simultaneously while continuous operation will make long term irradiations of targets feasible.

The BLIP project is designed to evaluate a number of unexplored capabilities in isotope preparation, including (1) the production of short- and long-lived activities by both fast and slow reactions simultaneously, (2) the discovery of new nuclides, (3) the preparation of both neutron rich and neutron deficient products, (4) greatly increased quantities or improved purity of certain existing nuclides with important application potential, and (5) the production of milligram quantities of long-lived materials presently available only in minute amounts. Part of the beam time is expected to be available for service irradiations.

Progress and Results

Since BLIP incorporates the first high-energy accelerator to be used for isotope development and production, much original effort has been required to realize the full capability of this unique machine. The design of a facility for utilization of the beam has been completed and construction should be nearly complete by the end of 1971. Work is being coordinated with the start-up of the Linac which is currently in preliminary operation but will not require beam dumping until early in 1972.

The water tank beam stop was fabricated and installed. A 32-ft depth of water will serve as a transparent, flexible shield against the high-energy neutrons generated during operation. A prefabricated operations building has been constructed over the beam stop tank. The remote target manipulation system has been fabricated and installed in the tank. This will allow the insertion into or removal from the beam of a number of individual targets, each with its own cooling system.

A standard target holder assembly has been developed and is shown in the figure. Preliminary studies of target processing, equipment development, and the modification of existing processing facilities have been started. Work has been started on several products, including Fe-52, I-123, Xe-127, and Ta-179. Several low-intensity bombardments have been carried out on target materials in the Linac tunnel to verify heat transfer calculations.

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BLIP target assembly. From left to right: 1. Target holder with inlet and outlet cooling water connections. 2. Two typical metallic targets. 3. Metal O-ring. 4. 0.005-in. stainless-steel window. 5. Retaining flange.

Plan for Next Period

Deflection of the 200-MeV proton beam into BLIP should start by the beginning of 1972. The facility, including the target handling equipment and target cooling system, should be completed and in operation early in the year. Experimental studies of radiation damage, heat transfer, and target manipulation problems will be undertaken.

Preliminary production parameters for certain nuclides will be evaluated for comparison with previously calculated data. The development of chemical processes for specific products will be under way and exploratory production should start during 1972. Some of the typical products to be investigated initially include (1)

I-123 (half-life, 13 hr), which has the best physical properties of all iodine isotopes for medical diagnostic scanning; (2) Fe-52 (half-life, 8 hr), which is the only isotope of iron suitable for clinical diagnostics and is expected to find use in studying the distribution of functioning bone marrow in various diseased states; (3) Ta-179 (half-life, 1.6 yr), which decays by electron capture, emits 55-keV x rays and should find important uses in radiography, transmission scanning, and as a source for gaging instruments; and (4) Xe-127 (half-life, 36 d), which, with its higher energy photons, appears to be superior to the presently used Xe-133 for studies of lung perfusion and ventilation.

Some irradiations of special targets for outside interests should also take place during 1972.

Reports Published

None

Estimated Date of Final Report

Continuing project

BNL CYCLOTRON PRODUCT DEVELOPMENT***Objective of Work**

The objectives of the BNL cyclotron program are twofold (1) to develop production technologies for radionuclides for potential application in collaborative studies with the BNL Medical Research Center and for cooperative programs with other institutions and (2) to develop production information useful to both medical and commercial cyclotrons, existing and planned. The BNL 60-in cyclotron can accelerate protons to 40 MeV, deuterons to 20 MeV, He-3 to 60 MeV, and He-4 to 40 MeV. Because of its relatively high energy and its ability to accelerate both hydrogen and helium isotopes, this machine has unique capabilities both to produce useful quantities of biomedically useful radionuclides and to develop production parameters for use on other machines with lesser capabilities. The program is also involved in areas of target development and materials, chemical processing, and purification procedures.

Plan for Next Period

See summaries of individual projects

Reports Published

- E Lebowitz, M W Greene, M Kinsley, and P Richards, The Development of New and Improved Isotopes for Nuclear Medicine with the Brookhaven Cyclotron — Dy-157 and Bi-204, *J Nucl Med*, 12(6) 376-7 (1971)

Estimated Date of Final Report

Continuing project

DYSPROSIUM-157***Objective of Work**

Hydroxyethylethylenediaminetriacetic acid (HEDTA) complexes of many rare-earth nuclides are good bone seekers, while many of the rare-earth colloids localize in bone marrow. Dysprosium-157 has excellent physical characteristics (half-life, 8.2 hr, photon energy, 326 kilovolts in 91% abundance). The objective of this study is to develop the necessary production technology and produce sufficient Dy-157 for evaluation as a bone and bone marrow scanning agent. This work is being carried out in cooperation with Dr. John McAfee of the State University of New York at Syracuse.

Progress and Results

A method has been developed for producing and separating by ion exchange Dy-157 in high yield (~30 mCi/ μ Ah), high-specific activity (~100 μ g of rare-earth material in sample), high radiopurity (<10⁻⁴ radio-contaminants), and high chemical purity. In addition, the excitation function has been measured and evaluation of methods of production carried out to assist potential producers of Dy-157.

Good results have been obtained at the Upstate Medical Center in Syracuse, using Dy-157 for both bone and bone marrow imaging depending on the chemical form in which the nuclide is administered.

The excitation function for the reaction Tb-159(p,n)Dy-157 was determined by irradiation of a stack of terbium foils and subsequent assay of the Dy-157 activity with a Ge(Li) detector. Copper flux monitors were included in each stack of foils to determine the proton beam current.

In the production method, a terbium foil (825 mg/cm²) is irradiated with 33-MeV protons in the external cyclotron beam. After bombardment and target dissolution the carrier-free Dy-157 is separated from the terbium target material by ion exchange chromatography on AG 50W-X12 resin with elution by α -hydroxyisobutyric acid (HIBA). Optimum conditions for the separation have been determined and Dy-157 of excellent purity has been achieved. The product activity is converted to the desired chemical form for administration.

This work has shown that, although very high yields are obtained at higher energy protons, the 22- and 26-MeV commercial cyclotrons can still produce practical quantities of Dy-157.

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Plan for Next Period

More full-scale production runs will be carried out to supply collaborating institutions with Dy-157 for clinical evaluation

Reports Published

- G Subramanian, J G McAfee, R J Blair, R E O'Mara, M W Greene, and E Lebowitz, Dysprosium-157-HEDTA for Skeletal Imaging, *J Nucl Med*, 12 558-61 (1971)
- G Subramanian, J G McAfee, M Rosenstreich, A Mehter, M W Greene, and E Lebowitz, Dysprosium-157 Gluconate A New Radiopharmaceutical for Bone Marrow Imaging, *J Nucl Med*, 12(6) 400 (1971)

Estimated Date of Final Report

July 1972

AUXILIARY CYCLOTRON BEAM MONITOR DEVELOPMENT*

Objective of Work

The objective of this project is to develop methods to accurately measure the amount of a cyclotron beam that is actually striking the target. Some causes of the uncertainty in this measurement may be the use of a target smaller than the beam, a misaligned beam, or measurement by the target current monitor of secondary particles introduced into the beam by its passage through a window or air gap. A simple technique to check the beam current consists in simultaneously irradiating a copper foil with an identically shaped target. The cross section for the production in copper of the measured reaction product must be known. If a Ge(Li) detector is used for the radioisotope assay, no chemistry is required and the beam current is calculated from the assay and cross section data.

Progress and Results

In the previous report (TID-4066) an auxiliary cyclotron beam monitor for use with He-3 and He-4 beams was described. A similar technique for monitoring proton beams using cross sections reported in the literature gave serious discrepancies between the 60-in

Excitation function for the reaction $\text{Cu-63}(p,2n)\text{Zn-62}$. Present work indicated by error bars and compared with other literature values

BNL cyclotron monitoring system and the foil monitors. Therefore, independent measurements of proton cross sections on copper were made. Polyethylene foils were bombarded as a check of the cyclotron beam integration system by using the very accurately measured ($\pm 3\%$) cross sections for the $\text{C-12}(p,pn)\text{C-11}$ reaction to determine beam current. Reasonable agreement with the cyclotron beam integration system was obtained. Our results are shown in the figures.

This auxiliary beam monitoring system offers a simple, rapid, and accurate method to cyclotron users for determining the beam current striking a target.

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Excitation function for the reaction $\text{Cu-65}(p,n)\text{Zn-65}$. Present work indicated by error bars and compared with other literature values

Plan for Next Period

Other foil monitors will be developed as the need for them arises in the accelerator program

Reports Published

E Lebowitz and M Greene, An Auxiliary Cyclotron Beam Monitor, *Int J Appl Radiat Isotop*, 21 625 (1970)

Estimated Date of Final Report

Continuing project

VAN DE GRAAFF GENERATOR TECHNOLOGY*

Objective of Work

The development of methods for the production of Mg-28 and F-18 by the bombardment of targets with tritons accelerated in a Van de Graaff generator was described in a previous edition of this report (TID-4066)

A study is currently under way to determine the feasibility of labeling potentially useful biological compounds with F-18 , a 110-min positron emitter, by recoil techniques. The method involves bombarding of a neon gas target in a container, whose walls are coated with a compound to be labeled, with deuterons accelerated in the Van de Graaff generator. The F-18 atoms produced by the reaction $\text{Ne-20}(d,\alpha)\text{F-18}$ recoil to the target walls and react with the compound to be labeled.

Progress and Results

The targetry and separation technology have been developed and a study is presently under way to determine the feasibility of labeling uracil by this technique. Collimation of the deuteron beam prevents it from hitting and destroying the compound to be labeled and the neon target gas is recirculated through a liquid nitrogen trap to cool the window and target. By having the compound to be labeled in the solid phase rather than as a gas, higher yields are achieved and the radiation damage done to the target compound and product compound is minimized. The $\text{Ne-20}(d,\alpha)\text{F-18}$ reaction is advantageous for F-18 recoil labeling because the target gas is inert and because the α -recoil energy gives the F-18 much initial kinetic energy.

After the deuteron irradiation, the compound coating the walls and the fluorine-labeled product are dissolved in distilled water and passed through a SrF_2 column to

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Separation of 1 mg of 5-fluorouracil from 300 mg of uracil.

remove F^- ions (decontamination factor $>10^4$). The effluent of the SrF_2 column is then eluted through an Amberlite CG-50 (200 to 400 mesh) cation exchange column (95 cm $\times$ 255 cm at $54^\circ C$) to separate the F-18 labeled compound from the unlabeled compound and degradation products (see figure). Since the amount of carrier fluoro-compound can be as small as desired, very high specific activity material can be produced.

The processing time is approximately two hours from the end of bombardment until the final product is ready for medical use.

The effect of the filling target gas pressure on the percentage of the F-18 produced which is in the form of 5-fluorouracil is shown in the figure. Fluorine labeling of uracil at other positions of the ring leads to an unstable molecule.

Since the yield stays approximately constant well beyond pressures corresponding to the F-18 recoil reaching the walls at higher than thermal velocity, the labeling is not done by fluorine of high kinetic energy. It would be expected that a mixture of neutral species and electronically excited species contribute to the labeling yield. The very low labeling yield obtained at 1

Yield of F-18-uracil as a function of target gas pressure.

atm indicates that the kinetically deexcited fluorine reactive species will become unreactive if enough collisions (which could lead, for example, to chemical reactions with impurities) are made before it reaches the labeling compound.

Plan for Next Period

Some preliminary measurements have been made which indicate that substantially more F-18 activity could be produced with higher energy deuterons (~ 7 MeV) to which we presently do not have access. When a higher energy deuteron beam becomes available next year on the Brookhaven Radiobiology Van de Graaff, we estimate that enough activity could be produced for clinical studies.

Reports Published

None

Estimated Date of Final Report

Continuing project

XENON-127*

Objective of Work

The objective of this study is to develop the production technology as well as to produce Xe-127 for clinical evaluation. Thirty-six-day Xe-127 with its higher energy photons (172 and 203 keV), lack of particle emission, and longer shelf life, should be superior to the presently used 5-day Xe-133 with its low-energy photons (which are heavily scattered) for

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studies of lung perfusion and ventilation. Initial development of the chemical processing and preliminary clinical evaluations will utilize the BNL 60-in cyclotron for production of Xe-127. Then large-scale production and clinical evaluation will be carried out on BLIP, based on the cyclotron results.

Progress and Results

A study has been initiated to develop the necessary technology for the production and separation of Xe-127 and to make the isotope available for medical evaluation. The chemical processing consists of melting the salt target (CsCl) and sweeping out the xenon with a stream of helium. The gas stream flows through a dry ice trap to remove any iodine activity and then through a liquid nitrogen cooled trap which catches the xenon. This xenon product can then be used as a gas for ventilation studies or can be dissolved in saline for perfusion and ventilation studies.

Plan for Next Period

The optimum technology for the production, separation, and clinical use of Xe-127 will be developed. Clinical studies with this isotope will be performed at the BNL Medical Research Center.

Reports Published

None

Estimated Date of Final Report

Continuing project

BISMUTH-204*

Objective of Work

The objective of this work is to study the feasibility of producing Bi-204 on the BNL cyclotron for medical evaluation studies. It has been reported that bismuth localizes in certain types of tumors with a relatively high tumor-to-nontumor ratio, suggesting its use in nuclear medicine for tumor diagnosis. Unfortunately, none of the radioisotopes of bismuth have very good physical properties for use as a scanning agent. Bismuth-204 appears to have the most potential. This study is being carried out in a cooperative program with Dr. Richard E. Peterson, University of Iowa Hospitals, Iowa City.

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Progress and Results

The preferred method for Bi-204 production is via the Pb-206(p,3n)Bi-204 reaction. Because of radiation dose considerations, the Pb target thickness and incident proton energy must be carefully controlled to maintain high radioisotopic purity of the Bi-204. A metallic Pb target has been developed which allows the water to impinge directly on the Pb, thereby allowing the use of relatively high proton beam currents. This method of production gives a yield of a few mCi/μAh and high product purity (<1% Bi-205, <10% Bi-203). A rapid electrolysis method has been developed to separate carrier-free Bi-204 in high yield from the Pb-206 target and also allow satisfactory recovery of the enriched lead for reuse. The figure shows the electrolysis rig. After electrolysis, the bismuth is leached off the platinum cathode. The following table gives the chemical analysis of a typical Bi-204 product solution.

	μg		μg		μg
B	0.1	Cr	<0.1	Hf	<1
Pb	0.5	Sn	<0.1	Ga	<0.1
Si	~3	Ni	<0.1	In	<0.1
Cu	0.5	Mo	<0.1	Os	<1
Fe	0.3	Ge	<1	Pd	<0.1
Ag	0.5	Zr	<0.1	Rh	<1
Mg	<0.01	Be	<0.01	Ru	<1
Bi	<1	Ti	<0.1	Co	<0.1
Hg	<1	Nb	<0.1	Bi	<0.1
Sb	<0.1	Pb	<0.1	Al	<0.1
As	<1	Ir	<1	Cd	<1
Mn	<0.01

Apparatus for electrolytic separation of Bi-204 from Pb target. 1. Screw link 2. 50 ml beaker 3. Pt foil 0.39 mm, 19 mm dia connected with 0.4 mm Pt wire 4. Magnetic stirrer 5. Electrolyte 6. Pyrex pipe 8 mm dia with fine frit 7. Teflon plug 8. Pt wire 0.5 mm dia

Clinical evaluations are under way at the University of Iowa

Plan for Next Period

Several full-scale production runs will be carried out to verify the production method and to continue clinical evaluations of Bi-204 for tumor diagnosis

Reports Published

None

Estimated Date of Final Report

June 1972

ORNL ACCELERATOR-PRODUCED RADIOISOTOPES PROGRAM*

Objective of Work

The objective of this work is the development of improved targets and target materials, of separation-process chemistry, and of recovery techniques for target materials for the production of isotopes in the ORNL 86-in cyclotron

Cyclotron production offers a broad range of available radionuclides of any element and in some cases offers the only practical radionuclide for a given element. The principal areas of application of neutron-deficient radionuclides have been radiobiology, nuclear medicine, chemistry, metallurgy, and physics. A primary source of carrier-free, neutron-deficient radioisotopes continues to be the ORNL 86-in cyclotron

Progress and Results

A number of cooperative programs were established, or continued, to assist cooperative participants to fill specialized requirements for specific radionuclides not usually available. A considerable increase in the Ga-67 pilot production operations was necessitated by product requirements for an Oak Ridge Associated Universities sponsored cooperative group study of the applications of Ga-67 in medicine. Indium-111 pilot production was also increased in response to growing interest in its medical applications.

Development work on Rb-83 and Rb-84 was completed during the previous reporting period with the exception of a new half-life determination of 86.2 days

for Rb-83. New development was initiated on the production of Cs-129 by proton reaction on enriched Xe-129. Considerable interest has been evidenced in evaluation of this radionuclide for heart studies.

Separate project summary reports describe the development activities for Ga-67, In-111, Ba-133m, Bi-206, and Pb-203.

Plan for Next Period

See summaries of individual radionuclides

Reports Published

See summaries of individual radionuclides

Estimated Date of Final Report

Continuing project

GALLIUM-67*

Objective of Work

The objective of this program is to determine the optimal proton production parameters, e.g., target configuration, chemical processing, etc., for the preparation of carrier-free Ga-67 (78.2 hr) in acceptable purity and quantity for clinical applications research and development. Interest in this isotope has been spurred by evidence of a high uptake of carrier-free Ga-67 by lymphoid tumors in both animals and humans. This discovery was made during the evaluation of carrier-free Ga-67 as a bone-scanning agent for a patient with Hodgkin's disease and has since been successfully extended to clinical studies of several types of malignant neoplasms.

Progress and Results

The chemical processing development stage, i.e., separation and purification of the product, has been completed and reported. The natural-isotopic-abundance zinc tube target (0.375 in OD $\times$ 0.020 in wall thickness) has been tested extensively and found acceptable for moderate-scale production (200 to 300 usable mCi/target). This target configuration has been used to produce more than 25 Ci of Ga-67 (reference time end of bombardment) during the past year for distribution through cooperative programs and for sale as an experimental product.

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Plan for Next Period

The current zinc tube target design also yields Ga-66 (9.5 hr) that is produced concurrently by the Zn-67(p,n)Ga-66 reaction. This activity must be allowed to decay, usually 3 to 4 days, before the Ga-67 product can be used. Further effort will be applied to develop enriched Zn-68 targets for increased Ga-67 yields and minimum Ga-66 contamination, thereby reducing the required decay time. Pilot production will be continued to supply Ga-67 to cooperative participants and as an experimental product for research and development evaluation.

Reports Published

L. C. Brown, Chemical Processing of Cyclotron-Produced Ga-67, *Int J Appl Radiat Isotop*, 22, 710-13 (1971)

Estimated Date of Final Report

Continuing project

INDIUM-111*

Objective of Work

The initial objectives of this program were to define and optimize the pertinent production parameters for the economical preparation of In-111 of satisfactory quality for radiopharmaceutical manufacture. The current objectives are to obtain practical operating experience of the procedures developed for pilot production and to provide this radionuclide to interested users as a special research material for their evaluation and application. Indium-111 has been either used for or suggested for such studies as spinal-cerebral cisternography, aerosol lung studies, delayed brain scanning, visualization of the lymphatic system, metabolic studies of indium-labeled macroaggregates and colloids, and tumor localization. Indium-111 has gamma emissions of 171 keV (89%) and 245 keV (94%) — ideally suited for external detection — and an optimal half-life (2.82 d) for labeling and distribution studies which must be carried out over 24 hr or longer.

Progress and Results

Development of the large-scale cyclotron production of In-111 by the Cd-111(p,n)In-111 reaction path and

of the associated chemical processing, i.e., separation and purification of the product, has been completed and reported. The product has a radionuclide purity of $>99\%$ and contains $<20\ \mu\text{g}$ Cd and $<5\ \mu\text{g}$ Fe per ml of solution. Additional operating experience has been obtained from pilot production during the past year that has shown that the process consistently yields a pyrogen-free, high-purity product acceptable for human use. More than 3 Ci of In-111 has been produced during this period for distribution as a special research material.

Plan for Next Period

Pilot production will continue to supply In-111 as a special research material in batch lots to customers interested in its use and evaluation.

Reports Published

None

Estimated Date of Final Report

Continuing project

BARIUM-133m*

Objective of Work

The medium to heavy alkaline earths (i.e., Ca, Sr, and Ba) have been used extensively as bone-scanning agents. Due to the high toxicity of barium, the preparation of low-dose, carrier-free Ba-133m by the $\text{Cs-133(p,n)Ba-133m}$ reaction has been initiated as a possible alternative to lower specific activity, neutron-produced Ba-135m (28.7 hr). Barium-133m has a single prominent gamma ray of suitable energy (276 keV) and intensity (17%) for scanning. Production of Ba-133m would be possible with conventional low-energy cyclotrons, whereas preparation of high specific activity Ba-135m requires a high-flux reactor.

Progress and Results

Two preliminary irradiations have been completed, with one product being retained for characterization and one being distributed to cooperative participants for evaluation. Thick-target production rates of $\sim 350\ \text{mCi/mA-hr}$ with $\geq 99\%$ Ba-133m radionuclidic purity (i.e., $\leq 0.1\%$ Ba-133) have been achieved for short

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irradiations of a CsCl target. Attempts to extend this target configuration to longer irradiation times resulted in premature target failure, presumably due to the ~15% volume expansion of CsCl at 496°C resulting from a change in crystal structure.

A preliminary ion-exchange separation scheme has been devised and initial testing begun with good results. Barium-133m is separated from a dilute HCl solution of the cesium target material by passage through a strong cation exchanger. The ion-exchange resin is then washed with 1M HCl to remove any remaining cesium ions and cobalt and strontium radionuclidic impurities before removal of the product with 4M HCl. The preliminary evaluation of this isotope by cooperating participants indicates that it is comparable to Ba-133m.

Plan for Next Period

Work will be continued on target design, chemical processing, and product characterization. Developmental production runs will be performed to verify production parameters and product purity, and to supply cooperating participants with material for product evaluation.

Reports Published

None

Estimated Date of Final Report

1972

BISMUTH-206*

Objective of Work

The objectives of this program are to evaluate the pertinent production parameters and to provide 6 24-day Bi-206 for evaluation as a diagnostic radionuclide in medicine. Bismuth-206 has been used to delineate brain tumors in patients with known brain tumors which are poorly visualized by more common radionuclides. Even though very favorable tumor-to-background activity ratio has been obtained by earlier workers, the lack of an appropriate scanning system suitable for the high-energy emissions of this nuclide [803 keV (99%), 880 keV (72%), 895 keV (19%), *et al*] has retarded the evaluation of the isotope. A cooperative program for the evaluation of Bi-206 as a diagnostic radionuclide using a high-energy photon

scanning system has been undertaken as part of this program with Vanderbilt University.

Progress and Results

Bismuth-206 has been prepared by both the Pb-207(p,2n)Bi-206 and Pb-206(p,n)Bi-206 reactions using isotopically enriched targets. The Bi-205 contamination levels of purified bismuth products obtained from these two production paths have been determined to be 1.1 and 1.2% for the (p,2n) and (p,n) reactions, respectively. The patient radiation dose from Bi-205, which is produced from proton-induced reactions on isotopic impurities in the enriched targets, is estimated to be ~2-1/2 times that from Bi-206. As a result of these findings, development of the (p,2n) production path will be pursued to provide a lower dose product. Excitation functions shown in the literature indicate that the (p,2n) production path should also offer larger production rates than the (p,n) path for the proton energies available, that is, ≤22 MeV. Preliminary work has begun on a PbCl₂ precipitation-cation exchange separation procedure to separate lead and bismuth. Preliminary data indicate greater than 95% bismuth recovery.

Plan for Next Period

Development of a high current, high yield, enriched Pb-207 cyclotron target will be pursued. Process development has been only preliminary and will be continued to obtain a rapid and efficient process which yields a product suitable for human use. Product characterization and specifications will also be determined concurrent to process development.

Reports Published

None

Estimated Date of Final Report

Continuing project

LEAD-203*

Objective of Work

The objective of this work is to investigate the proton production parameters of high-purity Pb-203 suitable for human use. Lead-203 (52.1 hr) has been suggested

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as a possible bone-scanning agent with an acceptable half-life for production and shipment and with low radiation dose decay characteristics (i.e., it decays solely by electron capture with the emission of two main gamma rays of 279 and 401 keV with intensities of 81% and 5%, respectively) Three medical cooperative programs have been established to perform the applications evaluation of this nuclide

Progress and Results

Lead-203 can be produced with ≤ 22 -MeV protons by two different reaction paths (1) $\text{Tl-203}(p,n)\text{Pb-203}$ or (2) $\text{Pb-204}(p,2n)\text{Bi-203} \xrightarrow{\text{EC}, \beta^+} \text{Pb-203}$ Preliminary experiments utilizing the first reaction path have been undertaken for the initial investigation due to the ready availability of a cyclotron target holder design and the lower cost of the enriched target material. A preliminary irradiation has been carried out, and the Pb-203 produced supplied to a cooperating participant for evaluation. A preliminary separation procedure based on the extraction of Tl(III) from a $2M$ $\text{HBr}-\text{Br}_2$ solution into isopropyl ether, followed by a cation exchange chromatographic separation of Pb-203 from the aqueous raffinate, has been employed A 94% chemical yield with $>99\%$ radiochemical purity was obtained from this process Additional chemical process development is indicated by a high total solids content of the first experimental product

Plan for Next Period

Additional target design and chemical process development will be undertaken The relative economics of reaction paths (1) and (2) will be evaluated to determine whether increases in cyclotron yield will offset the added cost of the more expensive Pb-204 target material Full-scale irradiations will be performed and the Pb-203 produced distributed to cooperating participants for applications evaluation

Reports Published

None

Estimated Date of Final Report

Continuing project

BNL BIOMEDICAL PRODUCTS DEVELOPMENT*

Objective of Work

The primary objective of the BNL biomedical development program is to increase the use and usefulness of radionuclides in biomedical applications Technology resulting from this program is used to develop new materials for biomedical research and to improve diagnostic tracers for many hospitals while furnishing technology useful to the commercial radioisotope industry Areas of work include (1) the incorporation of suitable existing radionuclides, as well as of new radioisotopes resulting from developments in the BNL accelerator development program, into special chemical forms for use in new or improved biomedical applications and (2) the development or improvement of radioisotope generator systems and their evaluation This program is carried on in collaboration with members of the staff of the BNL Medical Research Center as well as in cooperation with other medical institutions

At the present time, major emphasis is being placed on the continued development of Tc-99m, as it is considered to be the best radionuclide for use in most radioisotope procedures because of its ideal physical properties and its general widespread availability Technetium-99m studies relating to its use in nuclear medicine are being carried out on a broad front including technetium chemistry, technetium-labeling techniques, and the improvement and simplification of existing technetium procedures The development of analytical techniques can also be expected to play a greater role This work will later be expanded to include other radionuclides appearing to have potential use in biomedical applications

Plan for Next Period

See summaries of individual projects

Reports Published

H L Atkins, K. G. Cardinale, W C. Eckelman, W Hauser, J F Klopper, and P Richards, Evaluation of Tc-99m-DTPA Prepared by Three Different Methods, *Radiology*, 98 674 (1971)

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H L Atkins W C Eckelman, W Hauser, J F Klopfer, and P Richards Evaluation of Glomerular Filtration Rate with Tc 99m-DTPA, *J Nucl Med*, 12(6) 338 (1971)

Estimated Date of Final Report

Continuing project

CHEMICAL STUDIES OF TECHNETIUM-99m*

Objective of Work

The objectives of this project are to obtain a more thorough understanding of the chemistry of technetium in some of the presently used technetium labeled compounds and to develop information useful in the labeling of other potentially useful biological materials

Progress and Results

A number of analytical methods have been surveyed to determine if equilibrium constants can be obtained for technetium biomedical products. Various methods using spectroscopy were not successful because the technetium chelates studied did not have a strong absorption either in the visible or near ultraviolet region.

Potentiometric determination of the equilibrium constant was not possible because of the interaction of the chelates with the titrating solutions. Ceric ions were used to titrate the reduced states of technetium produced by stannous ion and ferrous ion. However, when chelate was added, the ceric ion oxidized the chelate and no equilibrium values could be obtained. The reversibility of the reactions taking place with technetium is also questionable, and therefore this approach was not carried further.

Another method used to determine equilibrium constants is that of partition chromatography. Solutions of various technetium-chelating agent ratios were eluted across Sephadex G25 and the partition of technetium was used to calculate equilibrium values. Studies at various pH values and concentration ranges were undertaken. These studies have the advantage that they can be performed with carrier-free technetium whereas spectroscopic methods require large amounts of carrier technetium. However, the analysis of the data showed that the system did not involve a simple equilibrium

between one technetium species and the chelating agent. The use of concentrated HCl to reduce the technetium resulted in a system which was complicated by slow hydrolysis of the reactive technetium species and by a possibility of mixed valence states in addition to the desired reaction. Attempts to reduce technetium by stronger reducing agents such as stannous ion eliminated the mixed oxidation state complication, but the system was apparently still susceptible to irreversible hydrolysis reactions which prevent the elucidation of equilibrium expressions.

Another area of work concerned with the chemical state of technetium involves the development of analytical techniques that can accurately identify various components in these products. Work here and at other laboratories has shown that technetium activity is associated with several components following the chemical manipulations commonly used in preparing labeled compounds. Technetium-HSA, for example, usually contains varying amounts of technetium activity as a chelate, as unreacted pertechnetate, and as a hydrolyzed form as well as that which is bound to HSA.

The recent literature contains much misleading analytical data concerning the identification of these various technetium components in biomedical products. Inasmuch as the different forms of technetium have different biological behavior, failure to identify and quantify them can result in misuse of the compound or give incorrect internal dose calculations. This study has compared various analytical methods now in use to determine the advantages and disadvantages of each method. Commonly used technetium-labeled compounds were analyzed by anion exchange chromatography, by paper chromatography with both 85% methanol and saline as solvents, and by gel chromatography (Sephadex G25).

Results show that the most widely used techniques, ion exchange and paper chromatography with methanol, give the least discriminating analysis. Ion exchange on anion resins will separate pertechnetate from the other components but does not identify other fractions. Paper chromatography with saline solvent gives somewhat better identification of components but there appears to be some oxidation or breakdown on the paper because of the small sample size. Gel chromatography, with nitrogen-purged saline as eluant, will separate and identify technetium-labeled proteins, chelates, hydrolysis products, and pertechnetate. Gel chromatography seems to be the single analytical method of choice but nevertheless it seems advantageous to use more than one analytical method whenever possible.

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Plan for Next Period

Further efforts will be made to determine the oxidation state and physical constants of Tc-99m in biomedical products. Additional work is also required to improve analytical techniques for Tc-99m biomedical products.

Reports Published

W. Eckelman, G. Meinken, and P. Richards, The Chemical State of Tc-99m in Biomedical Products, *J Nucl Med*, 12: 596 (1971)

Estimated Date of Final Report

Continuing project

INSTANT Tc-99m COMPOUNDS*

Objective of Work

One of the present goals of the Biomedical Development Program at BNL is to prepare technetium compounds which contain a single component, as demonstrated by various analytical techniques, and which can be easily prepared, ideally by adding pertechnetate to a prepared solution. Such a one-step, single-component product would enable the widespread use of this compound, especially if it were commercially available to those hospitals and clinics that have limited radio-pharmaceutical facilities. Such a system would also enable sterility and pyrogenicity to be determined before use.

During the development of a red blood cell labeling procedure, the effectiveness of stannous ion in carrying out the above mentioned objectives was noted. Therefore, single-component one-step products are being formulated using stannous ion as the reducing agent.

Progress and Results

An instant procedure for the preparation of the chelate TcDTPA (diethylenetriaminepentaacetic acid) has been reported previously (TID-4066). This instant compound has been used successfully in single injection methods for determining renal clearance rates. Infusion techniques are usually thought to yield more reliable clearance values because equilibrium of the infused substance is achieved between the plasma and the extravascular space.

To perform constant infusion studies, large volumes of instant TcDTPA are required. Therefore, a study was initiated to determine the factors involved in the use of 500-ml volumes of TcDTPA. The use of large volumes of stock SnDTPA did not seem advisable because of the toxicity of the stannous ion. Therefore, a minimum amount of stock solution (5 ml) was used to retain reducing power, and the overall stability of the TcDTPA was maintained by adding DTPA to maintain constant molarity. This combination of factors resulted in a solution of 500 ml of TcDTPA which showed a yield of 95% for a period of 5 hr as measured by gel chromatography.

Samples of urine and plasma from the infused dogs were assayed by gel chromatography to determine the stability of the chelate. Technetium was excreted in the urine exclusively as TcDTPA. The plasma samples contained a plateau value of 10 to 15% protein bound technetium as a result of breakdown of the constantly infused TcDTPA. The protein binding in a single injection of TcDTPA is ~5% as determined by gel chromatography. After correction for protein binding of the infused TcDTPA, the renal clearance values indicate that the TcDTPA is filtered exclusively by the glomeruli and not reabsorbed by the tubules.

The second instant procedure developed was that for TcHSA (human serum albumin). The approach was similar to that used in TcDTPA. Stannous HSA chelate was prepared at various pH values. However, unlike the DTPA case, the majority of the tin could not be chelated with up to 250 mg HSA (~3 micromoles). This presented problems because colloidal tin could compete with the HSA for the technetium. The elimination of this competitive binder effect was achieved with varying success by (1) using less initial stannous ion, (2) removing the tin colloid before addition of pertechnetate, and (3) adding the technetium at a pH sufficiently low to prevent tin hydrolysis. The procedures incorporating these three approaches are given in the first table. Both of the procedures which incorporate stock solutions at pH 6 give relatively high yields, but both were more variable and not as high as the low pH method. In this procedure, the technetium is added to a solution of $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$ and HSA at pH 2, which was determined to be the binding pH giving the highest yield. This preparation gives TcHSA of >95% yield and high reproducibility. It is not a one-step procedure, since the tin HSA cannot be stored at pH 1 because of gel formation and because the pH must be raised after pertechnetate addition. However, the manipulations necessary to add HSA, pertechnetate, and buffer are

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Low pH Method	Instant Procedures	
	Excess Tin Not Removed	Excess Tin Removed
1 1 ml of 250 mg/ml HSA is added to 0.5 ml of 2 mg/ml $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}^*$	1 4 ml of 250 mg/ml HSA is added to 2 ml of 1 mg/ml $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}^*$	1 4 ml of 250 mg/ml HSA is added to 2 ml of 1 mg/ml $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}^*$
2 0.1–2 ml $^{99\text{m}}\text{TcO}_4^-$ is added	2 The pH of the solution is raised to 6 with 1M NaOH**	2 The pH of the solution is raised to 6 with 1M NaOH**
3 The solution is mixed for one minute	3 0.1–1 ml $^{99\text{m}}\text{TcO}_4^-$ is added per ml stock solution as needed**	3 The entire solution is eluted across a 35 cm Sephadex G25 column with saline eluant
4 The pH of the solution is raised to 6 with 1M NaOH**	4 Terminal dilution up to 20 ml total can be made if lower HSA concentration is desired***	4 The protein fractions are collected and the solution assayed for HSA concentration
5 Terminal dilution up to 20 ml total can be made if lower HSA concentration is desired		5 0.1–1 ml $^{99\text{m}}\text{TcO}_4^-$ is added per ml stock solution as needed***

*Preparation of $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$ solution The appropriate amount of $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$ is dissolved in 2.5 ml concentrated HCl with heat. The solution is then diluted to 25 ml with N_2 purged distilled water.

**NaOH solution of $\leq 1M$ NaOH should be used to neutralize the solution to prevent localized concentrations of strong base which lead to formation of hydrolyzed technetium.

***All stock solutions are stored under nitrogen.

Yield of $^{99\text{m}}\text{Tc}$ HSA as Function of pH and Time

Variation of pH for 15-min mix			Variation of pH for 30 min mix			Variation of time at pH 2.5		
pH	Tc HSA	Tc hydride	pH	Tc HSA	Tc hydride	Min	Tc HSA	Tc hydride
0.9	63	37	2.4	96	4	5	82	18
2.2	80	20	2.9	91	9	15	87	13
3.3	59	31	3.5	82	18	30	96	4

Reaction of 1 ml of TcO_4^- with 25 mg HSA and 250 μg $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$ in appropriate solution of HCl. Final pH of 6.1 achieved with 0.15 ml 0.8M NaH_2PO_4 . Analysis performed on Sephadex G25.

well worth the increased yield and reliability obtained as compared to the two one-step instant procedures.

The critical variables were also determined for production of high specific activity HSA using 25 to 50 mg HSA instead of the 250 mg used in the original low pH procedure. The results of these experiments are given in the second table. The maximum yield seems to be obtained at pH 2.0 to 2.5, as in the case of 250 mg HSA. As expected, the labeling of HSA using 25 mg HSA was slower than the labeling using 250 mg HSA. Whereas the 250 mg HSA produces yields of >95% with one-minute mixing at low pH, the 25 mg HSA procedure requires 30-min mixing at low pH to achieve comparable yields. Use of 50 mg HSA did not increase the yields significantly and 30 min was still required to achieve yields of >95%.

Plan for Next Period

It appears that development of kits for other biologically interesting compounds may be possible using similar procedures. The major emphasis will be on lung, kidney, and bone scanning agents.

Reports Published

None

Estimated Date of Final Report

Continuing project

TECHNETIUM-99m LABELED RED BLOOD CELLS*

Objective of Work

The objective of this work is to develop an improved method for labeling red blood cells with Tc-99m. This product is valuable for spleen scanning and for blood pool studies. Chromium-51, presently used to label cells, does not have the favorable decay characteristics that technetium does.

Progress and Results

The development of methods for the labeling of red blood cells with Tc-99m and then altering the cells for use in visualization of the spleen has been previously reported. Further work in this area included additional studies of both the labeling procedure and the alteration or damaging procedure. The original labeling procedure, as a result, has been shortened substantially, and the abbreviated method makes Tc-99m labeling competitive with the more commonly used Cr-51 procedure. Studies of the glucose levels and fluoride exchange reactions using F-18 tracer indicate that the mechanism of labeling involves pertechnetate crossing the membrane by passive diffusion. The pertechnetate is then reduced by the stannous ion and bound intercellularly. Approximately 30% of the technetium appears to be associated with the hemoglobin whereas the remaining technetium is reduced and bound to a low molecular weight species as yet unidentified. There does not seem to be any alteration of the transport properties of the cell but rather simple diffusion of pertechnetate with subsequent chemical binding to the intracellular components.

A second aspect of the work involves alteration or damaging of the cells so that they will be removed by the capillary system of the spleen thus enabling the physician to visualize the spleen. The measurement of the osmotic fragility of the cells as a means of determining the degree of alteration indicated that increasing amounts of stannous citrate made the cells more fragile. Measurement of filterability of the red cells, perhaps a closer analogy to the action of the spleen in removing altered red cells, was also undertaken. A Millipore filter of 5 micron pore size was used to filter the cells altered to various degrees by the use of different amounts of stannous citrate and citrate alone. The results of this study are given in the table. From these results, animal experiments were set up. In-

Red Cell Filterability Studies

ml of 1 mg/ml $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}^*$ in ACD	Flow rate at 4 in Hg (ml/min)
0	0.033
0.1	0.033
2	0.027 (81%)
3	0.027

*Added to 4 ml packed red blood cells

creasing amounts of splenic uptake were observed, respectively, with the use of 2 ml of stannous citrate, 4 ml of citrate alone, and 4 ml of citrate mixed at 37°C — all added to 4 ml of packed red cells. Trials on humans, however, did not indicate such drastic differences and, therefore, the standard procedure of altering the cells with excess citrate (1 ml of citrate per ml of packed red cells) is now used.

Clinical evaluation involving double isotope studies has indicated that >5% of the cells accumulate in the spleen and that good spleen visualization is achieved at these levels. The alteration procedure contains only one additional step during the labeling procedure, making it easier to carry out than the more commonly used heat treatment method. Better uniformity of the citrate altered cells may also make dynamic studies more reproducible. The higher photon yield and imaging characteristics of Tc-99m make this procedure superior to the Cr-51 labeling and heat treatment method.

Plan for Next Period

Clinical evaluation of the red-cell labeling procedure will be completed.

Reports Published

- W. Eckelman, P. Richards, W. Hauser, and H. Atkins, Technetium Labeled Red Blood Cells, *J. Nucl. Med.*, 12: 22 (1971).
- W. Eckelman, P. Richards, H. L. Atkins, W. Hauser, and J. F. Klopper, Visualization of the Human Spleen with Tc-99m Labeled Red Blood Cells, *J. Nucl. Med.*, 12: 310 (1971).
- W. C. Eckelman, P. Richards, W. Hauser, and H. L. Atkins, Reply to a Letter to the Editor, *J. Nucl. Med.* Concerning Comparison of Tc-labeled Red Blood Cells Prepared by Two Methods, *J. Nucl. Med.*, 12: 577 (1971).

Estimated Date of Final Report

June 1972

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NEW CESIUM-137 SOURCE COMPOUNDS DEVELOPMENT AND EVALUATION*

Objective of Work

The objectives of this project are (1) the study and development of new Cs-137 source compounds (other than CsCl) having a high degree of stability in contact with aqueous solutions and in a high temperature environment, including preparation methods and product characterization, and (2) an economic assessment of the conceptual process designs resulting from the study

Progress and Results

The literature study and the preliminary experimental investigation continued in order to evaluate technical feasibility and economic potential of Cs-137 source compounds other than Cs(137)Cl. High temperature stability, low solubility in aqueous solvents, and good mechanical properties are among the criteria established for such compounds. The initial phase of the study was the development of preparation methods for cesium niobates and cesium tantalates. The three methods investigated were the molten $\text{CsOH}-\text{M}_2\text{O}_5$ reaction (M is either Nb or Ta), the $\text{CsOH}-\text{M}_2\text{O}_5$ (or $\text{Cs}_2\text{CO}_3-\text{M}_2\text{O}_5$) slurry reaction, and the simultaneous solvent extraction—chemical reaction. Of these methods, the one based on slurry reaction appears most attractive.

Characterization of the cesium niobates and tantalates included determination of the chemical composition (M/Cs ratio, M is either Nb or Ta), solubilities in seawater and distilled water, and stability at elevated temperatures. The volumetric specific activities (Ci/cm^3) of the products were computed from the chemical composition and estimated values of densities, assuming an isotopic purity of 36.5% Cs-137. Various properties of the cesium niobates and tantalates thus determined were correlated with the methods of preparation as well as the reaction conditions.

The volumetric specific activity of cesium tantalate products ranged from 60 to 82 Ci/cm^3 , and was primarily governed by the stoichiometry (i.e., Ta/Cs ratio) of the product which, in turn, is strongly influenced by the calcining temperature. For products from the molten $\text{CsOH}-\text{Ta}_2\text{O}_5$ reaction and the $\text{Cs}_2\text{CO}_3-\text{Ta}_2\text{O}_5$ slurry reaction, the amount of seawater insolubles (68 to 100%) is favored by a high Ta/Cs ratio which results in a low specific activity. The

melting points (temperature at complete melting) of most cesium tantalates were in excess of 1350°C . Runs with short slurry reaction times resulted in products with low melting points. Compounds with low Ta/Cs ratios had a tendency to display some degree of thermal instability, i.e., low melting point, incongruent melting, and thermal decomposition.

For cesium niobates, the trend in the variation of the specific activity (60 to 90 Ci/cm^3) with the product stoichiometry (Nb/Cs ratio) was the same as that for the tantalates. However, no simple relation was apparent between the stoichiometry, the amount of seawater insolubles, and the thermal stability. In general, cesium niobates were thermally less stable than cesium tantalates. Especially notable was the presence of several incongruent melting temperatures for most cesium niobate products. The melting point (790 to 1340°C) increased with increasing calcining temperature.

Both cesium tantalates and niobates have good potentials as Cs-137 gamma sources. The volumetric specific activity of these compounds compares favorably with that of Cs(137)Cl gamma sources ($\sim 100 \text{ Ci}/\text{cm}^3$). They exhibit considerably lower solubility in an aqueous solution and higher stability in a high temperature environment than cesium chloride. Cesium tantalates appear to be especially attractive since it is feasible to obtain a product having a specific activity greater than $70 \text{ Ci}/\text{cm}^3$, with extremely low solubility and with a melting point in excess of 1350°C . A summary of the characteristics of cesium tantalates and niobates obtained to date is given in the table. According to the preliminary economic evaluation (based on the conceptual flow diagram and estimated processing factors), the fixed capital costs and the manufacturing costs for the processes producing cesium tantalates (or niobates) would probably be of the same order of magnitude as those for the CsCl process based on the $(\text{NH}_4)_2\text{CO}_3-\text{NH}_4\text{OH}$ eluant.

Cesium vanadates were also prepared using methods similar to those for cesium niobates and tantalates. The volumetric specific activity of cesium vanadates was estimated to be about the same (74 to $83 \text{ Ci}/\text{cm}^3$) as those of niobates and tantalates of equivalent stoichiometries. Cesium vanadates have an advantage over cesium tantalates and niobates in that the mass absorption coefficient for the gamma radiation would be appreciably smaller. However, the low thermal stability (melting points are probably 700°C or lower) and the relatively high solubility ($\sim 70\%$ solubles) of cesium vanadates in an aqueous solution make them less desirable than either niobates or tantalates as a gamma source.

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Characteristics of Selected Cesium Tantalate and Niobate Products
(Tentative)

Composition (atomic ratio)	Estimated density g/cm ³	Sea water insoluble ^a wt %	Melting Temperature, °C ^b		Estimated volumetric spec act Ci/cm ^{3c}
			Incongruent	Complete	
Cesium Tantalates					
Ta/Cs					
1 28	6 7	93 7		>1350	67 8
1 10	6 6	90 0		>1350	73 8
1 55	6 9	100 0		>1350	60 5
Cesium Niobates					
Nb/Cs					
1 45	5 1	~90		885	64 9
1 49	5 1	~90	875, 1005	1325	64 3

^aPowder sample

^bBased on observation under high temperature microscope (maximum 1350°C)

^cVolumetric specific activity of a ¹³⁷CsCl gamma source is approximately 100 Ci/cm³

Studies are in progress on ceramic compounds of cesium, such as Cs₂O·Al₂O₃·nSiO₂ and Cs₂O·MgO·nSiO₂. These compounds were prepared by first forming the water-glass-like compound Cs₂O·nSiO₂, followed by reaction with Al₂O₃ or Al(OH)₃ and MgO, respectively, and finally calcining the products at elevated temperatures. The volumetric specific activity of the products is estimated to be in the range of 43 to 66 Ci/cm³ depending upon the value of n (1 to 4).

Plans for Next Period

The study and development of new Cs-137 source compounds will continue. More detailed and precise characterization will be carried out for selected products. The production process conditions will be defined more specifically and economic assessment made for most promising source compounds.

Reports Published

None

Estimated Date of Final Report

Continuing project

STRONTIUM SILICATE BETA SOURCE DEVELOPMENT*

Objective of Work

The objectives of this project are to develop methods for fabrication of high performance Sr-90 beta sources

that have thin windows to yield high radiation output, to characterize the physical properties of the sources, and to determine radiation output characteristics.

Progress and Results

Studies were carried out to determine high temperature compatibility of strontium silicate minibeads in aluminum matrix and encapsulated in 347 stainless steel. Fabricated sources were heated in air for a period of 1080 hr at temperatures ranging from 350 to 900°C. In all test source samples heated in the temperature range of 600 to 900°C, intermetallic diffusion between the matrix material and stainless steel capsule occurred. The reaction progressed to a depth of 0.006 in. into the capsule wall. Samples tested at 250°C showed no evidence of reaction between the capsule wall and the aluminum/strontium silicate, indicating sources prepared by this method can be subjected to a continuous use in temperatures up to 250°C. Evaluation of the test sources indicates that severe reaction between the source capsule and the aluminum/strontium silicate will occur above 500°C.

Graphite was substituted for aluminum as a matrix for strontium silicate beads. Beads were coated with pyrolytic carbon using a mixture of propane and helium at a temperature of 1050°C. A second method using a mixture of graphite flour and a phenolic binder was investigated. A mixture of strontium silicate beads and

*T. S. Mackey and F. N. Case, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, TN 37830, Contract W 7405-eng-26

the graphite mixture was extruded, and after baking had adequate mechanical strength for handling for encapsulation. The method was demonstrated to be feasible.

Measured surface dose rate from a source loaded to the maximum with strontium silicate beads in aluminum matrix of 38.7 Ci/linear inch in a capsule 0.085 in thick by 0.395 in wide and with 0.010 in stainless steel walls was determined to be 5×10^6 R/hr.

The output from a disc source 0.750 in in dia and 0.030 in thick containing 3 mCi of Sr-90 silicate beads distributed in an aluminum matrix and covered with 0.005 in stainless steel was measured in an extrapolation chamber to be 1.5×10^4 Rads/hr·cm² as a surface dose rate.

Plan for Next Period

The original objectives have been completed.

Reports Published

F. N. Case and T. S. Mackey, *Preparation of Strontium-90 Silicate Beta Sources*, USAEC Report ORNL-4340, February 1969.

Estimated Date of Final Report

1972

LOW-ENERGY PHOTON RADIATION SOURCES*

Objective of Work

The objectives of this project are to develop low-energy radiation sources that have high radiation intensity and useful energy spectra and to define safety characteristics of the source capsules. The development of prototype sources is essential to the development of devices to measure compositions or density by secondary x-ray emission, backscatter, and photo attenuation.

Progress and Results

The principal radiochemical impurities in gadolinium-153 produced by irradiating normal europium are high energy components of europium-152-154. Sources of gadolinium-153 used in x-ray fluorescence analysis must be essentially free of europium. A relatively

low-cost supply of high purity gadolinium-152 was developed as by-product to the operation of the HFIR (High Flux Isotope Reactor) by recovery of gadolinium-152 produced in the europium control plates. Sources were prepared in which the concentration of radionuclides of europium in the gadolinium-153 was less than 10 ppm. The sources were fabricated as point sources using stainless steel encapsulation and the radiation output was characterized. A method was developed in which the gadolinium-153 as nitrate was extracted into tri-butyl phosphate and this solution dispersed in polyurethane. When the polyurethane solution was applied to an aluminum substrate and allowed to polymerize, a uniform, large surface, flat plate source was achieved.

Cadmium-109 purification methods and source fabrication methods were studied to obtain very high radiochemical purity, high output sources for use in fluorescence x-ray analysis. To maximize the spectra purity and output, thin graphite substrates were used to stabilize the cadmium-109 activity into a fixed geometry. Carrier-free cadmium-109 prepared by proton bombardment of a silver target was used in preparing sources up to 55 mCi. The graphite, containing cadmium-109, was covered with a 4-mil aluminum window. Particular emphasis was directed toward extracting the 22 keV photon.

Methods to prepare high integrity americium-241 sources were studied. Photon sources were prepared by using americium metal alloys with aluminum and copper. Aluminum cladding of americium metal is under study as a method for obtaining thin window alpha sources. Output measurements and spectral characteristics are being correlated with alloy composition.

Literature reviews concerning low-energy and alpha sources were carried out to determine the state-of-the-art. Work was carried out to characterize source output parameters through the use of an extrapolation chamber and silicon detector system.

Plan for Next Period

Work will continue on state-of-the-art reviews to assemble existing information concerning low-energy sources. Study of sources and source collimators to improve the quantity and quality of radiation reaching a sample being analyzed will be continued.

Reports Published

F. N. Case, E. H. Acree, and N. H. Cutshall, *Production Study of Gadolinium-153*, USAEC Report ORNL-TM-2632, August 1969.

*R. S. Pressly, T. S. Mackey, and E. H. Acree, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, TN 37830, Contract W-7405-eng-26.

R. S. Pressly, The Development and Use of Low-Energy Gamma, Secondary X Ray, and Beta Sources, *Trans. Amer. Nucl. Soc.*, 14(2): 497-8 (October 1971).

Estimated Date of Final Report

Continuing project.

ADVANCED SOURCE DEVELOPMENT*

Objective of Work

The objective of this program is to evaluate present and future heat- and radiation-source application requirements and to relate them to present and future encapsulation technology. The goal of the current work is concerned primarily with the development of thin-film encapsulation and evaluation techniques for alpha-photon sources. Emphasis is on the technique of rf sputtering as a means to obtain thin films of radioisotope and containment materials.

Progress and Results

Alpha-photon radiation sources, having an Am-241-containing active layer, were fabricated by rf sputtering in several different source preparation runs. Both the

active layer and the containment layer were deposited by rf sputtering.

The rf-sputtering system located in a radioisotope glove box was modified by adding or replacing a number of components. The updated system is easier to operate and maintain, and with it two different materials may be deposited consecutively. Preliminary results indicate that the use of sequential rf sputtering available in these glove-box systems gives sources that have better radioisotope containment than the sources prepared in the original system.

The rf-sputtering system outside the glove box was used for nonradioisotopic sputtering work such as source preparation hardware development. Hardware for preparing ten radiation sources simultaneously (see figure) was developed and used to prepare Am-241 radiation sources. The hardware allows accurate positioning of a $\frac{1}{2}$ -in.-dia Am-241-containing active layer on $\frac{7}{8}$ -in.-dia substrates. The positioning mask is removed for deposition of the active layer.

An Am(241)O₂-copper sputtering target (the source of sputtered material) was used in preparing the active layer for radiation sources. The target was prepared by cold-pressing pellets of Am(241)O₂ and copper powders, and bonding the pellets to a copper backing plate. The radioactive layer sputtered from this target contained less than 5% Am-241. A target yielding a sputtered film containing a much greater portion (over 50%) of Am-241 was also prepared similarly. Techniques were developed for minimizing radioisotope

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Sputtering hardware for preparing 10 radiation sources in a single run.

inventory in the target while still providing a target face that contained a high proportion of $\text{Am}(241)\text{O}_2$. Experiments to demonstrate the production of high activity, high integrity alpha photon sources from this target were started.

Assistance in evaluating PNL prepared alpha photon sources was provided by Oak Ridge National Laboratory. The test results on a group of alpha-photon sources were encouraging since the demonstrated source integrity was greater in most tests than the source integrity determined on previous PNL prepared alpha-photon sources. However a problem in radioisotope containment at elevated temperature was indicated. Contamination was released in this test when the sources were heated to over 900°C in one hour in flowing air. The results of all tests were factored into the design and preparation of the next generation alpha photon sources. The ORNL evaluation involved boiling water leach testing, temperature-humidity testing, and the ORNL source capsule classification testing (Class I A).

The primary advantages of sputtering for preparing thin film radiation sources are:

- Most metals, ceramics and cermets may be deposited by sputtering.

- Sputtered films generally have high density, good integrity, and good uniformity.

- A number of similar sources may be prepared in a single run, and different geometry sources may be conveniently prepared.

- Source layer thicknesses and source activity may be accurately controlled.

- Source preparation by sputtering is well suited to the automated production of a number of similar sources per run on a continuing basis.

- Techniques for minimizing radioisotopic cross contamination when preparing sources containing different radioisotopes, and techniques for removing sources from the sputtering system without adding significant surface contamination to them have been demonstrated.

The disadvantages of sputtering are:

- Because of the radioisotope inventory required in the target, sputtering is not a convenient method for preparing a one of a kind source. The exceptions would be cases where a sputtering target containing the desired radioisotope is available from past source preparation work or where the radioisotope layer is deposited by a technique other than

sputtering, and sputtering is used to provide the encapsulation or containment layer.

Conventional sputtering systems are not fast. However, extended runs and unattended equipment operation have been successfully demonstrated. Metals may be deposited at about one to two microns per hour and ceramic materials have deposition rates of about an order of magnitude slower. As rates are increased, substrate cooling becomes a problem unless there is good path of thermal conduction between the substrates and the water cooled substrate plate.

Plan for Next Period

Another generation of alpha photon sources will be fabricated using rf sputtering. They will be given preliminary evaluation at PNL and then sent to ORNL for extensive evaluation. The preparation of high specific activity sources will be investigated and the preparation of various geometry sources for select applications will be considered. The rf sputtering systems will be further automated. Attempts will be made to correlate residual gas analysis (RGA) in the vacuum chamber, before, during and after a deposition run, with the properties of sputtered films. If successful, RGA could provide a useful quality control technique. Projected system economics will be briefly evaluated to give an indication of source costs on a production scale basis.

Reports Published

None

Estimated Date of Final Report

Continuing project

NONDESTRUCTIVE TESTING TECHNOLOGY IN SUPPORT OF DID CAPSULE DEVELOPMENT ACTIVITIES AND RADIATION SOURCE PROGRAMS*

Objective of Work

The objective of this program is to develop nondestructive test methods, devices and instruments that will assist the designer or fabricator of isotope encapsulation.

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Differential transducer technique.

lations by (1) evaluating the as-fabricated encapsulation for comparison with designed parameters, (2) helping to select and evaluate encapsulation materials and materials compatibility, and (3) developing faster, more economical, and more easily interpreted test methods. Cooperation of the nondestructive test (NDT) development and fabrication efforts assures the isotope user and the public of optimum encapsulation integrity.

Progress and Results

Construction and evaluation of a differential transducer system was completed (see figure). This technique was developed to permit the inspection of thin materials, or to locate anomalies lying close to the surface of thicker materials. It is based upon placing two electrodes on the back of a piezoelectric ultrasonic transducer, inverting the signal from one of the electrodes, and summing the inverted and noninverted signals. Common signals (such as those from the front and back surfaces of the object under test) will cancel, leaving only the signals due to any small anomalies present under only one of the electrodes. Evaluation established that the differential transducer worked quite well. Front and back surface reflections were reduced by a factor of 20 or more, allowing flaws close to the surface to be resolved.

Plan for Next Period

Due to budgetary limitations, further work on this program is presently curtailed.

Reports Published

- D. R. Newman and R. W. Steffens, *Multielectrode Ultrasonic Transducer Test Techniques for Radioisotope Encapsulation Inspection*, USAEC Report BNWL-1491, February 1971; also printed in the *Proceedings of the 8th Symposium on Nondestructive Evaluation in the Aerospace, Weapons and Nuclear Industries*, San Antonio, TX, April 1971 (Available from Western Periodicals, 13000 Raymon St., N. Hollywood, CA 91605).
- D. R. Newman, Radioisotope Bond Test Applications of a Pulsed Laser Schlieren System, in *Proceedings of the 8th Symposium on Nondestructive Evaluation in the Aerospace, Weapons, and Nuclear Industries*, San Antonio, TX, April 1971 (Available from Western Periodicals, 13000 Raymon St., N. Hollywood, CA 91605).
- D. R. Newman and J. C. Crowe, Recent Developments in Nondestructive Evaluation Applicable to Encapsulated Isotopes, in *Proceedings of the 3rd International Symposium on Packaging and Transportation of Radioactive Materials*, Richland, WA, August 16-20, 1971, pp. 138-65, USAEC Report CONF-710801, 1971.

Estimated Date of Final Report

Topical reports will be issued.

SHIPPING CASK DESIGN AND EVALUATION*

Objective of Work

The objective of this project is to determine whether packages for shipping large quantities of radioactive materials meet the performance standards developed by

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the Atomic Energy Commission, the Department of Transportation, and the International Atomic Energy Agency. These standards cover normal conditions during transport, as well as hypothetical accidents.

Progress and Results

Packages having several different designs were evaluated and approved as meeting the required performance

Neutron shielded fire and impact shield assembly.

standards. The packages were required to withstand a 30 ft drop, a 40-in puncture test (dropped onto a 6-in -dia pin from a distance of 40 in), and a 30-min open fire. The designs included the Puerto Rico Rain Forest Irradiator, a Canadian fissile package, and a British package utilizing a wooden overpack.

The overpack for Specification 55 packages developed last year, utilizing lithium hydroxide monohydrate as a thermal-conducting medium, was successfully subjected to the consecutive hypothetical accident tests (see figure).

Plan for Next Period

New shipping-package concepts and designs will continue to be evaluated under this program.

Reports Published

- R D Seagren, *Safety Analysis of ORNL Bulk Radioisotope Shipping Cask*, USAEC Report ORNL TM-3506, August 1971
- R D Seagren, *Safety Analysis of the ORNL Type B Gas Cylinder Protective Packaging*, USAEC Report ORNL-TM 3544, October 1971
- R D Seagren and L B Shappert, *Indexed Bibliography on Transportation and Handling of Radioactive Materials 2*, USAEC Report ORNL-NSIC 84, January 1971
- R D Seagren, Shipping Cask Evaluations, *Isotop Radiat Technol* , 8(3) 350-59 (1970)

Estimated Date of Final Report

Continuing project

RADIOISOTOPE SAFETY TESTING*

Objective of Work

The objective of this program is to determine the reliability of radioactive sources in containing the radioactive material under the conditions of normal use as well as in the event of an accident. The investigations produce data that will enable source manufacturers and regulatory agencies to evaluate existing and future source designs from the standpoint of safety to the users and general public. Test procedures are also developed that can be used by manufacturers for quality assurance purposes.

*R G Niemeyer, Oak Ridge National Laboratory, P O Box X, Oak Ridge, TN 37830, Contract W 7405-eng 26

Progress and Results

A study of tritium losses from commercial tritium foils used in gas chromatograph detectors was completed. These foils had nominal tritium concentrations of 250, 500, and 1000 mCi/in² as titanium tritide on a stainless steel backing. The corresponding measured total activities in each foil were 62.5, 125, and 250 mCi. Tritium losses were determined as a function of foil temperature, foil-heating rate, carrier-gas composition, carrier-gas flow rate, time that foil is exposed to temperature, and foil-cleaning procedure. The tritium losses as a function of temperature increased slowly with increasing temperature to a break point, above which the losses increased at a much higher rate. The tritium losses when new foils were heated at 150°C for 1 hr in an argon flow were 4.9, 51.1, and 14.9 μ Ci/in² respectively, for the 250, 500, and 1000 mCi/in² foils. The temperatures at which the break points occur are approximately 238, 198, and 200°C, respectively, for the 250, 500-, and 1000-mCi/in² foils (see figure).

Tritium losses as a function of operating time at 225°C (maximum temperature allowed by USAEC regulations) were obtained in an argon flow for each type of foil. The losses were relatively high during the first few hours of operation but quickly decreased with time and remained nearly constant for the remainder of the test. Since new foils were used for each of the tests

Tritium loss as a function of temperature in argon.

in the study, the test results represent the maximum losses that would be expected under the test conditions

A proposed procedure for accurately measuring the radiation dose rates from small commercial light sources containing Pm-147 and Kr-85 was developed as an aid to source manufacturers, standards groups, and individuals involved in their use. These dose-rate measurements are complicated by many variables including geometry, type and quality of the radiations involved, and energy dependence of the common survey instruments used in making the measurements. Over 2000 dose-rate measurements were made using eight different survey meters and thermoluminescent dosimeters to identify the problems involved in obtaining accurate dose-rate measurements of 16 commercial sources.

Additional data were obtained on the 10-year aging and drying effects study on the release of tritium from luminous phosphor paint and on the aging effect of alpha calibration sources. The paint samples have shown a steady physical deterioration over the years and they are extensively cracked and pieces have flaked off the surface. Over the past six years, the water leach tests of the samples have shown an increase of about a factor of three in the amount of leachable tritium; however, the total quantity is still below that permitted by present regulations. The plated uranium and plutonium alpha calibration sources have not shown any significant change since the start of the test.

Plan for Next Period

Tests will be performed on watches having luminous tritium-painted dials and hands to determine if any tritium is released during normal use.

The 10-year study of tritium paints will be terminated and fresh supplies of currently manufactured tritiated paints will be obtained for a new study in which more emphasis will be placed on determining losses of tritium in normal use and to observe any improvement in stability of the newer paints.

Studies will be initiated to determine the optimum leak test sensitivity for encapsulated sources by obtaining data on the amounts of activity released under various conditions from source capsules having known leak sizes.

Specific tests will be carried out when requested by the AFC.

Reports Published

None

Estimated Date of Final Report

Continuing project

RADIOISOTOPE CHARACTERIZATION, QUALITY CONTROL, AND STANDARDS*

Objective of Work

The objectives of this program are to select or determine radionuclide characteristics such as half-lives and modes and energies of decay, to review and recommend quality-control procedures and analyses, and to study radioisotope measurement methods and standards. Critically evaluated constants are essential in all aspects of analysis, handling, and use of radioisotopes, and in supplying information about them. Methods are reviewed and selected, and special analyses are performed when required. Both local and national needs are involved in the study of standard materials and measurement methods.

Progress and Results

Half-lives have been determined for 60 nuclides since our last publication of such data; these included the controversial nuclides Mo-99, Eu-155, W-181, and Hg-203. The reactor-neutron cross section of Ni-59 was estimated as 85 barns, and the cross section and resonance integral for production of Ag-110m were determined. Information, some of types not previously included, was collected for revisions to the *Isotope User's Guide*. The majority of recipients of the Guide who responded to a questionnaire found the sections on characteristics and standards "very useful."

Some needs in quality-control analysis were satisfied by recommendations of neutron activation to determine chlorine in elemental C-14, and gamma spectrometry for traces of Kr-85 in inactive fission-product xenon. With respect to raw material for Sr-90 sources, study of methods and results yielded the conclusion that present techniques are satisfactory, provided that characteristic elemental and isotopic compositions are taken into account.

The National Academy of Sciences published the report of the ad hoc panel on standard radioactive materials, based in large part on our survey of national needs. Over 700 copies of the panel report have been distributed to selected individuals and institutions. Its recommendations, supported by other groups, have influenced policies at the National Bureau of Standards (NBS) and actions by organizations concerned with radioactivity. With the conclusion of the survey, and in line with panel recommendations, emphasis is being placed upon making local calibrations traceable to NBS.

*S. A. Reynolds, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, TN 37830, Contract W-7405-eng-26

whenever practicable. A report was prepared on ORNL calibrations, especially as related to NBS, containing a brief history of the work during the past quarter century.

Plan for Next Period

Studies of radioisotope characteristics, quality-control procedures and standards, and nonroutine analytical problems will continue as required. Emphasis will be on nuclides of biomedical and environmental usefulness, and HFIR products. It is expected that the revised *Isotope User's Guide* will be issued. Collaborative efforts will include beginning preparation of a general manual on calibrations, acceptable to organizations requiring radioactivity measurements, and gathering and disseminating information on standards.

Reports Published

- S. A. Reynolds, *A Survey of Needs for Radioactivity Standards*, USAEC Report ORNL-TM-3239, September 1970.
- S. A. Reynolds, Radioactive Materials Problems Under Scrutiny, *Science*, 171: 955 (1971).
- S. A. Reynolds, Cooperative Isotopes Program, Radioisotope Characteristics and Measurements, in *Analytical Chemistry Division Annual Progress Report for Period Ending Sept. 30, 1970*, USAEC Report ORNL-4636, pp. 22-24, January 1971.
- National Academy of Sciences, *National Uses and Needs for Standard Radioactive Materials*, Washington, DC, 1970.

Estimated Date of Final Report

Continuing program

BLANK

THERMAL APPLICATIONS

Rotating arm impact test facility.

BLANK

Thermal Applications

The objective of the Thermal Applications Program is to develop uses of the thermal energy generated by radioisotope decay. (This does not include the development of radioisotope thermoelectric generators for use in space, which is supported by the Division of Space Nuclear Systems.) In a like manner, the development of thermoelectric generators for other than space use, e.g., the nuclear pacemaker and terrestrial thermoelectric generator units, is supported by the Division of Reactor Development and Technology.

The heat sources range from simple ones, requiring little or no temperature control, to more complex units distributing the thermal energy and controlling temperature within narrow limits. All require a high degree of

reliability and inherent safety. Current emphasis in this program is on (1) research related to the development of fuels for an implantable radioisotopic power source for heart assist devices, (2) evaluation of the practicality of developing a radioisotope thermal converter for an artificial heart having a minimum weight and volume and a minimum life of 10 years, (3) development and ground test of a waste-management/water-reclamation system for aerospace applications; (4) development and test of a large Co-60 heat source using an electric converter for test purposes; and (5) studies and development of small radioisotopic heat sources for thermal control.

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EVALUATION OF PRACTICABILITY OF A RADIOISOTOPE THERMAL CONVERTER FOR AN ARTIFICIAL HEART DEVICE*

Objective of Work

The objective of this program is to determine the practicability of developing a compact radioisotope thermal converter having a minimum weight and volume and a minimum life of 10 years. In this context, the converter is the mechanism by which thermal energy from a radioisotope heat source is converted to another energy form suitable for pumping blood. The program is divided into two phases, with the objective of Phase I being to evolve and evaluate innovative concepts for a variable powered thermal converter incorporating a vented radioisotope heat source. Pre-established selection criteria will be used to recommend the most promising concept for Phase II verification of theoretical and experimental practicability.

Progress and Results

The Phase I technical effort was completed mid-October 1971 with recommendations that a Stirling cycle power source providing pulsed input to a blood pump diaphragm by means of a mechanical drive train was the most promising approach. This selection was made with the aid of a decision modeling analysis of eight final candidate systems that had been screened from the approximately 25 concept variations investigated. Concepts included static conversion devices as well as dynamic systems representing several different thermodynamic approaches. Included in the eight contending concepts were systems using triple cascaded thermoelectrics, Stirling cycle gas dynamic systems, and vapor-cycle dynamic systems including both heat rate devices and high speed Rankine cycle devices. The three basic methods that were used for transforming and transferring the power source output to pulsed operation of the blood pump diaphragm were (1) mechanical drive, (2) direct hydraulic drive, and (3) accumulator hydraulic drive.

The recommended Stirling mechanical thermal converter concept shown in the figure has two major assemblies: a Stirling mechanical power source and a blood pump, interconnected by a flexible mechanical

shaft to transmit drive power and by coolant lines to carry waste heat rejected from the power source to a blood heat exchanger located in the blood pump assembly. System characteristics are:

Power to blood pump	4.45 W(c)
Initial isotope inventory	28.8 W(t)
Temperature peak	1520°F
Temperature sink	110°F
System weight	1646 g
System volume	706 cm ³

The power source is a Stirling cycle heat engine with rhombic drive connected through step up gearing to a magnetic coupling that provides a hermetic interface between the power source and the flexible drive shaft. The hot portions of the power source — the isotope capsule, the engine hot finger, and the combined displacer and regenerator — are enclosed in vacuum foil insulation. The heat sink is located above the power piston and below the displacer where waste heat is removed by a circulating isotonic cooling water mixture. Helium vented from the isotope capsule, which is enclosed in the engine volume, diffuses into the engine working space, into the crankcase, and out through a selective vent into the body cavity.

The blood pump and drive train are powered by a flexible shaft which enters the pump assembly from the left. It drives the flywheel through right angle bevel gears that transmit the power through a double reduction planetary gear system to two sets of right angle bevel gears that drive a set of Scotch yokes. The Scotch yokes, in turn, drive the actuating rod which pushes the blood pump diaphragm assembly to expel the blood on systole. The pushing motion of the two Scotch yokes is coordinated by connection to the actuation rod through a yoke pivot to insure balanced operation.

Plan for Next Period

During Phase II, parametric and trade studies will be conducted to optimize the preliminary design of an implantable version of the recommended concept. A realistically sized bench model will be designed, built, and tested in support of practicability assessment.

Reports Published

None

Estimated Date of Final Report

January 1972

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Stirling mechanical thermal converter schematic.

PREPARATION AND EVALUATION OF PLUTONIUM-238 RADIOISOTOPIC HEAT SOURCES*

Objective of Work

The objectives of this work are (1) prepare and evaluate medical grade Pu-238 compositions for life support applications, (2) fabricate and characterize heat source capsules (up to 50 W) for dosimetry, systems studies, and implantation studies, (3) provide heat source capsule on an interim basis which will also establish process reliability and product quality control for guidance in developing a commercial capability, (4) establish the compatibility of biomedical fuel forms with container materials, and (5) measure the rate of helium and radon release from biomedical Pu(238)O₂(16) fuel forms

Progress and Results

The preparation of potential Pu-238 fuel forms during 1968 and 1969 led to the development of four fuel forms having minimal radiation properties. These forms

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are the metal, Pu-238-3 at % Ga, Pu(238)N(15), and Pu(238)O₂(16). All four fuels were prepared from electrorefined metal of high chemical purity. Theoretical and experimental evaluations of these forms led to the conclusion that Pu(238)O₂(16) was the preferred form for high temperature application in the artificial heart program. During 1970 and 1971, emphasis was placed on the development of procedures for the ceramic fabrication of 20- and 50-W cylinders of Pu(238)O₂(16). Three 30-W and six 50-W cylinders were fabricated for delivery to the USAEC. Properties of two 30-W sources and three typical 50-W sources (80 at % Pu-238) are given in the table. The 30-W sources were prepared before the 50-W sources. A comparison of the neutron emission rates shows a small but steady decrease in the neutron emission rate as the procedures were improved. The neutron emission rates of sources 30-3, 30-6, 50-3, and 50-6 were 3608, 3519, 3488, and 3466 n/(sec)(g)Pu-238, respectively.

A study of the exchange of O-18 and O-17 with sintered pellets of Pu(238)O₂(16) in atmospheres of argon-plus-O₂, dry air, and air saturated with water vapor has shown that 50-W cylinders may be handled in air-filled glove boxes without any measureable oxygen exchange. This behavior is in marked contrast to that of Pu(238)O₂(16) powders, which must be handled in inert atmosphere glove boxes to avoid oxygen-isotope

Properties of Five Pu(238)O₂(16) Cylinders

	30-W Pu(238)O ₂ (16) cylinders		50 W Pu(238)O ₂ (16) cylinders (80 at % Pu-238)		
	LAS 30-3	PNL 30-6	NHI 50-2	NHI 50-3	NHI 50-4
Wt of Fuel Pellet, g	74.76	74.69	125.08	125.27	124.68
Wt of Pu, g	65.9	65.4	110	110	110
Oxygen/Pu Atom Ratio	2.01	2.01	1.99	1.99	1.99
Pu Isotopic Composition					
Wt % Pu-238 ^a	79.82	86.96	79.72	79.98	79.62
ppm Pu-236 ^b	0.70	0.25	0.76	0.75	0.58
Dimensions					
Diam, in	0.846	0.858	1.006	0.997	1.004
Height, in	0.803	0.808	0.996	1.003	1.013
Density, gPuO ₂ /cc	10.11	9.77	9.65	9.75	9.49
Power, watts	29.8	32.2	49.7	49.8	49.6
Power Density, watts/cc	4.03	4.20	3.83	3.88	3.78
Neutron Emission Rate					
n/(sec)(g) Pu	2880	3060		2790	2760
n/(sec)(g) Pu-238	3608	3519		3488	3466
Dose Rates at 10 cm					
Neutron, mrem/(hr) (g) Pu-238	0.36	0.35	0.35	0.35	0.35
Gamma, mr/(hr) (g) Pu-238	0.14	0.14	0.17	0.14	0.16

^ag Pu-238/100 g Pu

^bg Pu-236/10⁶ g Pu

exchange Two processes are still under study for the preparation of $\text{Pu}(238)\text{O}_2(16)$ powder In one, the oxide is prepared by converting electrorefined metal to $\text{Pu}(238)\text{O}_2(16)$ In the other, $\text{Pu}(238)\text{O}_2(\text{nat})$ is converted to $\text{Pu}(238)\text{O}_2(16)$ by isotopic exchange At present, the electrorefined metal route is preferred and has been used for the preparation of all 30- and 50-W sources

Work was started during 1971 on measurements of the rate of release of helium and radon from $\text{Pu}(238)\text{O}_2(16)$ cylinders

The characterization of Pu 238 fuel materials continues A nondestructive analytical method was developed for determining O 17 and O-18 in $\text{Pu}(238)\text{O}_2(16)$ by measuring the intensities of the (α, n) reaction gamma rays of O 17 and O-18 The reliability of the mass spectrometric measurement of O-18 in $\text{Pu}(238)\text{O}_2(16)$ was shown to be satisfactory by the new nondestructive technique Continuing study of the basic radiation properties of Pu 238 has revealed two previously unreported gamma rays having energies of 1428 and 1435 keV, the absolute intensities of these rays were measured

The experimental facility for the synthesis and fabrication of heat sources is operational One line of shielded glove boxes is used for the conversion of Savannah River $\text{Pu}(238)\text{O}_2(\text{nat})$ to bomb-reduced metal, two large inert atmosphere glove boxes are used for the preparation of metal and ceramic fuel forms, and another line of glove boxes is used for the encapsulation of sources

Plan for Next Period

The radiation properties of isotopically pure Pu-238 will be measured Fifty-watt $\text{Pu}(238)\text{O}_2(16)$ heat source capsules for implantations in miniature swine will be supplied The measurement of helium and radon release rates from $\text{Pu}(238)\text{O}_2(16)$ fuels will be continued, as will compatibility studies on vented and unvented capsules

Reports Published

- L J Mullins and J A Leary, Purification, Encapsulation, and Characterization of Pu-238 for Implantation Purposes, *Trans Amer Nucl Soc*, 13 496-97 (1970)
- L J Mullins and J A Leary, *Plutonium-238 Radioisotopic Heat Sources for the Artificial Heart Application*, Amer Inst of Chem Engineers, Symposium on Applications of Radioisotopes, December 1970

Estimated Date of Final Report

Continuing project

EVALUATION OF PRACTICABILITY OF A RADIOISOTOPE THERMAL CONVERTER FOR AN ARTIFICIAL HEART DEVICE*

Objective of Work

The objective of the program is to determine the practicability of developing a radioisotope thermal converter for an artificial heart device The thermal converter, including the radioisotope heat source, and all associated power conditioning and control systems are to be fully implantable and capable of functioning for 10 years with high reliability The device must supply the necessary mechanical, hydraulic, or pneumatic power for a blood pump which provides 100% of the left ventricular pumping function

Progress and Results

The program is divided into two phases The first phase is a 7-month evaluation of candidate concepts and the selection of a preferred design for further detailed evaluation The second phase, which is not currently a part of the contractual effort, will be a 17-month effort that will include detailed analyses of the preferred concept and culminate in the design, fabrication, and test of a realistically sized thermal converter using a radioisotope fueled heat source

During Phase I, a list of evaluation criteria was compiled for use in selecting a preferred thermal converter system for Phase II development The principal design criteria included a daily average power level of 2.8 W(e) delivered to the blood pump and the following maxima: 60 W(t) heat source, 1.5-liter volume, specific gravity of 2.0, and peak power level of 4.44 W(e)

The concepts evaluated included various mechanizations of reciprocating, rotary, and thermoelectric cycles, with both gas and vapor operating fluids, as well as various combinations of these devices Eight different thermal converter systems were found to be capable of meeting all the design criteria

These eight systems were compared, using the previously defined evaluation criteria and scoring methodology The preferred concept shown in the figure is

*I R Jones, M G Ridgway, and D R Snoke, TRW Systems Group, One Space Park, Redondo Beach, CA 90278, Contract AT(04 3) 857

Hybrid engine (left) and motor (right)

an all electric output hybrid engine that powers a motor that in turn, mechanically actuates a Kwan-Gett type blood pump. The total weight and volume of the engine, motor, and actuator are 1.83 kg and 1.33 liters. The hybrid engine includes cascaded Si-Ge(Te) thermoelectric modules mounted on each end of a 49 W(t) vented, cylindrical heat source with hot and cold junction temperatures of 1740°F and 460°F.

The cold side of the thermoelectrics and surrounding insulation system serves as the source of heat for the boiler of a rotary vapor turbogenerator which uses CP 34 as the working fluid, operates between 430°F and 116°F, and puts out 7.24 W(e) at 15 volts. The thermoelectric system, after dc to dc conversion, provides 2.61 W(e) at 15 volts. The motor uses the total of 9.85 W(e) to power a 15 volt, 9000 rpm brushless dc motor that is geared down to drive a rotary cam at 120 rpm. The rotary motion is converted to reciprocating motion by a cam-follower operating in a continuous elliptical groove on the inner diameter of the rotary cam. The reciprocator is mechanically coupled to an automatic actuator that provides passive autoregulation for a modified Kwan-Gett blood pump. This feature permits the actuator to operate at a constant 120 cycles per minute and accommodate blood flow rates ranging from 6 to 12 liters/min. Because the overall efficiency

of the motor and automatic actuator is 45%, the power delivered to the blood pump is 4.44 W(e), which corresponds to the peak activity requirement.

The hybrid thermal converter employs only fibrous thermal insulation with an inert gas fill. Some volume and weight can be saved by use of vacuum foil insulation but at the expense of reduced system reliability. Heat is rejected directly into the abdominal body tissues and fluids, although about half the heat can be rejected through the blood pump if required. When high energy, high density solid electrolyte batteries are developed, the hybrid concept offers considerable growth capability since the device could then be sized to provide the average power level (2.81 W) rather than the maximum power level (4.44 W). Options include a pure rotary vapor system (most compact), a pure thermoelectric system (most reliable), or a hybrid/battery system (smallest heat source).

Plan for Next Period

The Phase I Final Report will be completed and submitted to the AEC.

Reports Published

None

Estimated Date of Final Report

The Phase I Final Report will be issued in November 1971

RECIPIENT RADIATION EXPOSURE AND RADIOLOGICAL ENGINEERING STUDIES RELATING TO CIRCULATORY SUPPORT SYSTEMS*

Objective of Work

The major objective of this study is to determine the dose from radioisotopic heat sources suitable for circulatory support systems. This is accomplished by measuring and calculating, with suitable codes, the dose rates within and surrounding various tissue-equivalent phantoms. Another portion of the program concerns the development of radioisotopic blood heaters for use in an animal study aimed at assessing the combined effects of heat and radiation in miniature swine implanted with 50-W plutonium heat sources.

Progress and Results

Completed studies on shielding, dose prediction, and dose measurement with a 30-W Pu-238 medical-grade metal source demonstrated that the computer code,

*F. T. Cross, J. C. Sheppard, and B. D. Bingham, Battelle, Pacific Northwest Laboratory, Richland, WA 99352, Contract AT(45-1) 1830

QAD-P5A, when suitably modified, can be used to calculate the dose rates to various organs for a wide variety of source configurations.

For many organs, the center point dose rate is a satisfactory estimate of the average organ dose rate. The calculated dose rates to various organs from a 30-W Pu-238 metal source placed on the inside surface of the left ilium are shown in the table. The coordinate system is centered in the bottom plane of an elliptical cylinder that represents the body, with a Z-axis positive in the vertical direction, the X-axis positive to the left side of the body, and the Y-axis positive to the back of the body. The relevance of these dose rates is highly dependent on the source and organ coordinates simulating an actual implant. Any other source location would significantly change the dose rates to those organs proximal to the source.

Dose rates from Pu-238 and the daughters of Pu-236 contained in the Hittman-Rankine dosimetry subsystem, a prototypic power source, and various prototypic circulatory support systems have also been computed. Whereas calculation of the neutron contribution to the dose appears straight-forward and verifiable by measurements, calculation of the photon dose rates remains subject to the uncertainties of assigning buildup factors for multilayer systems. The measurements on the more complex power source systems provide guidance on the choice of buildup factors.

Measurements of the neutron dose rates from a 32-W Pu(238)O₂(16) source contained in the Rankine device

Total (n + γ) Dose Rates to "Mathematical" Organs^a

Organ	Center coordinates (X,Y,&Z)	Center to center distance cm	Average dose rate, mrad/hr ^b	Average dose rate, mrem/hr ^c
Spleen	11, 3, 37	19.5	1.4	2.1
Left kidney	6, 3, 32.5	15.8	2.7	4.4
Stomach	8, 4, 35	17.5	2.0	3.1
Small intestine		10	7.0	13
		(equivalent)		
Left ovary	6, 0, 15	3.61	44	100
Left testis	12, -2.2, 2.3	21.5	0.99	1.4
Uterus	0, 2, 14	9.17	8.3	16
Bladder	0, -4.5, 8	13.6	3.5	5.8
Left adrenal	4.5, 6.5, 38	21.3	1.1	1.6
Heart ^d	1, -3, 51	34.3	0.24	0.27

^aFrom a 30-W Pu-238 metal source shielded with 30 mil Ta and 63-mil Al centered at 8, 0, 18, and placed with its axis parallel to the body axis.

^bEstimated accuracy +25% (dose rates exclude Pu-236 contribution).

^cNeutron Quality Factor = 10.

^dIn an artificial heart implant, these values would represent the dose rates to the blood pump occupying the same location.

and placed in a large homogeneous phantom and Remab phantom filled with tissue equivalent fluid were completed. Californium-252 was used as a stand-in for the Pu-238 so that long measurement times would not be necessary. Small tissue-equivalent proportional counters were used for these measurements. In addition, measurement was completed of the photon dose rates from the oxide source placed in the Remab phantom. The combined ($n + \gamma$) dose rates on the phantom surface shown in Figs 1 to 4 can be compared with the dose rates reported in the 1970 Project Summary for the metal source, but, because the oxide source was contained in the Rankine device, it was physically too large to place in the same exact location. The source is still placed on the left ilium in line with the 5th lumbar vertebra, but its center is closer to the front surface of the body, and the effect of shielding by the device materials and the source relocation is reflected in the measurements. In addition, the photon dose rates from the daughters of Pu-236 in the metal

source were repeatedly measured. The measurements confirmed predictions of the growth of the dose rates from the daughters of Pu-236.

Engineering studies to develop radioisotopic blood heaters in support of a project for the Division of Biology and Medicine (Biological Effect of Intracorporeal Radioisotope Heat Source) were initiated this year. Heater geometry, materials, and temperature changes in the bulk stream and boundary layer of swine blood were investigated. Heat exchangers incorporating telemetry units for 50-W $\text{Pu}(238)\text{O}_2(16)$ sources and various biological shams were developed for interposing the thoracic aorta. The temperature analysis indicated that, upon the death of the animal and loss of blood as coolant, the plutonium heat source centerline temperature would reach about 972°F if the ambient temperature was 75°F.

Also initiated was the design of heat conduit systems for abdominal heat sources, with exchangers similar to the devices designed for thoracic implant but with an

Fig. 1.

Total ($n + \gamma$) dose-rates in mrad/hr on surface of remab phantom. (Excluding Pu-236 contribution)

Fig. 2.

Total ($n + \gamma$) dose-rates in mrem/hr on surface of remab phantom. (Excluding Pu-236 contribution QF = 1.0)

interconnecting heat conductor to allow dumping of waste heat into the larger blood vessels in the thoracic region

Fig. 3.

Total ($n + \gamma$) dose-rates in mrad/hr in bisecting radial plane of source. (Excluding Pu-236 contribution)

Fig. 4.

Total ($n + \gamma$) dose-rates in mrem/hr in bisecting radial plane of source. (Excluding Pu-236 contribution QF = 10)

Plan for Next Period

Measurements of the dose rates from the oxide source contained in the Rankine device will be completed and compared with the QAD-code predictions. A User's Manual for dose rate calculations from Pu-238 heat source will be written. The dose rates from future prototypic power sources will be determined, and guidance will be provided on the dose rates received by animals with implanted heat sources. The design and fabrication of the animal blood heaters and shams will be completed.

Reports Published

None

Estimated Date of Final Report

A topical report is scheduled for July 1, 1972

POPULATION RADIATION EXPOSURE FROM HEART DEVICE*

Objective of Work

The objective of this project is to identify the radiation dose to the population that would result from introduction of artificial heart devices fueled with Pu-238.

Progress and Results

The procedure in this study develops an estimate of the potential radiation dose from artificial heart devices through construction of time-distance relationships between persons typifying potential heart recipients and other members of the population. A profile of potential artificial heart recipients is being constructed based on previous National Heart Institute studies and US Public Health Service survey results, and an epidemiological study is being conducted to determine appropriate life-expectancy factors for recipients of artificial hearts.

A computer program is being written to combine the data on heart recipient profile, life expectancy, and device failure probabilities and to calculate the number of devices in use and the age and sex distribution of the users, under various implantation rate assumptions. The methodology for measuring the time-distance relationships of heart recipients to the rest of the population is

*R. W. McKee, Battelle, Pacific Northwest Laboratory, Richland WA 99352, Contract AT(45-1) 1830

being developed, taking into consideration studies in the field of environmental psychology relating to interpersonal distance requirements of individuals

Plan for Next Period

Substantial survey measurements of time-distance relationships of people in the principal activities of present day society (including occupation, leisure, travel, etc.) are anticipated. The time-distance relationships determined from the survey will be converted to radiation dose using a computer program. Results will be summed up for all contacts of heart recipients with the rest of the population to provide an estimate of total radiation dose to the population.

Reports Published

None

Estimated Date of Final Report

December 1972

ARTIFICIAL HEART PROGRAM*

Objective of Work

Monsanto Research Corporation's (MRC) participation in the Artificial Heart Program consists of developing, in conjunction with TRW Systems, the design for the 50-W heat source and a process for fabricating the heat source and of producing both test and fueled sources for TRW's use.

Progress and Results

Progress has been limited so far to conceptual discussions with the system's contractor.

Plan for Next Period

During the next year, MRC will have completed the objectives listed above. The hardware delivery quantities are twelve test capsules and three fueled heat sources.

Reports Published

None

*E. W. Johnson, Monsanto Research Corp. Mound Laboratory, Miamisburg, OH 45342, Contract AT(33-1)-GEN 53 and NASA, Ames Research Center, Moffett Field, CA

Estimated Date of Final Report

December 1972

INTEGRATED WASTE MANAGEMENT — WATER SYSTEM USING RADIOISOTOPES FOR THERMAL ENERGY*

Objective of Work

In the present Apollo spacecrafts water is not recovered from biological wastes because drinking water is produced as a by-product of the fuel cell electrical power system. The waste management system does not provide earth-type convenience because of the short mission and the spacecraft restrictions on weight and electrical power, but longer space missions and more personnel will require a more acceptable approach to waste management. Water must be recovered so that launch weight will be minimized. For example, four men on a 180-day space mission will need nearly 10,000 lb of water — which could cost as much as \$10,000,000 to supply.

The objectives of this program are to design, develop, fabricate, and test an engineering model of a completely integrated Waste Management Water System (WM-WS) using Radioisotopes for Thermal Energy (RITE). The complete system must be capable of collecting and processing wastes from four men for a 180-day simulated space mission. Subsystems include feces, trash, and urine collection, water reclamation, storing, heating, and dispensing the water, storage and disposal of the feces and urine residue and all other nonmetallic waste material by incineration.

Progress and Results

The system has been designed and fabrication has begun. A new solids pump-filter has been developed and is being tested in the breadboard system for an additional 30 days. The water recovery portion of the system is being operated at approximately 130% of capacity to ensure that excess water can be provided if necessary. The water recovery efficiency is in excess of 98%. The incineration process typically reduces solids weight and volume by 99%. A new packaging arrangement has been derived (see photo) which permits separation of the waste collection from the processing section. This arrangement also permits the use of several collection units with one central processing unit.

*R. W. Murray, General Electric Company, Space Division, P. O. Box 8555, Philadelphia, PA 19101. Contract AT(11-1) 3036

Integrates waste management water system.

Plan for Next Period

The total system will be assembled and tested. The radioisotope heater will be concurrently fabricated for use in a 4-man, 180-day test of the total system in a simulated space cabin. The selected fuel for the heater is Pu-238, with a thermal output of 400 W.

Reports Published

None.

Estimated Date of Final Report

February 1972.

INTEGRATED WASTE MANAGEMENT PROGRAM [RADIOISOTOPES FOR THERMAL ENERGY (RITE) HEAT SOURCE]*

Objective of Work

The objectives of the work are to develop and fabricate the RITE heat source for use in the integrated waste management—water system. Present plans are to

use the heat source in a system which will be used in a simulated space mission designed to accommodate four men for 180 days. Again, sources fueled with $\text{Pu}(238)\text{O}_2$ were selected because of their reliability and weight savings.

Progress and Results

None.

Plan for Next Period

A modified Pioneer heat source fueled with 400 W(t) of $\text{Pu}(238)\text{O}_2$ shards will be fabricated at Mound Laboratory for delivery in February 1972. The heat source will be placed in a Hastelloy-C container and will be used initially for ground-test studies.

Reports Published

None.

*C. E. Burgan, D. L. Coffey, and C. O. Brewer, Monsanto Research Corp., Mound Laboratory, Miamisburg, OH 45342, Contract AT(33-1)-GEN-53.

Estimated Date of Final Report

Continuing project

BIOMEDICAL ENGINEERING SUPPORT*

Objective of Work

The objectives of this project are (1) to determine the power required to drive an artificial heart under various conditions, (2) to determine the efficiency of a Kwan—Gett hemispherical artificial heart and the source of energy losses, and (3) to provide the USAEC with artificial ventricles and any information and expertise required in the development of an implantable power supply.

Progress and Results

The power requirements for an artificial heart with 60% pump efficiency in a 25-yr-old 68-kg male 173 cm tall — with a daily energy requirement of 3295 kcal, an arterial pressure of 100 mm Hg, and pulmonary artery pressure of 15 mm Hg — have been determined from the literature. The average power for the total artificial heart is 3.23 W with a peak power requirement of 4.9 W during moderate exercise. The average power requirement increases from 2.95 W at a body weight of 45 kg to 3.25 W at a body weight of 80 kg for the 25-yr-old male. The average power requirement decreases from 3.23 W at 25 years of age to 2.85 W at 70 years of age for the 68-kg male.

For measurement of the efficiencies of a Kwan—Gett artificial heart, the necessary equipment has been assembled to determine the energy input to the system at five locations (see figure). These measurements will be made under simulated circulation and verified in animal experiments. The rates of energy transfer at two points are calculated from the flow rate of air determined by a venturi meter and the air pressure. Pressure—volume diagrams are used to determine the energy transfers to the diaphragm in the air chamber and from the diaphragm to the blood chamber. The volume of blood in the heart at any instant is determined by volume transducers. The energy dissipated during simulated circulation is calculated from the fluid flow rate measured by an electromagnetic flow transducer and the arterial pressure. No energy measurements have been made at this time.

*F. M. Donovan, Jr. and W. J. Kolff, Department of Surgery, Division of Artificial Organs, Building 512, University of Utah, Salt Lake City, UT 84112, Contract AT(11-1)-2155

Schematic of blood pump system.

Plan for Next Period

The efficiencies and energy losses for the artificial heart will be determined during simulated circulation. These measurements will be verified by implanting artificial hearts in calves and measuring the energy transfer into the distal end of the air drive line under various cardiac outputs and arterial pressures in the animal.

Reports Published

None

Estimated Date of Final Report

July 15, 1972

RADIOISOTOPE HEATER DEVELOPMENT PROGRAM — PHASE II*

Objective of Work

Phase I of this program was performed under sponsorship of the National Aeronautics and Space Administration, Manned Spacecraft Center (Contract

*R. B. Goranson, McDonnell Douglas Astronautics Company, Donald W. Douglas Laboratories, Richland, WA 99352, Contract AT(45-1)-2166

NAS-9-8846), and was completed during the second quarter of 1969. Phase I effort concentrated on preparation of 10- and 50-W plutonia and promethia reference design heaters for manned spacecraft using current technology. Designs included the largest number of potential applications with a minimum number of heater configurations.

The Phase II program — completed August 30, 1971 — included both manned and unmanned spacecraft applications and was directed toward development of radioisotope heater technology using improved containment and ablation materials. A major objective was to establish practical design configurations so that identifiable applications could be studied rapidly and efficiently.

Progress and Results

Data from questionnaires sent to NASA aerospace centers, the Department of Defense, and commercial satellite prime contractors about potential applications of 1- to 50-W heaters in manned and unmanned spacecraft were used to update the Phase I design criteria and identify a wide range of potential applications.

Initial experimental effort was directed toward obtaining physical and thermophysical properties of candidate insulator and ablator materials. After a preliminary screening, efforts were concentrated on pyrolytic graphite (PG) and expanded pyrolytic graphite (EPG) insulators and on the DWDL-developed HfB_2 composite reentry ablator material. Thermal conductivity measurements gave values for hemispherical PG specimens lower than previously reported and confirmed the ideal conditions of thermal conductivity at normal temperatures. The very low thermal conductivity at reentry was confirmed by a series of arc-tunnel tests.

By application of the experimentally determined thermal and physical properties of candidate materials, optimized heater designs were developed. Computer techniques have been developed that permit rapid design of minimum-weight heaters with an array of optional materials and geometries. For a given heater size and geometry (spherical or cylindrical capsule, spherical or cylindrical insulator, and cubic, cylindrical, or prismatic ablator), the program determines pressure-vessel wall thickness and the required thickness of insulator and ablator layers for given lifetime and mission conditions.

Post-test view of HfB_2 composite cube and cylinder (left) and comparable ATJ-S models (right).

After initial design studies, 24 ablation test specimens were fabricated for testing in the arc-tunnel facility of Aerotherm Corporation, Mountain View, CA. The specimens included solid cubes and cylinders of ATJ-S and HfB_2 composite ablator material as well as a thermocouple-instrumented set of specimens containing ZrO_2 fuel simulant and PG insulator internal to the ATJ-S ablator. Tests were run using full scale models at calculated reentry conditions. The figure shows the HfB_2 composite ablators (left) and ATJ-S ablators following tests at identical reentry conditions. The material loss evident on the ATJ-S specimens was not observable on the HfB_2 composite.

Impact survival after reentry is another area of concern in heater safety. Impact-test specimens were prepared (fuel simulant, container shell, and insulator) and tested at various velocities using the unique rotating-bloc impact-test facility at DWDL, which permits precise control of impact temperature, velocity, and orientation, minimum turnaround time (15 min for a cold specimen), and greatly reduced test costs. A photograph of this apparatus is shown in the section frontispiece. Data eliminated any existing preference for EPG over PG as a bumper material and yielded the basis for semiempirical impact-survival prediction techniques for materials examined.

A brief series of simulated launch-abort debris tests was conducted at the MDC Aerophysics Laboratory, El Segundo, CA. Heater models were subjected to 2000 fps impact by .22 and 30/06 caliber bullets. Results showed less capsule deformation when the ATJ-S ablator is replaced by the DWDL HfB_2 composite.

Design, fabrication, and test capabilities — using advanced materials and techniques — have been developed to permit efficient response to specific heater questions.

Plan For Next Period

The final report will be published.

Reports Published

The Phase I Summary Report — published as McDonnell Douglas Report MDC-G2029 — was submitted to the USAEC in November 1970.

Estimated Date of Final Report

A draft of the Phase II Final Report was submitted to USAEC on Sept. 30, 1971.

DEVELOPMENT OF COBALT-60 HEAT SOURCES*

Objective of Work

The purpose of this project is to provide basic data that will be required to design, fabricate, and operate Co-60 heat sources. Primary emphasis is on selecting materials to encapsulate cobalt fuel forms and establishing temperature limits for long-term operation of capsules. Development of specific heat source concepts is not included, but assistance is provided when required for developing and testing demonstration units.

Progress and Results

Heating tests of four selected superalloys containing irradiated and unirradiated cobalt were continued. The four alloys are:

Haynes 25 (Trademark of Cabot Corporation)

Inconel 600 (Trademark of International Nickel Company)

Hastelloy X (Trademark of Cabot Corporation)

Hastelloy C (Trademark of Cabot Corporation)

Measurements on 13 capsules, initially containing ~13 thousand Ci of Co-60 each, after exposures up to 20 thousand hr at 900°C or 1000°C, showed maximum diameter changes of:

1. A decrease of 0.007 in on an Inconel-600 capsule heated 20 thousand hr at 900°C, the decrease probably reflects spalling of the oxide scale.
2. An increase of 0.008 in on a Haynes-25 capsule heated 18 thousand hr at 1000°C, this change probably reflects a density decrease in this alloy due to precipitation of Co_2W .

Metallographic examination of two Hastelloy-X capsules (10 thousand and 20 thousand hr at 1000°C) and two Haynes-25 capsules (5 thousand and 20 thousand hr at 1000°C) showed that the widths of the reaction zones and the oxidation-affected zones were in good agreement with those measured on "inactive" capsules.

*S. Mirshak, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC 29801, Contract AT(07-2)-1.

heated under similar conditions. For example, Hastelloy-X capsules heated 20 thousand hr at 1000°C had reaction zones between the alloy and the cobalt 0.0185 in wide for the active capsule and 0.017 in wide for the inactive capsule. The depth affected by oxidation was 0.014 in. on the active capsule and 0.010 in. on the inactive capsule. Examination of the remaining capsules is continuing.

These tests demonstrate that Haynes-25, Hastelloy-C, Hastelloy-X (20 thousand hr at 1000°C), and Inconel-600 (20 thousand hr at 900°C and 10 thousand hr at 1000°C) will perform satisfactorily as capsules at least at the indicated condition. However, an inactive Haynes-25 capsule oxidized severely after 29 thousand hr at 1000°C, resulting in loss of weld integrity. Nondestructive examination of this same capsule after 11,500 hr had shown that the capsule was intact. The oxidation reduced the wall thickness from 0.095 in. to <0.050 in. This amount of oxidation is essentially equivalent to the initial weld penetration and, therefore, caused the loss of weld integrity. Tests of these alloys to 50 thousand hr are continuing.

Satisfactory performance through 1000 hr at 1200°C was demonstrated with a tungsten capsule containing unirradiated cobalt metal wafers. Although the cobalt wafers bonded to each other, they did not bond tightly to the capsule and fell out during sectioning for metallographic examination.

Cobalt diffused into the wall of a W-Re capsule heated 1000 hr at 1200°C to a depth of 0.0065 in. In another capsule, heated 1000 hr at 1400°C, the total depth was 0.012 in. Extrapolation of these results to 50 thousand hr predicts penetrations of 0.045 in. at 1200°C and 0.085 in. at 1400°C. No more work on the compatibility of cobalt metal with refractory metal alloys is planned.

Construction of the new shielded cell to be used for long-term Co-60 capsule testing is complete. Cold runs are in progress prior to installation of the Co-60.

Technical assistance is being provided in the project of Westinghouse Astronuclear Laboratory to design, fabricate, and test an experimental heat source containing ~2 MCi (30 kW) of Co-60. Tests were completed to measure the oxidation resistance of Nickel-200, Hastelloy-X, and Haynes-188 at 1000° and 1125°C. Nickel-200 is similar to the stronger and slightly more oxidation-resistant Nickel-201 that will be used for the heat source core. Hastelloy-X and Haynes-188 were candidate materials for the fuel capsules and pins. Hastelloy-X was selected because of its better oxidation resistance.

Because of the potential application of high-activity-cobalt metal in heat sources, the USAEC has established a loan program for this material. About 10 MCi (150 kW) of Co-60 at over 350 Ci per gram of cobalt will be available for this program. Most of this material is in the form of 0.745-in.-dia wafers, plated with nickel. Individual companies or groups of companies are invited to participate by contacting the Savannah River Operations Office of the USAEC. Moderate activities and amounts of Co-60 can be obtained commercially.

Plan for Next Period

During the next year, destructive examination of capsules removed from tests will be continued and technical support of the 30-kW heat source project will continue. Heating tests of Co-60 capsules in the new cell will begin. Compatibility of oxidized cobalt fuel forms with iridium will be investigated.

Reports Published

C. L. Angerman and C. P. Ross (Compilers), *Savannah River Laboratory Isotopic Power and Heat Sources Quarterly Progress Reports*, USAEC Reports DP-1254, July-September 1970, DP-1267, October-December 1970, DP-1272, January-March 1971, DP-1279, April-June 1971.

Estimated Date of Final Report

Continuing project

COBALT-60 HEAT SOURCE DEMONSTRATION PROGRAM*

Objective of Work

The objectives of this project are to design, develop, fabricate, and test a terrestrial radioisotope heat source fueled with Co-60 for the purposes of establishing heat-source integrity and safety, obtaining engineering information to guide the design of future devices to power a variety of energy conversion systems, demonstrating reliability and freedom from maintenance, and showing that a heat source of this type can be economically attractive.

*J. W. Sadler, Westinghouse Electric Corp., Astronuclear Laboratory, P. O. Box 10864, Pittsburgh, PA 15236, Contract AT(38-1) 543.

Heat pipe for Co-60 emergency cooling subsystem.

Progress and Results

The Co-60 heat-source demonstration program has been divided into five phases: (1) preliminary design; (2) detail design; (3) fabrication; (4) demonstration test program; and (5) disassembly.

The Phase II design, completed in October 1970, was modified early in Phase III based on results obtained from the supporting SRL Co-60 Material Technology Program, which indicated that the previously selected Co-60 encapsulating materials, Haynes-188 and Haynes-25, were subject to breakaway oxidation at the elevated temperatures corresponding to the emergency cooling mode. The heat source design was therefore modified to allow the use of Hastelloy-X as the encapsulating material and to provide a means of controlling the fuel pin cavity environment. The modified design provides for an argon coolant outlet temperature of 1350°F.

Extensive testing of the heat pipes (see figure), which function as the passive emergency cooling subsystem, has confirmed the operating characteristics of the inert-gas-controlled heat pipe, specifically a switching temperature difference of 55°F from the 1040°F "off" condition to the point at which 1 kW is transferred. The upper limit on heat pipe capability is approximately 1.5 kW. Over 4000 hours of heat pipe testing, including weekly operating cycles, has been compiled. Procurement of the heat source and demonstration loop is continuing, and encapsulation of the 2×10^6 Ci of Co-60 has been initiated.

Plan for Next Period

Heat source assembly and test-loop checkout will be completed. The demonstration program, including thermal characteristics and subsequent endurance testing, will be initiated.

Reports Published

J. W. Sadler and G. H. Parker, Large Cobalt-60 Heat Source Program, in *Intersociety Energy Conversion Engineering Conference*, Boston, MA, August 1971, pp. 741-50, Society of Automotive Engineers, Inc., New York, 1971.

Estimated Date of Final Report

The Phase III Summary Report will be issued April 1972.

PIONEER RADIOISOTOPE HEATER UNIT (PRHU) PROGRAM*

Objective of Work

The objective of this program is to design and fabricate small heat sources of $\text{Pu}(238)\text{O}_2$ /Molybdenum Cermet (PMC) fuel for use on the Pioneer spacecraft as heaters for the tanks and nozzles handling the hydrazine propellant which freezes at 1°C. The use of the sources obviates the need for the spacecraft's electrical system to provide the necessary heating.

Progress and Results

Thirteen heat sources (see figure) were fabricated at Mound Laboratory and delivered for use on Pioneer Flight F.

Plan for Next Period

Fifteen heat sources are scheduled for fabrication in 1972 for Pioneer Flight G.

*E. W. Johnson, Monsanto Research Corp., Mound Laboratory, Miamisburg, OH 45342, Contract AT(33-1)-GEN-53 and NASA, Ames Research Center, Moffett Field, CA.

Pioneer Radioisotope Heater Unit (PRHU).

Reports Published

None.

Estimated Date of Final Report

Continuing project.

**A REVIEW AND EVALUATION OF
TRW SAFETY ANALYSIS REPORT,
VOLUMES I AND II, FOR THE
PIONEER RADIOISOTOPE HEATER UNIT***

Objective of Work

The objective of this review was to verify the technical adequacy of the TRW Safety Analysis Report (SAR) on the 1-W Pu(238)O₂ Radioisotope Heater Unit (RHU). Twelve heaters were considered for use on each Pioneer F and G spacecraft.

Progress and Results

As stated in Volume I of the SAR, under certain unlikely conditions, PuO₂ may be released from the RHU's as a result of (1) a launch pad abort which involves burning of the third-stage propellant; (2) an

ascent abort which leads to a grazing reentry 25 months or more after fuel encapsulation; or (3) a random orbital decay 20 years or more after fuel encapsulation. Weiner Associates, Inc., feels that a many-skip reentry could cause excessive ablation of the graphite heat shield, especially with the uncertainty of high velocity ablation data. However, all material released during reentry would not produce a hazard greater than 1/10,000 of the maximum permissible burden (MPB) in human lungs, and whether additional releases can occur from other origins should not detract from the overall safety evaluation of the RHU's. While pointing out a few specific areas where further investigation might be in order, WAI generally finds the TRW Safety Analysis Report to be adequate.

Plan for Next Period

None.

Reports Published

A Review and Evaluation of TRW Safety Analysis Report, Volume I and II, for the Pioneer Radioisotope Heater Unit, USAEC Report WAI-TR-70.6-AEC, Weiner Associates, Inc., December 1970.

Estimated Date of Final Report

The project has been completed.

*R. I. Weiner and D. W. Pyatt, Weiner Associates, Inc., 107 Industry Lane, Cockeysville, MD 21030, Contract AT(30-1)-4145.

ENVIRONMENTAL AND OCEAN SCIENCES

Layout on beach for radioisotope sand tracking.

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Environmental and Ocean Sciences

The objectives of the Environmental and Ocean Sciences programs are to develop isotope techniques to aid in environmental pollution control and to assist in the exploration and economic development of the oceans.

Technological achievements and the concurrent advances in standards and patterns of living have brought with them a myriad of environmental problems, some of the more notable being air and water pollution, i.e., the unwanted presence of chemical substances in low but often noxious concentrations. To determine the damage pollutants may do to plants and animals, the presence of such pollutants must be detected, measured, and followed. The measurement problems invite a consideration of isotopic tracers and related analytical techniques, which, because of their extreme sensitivity and specificity, are potentially very valuable. In addition, low-energy x and gamma rays from isotopic sources can be used for x-ray-fluorescence, Moessbauer, and absorptiometric determinations of very low concentrations of substances.

Ocean science offers hope to the growing populations of the world. Population increase and rising living

standards are exerting increasing pressure on land resources, and man can no longer ignore the cultivation and mining potentials of the oceans. Techniques for extracting resources from the seas have not been developed because they have been uneconomic compared with land farming and mining and because knowledge of location of the sea's resources has been restricted. However, imminent shortages of food and minerals are rapidly making development of the seas essential. Isotopic and radiation techniques hold many possibilities for obtaining oceanographic data that are difficult or impossible to obtain by other methods.

The environmental and ocean-science projects are being conducted in collaboration with other government agencies that have primary responsibilities in these areas, and many are being cofunded by these agencies. In particular, ocean-science projects are being selected and technically monitored with the assistance of the Navy's Oceanographic Office.

The environmental-science projects are presented first and are followed by the oceanographic projects.

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ATMOSPHERIC DIAGNOSTICS PROGRAM*

Objective of Work

The atmospheric diagnostics program has the objective of developing tracing methods and instrumentation and applying them to the study of pollution and meteorology. The chief emphasis has been placed on the isotope-ratio and sulfur hexafluoride tracer methods. The sulfur-isotope-ratio method, based on variations in the ratio of the stable sulfur isotopes — S-32/S-34 — in sulfur from different sources, uses an isotope-ratio mass spectrometer. It provides both a sensitive means of physically tracking SO_2 from a discrete source and a way of measuring the degree of conversion that sulfur has undergone in a chemical reaction (since the ratio change resulting from isotopic exchange after the reaction is quantitatively related to the degree of conversion). The chemically stable SF_6 is a gaseous tracer for stack emissions whose concentration in air can be determined with extreme sensitivity and precision by gas chromatography. These two tracer methods can be used either individually or as a dual system in which SF_6 defines the shape of a stack plume, against which measurements of chemical changes determined by the sulfur-isotope-ratio method may be normalized.

Progress and Results

The sulfur-isotope-ratio and SF_6 methods are being applied to the study of sulfur pollutants in power-plant stack plumes at two tall-stack stations in low-background rural areas — the 370-MW oil-fueled Northport Station of the Long Island Lighting Company and the 900-MW coal-fueled Keystone Station of the Pennsylvania Electric Company. Primary interest is in the oxidation of SO_2 to sulfuric acid and sulfates as a function of distance of plume travel from the stack and of meteorological conditions. Sulfur hexafluoride is injected into the base of the stack, and samples are taken, aloft, of the plume and of the background. At the same time, samples of the flue gas are taken isokinetically as it enters the stack. The SO_2 , sulfate, and SF_6 concentrations and the S-32/S-34 ratios are then determined. The laboratory instrument used for

determining SF_6 concentrations is based on a direct gas-chromatographic method and can detect 1 part of SF_6 in 10^{13} parts of air with no preconcentration. The precision of the S-32/S-34 ratio determination is ± 0.2 per mil.

Ten to 20% conversion of SO_2 to sulfate at distances of up to 10 miles under the conditions prevailing has been observed. A second-order mechanism is postulated for the oxidation, with a rate constant of approximately $1 \text{ hr}^{-1} \text{ ppm}^{-1}$, possibly indicating an interaction between SO_2 and particles in the plume or moisture absorbed on such particles.

Development of a real-time portable gas chromatograph for airborne detection of SF_6 has been completed, and it is now in routine use in plume-definition studies (see figure). It is stable in operation and can detect 1 part of SF_6 in 10^{12} parts of air. It is being used in conjunction with an optical densitometer on a single plane in studies of the correspondence of the particulate and gaseous plumes from a stack.

The small-city SO_2 -distribution analyzer developed for New Haven, CT, indicated that the general background level of SO_2 , either emitted from low-lying sources or carried in from outside the city, was sufficiently high to obscure the contributions from individual high-stack sources in the city.

Plan for Next Period

Work will continue along the lines already started: establishing of definitive patterns for SO_2 conversion in plumes, mode of removal of sulfur oxides from plumes, the structure of plumes at long distances from a stack, and the nature of particulate sulfates formed in a plume. Development of the techniques, equipment, and instrumentation required for the study of pollutant problems in the atmosphere will be continued. Application of the SF_6 tracer method to tall stack plume-dispersion studies in an urban area is being initiated in New York City.

Reports Published

- B. Manowitz, L. Newman, and W. D. Tucker, *The Atmospheric Diagnostics Program at BNL. Third Status Report*, USAEC Report BNL-50280, December 1970.
- B. Manowitz, M. E. Smith, M. Steinberg, L. Newman, and W. D. Tucker, *The Isotope Ratio Tracer Method. Applications in Atmospheric Sulfur Pollutant Studies*, Second International Clear Air Congress, Washington, DC, December 1970, USAEC Report BNL-15095.

*B. Manowitz, M. Smith, L. Newman, and W. Tucker, Brookhaven National Laboratory, Upton, NY 11973, Contract AT(30-2) GLN 16. The project is jointly supported with the Division of Biology and Medicine and the Environmental Protection Agency's Division of Chemistry and Physics and Division of Meteorology.

Instrumentation for plume definition studies in place on aircraft, showing optical densitometer in rear, real-time SF_6 analyzer in front, together with inverters and storage batteries in case.

- M E Smith and D A Singer, Application of a Multisonic Model to an Urban Study, *Ibid* , USAEC Report BNL-15455
- G S Raynor, *An Isokinetic Sampler for Use on Light Aircraft Atmospheric Environment*, USAEC Report BNL-15374, November 1970
- L Newman, M Smith, J Forrest, W Tucker, and B Manowitz, *A Tracer Method Using Stable Isotopes of Sulfur*, American Nuclear Society Topical Meeting on Nuclear Methods in Environmental Research, University of Missouri, August 1971, USAEC Report BNL-16100
- B Manowitz and W E Mott, *Nuclear Techniques for Studying Environmental Pollutants*, Fourth UN International Conference on Peaceful Uses of Atomic Energy, Geneva, September 1971, USAEC Report BNL-15843

Estimated Date of Final Report

Continuing project

MEASUREMENT OF SULFUR DIOXIDE IN STACK GASES*

Objective of Work

The objective of the SO_2 stack-gas monitoring program is to develop and test an instrument for determining SO_2 in stack gases, based on a mercury substitution nucleonic detection principle. The current phase is directed toward the integration of an improved reaction cell (developed by Battelle Memorial Institute in Columbus, OH) into the gage and the testing of the instrument in the laboratory and in the field.

Progress and Results

In the system developed, SO_2 reacts stoichiometrically with insoluble mercuric oxide in a buffered

*R J Pfeifer, Industrial Nucleonics Corporation, Federal Systems Division, 650 Ackerman Road, Columbus, OH 43202, Contract AT(30 1) 3882

aqueous medium to produce a soluble sulfite mercury complex. The complex flows into a measurement cell where the mercury concentration is indicated by the absorption of low-energy x rays from Co-60. An improved cell was developed following initial field testing of a working model, which indicated several performance problems with the reaction cell. The new cell has been integrated into the previously developed working model, and preliminary laboratory testing indicates that both the response time and system reliability are improved.

Plan for Next Period

The working model will be extensively field tested, and efforts involving the cooperation of private industry as much as possible are envisioned for the near future. Instrument commercialization, in which instrument size, reliability, performance, and cost factors will be optimized, will be accomplished concurrently with the extended testing programs.

Reports Published

- B. Y. Cho, H. J. Evans, and L. B. Anderson, Radioisotope Methods of Sulfur Dioxide Measurement, *Trans Amer Nucl Soc*, 11: 55 (1968)
- B. Y. Cho, Measurement of Sulfur Dioxide in Stack Gases, *Studies of the Measurement of Sulfur Dioxide in Stack Gases*, USAEC Report TID-25137 (1968)
- R. J. Pfeifer and B. Y. Cho, Mercury Substitution—Nucleonic Detection Instrument for Sulfur Dioxide Measurement, *Analysis Instrumentation*, 7: 177 (1969)
- R. J. Pfeifer, B. Y. Cho, and O. L. Utt, Mercury Substitution—Nucleonic Detection Instrument for Sulfur Dioxide Measurement, *ISA Transactions*, 9(1): 9 (1970)
- R. J. Pfeifer, *Measurement of Sulfur Dioxide in Stack Gases: Demonstration Instrument Design, Fabrication, and Test*, USAEC Report NYO-3882-2 (1970)

Estimated Date of Final Report

June 1972

SNOW-MANAGEMENT RESEARCH*

Objective of Work

The purpose of the snow-management project is to assist industrial contractors in designing and testing a remotely operated telemetered version of the profiling

isotopic snow gage developed in this project (described on page 105 of the 1968 edition of *Research and Development Projects*, TID 24823).

Progress and Results

Performance specifications for a remotely operated, telemetered gage were developed and furnished to the scientists designing the gage at the Materials Testing Reactor site at Arco, ID.

A new gage was built and delivered to the Central Sierra Snow Laboratory at Donner Pass in the Sierra Nevada of California. The gage, described in detail in the next section of this publication, was intensively tested in round-the-clock operation during a part of the 1971 snow season. No major problems were encountered in operation of the equipment, and several opportunities were found to decrease operation time and thus data-gathering costs. A snow gage used in near by snow-hydrology studies was used for comparisons.

In January 1971, a small burst of rain — 0.2 in. — fell onto a deep snowpack and froze into an ice layer at about 40 in. depth. Two inches more rain fell, beginning about 8 hr later, and ponded above the impenetrable ice layer, melting the top 20 in. of snow and flowing over the snow surface to create rivulets. Ponded water was released from the snow in a very short time span. The entire sequence of events was easily followed with the gage. Other types of sensors in the mountains indicated the increase and sudden decrease in water at measurement sites. The cause of this and the effect on the snow and stream flow could not be found with any other type of equipment.

Plan for Next Period

During the snow year 1971-72, four telemetered snow gages will be installed — and operated automatically from one location — by Aerojet Nuclear Co. We will train the data-analysis crews for each gage-user group and will select sites for the snow gages.

Reports Published

None

Estimated Date of Final Report

June 1972

*J. L. Smith, Water Yield Improvement, Conifer Zone, Pacific Southwest Forest and Range Experiment Station, US Forest Service, P. O. Box 245, Berkeley, CA 94701, Contract AT(49-11)-2773. This project is jointly supported with the US Forest Service and the US Department of Agriculture.

Artist's sketch of the profiling isotopic snow gage.

TELEMETERED PROFILING ISOTOPIC SNOW GAGE*

Objective of Work

The major objectives of the telemetered snow gage project are to demonstrate the feasibility of a telemetered gage, to fabricate and install three additional gages at user selected sites, to operate all four gages as a telemetered network, and to effectively transfer the technology to users

Progress and Results

The gage consists of two parallel vertical tubes that extend from below ground level up through the snow surface (see figure). One tube contains a Cs 137 source and the other a scintillation gamma detector. As the tubes are lifted simultaneously in fixed steps, the gamma attenuation by the snow, and hence the snow density, is determined at various levels, yielding a density profile of the snow pack.

An engineered telemetered version of the gage was designed and fabricated and then installed in California in March 1971 at the Centia Sierra Snow Laboratory on Donner Pass. The base station was at Berkeley, 140 miles away. The telemetry data link was by leased telephone line. Data were sent to the base station at the end of each lift step. The field control unit that operates the gage under base-station control is designed to operate from -30 to $+70^{\circ}\text{F}$. It uses 15 W when operating and less than 2 W when on standby. The first gage was installed without difficulty and operated quite successfully.

With completion of tests at Donner Pass, three more gages were fabricated and were installed in Colorado, in Idaho, and in Oregon — in November for use during the 1971-72 snow season. They are being operated, together with the Donner Pass gage, as a network from a central base station at the National Reactor Testing Station in Idaho. After each run, snow density vs. snow height data are sent by telephone to teleprinters in each user agency's office. In normal operation, snow profiles for each site are taken automatically at night and the data stored for transmittal to the user agency upon request. However, the user can request a run at any time and there is the additional option of the user

running his own gage from his office via the central base station.

Plan for Next Period

Operation of the 4-gage network will be continued for the duration of the 1971-72 snow season. Detailed specifications and drawings of the snow gage will be prepared for use by commercial vendors, and user agencies will be assisted in operation and maintenance of gages for the 1972-73 snow season.

Reports Published

None

Estimated Date of Final Report

July 1972

ROLE OF PAPER MILL ADDITIVES AS POTENTIAL STREAM POLLUTANTS DEVELOPMENT OF NUCLEAR ANALYTICAL TECHNIQUES*

Objective of Work

The principal aim of this work is to determine how much of a specific additive is actually required by the fibers to fulfill a prescribed function and how much of it leaves the paper mill as a potential pollutant. The information obtained will, hopefully, decrease both pollution from paper mills and the paper industry's economic loss when unused additives leave the production lines in the waste.

Progress and Results

The major effort has been applied to the determination of in-plant process and effluent waters containing glucose and starch, which have very high potentialities as stream pollutants.

For the determination of glucose, alkalized water samples were treated with a mixture of a mercury salt and diatomaceous earth. The salt was reduced by the sugar to mercury, which was sorbed by the diatomaceous earth and determined by neutron activation. As an alternative method, a mercury salt containing

*P. D. Randolph and R. L. Heath, Aerojet Nuclear Co., National Reactor Testing Station, Idaho Falls, ID 83401, Contract AT(10-1) 1375. This project is jointly supported with the Soil Conservation Service, the National Weather Service, and the Bureau of Reclamation.

*R. M. Chatters, Radioisotopes and Radiations Laboratory, College of Engineering Research Division, Washington State University, Pullman, WA 99163, Contract AT(45-1) 2221, Task 3. This project is jointly sponsored with Washington State University and Northwest Pulp and Paper Association.

Hg-203 was used, thus obviating the reactor-activation step and shortening the analytical time to less than 1 hr for an individual sample. Each test involves less than 10^{-2} μ Ci of Hg-203.

Starch was determined by treating the effluent with enzymes to convert starch to glucose, which was determined as above.

A suitable tag for the common paper-making clay, kaolin, was found to be iridium as ammonium chloroiridate, which was reduced, by hot sodium formate solution, to free iridium or iridium oxide, which then bonded to the clay. Iridium was determined by neutron activation analysis.

At the request of a western paper mill, a lanthanum salt tracer method was developed for tracing an adhesive through the plant process lines. A lanthanum salt was added to the heated adhesive, which was then mixed with other stock, and samples from several places in the mill were analyzed by neutron activation analysis.

Samples for study have been collected in cooperating paper mills under normal conditions, and test materials used in the laboratory have been obtained from mill stocks or from the suppliers of paper mill additives.

Plan for Next Period

A detailed investigation will be made of sizings (other than starch), pigments, dyes, bleaching compounds, and coatings, and analytical techniques will be studied.

Reports Published

R. M. Chatters, G. S. Clifton, S. D. Slater, and J. E. Kimbrell, *The Role of Paper Mill Additives as Potential Stream Pollutants: Development of Nuclear Analytical Techniques*, June 1, 1970–June 1, 1971, USAEC Report RLO-2221-T3-1, December 1971.

Estimated Date of Final Report

Continuing project

DEVELOPMENT OF NUCLEAR ANALYTICAL TECHNIQUES FOR OIL-SLICK IDENTIFICATION*

Objective of Work

The main objective of the oil-slick identification work was to define the methodology and utility of neutron

activation analysis (NAA) for the identification of oil slicks.

Progress and Results

Marine oil slicks and potential sources of slicks were examined with respect to replicate (matched) and different (unmatched) samples, and the NAA data on them were used to define a statistical basis for interpreting a pair of samples as matched or unmatched. The effect of exposure and the preparation of slick and suspect source samples for analysis have been defined.

A difference-score (DS) technique was developed for quantitative sample comparisons, the observed concentration values for each element being codified so that the code scores have equal weight. In a pair comparison, the differences in code scores for each element in the two oils are summed, and a constant is added for each case in which a given element is found in only one sample of the pair. The DS scores of matched pairs follow a binomial distribution in which there is a 0.28% chance of exceeding $D = 8$ and a $<0.0001\%$ chance of exceeding $DS = 15$. Unmatched pairs have an average $DS = 54$, 0.5% chance of being ≤ 15 , and essentially zero chance of being ≤ 8 . Therefore, in the vast majority of cases, the trace element patterns of two oils indicate immediately whether the oils are matched or unmatched. In a very few cases where $DS = 9$ to 15 , the exact odds for each interpretation (which depend on the number of elements observed) may be computed to arrive at the most probable designation.

Plan for Next Period

Phase II of project completed

Reports Published

H. R. Lukens, D. Bryan, M. A. Hiatt, and H. L. Schlesinger, *Development of Nuclear Analytical Techniques for Oil-Slick Identification (Phase IIA)*, USAEC Report Gulf-RT-A-10684, June 1971.

Estimated Date of Final Report

Already issued (see above)

*H. R. Lukens, D. Bryan, M. A. Hiatt, and H. L. Schlesinger, Gulf Radiation Technology, Division of Gulf Energy and Environmental Systems, Inc., San Diego, CA 92112, Contract AT(04-3)-167.

RADIOISOTOPIC SEDIMENT TRACING (RIST) STUDY SYNOPSIS STUDY OF SEDIMENT TRANSPORT*

Objective of Work

The basic objective of the radioisotopic sand tracer (RIST) study is to develop data on littoral transport, with the major effort concentrated on a synoptic study of the mechanics and rate of sediment transport in the offshore, inshore, and beach zones and to develop a simplified survey system directly applicable to such studies as determining the rate of littoral drift and patterns of sediment movement in the vicinity of harbor- and shore-protection structures

Progress and Results

Efforts during the past year have been concentrated on the development of improved methods of environmental data collection to provide better correlation between tracer results and oceanographic conditions, and of theoretical models to determine the volume rate of littoral drift from tracer results

An instrumented sled for measuring currents in various water depths at various distances below the surface in several planes was constructed and tested. Other devices such as pressure transducers for wave measurement will be added at a future date. ORNL designed a bottom-elevation monitor using a 100-mCi Cs-137 source at a fixed distance below the detector. The monitor was recently tested at Point Mugu, CA, and elevation changes of 1 to 2 in. were detected over a period of a few hours at a 15-ft water depth.

A method is being investigated for using tracers in quantitative measurement of the littoral volume drift rate. Tracer is continuously injected in the surf zone. When an equilibrium concentration is attained at an appropriate distance downdrift from the injection site, the drift rate, Q , is estimated from $Q = I/c$, where I is the injection rate and c is the steady-state tracer concentration. Further details of this project are reported in the ORNL project which follows.

Plan for Next Period

A method of quantifying littoral drift will be finalized, and efforts will then be directed toward correlat-

ing the determined drift rates with various oceanographic conditions for particular areas and estimating long-term drift rates from an analysis of a time sequence of oceanographic conditions. Construction of the simplified data-collection system will be completed, and personnel will be trained by ORNL in its operation. Tests involving the unit as a total operating package will begin at particular sites where data are required for engineering purposes. Work will also begin on designing modifications to the RIST system for tracing water masses and ocean disposal of dredge spoils.

Reports Published

None

Estimated Date of Final Report

Continuing program

RADIOISOTOPIC SEDIMENT TRACING (RIST) STUDY FUNDAMENTAL STUDIES OF SAND TRANSPORT AND EQUIPMENT DESIGN*

Objective of Work

The objective of the radioisotopic sediment tracing work is to develop an underwater radiation survey system capable of operating in the surf zone, in deep water, and on the dry beach and to apply the system to coastal engineering problems involving sand transport and the determination of volume rates of sand transport.

Progress and Results

A PDP 8 computer was integrated with the radiation detection system and the range-positioning equipment. Data introduced to the computer were processed in the field to yield a display of the survey vessel track and the radiation intensity along the track. Formerly, data processing to determine the completeness of the survey required several weeks. The on-board plotting was demonstrated at Masonboro Inlet, NC, where a survey was made to determine the operation of a weir jetty that had been installed to produce a ship channel. A second demonstration was made at Point Mugu, CA. Real-time plots of the survey were displayed on an

*D. B. Duane, C. W. Judge, W. R. James, and P. G. Teleki, U.S. Army Corps of Engineers, Coastal Engineering Research Center (CERC), 5201 Little Falls Road, NW, Washington, DC 20016, Contract A1(49-11) 2988. This project is jointly sponsored with CERC and Oak Ridge National Laboratory (ORNL).

*F. N. Case, E. H. Acree, and H. R. Brashear, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, TN 37830, Contract W-7405-eng 26. This project is jointly supported with the Army, Navy, Air Force, and State of California.

oscillograph, and contour maps were prepared from the plots

A compact survey system was designed and fabricated for use by the U S Army Corps of Engineers in routine sediment tracing. The new system was designed with all the required functions of the preliminary system, but it is smaller and has less power demand. The new unit can be integrated with a PDP-8 computer to provide real-time data processing for display and can accept data from three different range finders, eliminating the need for a specific navigation system.

Measurement of the volume rate of sand transport is progressing through field tests and mathematical modeling. However, normal sand-injection techniques cannot be used for labeling the system because of the slow equilibrium distribution of the labeled particles, and, once equilibrium is reached, the starting point for a measurement has not been determinable. Further, the depth of the moving layer cannot be accurately measured. To overcome these problems, a plug-injector model was constructed, and experiments were designed to test the model. A device was fabricated for inserting a 12-in -dia by 10-in -deep plug of labeled sand into the ocean floor. The plug is sampled by coring, and data from radioisotope detection in the plug are used to test the mathematical model.

Plan for Next Period

Project to be terminated

Reports Published

F N Case, E H Acree, and H R Brashear, A Survey System for Use in Tracing Radionuclide-tagged Sediment in the Marine Environment, in *Nuclear Techniques in Environmental Pollutions*, Proc Symp Salzburg, Austria, Oct 26-30, 1970, pp 603-28, IAEA, Vienna, 1971 (STI/PUB/268)

Estimated Date of Final Report

FY 1973

EVALUATION OF TREATMENT PLANTS BY TRACER METHODS*

Objective of Work

The objective of this project is to study the application of tracer techniques to control of process conditions in multistage treatment plants for municipal and industrial wastes.

Progress and Results

In close cooperation with sewage-treatment plants in the Metropolitan Atlanta area, tracer tests have been run to study the performance of sedimentation basins, trickling filters, and sludge-treatment systems. Post-sampling activation analysis, using nonradioactive indium salts as tracers and neutron activation in the Georgia Tech Research Reactor as the method of analysis, has been shown to be a simple, effective, and environmentally acceptable procedure.

Improper operating conditions, such as short-circuiting and excessive recirculation, were shown to exist at some of the plants. Labeling of sludge by indium salts was found to be relatively simple, and the labeled compounds were chemically stable. Much of the information obtained was clearly unobtainable by other means, and the cost per test seemed reasonable.

Laboratory tests were conducted to explore alternative tracer elements and different methods of labeling solids and to establish background concentrations in the water and in suspended matter for several potentially usable trace elements, including cobalt, scandium, and manganese. Manganese was found to occur in fairly high concentrations in most local waters.

Methods were worked out for adequate sampling procedures and for semiautomatic activation analysis using a Ge(Li) gamma-ray spectrometer.

Plan for Next Period

An important part of the project constitutes the transfer of the technology to plant operators and to industry at large. This will involve demonstration tests over a larger geographical area as well as further work to minimize the cost of the test runs. Additional work will also be done on sludge driers and other aspects of sludge treatment to monitor the attainment of optimum operating conditions.

Reports Published

None

Estimated Date of Final Report

Continuing project

*G G Eichholz and T F Craft, Engineering Experiment Station and School of Nuclear Engineering, Georgia Institute of Technology, Atlanta, GA 30332, Contract AT(40-1) 4156

NUCLEAR TECHNIQUES FOR SEABED MINERAL EXPLORATION*

Objective of Work

The objective of this program is to design, develop, test, calibrate, and apply a nuclear probe to seabed mineral exploration

Progress and Results

During the past year, the feasibility of using a 2-mg Cf-252 neutron source and a Ge(Li) gamma-ray spectrometer for the study of elemental concentrations in terrestrial and seabed materials was shown. An investigation of the detection sensitivity of a remote probe system for some 40 elements in matrixes of varying density, water content, neutron absorption cross-section, and overburden was completed using a developed seabed-simulation facility capable of handling samples of up to 90 kg. In addition, a subcritical assembly for Cf-252 neutrons which provides a 40-fold neutron multiplication was developed and evaluated for use as a neutron radiation source for seabed mineral analysis. This subcritical multiplier mockup was composed of a homogeneous mixture of PuO_2 in polystyrene reflected with Plexiglas. Simulated ocean floor

mineral samples were irradiated with a 30- to 40-fold flux increase over that available without multiplication. The investigation demonstrated that a subcritical assembly could be used to enhance the thermal flux from a Cf-252 neutron source in the seabed probe design. These calibrations, standardizations, and model developments provided the necessary criterions to complete the prototype design of an in-situ ocean floor mineral analyzer. The system, which is being constructed, uses a 2-mg or a 50- μg Cf-252 source in the subcritical assembly with a multiplication factor of at least 40 (see figure). The gamma-ray detector consists of a solid cryogen-cooled Ge(Li) diode of about 15% efficiency with a resolution of less than 25 keV. The entire underwater diode package contains the diode, preamplifier, bias supply, and cryogenics, with a cooling time of about 48 hr. The probe assembly provides sample stabilization, marker buoy storage, and vertical and horizontal positioning and has underwater lights, Ge(Li) control systems, and an underwater TV camera to permit viewing of the sample area and placement of both neutron irradiator and Ge(Li) probes.

Plan for Next Period

Following final assembly of the Cf-252 seabed probe, it will be tested and calibrated in shallow ocean water — 50-ft depth — at the Battelle-Northwest oceanography site at Sequim, WA. The system will then be used to

*N. A. Wogman and R. W. Perkins, Battelle Pacific Northwest Laboratory, Richland, WA 99352, Contract AT(45-1)-1830

In-situ ocean floor mineral analyzer system.

determine the element concentrations in minerals at ocean depths approaching 300 ft.

Reports Published

R. W. Perkins, W. A. Haller, H. G. Rieck, L. A. Rancitelli, and N. A. Wogman, Cf-252 Neutron Activation for Remote Surface Analysis, in *Neutron Sources and Applications*, Proc. Amer. Nucl. Soc. National Topical Meeting, Augusta, GA, Apr. 19-21, 1971, Vol. 3, pp. III-20-29 [summarized in *Isotop. Radiat. Technol.*, 9(3)].

Estimated Date of Final Report

Continuing project.

RADIOISOTOPE DENSITY GAGE FOR THE DEEP-OCEAN SEDIMENT PROBE*

Objective of Work

A gage incorporating both transmission and scatter modes was designed and built for attachment to the Deep-Ocean Sediment Probe (DOSP). Collimation and discrimination are used to provide a narrow beam for accurate profiling.

Progress and Results

In laboratory tests, the scatter-section results indicated density changes. The probe is now being tested in the bay, having been attached to sections of free flooding pipe to permit operation from a boat in waters up to 25 ft depth. Under a coordinated program, cores and in-situ electrical resistivity and formation factor readings are being taken in the same locations as the density readings.

Plan for Next Period

The gage will be attached to the DOSP for further testing in the bay.

Reports Published

None.

Estimated Date of Final Report

January 1972

*V. C. Rose, 218 Crawford Hall, Department of Ocean Engineering, University of Rhode Island, Kingston, RI 02881, Contract AT(30-1)-4193. This project is jointly supported with the U.S. Navy Underwater Sound Laboratory.

RADIATION ANALYSIS AND CONTROL

Moessbauer spectroscopy of an 18th-century painting.

The Moessbauer spectrum shown was obtained by backscattering, as may be seen in the photograph.

The area analyzed is a yellow-brown color, and the spectrum is typical of natural iron ochre.

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Radiation Analysis and Control

The research and development objectives of the Radiation Analysis and Control program are (1) to provide improved technology for the detection and analysis of radioisotopes and nuclear radiation, and (2) to encourage and demonstrate the beneficial application of radioisotopes and nuclear technology to problems of national importance in such areas as consumer protection, quality control, public health and safety, natural resource management and law enforcement and national security.

Projects to develop instrumentation technology are directed toward establishing and upgrading basic capabilities or broadening the technological base for the use of radioisotopes or nuclear radiations from radioisotope sources. The efforts are usually related to multiple-use goals and are not necessarily associated with specific hardware or system requirements. Projects to establish the applications are closely related to specific requirements or problems in national security, industry, or aerospace.

Emphasis has continued on the application of nuclear technology to the problems of criminal justice and

forensic analysis. Close liaison is maintained with government agencies that have the responsibility and operational requirements in these areas. Special attention has been given to critical problems of industrial development such as improved methods for minerals exploration and the application of nuclear technology to the field of construction materials and to civil engineering. In general, the projects are carried through to an in-plant or field evaluation to clearly demonstrate their capabilities. At this point, those industries or organizations who are interested in producing equipment or applying the technology are given full access, on an impartial basis, to the data and the reports that result from the development effort.

Additionally, radioisotopes and nuclear technology may offer unique solutions to aerospace problems. Most contracts in this area are jointly supported with other government agencies that have the specific operational requirements.

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RESEARCH ON SINGLE-CRYSTAL SEMI-INSULATING CADMIUM TELLURIDE AS A GAMMA-RAY SPECTROMETER*

Objective of Work

The objective of this work is to develop a room-temperature gamma-ray spectrometer from single crystals of semi-insulating cadmium telluride (CdTe).

Progress and Results

The performance attained with CdTe was improved. Precipitates and an excess density of hole traps — the factors that limit resolution of devices — were analyzed. To overcome these obstacles, the Bridgman- and vapor-growth techniques were refined, and crystals are being grown from solution.

Semi-insulating CdTe gives good gamma response around room temperature. The first figure shows the room-temperature response of a 5-mm³ device to Cs-137 (662 keV) for a bias of 1000 V. The photopeak, Compton edge, and backscatter peak are at 95% charge-collection efficiency. The resolution is 30 keV (FWHM) and is limited by incomplete charge collection of holes. The second figure shows the response of the same device to a Co-57 source at 0°C for the same bias. The 136-keV line can be resolved clearly from the 122-keV line; the resolution is ~6 keV (FWHM). For the linear plot (top), generators were inserted at 150

keV with the bias off and at 175 keV with the bias on. The similarity of line broadenings indicates that leakage current does not degrade the resolution. Here, the resolution is limited primarily by the preamplifier noise and secondarily by incomplete charge collection. The log plot resolves the backscatter and escape peaks and the Pb x rays. Although spectrums from vapor- and Bridgman-growth modes are comparable, the best results achieved to date were obtained with Bridgman-grown material. However, much more Bridgman-grown material was sampled, and Bridgman growth received considerably greater emphasis. As the size of the device increases, the quality of the photopeak degrades; with a 1-cm³ device at 40% charge-collection efficiency, a photopeak could barely be resolved. The loss of

*K. Zanio, N. Kyle, and W. Akutagawa, Hughes Research Laboratories, 3011 Malibu Canyon Rd., Malibu, CA 90265, Contract AT(04-3)-549.

Room-temperature spectral response of a 5-mm³ CdTe detector (linear plot) to Cs-137.

Spectral response of a 5-mm³ CdTe detector at 0°C to Co-57. Generator markers are inserted in the linear plot (top) at 150 keV, with the bias on, and at 175 keV, with the bias off. In the log plot (bottom), the Compton peak, Pb x rays from the collimator, backscatter peak, escape peak, and 136-keV photons are apparent.

resolution is due primarily to the presence of traps and precipitates with increase in device volume their effectiveness in degrading the resolution increases

By analysis of the transient response† of devices to ionizing radiation the density and depth of trapping levels have been determined. A shallow electron trap with 0.06-eV activation energy and $\sim 10^{17}$ centers/cm³ density was found and may be due to indium. A deeper trap with 0.6 eV activation energy has again been measured. Only the deep traps limit the charge transport, because rapid thermal emission occurs for electrons that are trapped at shallow levels. In some cases, material has been analyzed that shows no deep electron trapping. Because the mobility of holes (70 cm²/V sec) is less than that of electrons (1000 cm²/V sec), the charge transport of holes is generally poorer than that of electrons. An analysis of hole transport showed a level at 0.13 eV with a density of $\sim 5 \times 10^{16}$ centers/cm³. At room temperature and at moderate electric fields, the charge transport is inhibited primarily by the deeper-hole traps. To learn more about these deeper levels, improved mobility-trapping-time products ($\mu\tau^+$) must be attained.

Precipitates also inhibit both electron and hole transport. Infrared microscopy of material after it was subjected to thermal-annealing cycles indicates that the size of precipitates can be reduced to less than the resolving power of the microscope (1 μ m) and that, most likely, the precipitates are due to deviations from stoichiometry and not to foreign atoms. Pulse-height spectrums of alpha particles and of gamma rays are being used to analyze submicron precipitates and gross fluctuations in trap densities.

The resolution and the size of the detectors will be improved further when the concentrations of hole traps and precipitates are reduced. The hole traps are either acceptor native defects or foreign atoms introduced into the lattice during crystal growth. Several methods are in process to reduce the foreign-atom content. In a separate research program, multiple distillation of synthesized CdTe is being undertaken. Crystals are also being grown from the vapor and from solution to reduce not only the foreign-atom content but also the density of native acceptors. Also to minimize precipitation, material is being vapor grown within the phase field of CdTe. Solution growth is being undertaken at the tellurium rich liquidus below 500°C under conditions that avoid precipitation.

†Collaboration with A. Alberigi Quaranta, M. Martini, G. Ottaviani, and C. Canali, University of Modena, Italy, gave the results on transient response presented here.

Plan for Next Period

The evaluation of the spectral response of semi-insulating CdTe to ionizing radiation will be continued, and those material imperfections that degrade resolution will be analyzed further. More importantly, effort will be made to lower the foreign atom and native-defect concentrations and to reduce precipitation by refining further the crystal growth processes.

Reports Published

- N. R. Kyle, Growth of Semi-Insulating Cadmium Telluride, *J. Electrochem. Soc.* 118(11) 1790-7 (1971)
- C. Canali, M. Martini, G. Ottaviani, and K. Zanio, Time of Flight Measurement of the Differential Negative Mobility in CdTe, *Phys. Lett.* 33A(4) 241-2 (1970)
- A. Alberigi Quaranta, C. Canali, G. Ottaviani, and K. R. Zanio, Electron-Hole Pair Ionization Energy in CdTe Between 85°K and 350°K, *Nuovo Cimento Lett.* 4(19) 908-10 (1970)
- C. Canali, M. Martini, G. Ottaviani, and K. R. Zanio, Transport Properties of CdTe, *Phys. Rev. B*, 4(2) 422-31 (1971)
- C. Canali, M. Martini, G. Ottaviani, and K. R. Zanio, Dependence of Electron Trapping Time on the Electric Field in Semi-Insulating CdTe, *Solid State Commun.* 9(2) 163-6 (1971)
- W. M. Akutagawa and K. Zanio, Vapor Growth of CdTe, *J. Cryst. Growth*, 11 1 (1971)
- C. Canali, M. Martini, G. Ottaviani, A. Alberigi Quaranta, and K. Zanio, Characterization of High Resistivity CdTe for Gamma-Ray Detectors, *Nucl. Instrum. Methods*, 96(4) 561-71 (1971)
- C. Canali, G. Ottaviani, M. Martini, and K. Zanio, Hole Drift Velocity in Semi-Insulating CdTe, *Appl. Phys. Lett.* 19(3) 51-3 (1971)

Estimated Date of Final Report

Continuing project

CADMIUM TELLURIDE DETECTORS*

Objective of Work

The objective of this project is to grow single crystals of cadmium telluride (CdTe), by the traveling-heater

*F. Wald and R. O. Bell, Tyco Corporate Technology Center, 16 Hickory Dr., Waltham, MA 02154. Contract AT(30-1) 4202.

method (THM) of solution growth, for use in gamma-ray semiconductor detectors that can be operated at room temperature or above

Progress and Results

By the THM, primarily with Te as the solvent, CdTe is being grown which has the excellent homogeneity, low trap density, and low carrier concentration necessary for nuclear-radiation detectors. The primary effort has been in three areas

- 1 **Temperature control.** To successfully grow large single crystals of CdTe, the temperature distribution must be controlled carefully. The heat flow must be adjusted, along with the length of the solvent zone, to produce a flat or slightly convex solvent-solute interface. Detailed calculations of the thermal distribution show that at least one reasonably flat isotherm exists in the cylindrical geometry used in the THM furnace (first figure). Experimental modifications of the growth apparatus, such as the use of a water-cooled section below the heater and rotation (~ 2 rpm) of the charged ampule to ensure uniform radial temperature distribution, have shown promise in achieving the goal of single-crystal growth.
- 2 **Control of native defects.** After proper purification to remove electrically active impurities, a primary necessity of any semiconductor-growth technique is the control of native defects to minimize thermally generated carriers and traps. In the CdTe system, the electrical properties can be controlled by adding Group III elements (e.g., In) and Group VII elements (e.g., Cl). For example, large amounts of indium (up to 10^{20} atoms/cm³) can be added to CdTe without drastic reduction of the mobility trapping-time product ($\mu\tau$) while maintaining a low carrier concentration ($< 5 \times 10^9$ atoms/cm³). A tentative explanation of this behavior has been developed by considering the doping in terms of the CdTe-In₂Te₃ solid-solution system.
- 3 **Hall-effect measurements.** It has been determined that dc Hall-effect measurements can be performed successfully on high resistivity (10^6 to 10^7 Ω cm) CdTe, although great care must be taken to isolate and shield the leads and sample. A completely guarded and Teflon-insulated measurement circuit is used. The refinement of techniques for making ohmic low-resistivity contacts to both p- and n-type samples also contributes to the successful measurement. Results to date are in good agreement with parameters derived from nuclear-detector measure-

Positions of selected isothermal lines as a function of ingot radius and position along the ingot.

ments. The second figure shows a pulse height spectrum of a CdTe detector.

Plan for Next Period

Effort will be made to improve the fabrication of CdTe detectors, to study doping, including the verification of present models, to increase crystallinity by seeding and by better temperature distributions, and to make detailed electrical, optical, and nuclear measurements on CdTe.

Semilogarithmic plot of the pulse-height spectrum of a CdTe detector with a Cs-137 and with a Co-60 source.

Reports Published

R. O. Bell, Calculation of Gamma-Ray Pulse Height Spectrum in a Semiconductor Detector in the Presence of Charge Carrier Trapping, *Nucl. Instrum. Methods*, 93(2): 341-8 (1971).

Estimated Date of Final Report

Continuing project.

EVALUATION OF CADMIUM TELLURIDE FOR RADIATION DETECTORS*

Objective of Work

The objective of this project is to evaluate presently grown cadmium telluride (CdTe) for use as radiation

detectors. This is part of a continuing study to find a high-Z semiconductor material that will operate satisfactorily at room temperature and above as a counter and/or high-resolution spectrometer.

Progress and Results

Cadmium telluride grown according to current state-of-the-art procedures has been evaluated by nuclear techniques for use as radiation detectors. The material was supplied by General Electric Co., Hughes Research Laboratories, and Tyco Laboratories, Inc. The material had mobility-trapping-time products ($\mu\tau^+$) of $(2 \text{ to } 8) \times 10^{-4} \text{ cm}^2/\text{V}$ for electrons and $(2 \text{ to } 5) \times$

*J. M. Palms, Department of Physics, Emory University, Atlanta, GA 30322, Contract AT(40-1)-4146

Low-energy gamma-ray spectrum of Co-57.

Detector CdTe, n-type
Al surface-barrier
Dia, 2 mm
Thickness, 7 mm
Bias -400 V
Preamplifier Ortec 118A
Amplifier Tennelec TC-200
Temperature 50°C

10^{-5} cm²/V for holes. Detectors usable as alpha, gamma, and x-ray spectrometers were fabricated from high-resistivity (semi-insulating) and low-resistivity n-type CdTe (vapor-grown and grown from the melt)

These devices were aluminum and gold surface-barrier detectors having 0.8- to 1.5-mm thicknesses and 2- to 25-mm² areas. Their pulse-height responses to low-energy gamma and x rays were studied as a function of detector bias (0 to 1000 V), temperature (-75° to +52°C), and amplifier time constants (0.25, 1, and 4 μ sec). The pulse-height spectrums of all devices were affected by the presence of trapping sites and material inhomogeneities. All devices showed evidence of the mechanisms of trapping and detrapping of charge carriers; however, the total counting efficiency for most devices was high (>85%). The figures show some typical x-ray spectrums taken with a low-resistivity CdTe detector. The first figure gives the gamma-ray and x-ray spectrums from Co-57. The energy resolution (FWHM) of the 122-keV line is 10.7 keV, and the pulse-peak resolution at the same energy is 8.6 keV. The K x-ray escape peaks of Cd and Te, which lie ~25 keV below the 122-keV line, are clearly evident. The peak at 67

keV is due to fluorescence K x rays from a piece of gold used as a pressure contact to the aluminum electrode. The peak at ~22 to 24 keV is due to fluorescence K x rays from indium, which is used as a backing pad for the detector. The second figure shows the low-energy gamma and x-ray spectrums from Am-241. Even though the electronic resolution was not optimized for the spectrums shown, these spectrums demonstrate clearly that material suitable for x-ray spectrometers is now being grown. However, considerable improvement in detector performance is expected if material can be grown that has reduced trapping-site concentration as well as more-homogeneous structure. The last property is particularly important if larger detectors suitable for higher-energy gamma-ray spectrometry are to be fabricated.

Plan for Next Period

State-of-the-art material will be evaluated continuously by nuclear techniques. This work should lead to an understanding of the correlations between material

X-ray spectrum of Am-241. (Conditions same as for first figure)

characteristics and crystal-growing techniques. In addition, to obtain a more complete material characterization, with emphasis on understanding the trapping sites, electron paramagnetic resonance and far-infrared Fourier spectroscopy will be used to determine the location, structure, and population density of the trapping sites, as well as the free-carrier concentration in the material. Along with these measurements, radiation detectors will be continuously fabricated and evaluated.

Reports Published

J. M. Palms, Newer Developments in Detector Design and Materials, in *Semiconductor Detectors in the Future of Nuclear Medicine*, P. B. Hoffer, R. N. Beck, and A. Gottschalk, Eds., Society of Nuclear Medicine, 1971.

Estimated Date of Final Report

Continuing project, annual report will be submitted in early 1972.

LOW-NOISE FIELD-EFFECT TRANSISTORS*

Objective of Work

The objective of this work is to develop low-noise field-effect transistors (FET's) particularly suited for the amplification of signals from radiation detectors.

Progress and Results

The specific problem of interest is the noise generated by "lossy" dielectrics. The source of this noise is indicated by the shaded area in the equivalent diagram of radiation detector charge amplifier combination in the figure. The noise due to dielectric losses is generated in the insulating materials used for FET encapsulation. This noise source was obscure until observed and analyzed quantitatively. It is not inherent to the amplification mechanism, it represents a limit in noise

and therefore in energy resolution — that could be achieved with an otherwise ideal noiseless FET. The properties of this noise are summarized here, and some relations in terms of easily measurable parameters are given.

A lossy dielectric is shown as source 4 in the figure. If a dielectric with low dielectric losses is used, its dissipation factor is a constant

$$D = \frac{G(\omega)}{\omega C_d} \quad (1)$$

where $G(\omega)$ and C_d are the loss conductance and the capacitance of the dielectric as measured on an impedance bridge at an angular frequency ω . According to the dissipation-fluctuation theorem, the conductance

*V. Radeka, Brookhaven National Laboratory, Upton, NY 11973, Contract AT(30-1)-16.

Noise sources in the charge amplifier and their equivalent noise charge (ENC) as a function of filter time constant, τ , and input capacitance, C_{in} [Source 1 (FET channel noise) is of principal interest, source 2 (dielectric noise) sometimes exceeds and masks other sources]

$G(\omega)$ generates Johnson noise (current) at the frequency ω ,

$$\overline{i^2(\omega)} = 4 kT G(\omega) d\omega = 4 kT (DC_d) \omega d\omega \quad (2)$$

Where $k = 1.38 \times 10^{-23}$ joules/°K, $\omega = 2\pi f$, and T is the temperature of the dielectric. This noise current is converted into a noise voltage on the input capacitance,

$$\overline{v^2(\omega)} = \frac{\overline{i^2(\omega)}}{\omega^2 C_{in}^2} = 4 kT \frac{G(\omega)}{\omega^2 C_{in}^2} d\omega = 4 kT \frac{DC_d}{\omega} \frac{d\omega}{C_{in}^2} \quad (3)$$

Thus, the lossy dielectric generates an $1/|f|$ noise-current spectrum in parallel with the input, which integrated on the input capacitance becomes $1/|f|$ noise. The equivalent noise charge due to this noise can be calculated conventionally by integrating the noise voltage at the output of the filter with respect to frequency and comparing it with the signal output. One obtains for the equivalent noise charge,

$$(ENC_f)^2 = \frac{\epsilon^2}{\pi} kT (DC_d), \quad (4)$$

where $\epsilon = 2.718$

The most interesting characteristic of this noise source is that its contribution is independent of both the input capacitance and the filter time constant. Also, it depends little on the type of the filter.

Two numerical relations useful for evaluating various dielectrics and FET packages follow from Eq 4. In the first, the equivalent noise charge is expressed in root-mean-square electron charges as a function of dielectric loss factor D and sample capacitance C_d ,

$$ENC_f \approx 6 \times 10^8 \left(\frac{T}{295} \right)^{1/2} (DC_d)^{1/2} \quad [\text{rms } e] \quad (5)$$

In cases where the dielectric and/or its capacitance are not known, it is more convenient to use the loss conductance as measured on an impedance bridge at an arbitrary frequency f_o in the range of interest. By substituting Eq 1 and $\omega = 2\pi f$ in Eq 5, one obtains

$$ENC_f = 2.44 \times 10^8 \left(\frac{T}{295} \right)^{1/2} \left[\frac{G(f_o)}{f_o} \right]^{1/2} \quad [\text{rms } e] \quad (6)$$

The dielectric noise is an unnecessary component of the amplifier noise and can exceed the channel noise of the FET. The channel noise is inherent in the amplification mechanism, and its reduction is the main target in this project. Thus, any proposed FET package should first be examined for dielectric noise. The effect of dielectric noise on energy resolution for several different types of FET packages is given in the table. It is essential to use materials with low dielectric losses, such as beryllium oxide and high-purity alumina. Aluminas

Noise of Field Effect Transistors Arising in Dielectric Supports
($T = 295^\circ\text{K}$)

Type of package and materials	Loss conductance $G(f_0)$ at $f_0 = 10^4$ Hz nmhos	Equivalent noise charge ENC_f rms e	Lower limit for noise line width FWHM, eV (Si-equivalent)
TO 5, epoxy	3.19	138	1.2×10^3
TO 5, glass metal	0.28	41	3.6×10^2
BEO	0.025	12	1.04×10^2
Alumina, 995 Al_2O_3	0.02	11	0.96×10^2

are easier to work with in production. The test samples of new FET's are being mounted in an alumina package. Once the material is found to be satisfactory, the package geometry should be optimized so that the gate capacitance is minimized.

Plan for Next Period

New devices with a smaller gate-channel geometry will be evaluated, they were developed with Texas Instruments, Inc., as a part of this program.

Reports Published

- V Radeka, State of the Art of Low Noise Amplifiers for Semiconductor Radiation Detectors, in *International Symposium on Nuclear Electronics*, Vol. I, pp 46-1 46-28, Versailles, 1968
- V Radeka, $1/f$ Noise in Physical Measurements, *IEEE Trans Nucl Sci*, NS-16(5) 17-35 (1969)

Estimated Date of Final Report

Continuing project

NEW RESEARCH APPLICATIONS OF ČERENKOV RADIATION*

Objective of Work

The objective of this project is to develop and evaluate new applications of Čerenkov radiation. Potential areas for study include fission-product analysis, environmental radioactivity, biomedical assay, and special counting systems.

*H. H. Ross, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, TN 37830, Contract W 7405-eng-26

Progress and Results

Dual-isotope experiments make up an important segment of biomedical tracer applications. A study was completed which shows that the Čerenkov counting technique can be used to assay two radionuclides simultaneously. The technique is rapid, precise, and inexpensive. Also, a simple computer program was developed that models the Čerenkov system and that can be used both to establish the optimum operating conditions of the counting system and to estimate the error in the assay measurement. A description of this work is being prepared for publication.

The first part of a program to develop Čerenkov techniques for monitoring radionuclides in the environment was completed. The study illustrates the very high sensitivity attainable for aqueous environments; typical results show that many radionuclides in concentrations < 2 pCi/liter can be detected. This sensitivity compares favorably with the sensitivities of other slower and more expensive low-level techniques.

Plan for Next Period

Project terminated

Reports Published

- H. H. Ross, Performance Parameters of Selected Wave-shifting Compounds for Čerenkov Counting, in *Organic Scintillators and Liquid Scintillation Counting*, pp 757-69, D. L. Horrocks and C.-T. Peng, Eds., Academic Press, New York, NY, 1971

Estimated Date of Final Report

All reports are or will be published in the open literature.

EVALUATION OF THE 200-mCi IRON-55 X-RAY DIFFRACTION SOURCE IN FOCUSING AND DEBYE-SCHERRER ANALYTICAL APPLICATIONS*

Objective of Work

The objective of this work is to evaluate the feasibility of using a 200-mCi Fe-55 source for x-ray analytical powder diffraction. Both Debye-Scherrer and focusing back-reflection camera geometries are to be studied. In each case, the exposure times required to obtain well-resolved x-ray diffraction patterns will be evaluated critically. The major advantages of an isotopic x-ray source for diffraction are that no power and no cooling are required and the source is very small, lightweight, and stable. The major disadvantage is the relatively low intensity of the x rays available.

Progress and Results

Work continued with the 2-in.-dia Debye-Scherrer diffraction camera and the Fe-55 x-ray source. Because of self-absorption and natural decay, the effective output of the source was only ~110 mCi. A new collimator and beam stop were designed and constructed in an attempt to reduce the amount of scattering inside the camera chamber. The collimator has a 0.5- by 10.0-mm entrance slit. A second slit of the same dimensions defines the x-ray beam. A final scatter slit, which does not intersect the main beam, captures a

large portion of the radiation scattered by the first two slits. With the new collimator and beam stop, the background scattering in the diffraction patterns has been reduced to an acceptable level. Usually, LiF powder-diffraction patterns are obtained in 20 to 30 hr, and the line width of the reflection from the (200) plane is ~0.7 deg θ (first figure). Since the active area of the source is 2 by 10 mm, only 25% (28 mCi) of the radioactivity is actually used with this collimator. Use of a wider entrance slit would adversely affect the resolution.

To improve the useful intensity, the 2-mm-wide source was mounted obliquely behind the 0.5-mm entrance slit. The mounting angle of 14.5 deg allowed radiation from the entire source to pass through the entrance slit. In this way, exposure times were reduced by ~1/3 without loss of resolution.

A 2-in.-dia focusing back-reflection camera was designed and constructed. The source is mounted on the circumference of the camera, a set of apertures in front of it defines the beam. The powder-diffraction sample is placed directly opposite the source on a 30-deg arc of the camera, which can record reflections in the θ range 55 to 73 deg. With this camera, a series of exposures was made using several apertures and powder-diffraction samples. To avoid loss of resolution, only part of the active area of the source could be used.

With a 0.25- by 3.7-mm entrance slit, diffraction patterns of LiF powder could be obtained in 8 hr (second figure). With this slit, only 5.7% of the active area of the source (~6 mCi) was being used. The line width was usually ~0.5 deg θ . The camera was equipped with a motor to continuously oscillate the

*L. E. Preuss, Edsel B. Ford Institute for Medical Research, 2799 West Grand Boulevard, Detroit, MI 48202, Contract AT(11-1)-2063.

Densitometer trace of the x-ray diffraction pattern obtained from LiF powder in the Debye-Scherrer camera in 20 hr. [From the extreme edge to the center of the trace, the five prominent peaks correspond to reflections from the (222), (311), (220), (200) and (111) planes.]

Densitometer trace of the x-ray diffraction pattern obtained from LiF powder in the back-reflection camera in 8 hr. [The peak near the edge of the film is from the K_{α} reflection on the (311) plane, the other peak is from that on the (222) plane. The asymmetry of the pattern is due to anomalous scattering from the aperture.]

sample through a few degrees. With the motor in operation, exposure times could be reduced by about one-half.

Plan for Next Period

This project has been completed.

Reports Published

- W. S. Toothacker and L. E. Preuss, Developments in Analytical Diffraction Using Isotopic X-Ray Sources, in *Advances in X-Ray Analysis* Vol 14, pp 139-45, C. S. Barrett, J. B. Newkirk, and C. R. Rudd, Eds., Plenum Press, New York, NY, 1971.
- L. E. Preuss and W. S. Toothacker, X-ray Diffraction Using Fe-55 Sources, *Nature Phys Sci*, 231(18) 19-20 (1971).

Estimated Date of Final Report

December 1971

GAMMA-RAY SPECTROMETRY OF NEUTRON-DEFICIENT NUCLIDES*

Objective of Work

The major objectives of this program continue to be the generation of data about neutron-deficient nuclides

produced by particle accelerators, the development of instrumentation and techniques for a variety of applications of x-ray and gamma-ray spectrometry, and the study of techniques that use x-ray, gamma-ray, and isotopic-neutron sources. The basic data consist of gamma-ray spectrums, precision energies, intensities, and branching ratios for the gamma rays emitted in the decay of radionuclides. Such data are essential to nuclide identification, use of the nuclides as standards, and both industrial and biomedical applications of tracers.**

In view of the broad applications developing for high-resolution x-ray and gamma-ray spectroscopy, considerable emphasis is being placed on the establishment of usable criteria for testing and calibrating equipment used in these techniques and on the continued development of on-line computer systems and software for automated analysis of pulse-amplitude spectrums. Current emphasis also includes the evaluation of geometries for containment of Cf-252 isotopic neutron

*R. L. Heath, J. E. Cline, and R. J. Gehrke, National Reactor Testing Station, Aerojet Nuclear Co., Idaho Falls, ID 83401, Contract AT(10-1)-1375.

**These data are published in the *Gamma Ray Spectrum Catalogue* and appear tabulated in the *Handbook of Chemistry and Physics*.

sources for both prompt and conventional activation analysis

Progress and Results

Gamma-ray spectrums, energies, and intensities were obtained for >30 radionuclides produced by reactions such as (γ, xn, yp) , (p, xn, yp) , or (n, xn, yp) on the 100-MeV electron linear accelerator at the Lawrence Livermore Laboratory and the 600-MeV synchrocyclotron at the SREL † These data will be included in the *Ge(Li) Gamma-Ray Spectrum Catalogue* The precise measurement of energies of gamma rays for use as energy-calibration standards has been extended up to 3 MeV, thus providing a variety of well-established reference lines for laboratory use To obtain precise relative and absolute intensity measurements, techniques have been developed and documented for the analysis of pulse-amplitude spectrums obtained with Ge(Li) and Si(Li) detectors This work has included evaluation of commercial amplifiers, analog-to-digital converters, and associated electronics used in high-resolution x-ray and gamma-ray spectrometers The results will be published, and the criterions for establishing meaningful performance specifications for such equipment will be made available to vendors and experimenters

Development of techniques for automated data analysis of x-ray and gamma ray spectrums has continued Automated techniques for peak location, energy determination, and nuclide identification have been developed for small on-line computer data systems for laboratory use The first figure is a block diagram of a computer data system for the measurement and analysis of data for application in x-ray fluorescence This system, now in routine use, permits the complete automatic analysis and elemental identification of materials present in unknown samples The use of interactive techniques, including real-time data display on an oscilloscope screen, affords the laboratory analyst much flexibility in analyzing experimental data The second figure illustrates the real-time analysis of data, a segment of a radioisotopic x-ray fluorescence spectrum is shown The screen display gives the elemental identification of the x-ray line (by the arrow), the energy of the line, and the channel number

Significant progress was made in applying x-ray fluorescence to the measurement of manganese in carbon steels With this technique, hot ingots of steel in rolling-mill operations can be characterized

†Space Radiation Effects Laboratory, Virginia Polytechnic Institute, 11970 Jefferson Ave, Newport News, VA 23600

Block diagram of automated x-ray fluorescence analyzer.

Graphic display of automated analysis of x-ray spectrums.

The availability of Cf-252 isotopic neutron sources at reasonable cost has greatly expanded interest in the possible industrial-laboratory and field use of neutron techniques Extensive experiments have been made to establish recommendations for optimizing design geometries for both internal-sample and external-beam arrangements of these neutron sources The availability of this information and the experimental verification of elemental sensitivities for both prompt and conventional activation techniques that use these neutron sources should be of value in assessing the potential of the activation technique in this mode

Plan for Next Period

The first edition of *Ge(Li) Gamma-Ray Spectrum Catalogue* will be published in February 1972

Reports Published

- R J Gehrke, J E Cline, and R L Heath, Determination of Relative Photopeak Efficiency and System Linearity for Ge(Li) Gamma-Ray Spectrometers, *Nucl Instrum Methods*, 91 349-56 (1971)
- R J Gehrke and R A Lokken, Efficiency Calibration of a Low-Energy Si(Li) Photon Spectrometer, *Trans Amer Nucl Soc* , 14(1) 126-7 (1971)
- R L Heath, Table of the Isotopes, *Handbook of Chemistry and Physics*, 52nd Ed , Chemical Rubber Co , Cleveland, OH, 1971
- J E Cline, *Gamma Rays Emitted by the Fissionable Nucleides and Associated Isotopes*, USAEC Report IN-1448, December 1970
- J E Cline, P D Randolph, E L Murri, M J Laird, and R L Heath, Elemental Determinations in Idaho Wildlife Samples, *J Idaho Acad Sci* , Special Issue No 2 28 (Aug 1, 1971)
- R C Greenwood, Prompt-Neutron Activation Analysis with a Cf-252 Source, *Trans Amer Nucl Soc* , 14(2) 932 (1971)

Estimated Date of Final Report

Continuing project

APPLICATION OF NUCLEAR TECHNOLOGY TO ART-IDENTIFICATION PROBLEMS*

Objective of Work

A number of nuclear techniques offer promise in solving problems of art identification, including the objective detection of forgeries and the objective characterization of genuine works of art. The purpose of this project is to develop such techniques for use by museum authorities, appropriate government agencies such as the Internal Revenue Service, or interested private individuals.

Progress and Results

By methods developed earlier in the history of the project, neutron activation analysis (NAA) was shown to be usable for distinguishing among artists on the basis of the measured trace-element content of the lead white used in their works. The procedure necessarily involves the separation of the pigment and its impurities from extraneous material also present in the usual work of art. Data analysis based on the establishment of probable ranges of trace-element content is used to calculate the overall probability that a questioned work belongs to a particular source.

Neutron activation analysis was also used to help determine the commonality of sources of various terra-cotta statuary, as well as various bronze and brass objects, at the National Gallery of Art.

The technique of identifying man-made C-14 in linseed oil as a means to detect forgeries of oil paintings produced since the 1950's was extended to other artists' materials such as wood and paper. Methods for the simple but efficient conversion of such materials into a form suitable for liquid scintillation counting were explored. A new method, that of dissolving sample-derived acetylene in a toluene-base liquid scintillation solution, was developed. This method shows great promise in its ability to handle samples of intermediate size (e.g., 1 to 2 g of paper).

The use of the Moessbauer effect to analyze works of art was explored. A variety of frequently used artists' pigments contain iron compounds. There is, at present, no satisfactory way to distinguish among ochres, siennas, and various red, black, and yellow synthetic compounds and mixtures thereof. In a series of measurements (by transmission) performed on samples of pigment from a variety of sources, it was demonstrated that (1) meaningful classification can be made, (2) the composition of mixtures of various oxides can be determined semiquantitatively, (3) particle sizes can sometimes be determined on the basis of observation of superparamagnetism, and (4) natural materials can often be distinguished from synthetic. It was shown that, with a backscattering configuration, such measurements can be made in situ on actual paintings (see figure at start of this section), from which the same information as described above can be derived without sampling or harming the work in any way.

It was also shown that the temperature at which terra-cotta, or alleged terra-cotta, statuary had been fired can be estimated. This information may help with the authentication of a number of questionable works. The application of Moessbauer-effect spectrometry to other iron-bearing pigments, such as green earth and

*B. Keisch, Carnegie-Mellon University, 4400 Fifth Ave., Pittsburgh, PA 15213, Contract AT(30-1)-3953. This project is jointly supported with the National Gallery of Art, Washington, DC 20565.

Prussian blue, was also explored as a means to determine the composition (for the former) or the mechanism involved in the process of fading (for the latter)

Plan for Next Period

Since the exploratory and demonstration phases of this research project are essentially completed, the U S Atomic Energy Commission's support of this work will be terminated at the conclusion of the current contract year. However, because of the successful nature of the work accomplished, much of the work will be continued by personnel of the National Gallery of Art Research Project. This work will include the gathering of large quantities of data to make the application of these various techniques into a practical and useful methodology.

Reports Published

- B Keisch, *Application of Nuclear Technology to Art Identification Problems, Third Annual Report, February 1, 1970–February 28, 1971*, USAEC Report NYO-3953-3, September 1971
- B Keisch, *X-Ray Diffraction and the Composition of Lead White, Studies in the History of Art*, National Gallery of Art, Washington, DC, 1970

Estimated Date of Final Report

March 1972

STUDY OF TOOTH-ENAMEL DENSITY*

Objective of Work

The objective of this project is to apply the avalanche-type beta-particle detector, developed under a previous USAEC contract with the General Electric Co., to the problem of accurately determining the density of tooth enamels in vivo. Accurate density measurements are needed to study tooth-density differences as related to the onset of dental caries and to provide a means of measuring the rate of etching and solubility of tooth enamels in various solutions under in-vitro conditions. It was proposed to study tooth-enamel density in porous regions created during tooth formation, in regions demineralized due to dental decay, in areas of

increased mineralization due to aging, and in teeth susceptible to acid dissolution and teeth resistant to dissolution.

Progress and Results

The detector was obtained to measure tooth density. The instrument is unique in that both the radiation source and the backscatter detector are located in the probe head. The probe was constructed initially with Tl-204 (20 μ Ci), brass cones were placed over the probe head to hold the sample at a fixed distance from the radiation source. To eliminate abnormal backscatter obtained with the brass cones, they were replaced with five Lucite cones. However, count rates produced from crystalline material by the radiation source were too low to be useful. To increase count rates, the radiation source was changed to Pm-147 (5 mCi). The probe again gave mechanical problems. The main tunnel diode was replaced but operated for only a few minutes thereafter. The control systems ceased to function and were replaced.

The on-going work consists in the following:

- 1 The construction of a stabilizer–holder for the probe prior to its assembly at a determined optimum source–specimen distance. A graduated microscopical stage, designed for precise calibrated travel, is being incorporated into the system to allow exact positioning and adjusting of the probe–specimen for obtaining desired parameters.
- 2 The use of the probe (minus the radiation source in the probe head) as a tool to monitor the uptake of F-18 applied topically to the surfaces of extracted teeth. Experiments are being designed to show the relation between the observed F-18 and the total F, measured with an electron microprobe, in the sample teeth.

Plan for Next Period

The contract has been terminated. However, the contractor proposes to continue the project, at his own expense, in order to obtain an effective approach to determining differences in tooth-enamel densities, a measurement that may yet be possible when the new radiation source is properly positioned in the probe head. Some of the recent laboratory studies, exclusive of the beta-probe project, indicate that the density of the enamel surfaces is related to their solubility. If this is so and if such critical densities can be measured with the probe, teeth vulnerable to dental decay could be detected *before* cavities begin to develop.

*F. C. Besic, R. Zechman, P. Johnston (General Electric Co., Philadelphia, PA), P. Hoffer, M. R. Wiemann, Jr., and O. Keller, Zoller Dental Clinic, University of Chicago, Chicago, IL 60637, Contract AT(11-1)-2095

Reports Published

None.

Estimated Date of Final Report

September 1971.

RADIOISOTOPIC X-RAY FLUORESCENCE SPECTROMETRY FOR MULTIELEMENT ANALYSIS OF AIR POLLUTANTS*

Objective of Work

The purpose of this project is to investigate the sensitivity of energy-dispersive radioisotopic x-ray fluorescence for the analysis of particulate air pollutants and to develop a technique for their routine multi-element analysis that is rapid, nondestructive, and inexpensive. The elements provisionally selected for

study are: Mg, Al, Si, P, S, Cl, Ca, V, Fe, Cu, Zn, As, Br, Mo, Ag, I, Hg, and Pb.

Progress and Results

Standards for 20 elements were prepared by depositing standard solutions and solid compounds (mainly oxides as fine powders) on separate filter-paper disks of the same brands as those on which air particulates are collected [Whatman 41 and Schleicher & Schuell (S&S) 589].

A laboratory system and a routine procedure for the measurement of 17 elements in samples of air-pollution particulates were set up (see first figure). In cooperation with the Texas State Air Pollution Control Board, two batches of samples collected on Whatman 41 filter paper in 43 different locations in Texas were analyzed (see second figure).

Another batch of 14 samples collected on S&S 589 filter paper in New York City was measured for 14 elements, including cadmium and barium. The data were compared with the results of atomic absorption

*J. R. Rhodes and A. H. Pradzynski, Columbia Scientific Industries, P. O. Box 6190, Austin, TX 78762, Contract AT(40-1)-4205.

Facility for x-ray fluorescence analysis.

X-ray spectrum of an air particulate sample collected in an industrial site in west Texas.

analyses. The majority showed good agreement, the differences are being investigated.

The grain-size effect is an important potential source of error in x-ray fluorescence analysis of particulates. The most accurate theory on particle-size effects^{2,4} assumes "infinitely" thick samples that contain particles of a single size. The theory was generalized to include samples of arbitrary finite thickness with continuous distributions of particle sizes. Useful formulas have been obtained for practical correction factors.

^{2,4}P. I. Berry, T. Furuta, and J. R. Rhodes, Particle Size Effects in Radioisotope X-ray Spectrometry, in *Advances in X-Ray Analysis*, Vol. 12, pp. 612-32, C. S. Barrett, J. B. Newkirk, and G. R. Mallett, Eds., Plenum Press, New York, NY, 1969.

Plan for Next Period

Different standard-preparation techniques and types of sample collection filters will be compared. The validity of the derived formulas for particle size effects will be tested experimentally.

The investigation will also include research into the possibility of using simple chemical techniques for the indirect determination of Be, C, F, Mg, Al, Si, P, and S in particulates.

Reports Published

None

Estimated Date of Final Report

Continuing project, annual report, August 1, 1972

PORTABLE RADIOISOTOPIC X-RAY FLUORESCENCE DETECTOR OF BULLET HOLES*

Objective of Work

The objective of this work is to determine the field applicability, in law enforcement, of radioisotope x-ray fluorescence for detecting lead residues from a projectile.

Progress and Results

In a field examination where the types of firearms and projectiles may not be known, a control procedure must be established to evaluate the primary means of examination — in this case, the radioisotopic probe with

*M. Brewer, Dade County Public Safety Department, 1320 NW 14 St., Miami, FL 33125. This work is related to the work at Panametrics, Inc., under Contract AT(40-1)-3561.

connecting console and counter used to identify lead residues around a suspect bullet hole. The sodium rhodizonate colorimetric method for lead was used as a chemical control. By this method, as little Pb as $\sim 0.1 \mu\text{g}$ can be determined. The lead was extricated from the suspect bullet hole with a cotton swab and was determined colorimetrically. This chemical control was laboratory tested on several micro and semimicro quantities of lead standards prior to use as a control in the field. The materials examined in the field included wooden surfaces, cement, screen, plastered cement, Sheetrock, glass, and cloth. Soft-solid materials (mostly crystalline) frequently gave negative results by both the colorimetric and x-ray fluorescence methods. Pulverization of the material that receives the Pb wipe may partially explain the negative results; the pulverized material falls from the crater produced by the projectile and causes what is tentatively called "crater wipeout" (see figure). The shock wave that results from the

Photograph showing crater "wipeout" effect on wall.

impact of the projectile and that precedes the projectile's passage through the solid material may be a cause of this wipeout

Sheetrock walls often gave negative results for both the entry and the exit sides of the holes. If complete entry or passage through the wall did not occur, a positive result was more frequently obtained.

Cloth composed of both synthetic and natural fibers gave positive results. The cloth was not examined extensively, because a microscopic study of the suspect cloth is frequently conducted in many gunshot cases. Positive results obtained by the x-ray fluorescence method were reliable and, in all cases, correlated with wet-chemical test results.

Negative x-ray fluorescence results (which also agreed with wet-chemical test results) indicated an absence of lead residue. However, in some instances, the suspect hole was definitely caused by a lead projectile. Thus, the instrumental x-ray fluorescence examination of suspect bullet holes is a reliable indicator if the results are positive but not if they are negative. Research completed to date indicates a need to study further the mechanism of the transfer of lead from projectiles to surfaces. Further research is needed on a greater variety of materials that may receive the Pb residue.

Plans for Next Period

None

Reports Published

None

Estimated Date of Final Report

February 1972

X-RAY FLUORESCENCE ANALYSIS SYSTEM FOR LEAD IN BLOOD*

Objective of Work

The purpose of this work is to develop an x-ray fluorescence system for measuring lead in blood. In the health-department laboratories of most large cities, blood lead is now determined by atomic absorption spectrometry — a method that requires at least 5 cc of blood and certain chemical manipulations. Obviously, there is

reluctance to taking serial 5-cc samples from a 2- to 3-year-old child. Nevertheless, this is necessary for sound diagnosis and treatment planning, especially where chelation is being considered. To minimize delay and the number and size of samples, a portable analysis system is needed — one that can be used at regional health clinics or in a mobile van.

Progress and Results

For children's blood, a lead concentration of ~ 0.3 ppM is considered to be normal, a concentration as high as 0.4 to 0.8 ppM is abnormal, 0.6 ppM being generally accepted as the level at which treatment should be given. X-ray fluorescence analysis for lead in paint indicated that this method, with sufficient increase in sensitivity, might detect lead at sub-ppM levels in blood. Increase in sensitivity means essentially an increase in the signal-to-noise ratio. Consequently, the emphasis has been to maximize both the production of characteristic x-rays in the sample and the detection of these x-rays. Thus, the variables selected to be optimized were (1) energy and half-life of the excitation source and (2) source-sample-detector geometry.

Since the most efficient production of characteristic x-rays arises from absorption of photons just above the absorption edge, Pu-238 was chosen, it emits x-rays (uranium-L x-rays) just above the L_1 and L_3 absorption edges of lead. Although this x-ray emission is not as abundant as that from some other radioisotopes, e.g., Cd-109, it is essentially free from any higher-energy gamma rays and hence does not present shielding problems. Also, it has a long half-life (86 yr) and is relatively inexpensive.

The primary factor in the design of the source-holder-collimator was the size of the blood sample, chosen to be 0.05 ml — an amount easily obtainable with a hematocrit tube from a finger prick. Several source-sample-detector configurations were designed, and the efficiency of each was estimated. The first figure shows the one selected, a design inspired by the availability of $\text{Pu}(238)\text{O}_2$ as 250- μ -dia microspheres. Each microsphere contains 1.0 mCi of Pu, which allows 25 mCi of radioactivity to be arranged in a 2-mm-dia circle. With this arrangement, the sample in the form of a 1-mm-cross-section "doughnut" can be irradiated throughout 360 deg of the source plane with no external scattering to other material. Also, two detectors can be used face-to-face. This configuration allows the closest possible proximity of both source to sample and sample to detector, the estimated geometrical efficiencies of which are $\sim 11\%$ and $\sim 10\%$, respectively.

*G. R. Laurer and T. J. Kneip, New York University Medical Center, 550 First Ave., New York, NY 10016, Contract AT(30-1)-4252

SECTION B-B

Source-holder – collimator (top view).

Source-holder – collimator in place on cryostat with sample in position.

The blood doughnut is formed by dropping the red-blood cells, after centrifugation, onto a precut Millipore backing filter placed on a Teflon form. Heat-lamp drying shrinks the doughnut to approximately the dimensions shown.

A 200-mm² Si(Li) detector will be placed 5 mm below the beryllium entrance window (see second figure). This detector size is a balance between resolution and detection efficiency. Since only the 10.5-keV $L\alpha$ peak of lead will be observed and since the closest large interfering peak will be the 11.9-keV $K\alpha$ of bromine, the 270-eV resolution attainable with a 200-mm² detector should be adequate and still maintain a reasonably narrow lead peak.

Plan for Next Period

When the source and detector system are received, the reproducibility of results, obtained with the doughnut geometry, will be determined with blood samples of known lead content. Also, the limit of detection of the method will be established; it should be ~ 0.1 ppm for an ~ 15 -min counting time. Two background interferences are foreseen that may further limit the sensitivity: (1) scatter-peak tailing due to edge effects in the detector, and (2) $L\alpha$ x rays from the detector's gold entrance window. Both of these could be limited by use of a facing metal other than gold and/or use of a guard-ring detector. For a large-area detector, the improvement possible with the latter is uncertain.

The use of a Cm-244 (half-life, 17.6 yr) source, also available in microspheres, is planned. The radioactivity of a 250- μ -dia Cm-244 sphere is about five times that of a Pu(238)O₂ sphere of the same size. The source configuration for Cm-244 has not been decided.

Reports Published

None

Estimated Date of Final Report

Continuing project

KRYPTONATES FOR THE ANALYSIS OF AUTOMOBILE EXHAUSTS*

Objective of Work

The objective of this project is to develop an instrument that uses Kryptonates** in the analysis of air pollutants in automobile exhausts. Instruments now accepted for use have the disadvantages of being

sophisticated laboratory devices that require trained operators. In the instrument being developed, the same principle is used to detect carbon monoxide, hydrocarbons, and nitrogen oxides. The prediction is that such an instrument could be operated by service personnel and be suitable for monitoring air pollutants emitted by automobiles during periodic or random inspections.

Progress and Results

Kryptonates have been prepared from a number of substrates. The Kr-85, which is introduced and stably incorporated into a solid substrate, is located in the surface layers only. Upon reaction of the solid with a gaseous air pollutant, Kr-85 is released at a rate that depends on the concentration of the pollutant and on the Kr-85 concentration in the surface layers. By use of small-particle-size powders and long times for the Kryptonate preparation, sensitivity to a particular gaseous species can be maintained constant during the detection lifetime of the Kryptonate.

Phase I of this program (a feasibility study) illustrated that adequate sensitivity can be achieved easily to meet the projected 1975 Federal standards for automobile-exhaust emissions (50 ppm for hydrocarbons, 250 ppm for NO_x, and 5000 ppm for CO).

The current effort (Phase II) is devoted to constructing and testing a breadboard instrument (see figures). No effort was spent on package design; nevertheless, the potentiality for developing a portable instrument is demonstrated by the fact that the breadboard instrument can be housed in a container having ~ 1.25 -ft³ volume and ~ 20 -lb weight. The front panel contains the sampling inlet and all the necessary controls.

The Kryptonate sensors selected are PtO₂ Kryptonate for hydrocarbons, I₂O₅ Kryptonate for CO, and hydrocarbon clathrate for NO₂. Since NO does not react with the hydroquinone sensor under the operating conditions, it is oxidized to NO₂ by catalytic conversion. The catalyst is Draeger CH294 (Draegerwerk, W. Germany), which is effective at room temperature. In an alternate method, a heated copper helix is used. Each method has advantages and drawbacks, but catalysis is currently used for simplicity.

*P. Goodman, Panametrics Inc., 221 Crescent St., Waltham, MA 02154, Contract AT(30-1)-4069.

**Registered trademark of Panametrics, Inc.

Block diagram of auto-exhaust quality monitor.

Photograph of auto-exhaust quality monitor.

The exhaust gases from a "dirty," laboratory, 4-cycle test engine were examined with the breadboard instrument. The results were compared with measurements of hydrocarbons by flame-ionization detection and of CO by nondispersive infrared spectrometry. The agreement was excellent. Automobiles were also tested under various conditions and at several installations, both private and governmental. Again, the results agreed excellently with those of the other two methods. No comparison instrumentation for determining NO_x was available, however, in all test situations the NO_x reading was consistent with that expected in view of CO and hydrocarbon responses. Signal levels remained constant over the duration of the test cycle and did not deteriorate on exposure to the exhaust gases.

Plan for Next Period

From experience with the breadboard unit, the design configurations will be optimized in terms of sensitivity, response and recovery times, and sensor lifetime. Several prototype instruments will then be constructed and presented to independent groups in the field of pollution control for critical assessment. After that, further changes may be needed. The resulting exhaust monitor should be a state-of-the-art, portable, prototype instrument suitable for a wide variety of applications.

Reports Published

None

Estimated Date of Final Report

Phase II report: Early 1972

NUCLEAR INSTRUMENT LANDING SYSTEM*

Objective of Work

The objective of this project is to develop a short-range guidance system for landing aircraft. The recent efforts by the U.S. Federal Aviation Agency and the aircraft industry to decrease the minimum-visibility and ceiling requirements for landing of aircraft have created a need for a very accurate and reliable aircraft-guidance system. Phase I, which was completed in July 1971,

includes the design and laboratory test of a breadboard system. The Nuclear Instrument Landing System (NILS) is designed as a short-range guidance system that will augment the Instrument Landing System (ILS) and will provide highly accurate aircraft-position information during the last phases of descent and touchdown, where the present landing aids fail to function adequately. In conjunction with the ILS, the NILS provides a means to guide aircraft during completely automated landings in zero-visibility weather. The NILS can also serve as an independent monitoring system to check the position of the aircraft before it is committed to a landing.

Progress and Results

The NILS consists of ground and airborne equipment. The ground equipment comprises four beacons, permanently installed at the airport near the runway as illustrated in Part A of the figure. The airborne equipment is carried in each aircraft. The operating principles of the NILS are similar to those of the ILS except that the rf beams are replaced by gamma radiation from Co-60 radioisotopic sources. The gamma radiation emitting from the beacons is collimated and is modulated with four characteristic frequencies by mechanical shutters to provide radiation beams similar to those used by the present ILS.

The mechanical shutters consist of a thick metal cylinder having three equally spaced slots in the top half and two slots in the lower half. The radioisotopic source is placed in the center of the cylinder. When the shutter is rotated about its axis, it modulates the emanating radiation at one frequency in the top region above the center plane perpendicular to the axis of rotation and at another frequency below the plane.

All beacons are identical in construction, however, the shutters of beacons on the right of the runway are rotated at 1800 rpm, and those on the left at 720 rpm so that four frequencies are produced.

Part B of the figure gives a block diagram of the airborne equipment. A radiation detector detects the radiation, and the frequency content of the detector output is recognized by four frequency detectors tuned to receive frequencies F_1 , F_2 , F_3 , and F_4 . The position of the aircraft in relation to the NILS beacons is then determined by comparing the frequencies to give lateral and vertical displacements as shown in Part C of the figure. The aircraft-position information is displayed to the pilot on a standard ILS indicator in the cockpit.

By use of cone-shaped shutters in the NILS beacon, it is possible to provide a curved flight path and to incorporate a flare before touchdown.

*R. A. Kaminskas, TRW Systems, One Space Park, Redondo Beach, CA 90278, Contract AT(04-3)-828

Nuclear Instrument Landing System.

Plan for Next Period

Phase II was started in July 1971, and will result in construction and flight-test demonstration of a complete NILS

Reports Published

Phase I Final Report, October 2, 1971

Estimated Date of Final Report

Not estimated

NEUTRON RADIOGRAPHY WITH CALIFORNIUM-252*

Objective of Work

The objective of this program is to develop and apply low flux neutron radiography with Cf 252 as a neutron

source. The portable shield and exposure facility that evolved from earlier work are being made more portable and efficient. Fast neutron radiography will be explored further. Low flux imaging techniques will receive emphasis. A position sensitive proportional counter is being developed to give instantaneous readout of data that represent the imaged objects. Various converter foils and films are under study as means to radiographically record images.

Progress and Results

The first neutron source holder (NC 1) was evaluated and a second source holder and exposure system were developed. The second unit (NC 2, see first figure) has the same egg shaped Fiberglass shell used in NC 1, but

*K. L. Swinth, Battelle Pacific Northwest Laboratory, Richland, WA 99352, Contract AT(45-1) 1830

Sketch of the NC-2 source holder designed for ease of positioning.

Position-sensitive counter for imaging in thermal-neutron fields.

the internal moderators were replaced with water. Thus, the weight that must be handled when the facility is positioned is only that of the shell and miscellaneous hardware (75 lb); the NC-1 weighs 350 lb. After the unit is positioned, it is filled with water and the source is installed. To facilitate the handling of large sources, this procedure could be done remotely. This unit has a fixed collimator with a length-to-diameter ratio of 15:1; however, other collimators can be installed. The thermal flux at the face of the unit is 9.4×10^2 n/cm²·sec, the cadmium ratio is 1.8, and the neutron-to-gamma ratio is 4.2×10^4 n/cm²·mr. Radiographs produced with this unit are better than those produced with NC-1. With NC-1 in a position where the geometrical unsharpness is comparable with that of NC-2, the thermal flux is 2.8×10^2 n/cm²·sec and the cadmium ratio is 1.45.

Direct imaging in low-flux beams can be performed satisfactorily with standard techniques, such as the use of gadolinium foils; however, use of the fastest technique possible is desirable. Gadolinium oxysulfide and fiber-optics-coupled Li(6)F-ZnS(Ag) have been investigated as means to enhance the resolution and sensitivity. The gadolinium oxysulfide screens with Kodak Royal Blue film are much faster than gadolinium metal foils, but Li(6)F-ZnS scintillators are still the fastest means. Although the resolution achieved is better than that with Li(6)F-ZnS scintillators, the gamma discrimination is not as good. The primary emission peaks of gadolinium oxysulfide are above 4800 Å, which suggests the use of a green-sensitive film such as Kodak Type O. Comparison of the foil with Type O and Royal Blue films showed that the Royal Blue film is faster.

Fiber-optics face plates in conjunction with Li(6)F-ZnS scintillators were studied as a means to increase the resolution obtainable with Li(6)F-ZnS scintillators. Because of light attenuation in the fiber-optics face plates, this technique increased the required exposure time 50 to 400%, the increase depending on the fiber optics used. The resolution and contrast of neutron radiographs made by this technique were improved.

Proportional counters that have the ability to read out the position of interaction of radiation are being investigated for imaging thermal neutrons. Their use will result in a device that can determine neutron transmission with resolution in the range 0.5 to 1 mm. The information will be stored in digital form. The preliminary results with this counter are promising. The second figure shows the prototype counter, which is so designed that the gas pressure, degree of collimation, converter foil, and anode-to-cathode spacing can be

varied to determine their effects on imaging. The counter now has a geometrical unsharpness of ~3 mm.

Plan for Next Period

Information derived from the prototype imaging counter will be used to fabricate a large-area device for further evaluation. Investigations will continue on the counter, image intensifiers, and foil-film techniques to optimize imaging in low-flux neutron beams. Techniques and source holders will be developed for fast-neutron imaging in beams from Cf-252 sources.

Reports Published

- J. L. Cason, *Design and Fabrication of a Portable Device for Neutron Radiography*, USAEC Report BNWL-1329, March 1971.
- C. B. Shaw and J. L. Cason, Portable Neutron Radiographic Camera Using Californium-252, *Mater. Eval.*, 29(2): 40-4 (1971).
- K. L. Swinth, Developments in Low-Flux Neutron Radiography at Battelle-Northwest, *Trans. Amer. Nucl. Soc.*, 14(2): 516-7 (1971).

Estimated Date of Final Report

Continuing project.

INDWELLING INTRACRANIAL PRESSURE SENSOR*

Objective of Work

The basic objective of the Intracranial Pressure Sensor (IPS) Program is to demonstrate the feasibility of a fully implantable, long-term, intracranial pressure sensor using a radioisotope to transmit a pressure signal without the use of signal leads. The IPS unit is intended primarily for the use in hydrocephalic children.

Progress and Results

The device employs three fluid chambers, of which two contain a radioactive fluid to give the pressure reading. The IPS device is presented in the figure. Chamber one contains an inert fluid and is implanted within the cranium so as to be exposed to the intracranial pressure. The pressure is transferred to the second chamber, which has rigid walls and a flexible

*T. S. Bustard, Hittman Associates, Inc., 9190 Red Branch Rd., Columbia, MD 21043, Contract AT(11-1)-3206.

Indwelling, intracranial pressure sensor.

diaphragm. A radioactive fluid is contained in the upper portion of the second chamber. Radioactive fluid above the diaphragm is displaced into a third chamber, which also contains a flexible diaphragm. It is this displacement of radioactive fluid that serves as a measure of the intracranial pressure.

Americium-241 emits a 59-keV gamma ray such that the quantity of radioactive fluid in the third chamber can be monitored from above the scalp by a scintillation detector. The quantity of fluid in the third chamber is directly related to the intracranial pressure.

The initial effort on the program has included selection of a suitable radioisotope, selection of an inert fluid and radioisotope form, and materials selection such that they will be compatible with body tissue. Detail design has been completed and components fabricated for assembly. Initial bonding studies have also been completed.

Plan for Next Period

The components fabricated during the initial period will be assembled. Fabrication procedure will be developed by the assembly of a unit completely filled with an inert fluid to determine filling procedure and possible problem areas. After assembly procedures have been developed, a unit that contains an Am-241 salt solution will be assembled and tested. Both static and dynamic pressure testing are planned.

Reports Published

None.

Estimated Date of Final Report

February 1972.

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PROCESS RADIATION

Glass polymer composite material

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Process Radiation

The Process Radiation Program is concerned with the preparation and processing of products employing radiation as an energy source to effect chemical, physical, and biological change. The primary objective is the development of technology necessary to advance large-scale uses of radiation important to the national economy. Food preservation by irradiation is not included in this program.

The process radiation effort is divided into three categories: radiation synthesis of chemical products, modification of materials, and radiation technology area support. New and promising synthesis systems are investigated in the first category, and composite material development and grafting methods are investigated in the second category. The third category includes the acquisition of a broad base of radiation technology useful to industry and encompasses dosimetry studies, design and evaluation of radiation sources, and engineering development for radiation energy utilization.

Three BNL projects related to the food irradiation program are given in this section:

Portable Irradiator Support,

Field Dosimetry Standardization, and

General Engineering and Technical Assistance

To be supported, a concept of the proposed process should offer a unique advantage in the use of ionizing radiation or indicate a cost less than that of the competitive process currently in use. Unique advantages in the use of ionizing radiation include one or more of the following:

Low-temperature initiation of a chemical or polymerization reaction

Solid-phase initiation of a chemical or polymerization reaction

Production of a product with no catalyst impurities

Control of the reaction rate in exothermic processes by varying the dose rate

Irradiation of material either in its final container or in its final shape

Initiation of a reaction that cannot be brought about by other means

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CONCRETE-POLYMER MATERIALS*

Objective of Work

The objective of this project is to develop concrete-polymer composites for use as improved materials of construction. Current program priorities are:

1. Developing systems for yielding optimal product quality and process technology at minimal cost.
2. Providing information and obtaining an understanding of the major factors that control the physical, chemical, and mechanical properties of concrete-polymer materials.
3. Obtaining an understanding of the fundamental nature of concrete and concrete-polymer materials through theoretical and experimental studies.
4. Aiding in the transfer of laboratory techniques and materials development to potential full-scale applications.

Progress and Results

Three distinct types of materials are being investigated in the concrete polymer materials program. These are (1) polymer-impregnated concrete (PIC) precast portland-cement concrete impregnated with a monomer that is subsequently polymerized in situ; (2) polymer-cement concrete (PCC) - a mixture of water, portland cement, and aggregate to which a monomer is added and is subsequently polymerized after pouring of the mix; and (3) polymer-concrete (PC) - an aggregate mixed with a monomer that is subsequently polymerized after pouring.

In general, all of the composites show significant improvements in strength and durability. Monomers currently being tested for normal-temperature applications are concrete with methyl methacrylate (MMA), styrene, methyl methacrylate-10 wt% trimethylolpropane trimethacrylate (TMPTMA), acrylonitrile, chlorostyrene, and 90 wt% styrene-10 wt% polyester. The MMA-impregnated concrete has given the best results.

The strength improvements appear to be a function of polymer loading, and radiation polymerization gives slightly better strength properties than thermal-catalytic polymerization. Improvements in durability appear to be mainly a function of polymer loading and effectiveness of sealing the surfaces of the concrete.

A number of preparation variables, including concrete composition, drying temperature, curing time, and age at test, were investigated. The age of concrete at impregnation or its initial strength does not appear to significantly affect the ultimate strength.

The potential application of concrete-polymer composites as a material of construction for desalting plants is being evaluated. Two monomer mixtures - 60 wt% styrene-40 wt% TMPTMA and 90 wt% diallyl phthalate (DAP) 10 wt% MMA have been studied in detail. Structural properties such as compressive strength, Poisson's ratio, modulus of elasticity, tensile splitting strengths, creep, and steel rod pullout strengths have been measured at temperatures up to 290°F. The resistance to chemical attack by hot brine and distilled water is also being determined. Based upon these tests, the 90 wt% DAP-10 wt% MMA system was found to be unsatisfactory. Other candidate materials are currently being surveyed.

Significant advances have been made with PC. Monomers and monomer mixtures investigated include MMA, styrene, acrylonitrile, MMA-TMPTMA, polyester styrene, and styrene-TMPTMA. With the use of silane coupling agents to obtain bonding between the organic and inorganic phases, and the use of graded aggregate to obtain minimum void volume, compressive strengths as high as 20,000 psi have been obtained for samples containing 6 wt% polymer.

Only limited success has been achieved with PCC because of the inherent incompatibility of organic monomers with water and alkaline inorganic silicate cement materials.

A number of potential applications for concrete polymer materials are being evaluated as part of cooperative programs with other governmental agencies. The field testing of impregnated sewer pipe by the Water Utilities Department of the City of San Diego has been continued but data are not yet available. Ten additional lengths of 8-in.-diameter by 4-ft-long pipe have been impregnated and will be installed at San Diego during the current year. Laboratory tests are being performed at BNL. Compared with controls that failed after 17 days (25% weight loss), pipe impregnated with polyester-styrene has shown no attack after exposure to 5 wt% H₂SO₄ for 60 days.

*B. Manowitz, M. Steinberg, L. E. Kukacka, and P. Colombo, Brookhaven National Laboratory, Upton, NY 11973, Contract A1(30-2)-GEN-16. This project is jointly supported with the U.S. Bureau of Reclamation, Office of Saline Water, Federal Highway Administration, and Bureau of Mines. Cooperative efforts are also in progress at the Naval Civil Engineering Laboratory, Port Hueneme, CA, Army Corps of Engineers Construction Engineering Laboratory, Champaign, IL, and with the American Concrete Pipe Association.

The initial stage of a cooperative effort among the American Concrete Pipe Association, USAEC, BNL, and the Bureau of Reclamation to determine the value of concrete-polymer composites for pipe applications has been completed. Test results showed a sufficient improvement in strength from both the three-edge bearing and the hydrostatic tests to warrant further investigation. This work is in progress.

Experiments are being performed in conjunction with the Federal Highway Administration to determine the feasibility of using concrete-polymer materials for highway applications. To date, these studies indicate potential applications in bridge decks and other structural elements.

Plan for Next Period

Development of concrete-polymer materials will be continued. Emphasis will be on the development of process technology and the transfer of laboratory techniques and materials development to potential full scale applications.

Reports Published

- M Reich and J M Hendrie, *Report on Polymer Concrete Applications Development for the Bureau of Public Roads Design Studies on Highway Bridge Decking and Luminaire Supports*, January 1969–June 1970, USAEC Report BNL-15139, August 1970
- J T Dikeou, W C Cowan, G W DePuy, W G Smoak, G B Wallace, M Steinberg, L E Kukacka, A Auskern, P Colombo, J M Hendrie, and B Manowitz, *Concrete-Polymer Materials, Third Topical Report*, USBR Report REC-ERC-71-6 and USAEC Report BNL-50275, January 1971
- L E Kukacka, P Colombo, M Steinberg, and B Manowitz, *Concrete-Polymer Composites*, Journal of the Structural Division, ASCE, September 1971

Estimated Date of Final Report

Fourth topical report scheduled for January 1972

DEVELOPMENT OF CONCRETE-POLYMER MATERIALS*

Objective of Work

The Bureau of Reclamation and Brookhaven National Laboratory are engaged in a joint program to develop concrete-polymer composites for use as new materials

of construction. The program objectives are (1) development of process technology for economic production of a uniform material having exceptionally high strength and durability properties, (2) investigation of the physical, chemical, and mechanical properties, and the fundamental nature of the materials, (3) evaluation of these materials and transfer of the laboratory technology to potential full scale production applications, and (4) development of reliable design and construction criteria.

The work sponsored by the USAEC is directed primarily to the production of materials for normal temperature applications and preferably to using radiation-initiated polymerization. The work sponsored by the OSW is directed to development of materials for saline-water desalting plants; these materials must be capable of withstanding exposure to concentrated brines and distilled water at high temperatures.

Progress and Results

Three distinct types of materials for normal temperature applications are under investigation: (1) polymer-impregnated concrete (PIC) – precast portland-cement concrete impregnated by a monomer that is subsequently polymerized in situ, (2) polymer-cement concrete (PCC) – a premixed combination of portland cement, aggregate, water, and monomer, in which the monomer is added during conventional concrete mixing and polymerized after pouring, and (3) polymer-concrete (PC) – a composite material formed by polymerizing a monomer with an aggregate mixture. The monomers are polymerized by gamma radiation and by thermal catalytic techniques.

Investigations to date indicate that the most successful concrete-polymer material for construction is PIC, and it is now receiving the major emphasis in the program. Five impregnating materials are presently under test – methyl methacrylate (MMA), MMA plus 10% trimethylolpropane trimethacrylate (TMPTMA), styrene, acrylonitrile, and chlorostyrene. In addition, there is a continuing survey program to select other promising monomers for testing.

All monomers used for impregnation have produced improvements in the properties of concrete – MMA and MMA-TMPTMA showing the greatest improvements. Compressive strengths of over 25,000 psi and tensile

*J T Dikeou, U S Bureau of Reclamation, Denver, CO 80225, Contract AT(49-11) 3003. This project is jointly supported by the Bureau of Reclamation, Office of Saline Water, Washington, DC and Brookhaven National Laboratory.

strengths of 1700 psi (increases of more than 300% over unimpregnated concrete) have been obtained. Durability tests still in progress show improvements of 1000% or more in resistance to 15% HCl and to freezing and thawing, and essentially no sulfate attack occurs after an exposure three times as great as that required to cause failure in unimpregnated concrete.

Development work in process technology has included studies on the effects of both concrete-mix design parameters and concrete-curing conditions on the properties of polymer-impregnated concrete, and also on improvements in specimen fabrication and polymerization techniques. The specimen-fabrication technique may be improved through elimination of a wrapping step. Preliminary studies have indicated the feasibility of eliminating this step by polymerizing under water or by using live steam for thermal-catalytic polymerization inside an impregnation vessel.

Work on PC and PCC has been limited, but the results obtained thus far look promising. The PC studies have shown that mechanical packing of especially selected, sized aggregate has reduced monomer weight requirements from as much as 25% to 5 to 6%. The PCC studies are in very preliminary stages.

Plan for Next Period

The testing program for evaluating various monomers and the properties of PIC will continue. Two new PIC's are to be tested—one will contain polyester-styrene and the other MMA polymerized under water. The effects of concrete-mix design parameters on properties of PIC will be further explored, and new polymerization techniques using a promoter-catalyst system will be studied. Plans are being made to develop impregnation and polymerization techniques for treating variously shaped concrete members, such as large slabs and long beams.

Investigations are continuing with PCC and PC because these materials may have many potential applications if suitable properties and feasible fabrication methods can be developed. Mechanical packaging of aggregate for PC will be further explored and the feasibility of incorporating several acrylic monomer and polymer emulsions, and furfural-aniline polymers in the concrete mix will be investigated in the PCC studies.

Reports Published

J. T. Dikeou, W. C. Cowan, G. W. DePuy, W. G. Smoak, G. B. Wallace, M. Steinberg, L. E. Kukacka, A. Auskern, P. Colombo, J. M. Hendrie, B. Manowitz, *Concrete-Polymer Materials*, Third Topical Report,

USBR Report REC-ERC-71-6 and USAEC Report BNL-50275, January 1971.

Estimated Date of Final Report

Fourth topical report scheduled for January 1972.

RADIATION CHEMICAL ENGINEERING*

Objective of Work

The purpose of these investigations is to study the interaction of the kinetics of radiation-initiated chemical reactions with phenomena of radiation physics, fluid dynamics, and mass and heat transfer. The results of the theoretical and experimental work are useful in devising scale-up procedures, in designing irradiators for optimum source utilization or optimum product arrangement, and in interpreting radiation-chemical experiments, where chemical and physical processes interact. The results should apply to irradiators for the radiation inactivation of contaminants in liquids and particulate materials as well as to radiation-chemical reactors.

Progress and Results

Photoactivation is being used in the preliminary work. An experimental study of mixing effects in chain reactions is underway. The reaction is the benzoin-sensitized bulk photopolymerization of methyl methacrylate and it is carried out in a partially illuminated, stirred, quartz dilatometer. The rate of reaction and the molecular weight distribution of the polymer are measured as a function of the size of the illuminated region and the stirrer speed. The most interesting result of these measurements to date is the finding of a bimodal molecular weight distribution. The relative amounts of polymer associated with the two peaks and, in certain circumstances, the average molecular weight vary with stirrer speed and size of the illuminated region.

Two supplementary studies have been started which may disclose the origin of the bimodal distributions. In the first, experiments designed to reveal the flow paths of propagating chains are being performed. The reaction vessel is filled with a monomer containing a photochromic dye (1,3,3-tri-methylindolino-6'-nitrobenzopyrylospiran), and the vessel is partially illuminated, as in the experiments without dye. Under illumination the

*F. B. Hill and A. C. Muller, Brookhaven National Laboratory, Upton, NY 11973, Contract AT(30-2)-GEN-16

dye assumes its colored form, which persists after illumination for periods of seconds to minutes depending on the temperature. Thus the flow pattern of the dye as it leaves the illuminated region may be followed and is presumably the same as that of the propagating chains in photopolymerization. Initial results of these experiments include the finding of considerable flow by natural convection in the absence of stirring and the observation of highly stratified flow patterns at low and intermediate stirrer speeds.

The second study is theoretical and is concerned with the calculation of the molecular weight distribution in a partially illuminated vessel. Steady-state kinetics is assumed, with free radicals being produced in the illuminated region and diffusing into the dark region. It is reasoned that a bimodal distribution may be associated with the situation in which a high concentration of free radicals exists in the illuminated region and a low concentration in the dark.

Apparatus has been completed for the study of radiation-initiated polymerization in flow systems. A 20-Ci Sr-90-Y-90 line source of beta particles is used on the axis of an annular flow reactor. The first runs have been made using methyl methacrylate in single-pass flow and in recirculating flow.

A series of experiments has been conducted on a gas-liquid radiation-chemical reaction. Measurements were made of the apparent rate of oxidation of aqueous ferrous sulfate in the presence of interphase gas-liquid transfer of oxygen. Oxygen was absorbed by or stripped from the liquid phase, and mass-transfer coefficients were calculated from the data. This study is a forerunner for a later study of gas-liquid chain reactions.

A comparison of the relative advantages of chemical and radiation initiation of chemical reactions conducted on a large scale is being made. Stability and ease of control are the characteristics emphasized. Initial calculations have shown a larger region of stable operation of continuously stirred tank reactors when using radiation initiation. Experimentally, the stability of batch radiation-chemical reactors is being studied in a series of experiments on the production of poly(methyl methacrylate) sheet by a radiation process.

Plan for Next Period

Additional work as required will be done on the experimental and theoretical aspects of the batch photopolymerization to determine the cause of the bimodal molecular weight distributions.

Experimental study of conversion and molecular weight distribution in continuous radiation-polymerization reactors will continue.

Calculations of batch-reactor stability and of the effect of dose-rate variation on stability in batch reactors will be made. These calculations will be used to guide experimental work.

Reports Published

- R. M. Felder and F. B. Hill, Mixing Effects in Chemical Reactors. Chain Reactions in Batch and Flow Systems, *Ind Eng Chem Fundam*, 9, 360-7 (1970).
- L. H. Shendalman and F. B. Hill, Thermal Ignition and Stability of Batch Reactors with Nonuniformly Initiated Radiation-Induced Reactions, in *Chemical Reaction Engineering*, pp. 449-57, Pergamon Press, Oxford, 1971.
- H. T. Chen and F. B. Hill, Radiation-Induced Polymerizations in Continuous Stirred-Tank Reactors, *AIChE (Amer Inst Chem Eng) J*, 17, 933-9 (1971).

Estimated Date of Final Report

Continuing project

SURVEY EXPERIMENTS APPLICATION OF ISOTOPIC RADIATION FOR CHEMICAL PROCESSING*

Objective of Work

The objective of this project is to investigate the significant effects of radiation on a variety of chemical reactions that have a potential for chemical processing application.

Progress and Results

Effort has continued to center upon the synthesis and, more recently, the decomposition of ozone under the influence of ionizing radiation from Co-60. Experiments involving irradiation of carbon monoxide to evaluate the G value for carbon black formation and irradiation of metal salts in aqueous solution for possible reduction to pure metal have also been performed.

A detailed study of the kinetics of O₃ formation and decomposition was continued at temperatures from 27° down to -100°C, dose rates of 0.4 to 4 Mrads/hr using Co-60, and pressures primarily at 2 atm. The findings

*R. N. Dietz and M. Steinberg, Brookhaven National Laboratory, Upton, NY 11973, Contract AT(30-2) GEN 16

from this study are

- 1 The condition of the walls of a stainless steel vessel has a marked effect on the ozone steady-state concentration. An ultrasonically cleaned vessel in the presence of ultrapure O_2 gave a much lower O_3 concentration than one previously exposed to air. The wall effect was more significant at lower temperatures and pressures.
- 2 With increasing amounts of NO_2 (N_2O_4) adsorbed on the wall of a 30-ml vessel, the ozone steady-state concentration at a pressure of 2 atm of ultrapure O_2 , a temperature of $-100^\circ C$, and a dose rate of 0.4 Mrad/hr markedly increased: 175 ppM O_3 with ~ 0.001 mg N_2O_4 , 475 ppM with 0.010 mg, and >1600 ppM with 0.39 mg. The initial G value increased from 6.6 to 9.3 over the same range.
- 3 The addition of a small amount of sulfur hexafluoride (0.64%) caused the yield-dose curves at temperatures above $-20^\circ C$ to exhibit sharp maxima occurring near 1 to 2 Mrads of absorbed dose. At $-60^\circ C$, typical yield-dose curves were obtained, but the O_3 steady-state concentration was from 12 to 36 times that in the absence of SF_6 .
- 4 The decomposition of 3.65% O_3 in ultrapure O_2 was studied at $27^\circ C$, 2 atm, and 0.4 to 3.5 Mrads/hr. The rate of decomposition, $G_g(-O_3)$, was proportional to the O_3 concentration and inversely proportional to the square root of the radiation intensity. The initial G values of O_3 decomposition ranged from 2700 to 7000 at the conditions given, indicating a chain reaction.

In the nuclear industry, it is generally conceded that a no-loss radioactive off-gas system is technically feasible and most economical through cryogenic processing. One method proposed involves distillation of the off-gases at $-180^\circ C$. The radioactive xenon and krypton are concentrated in a reboiler containing several gallons of liquid O_2 . A preliminary calculation has shown that, depending on the size of the nuclear reactor, intensities ranging from 10 to 1000 Mrads/hr could be attained. Experiments with liquid O_2 at $-170^\circ C$, 15 atm, and 5.8 Mrads/hr in a 30-ml stainless steel vessel have given steady-state O_3 concentrations of >7 mole %. The potential hazard of this mixture was demonstrated by an explosion in one vessel, which fused part of the stainless steel. Catalysts for destroying the O_3 in situ are being investigated.

A study of radiation-induced reactions of CO is being conducted to find possible ways of utilizing the 6 to 60 million tons per year of CO currently produced as a

by-product of the steel industry's basic-oxygen-furnace process. The radiation and thermal decomposition of CO to produce carbon black has been investigated. At temperatures above $100^\circ C$ the products are predominantly C and CO_2 , with the G value for carbon black being about 10.

Recent experiments by others have shown that metals can be prepared from their aqueous salt solutions by irradiation with 2-MeV electrons. In a series of experiments using a 25% ferrous chloride solution, suppression of the rate of oxidation of ferrous to ferric iron by dissolved O_2 was achieved with Co-60 radiation. There was no indication of reduction to metallic iron, however.

Plan for Next Period

The study of the formation and decomposition of O_3 will continue and will include the effects of solid additives as well as different vessel materials. More details of the formation of O_3 in liquid O_2 will be investigated.

Other reactions of carbon monoxide will be examined, but reactions of salt solutions in the presence of radiation will be terminated shortly.

The treatment of sewage and waste waters with ozone has been studied by others in detail, as have the effects of radiation on such treatment. Ozone in combination with radiation will be examined in view of the growing awareness of possible toxicity problems associated with chlorine disinfection.

Reports Published

None

Estimated Date of Final Report

Continuing project

COMPOSITE MATERIALS*

Objective of Work

The objective of this work is to develop composite materials for structural applications from inexpensive raw materials and waste products. Various aggregates are mixed with liquid monomers that are subsequently polymerized by either radiation or thermal catalytic methods.

*M. Steinberg, P. Colombo, and A. Auskern, Brookhaven National Laboratory, Upton, NY 11973, Contract AT(30-2) GEN 16.

Progress and Results

Basic studies have centered on the mixing of silica sand of various sizes and stone aggregates with methyl methacrylate monomer prior to polymerization. Samples of Ottawa sand from 20 to 270 mesh were used to determine the effect of single size particles on packing density. The main conclusion was that with each sand-particle size used, sand occupied ~60% of the volume of the packing. This was determined by measuring the volume of monomer required to fill the voids. The compressive strengths of all specimens prepared with various single sizes of sand were essentially the same — ~11,500 psi.

Based on both experimentation and theoretical calculations, a sand mixture was selected that reduced the monomer requirements to 10% by volume (5% by weight of sand). This mixture consists of sand of mesh sizes 170–270, 40–60, 20–30, and of 0.5-in. stone in weight proportions of 6.1, 10.2, 23.0 and 60.7%, respectively. For practical purposes, only the four size ranges were used. With the above sand sizes and sand-size distribution, the compressive strength for cured specimens requiring 5% by weight of monomer was ~11,000 psi.

A simple technique has been developed for preparing composite mixes without use of vibrators, pressure or vacuum. The aggregate-monomer mix is contained in a shaped mold, which is placed in a radiation field; after curing, the composite is released from the mold. Various additives such as colorants, flame retardants, plasticizers, ultraviolet absorbers, etc., can be mixed with the monomer before adding the aggregate.

Effect of Method of Silane Coupling Application
on Compressive Strength of Polymer Concrete

Sample No	Compressive strength, psi	Method of silanization
1	13,875	1% by wt. of silane ^a blended with methyl methacrylate prior to contact with aggregate and sand
2	13,790	
3	13,800	
4	15,750	Aggregate and sand treated with 1% silane hydrolyzed in water, dried, and contacted with MMA
5	15,825	
6	15,790	
7	15,900	Aggregate and sand exposed to silane vapor before contact with MMA
8	16,150	
9	16,000	
10	11,200	No silane coupling agent added
11	10,950	
12	10,750	

^aUnion Carbide A-174 *gamma*-methacryloxypropyltrimethoxysilane

Silane coupling agents as additives to enhance bonding between the inorganic substrate and organic monomer are under study. In particular, *gamma*-methacryloxypropyltrimethoxysilane has been used with methyl methacrylate monomer. This bifunctional material is capable of reacting with and forming covalent chemical bonds between an inorganic substrate, such as silica, and an organic monomer. The various methods for applying the coupling agent and its effect on compressive strength are given in the table. Experiments are underway to optimize the method of application and the concentration of coupling agent required.

A study is also underway to determine the potential for use of solid wastes in structural materials such as pipe and building block. Initial efforts have included visits to various Federal and industrial facilities to determine raw material availability and product desirability. One potential application — sewer pipe — employs glass residue or waste as an aggregate and requires only a small percentage of a monomer. Specimens have been prepared from crushed glass that has been screened and graded to provide optimum void density for minimum monomer requirements. The graded glass is placed in a form, and methyl methacrylate containing 1% silane as a binder is diffused up through the glass. With benzoyl peroxide as a thermal catalyst, the monomer in the sample is polymerized with heat. The resulting product, designated as "Glastic," is impervious to water, resists 5% sulfuric acid solution, and has compressive and tensile strengths of 16,000 and 1200 psi, respectively. Glastic contains 93% glass and 7% methyl methacrylate by weight. A sample of sewer pipe, called "Eco-pipe," has also been produced from this material.

Work on development of a ceramic-polymer bone substitute of porous Al₂O₃ in which the pores are filled with poly(methyl methacrylate) is continuing. Methyl methacrylate is polymerized with Co-60 gamma radiation to avoid possible problems of toxicity from the use of a promotor and catalysts. A high-pressure mercury porosimeter is being used in the development of Al₂O₃ ceramics with the appropriate pore size.

Plan for Next Period

Attempts will be made to optimize the aggregate mixture to further lower monomer requirements. It has been observed that the stone aggregate is the weak link in the system, and stronger stone aggregates will be tested. Work with waste materials will be expanded and directed as review of the literature and of the information gathered from plant visits indicates. A preliminary cost analysis of material applications will be made.

Reports Published

None

Estimated Date of Final Report

Continuing project

**BASIC AND ENGINEERING STUDIES
OF RADIATION-INDUCED REACTIONS
IN THE LIQUID PHASE***

Objective of Work

The objective of this work is to explore in depth the basic and engineering features of a number of radiation-induced liquid-phase reactions of potential industrial significance

Progress and Results

The basic aspects of the work have been concentrated on the radiation-induced grafting of styrene to polybutadiene in latex form, the polymerization of vinyl chloride under precipitating conditions, and the ionic polymerization of vinyl ethers

The grafting of styrene to polybutadiene latex using radiation initiation has been studied, together with parallel chemically initiated graft polymerizations. The resulting grafted latexes are of the type used for high-impact polystyrene plastics, and the corresponding styrene-acrylonitrile grafts form the basis for acrylonitrile-butadiene-styrene (ABS) plastic materials. It is hoped that the radiation initiation will show advantages over chemical catalysis in the efficiency of graft formation and in the structure of the graft copolymers. The results obtained thus far show somewhat parallel yields of grafts with the two initiation systems. However, preliminary analyses of the graft copolymers gave an indication that the grafted side chains are shorter and more numerous with the radiation process.

Studies in the radiation polymerization of vinyl chloride under precipitating conditions have been continued, in certain nonsolvents, poly(vinyl chloride) (PVC) remains separated as a fine powder up to very high conversions. A reasonable mechanism for the reaction and the detailed kinetics have been elucidated.

The radiation-induced ionic polymerization of super-dry monomers has continued to center around the vinyl ethers. Tertiary-butylvinyl ether and isopropylvinyl

ether have both been polymerized under apparently optimum conditions. This was established by the square-root dependence of the polymerization rate on the dose rate. High molecular weight products were obtained from both ethers, and it seems clear that the molecular weight is governed by a chain transfer to the monomer.

Work with isobutylvinyl ether has been concentrated on the use of Molecular Sieves for drying to provide data for the engineering research. The effect of two solvents — methylene chloride and diethyl ether — has also been studied under these conditions. Considerable increases in the G values were obtained with diethyl ether. The polyvinyl ethers seem to be degraded by radiation, and some details of this concurrent process are now being determined.

The engineering aspects of the work have centered on the three separate pilot plants and their backup engineering studies. Small-scale research pilot plants of approximately 8 liters reaction volume have been operated for the study of both ionic and emulsion polymerization. A third system has been under design and construction for the bulk polymerization of vinyl chloride under precipitating conditions.

Successful operation of the pilot plant for dry ionic polymerization has continued with an in-depth study of isobutylvinyl ether, both in bulk and in methylene chloride solution. Engineering analysis has yielded a combined chemical-kinetic and flow model that fits performance well.

The pilot facility is being converted for processing gaseous monomers under continuous medium-pressure conditions. The initial program is the ionic polymerization of isobutylene, with future work centering about copolymer rubbers of this monomer.

Successful operation of the emulsion pilot plant has continued after modification to a continuous moderate-pressure system for the study of vinyl chloride. A complete study of parameters of the emulsion PVC system employing recycle has disclosed interesting interactions that lead to enhanced polymerization rates. Additional equipment is being constructed for continuous copolymerization of other monomers with vinyl chloride.

The design of an integrated pilot plant for bulk polymerization of PVC under precipitating conditions was completed, and the construction is nearly finished. The plant is centered about a recirculating two-phase reactor, with the discharge product stream being separated into PVC and unreacted monomer. The use of a polymer nonsolvent retards the agglomeration tendency and aids in the removal of residual monomer from the

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PVC powder Hydrocyclones and a solvent recovery unit are integral parts of the separation systems, making the pilot plant design complete with only a vinyl chloride continuous feed and a pure powder PVC product withdrawal

Plan for Next Period

The studies of the radiation-induced ionic polymerization of the vinyl ethers will be continued and extended to copolymerization systems

The grafting of vinyl monomers to diene polymers in emulsion will be extended to acrylonitrile-styrene systems (ABS type polymers)

The vinyl chloride studies in precipitating systems will be continued with a search for more efficient non-solvents

The engineering studies will continue, centering around the three pilot plant systems described above

Reports Published

- S Russo and V Stannett, The Mechanism of the Gamma-Ray Polymerization of Vinyl Chloride in Bulk I Dependence of the Degree of Polymerization on Temperature and Conversion, *Makromol Chem*, 143 47-56 (1971)
- S Russo and V Stannett, The Mechanism of the Gamma-Ray Polymerization of Vinyl Chloride in Bulk II Polymerization Rates and Kinetic Approaches, *Makromol Chem*, 143 57-66 (1971)
- F T Osborne, S Omi, V Stannett, and E P Stahel, Radiation-Induced Solution Polymerization of Styrene in an Engineering Flow System, *J Polym Sci, Part A 1*, 8 1657-74 (1970)
- V Stannett, E P Stahel, J Kohler, E Oda, S Omi, A Goineau, H P Shah, and F T Osborne, *Research and Development Studies into the Basic Technology of Radiation-Induced Ionic Polymerization Including Small-Scale Pilot Plant Systems*, Final Report, USAEC Report ORO-3667-1, June 1971

Estimated Date of Final Report

Vinyl Chloride in Precipitating Media, Topical Report, June 1, 1972

Emulsion Polymerization of Vinyl Chloride Continuous Engineering Systems, June 1, 1972

RADIATION-INDUCED CHAIN REACTIONS IN HETEROGENEOUS SYSTEMS*

Objective of Work

The primary aim of this program is to study chemical engineering problems associated with the design and operation of a system for conducting chemical reactions in radiation fields. In addition, relationships between the physical and mechanical characteristics of the product and the reaction conditions are being developed

Progress and Results

The reaction system for grafting gaseous butadiene (BD) to polyvinyl chloride (PVC) powder has received major interest. Factors studied included the effects of dose rate, BD concentration, and PVC particle size on grafting rate, postirradiation graft yields, and the effect of BD solubility in the grafted polymer. The results of major interest in the study of grafting of BD and PVC particles in a fixed-bed reactor were

- 1 Under all conditions the grafting rate increases to a maximum of ~ 10 wt%/hr and then decreases to a limiting steady-state value of ~ 2.5 wt%/hr. The limiting value of the reaction rate is insensitive to dose rate in the region 1×10^4 to 8×10^4 rads/hr. However, the maximum graft rate achieved depends strongly on dose rate.
- 2 In the transient region, wet BD grafts at a higher rate than does dry BD, but dilution of the BD gas with N_2 monotonically decreases the graft rate.

The continuous chemical-reactor system for the study of heterogeneous reactions employing gamma radiation was completed in June 1971. The reactor section in the gamma-radiation field is similar to a plug-flow reactor. This portion of the loop reactor will be analyzed by techniques developed for classical plug-flow chemical reactors. The system has been so designed that residence times inside and outside the radiation field can be varied independently. Thus, the desired degree of grafting can be obtained with a minimum radiation dose.

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To determine the effect of process variables on the reaction kinetics and the products produced, the products must be characterized. In the batch studies to date, a "weight-gained" measurement has been used to determine the degree of grafting. This method cannot be used for the continuous-loop reactor because a sensitive mass balance is not possible. A linear-density-gradient column was constructed, and polymer produced in batch runs was used to evaluate the efficacy of density measurements for determining the degree of grafting.

Plan for Next Period

Demonstration runs will be made on the continuous reactor system with the goal of determining how reaction conditions — e.g., temperature, butadiene concentration, dose, and postirradiation grafting time — affect the grafted material produced. Products are expected to be characterized by using a chemical method for determining the degree of grafting and dynamic mechanical testing and spectroscopic analysis to obtain information on the polymer fine structure. Kinetic models will be developed from these studies.

Reports Published

- I Block and J Silverman, Radiation-Induced Grafting of Butadiene to Chlorinated Polyethylene and Poly(Vinyl Chloride), *Trans Amer Nucl Soc*, 14 503 04 (1971)

Estimated Date of Final Report

Continuing project

INFLUENCE OF RADIATION UPON HETEROGENEOUS CATALYTIC SELECTIVITY*

Objective of Work

The objective of this work is to ascertain whether or not irradiation of a catalyst prior to or during a catalytic reaction will enhance the selectivity in complex reaction networks.

Progress and Results

Irradiation of supported silver catalysts with γ rays from a 12-kCi Co-60 source followed by measurements of ethylene oxide yield in the silver-catalyzed oxidation

Ethylene oxide yield vs. ethylene conversion for γ -irradiated (Co-60) and nonirradiated Engelhardt supported silver catalyst.

of ethylene was undertaken for two catalysts — a commercial Al₂O₃-supported silver catalyst and a similar catalyst prepared in this laboratory.

To assess the influence of γ irradiation upon a supported silver catalyst, a sample of commercial supported silver catalyst was placed in a laboratory chemical reactor, and steady-state values of ethylene conversion and yields (A in figure) of ethylene oxide and carbon dioxide were measured at a temperature of 280°C in excess oxygen. The sample was removed from the reactor and exposed to γ rays (Co-60) for 10 hr at a dose rate of 10^{18} eV/g·min. The irradiated catalyst sample was then returned to the reactor, and measurements of conversion and yield (B) were made as before. The irradiated catalyst was then reduced in situ in H₂ for 3 hr at 290°C, swept with N₂, and then exposed to air. The reduced and reoxidized sample was again tested for ethylene conversion and yield (C). Then the catalyst sample was again removed from the reactor, reirradiated, and tested for conversion and yield (D).

The data suggest that the influence of radiation on conversion and yield, which is eliminated by reduction and reoxidation, is obviously reversible since a cycle of reduction, oxidation, and reirradiation restores the enhancement of yield.

Ethylene conversion and yields of ethylene oxide and carbon dioxide for the normal and the irradiated catalyst at 280°C both indicate that irradiation of the

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catalyst increases the total conversion of ethylene, does not change the yield of CO_2 , and increases the yield of ethylene oxide

Laboratory-prepared alumina-supported silver catalyst containing calcium and Na_2SO_4 as imposed impurities, was tested at 240°C for conversion of ethylene and yield of ethylene oxide before and after γ irradiation. The results were

Conversion of ethylene, %	Yield of ethylene oxide, %	
	unirradiated	irradiated
10	43	67.5
20	46	67
30	44	65
35	38	62

As yields of ETO are 60 to 65% with irradiated commercial catalysts containing moderators, the significant benefits of irradiation are manifest. The data at 240°C show dramatic improvement of the laboratory catalyst by irradiation to give an industrially competitive yield for a "homemade" catalyst.

Samples of ZnO catalyst were prepared for studying the dehydration (desired reaction) and dehydrogenation of ethanol. Catalysts were sintered for different lengths of time and temperatures, and under various atmospheres, i.e., air, hydrogen, and vacuum. The catalysts were used prior to and after γ irradiation and over a range of reaction temperatures. The following observations were made:

1. Irradiation of the catalysts has no effect on dehydrogenation of ethanol.
2. Irradiation of the catalysts enhances the dehydration reactions, with the magnitude of the effect depending strongly on previous reduction treatment of the catalyst.
3. Enhancement of catalyst activity is completely eliminated after the catalyst is heated to about 350°C in the reaction environment. The catalysts heated in a reducing atmosphere (irradiated or not) behave the same as those heated in air.

Plan for Next Period

A number of industrially important catalytic reactions need study to determine the influence of γ irradiation of the catalyst upon selectivity. Dehydration selectivity of oxides of copper will be explored, and studies will continue on ethylene oxidation over our prepared catalysts to define precisely those kinetic

parameters that are altered by irradiation. Experiments will be continued to identify alterations in surface chemistry.

Other systems of prime industrial importance needing study are methanol oxidation over silver, propylene oxidation over silver and copper oxide, and xylene and naphthalene oxidation over V_2O_5 .

Reports Published

Because patent application filing on behalf of the AEC required withholding of our experimental results from the literature until all patents were filed, only recently have papers for publication been prepared.

Estimated Date of Final Report

July 1972

RADIATION CROSS-LINKING OF POLYMERS WITH SEGREGATED METALLIC PARTICLES*

Objective of Work

The objective of this project is to optimize conductive polymer-metal composites, design economic fabrication techniques, and demonstrate potential applications.

Progress and Results

During the past two years, the principle of segregated conducting networks was established. Conductive polymer-metal composites with low metal loadings are now possible. Thus 6 vol% of nickel powder (4 to $7\ \mu$) in poly(vinyl chloride) reduced the resistivity from 10^{14} to 1 ohm-cm. If the networks are formed by random mixing, a metal loading of 20 to 30% is necessary for an equivalent resistivity.

In essence, the principle is the compaction of large polymer particles with much smaller metal particles, which leads to a continuous conductive network throughout the material (see figure). For example, poly(vinyl chloride)-nickel composites were made by compacting $150\text{-}\mu$ polymer particles with $5\text{-}\mu$ nickel particles (10 vol%) at about 120°C under 10,000 psi pressure. The resulting compacts showed a resistivity of about 1 ohm-cm.

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A fracture surface of a polyurethane/nickel composite.

With the development of conductive polyurethane–nickel composites, the principle of segregated conductive networks has been shown feasible for all classes of polymers — semicrystalline, glassy, and elastomeric. Experience to date has shown that the critical fabrication condition is compaction without shearing. Shearing prevents the continuous network formation necessary for conductivity. Preliminary work with extrusion, rolling, and compression molding with open dies led to nonconductive compacts. All these processes may yet be used, but considerable modification will be necessary. However, preliminary review of the process economics indicates that rapid, semiautomated compression–molding cycles are competitive, particularly for small parts. Hence this limitation is probably not a serious deterrent to future applications.

Prototype construction and demonstration of feasibility is critically important to the rapid use of these conductive composites. After a series of meetings with

private organizations to extend this technology, the development of a transparent conducting window is underway in cooperation with one company and a conductive electrical shield for a connector is being considered by another.

This process has been extended to polyurethane composites. The difficulty with these rubbery materials is the movement of the metal particles into the soft polymer rather than forming a continuous network. This was prevented by polymerizing a glassy polymer within the polyurethane particle before compaction. The conductive polyurethanes were obtained by the following two-step process.

1. Irradiation of polymer powder and monomer with a 1-Mrad dose. Polymer powder is stabilized by polymerizing the allyl methacrylate within the polyurethane matrix, thus preventing metal penetration during compacting.

2. Irradiation of compacted polymer-metal composite with a 14-Mrad dose. Metal powder is mixed with the treated polymer particles, compacted, and then irradiated, increasing the gel content and hence the strength.

Plan for Next Period

The next phase of work involves definition of critical fabrication conditions, cost analysis, optimization of properties, and demonstration of applications. Further attempts will be made to extend this technology to interested parties in industry. Attempts will be made to convert the batch process of preparation to a continuous preparation method.

Reports Published

- R. P. Kusy and D. T. Turner, Electrical Resistivity of a Polymeric Insulator Containing Segregated Metallic Particles, *Nature*, 229: 58-9 (Jan. 11, 1971).
- A. Malliaris and D. T. Turner, Influence of Particle Size on the Electrical Resistivity of Compacted Mixtures of Polymeric and Metallic Powders, *J Appl Phys*, 42: 614-18 (1971).
- R. P. Kusy and D. T. Turner, Radiation Chemistry of Polymers Studied by Depression of Melting Temperature, *Macromolecules*, 4: 337-41 (1971)
- R. P. Kusy and D. T. Turner, Increased Energy Deposition in a γ Irradiated Polymer via a Metallic Microphase, *Macromolecules*, 4: 266-68 (1971).

Estimated Date of Final Report

Report of work from June 1, 1971 to July 31, 1972 will be submitted September 1972

ELECTRON AND X- AND GAMMA-RAY RESEARCH FOR RADIATION PROCESSING*

Objective of Work

The objectives of this project are (1) to perform experimental research and development on methods of measurement of electron and x- and gamma-ray dose distributions in materials of interest in practical radiation processing, (2) to continue development of radiochromic dye dosimeters as accurate and precise measurement devices for radiation doses between 10^2 and 10^8 rads and as qualitative indicators for dose levels in

this range, (3) to evaluate, develop, and make available new radiation measurement techniques for specific applications by radiation-process users; and (4) to coordinate this work with associated theoretical and experimental projects (funded separately) of the National Bureau of Standards, Center for Radiation Research.

Progress and Results

During the past year extensive dose-distribution studies were made with particular emphasis on radiation-processing geometries involving electron beams of 0.3 to 10 MeV. Target geometries included seminfinite and finite slabs of one material and of more than one material, so that dose distributions across the interface of two materials could be investigated. The effect of various angles of incidence was also studied. The results show that experiments using thin radiochromic-dye films give results that agree well with theoretical calculations, even close to interfaces between materials of widely differing atomic numbers, e.g., plastic and gold. Since the response of the dosimeters does not vary with dose rate, energy, temperature, etc., the present Monte Carlo calculations may be considered to be

Spectrophotometer traces of the radiochromic dye solutions for Co-60 γ -ray doses from 0 to 100 kilorads.

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sufficient for indicating dose distributions with electron beams and x and gamma rays in complex geometries

New radiochromic-dye dosimeters were developed for monitoring profiles of high-intensity beams. These include inexpensive impregnated papers and "grainless" white plastic films, which give a permanent high-resolution image of the radiation field. Also, the development of liquid dye dosimeters (see figure) for use in radiation measurements for insect population control, food shelf-life extension, mutation breeding, blood irradiation, etc., has been completed. The response range of the liquid systems is now from 1 to 1000 krads, and, because of ease of handling and good reproducibility, they have distinct advantages over the ferrous sulfate dosimeter. The film, paper, and liquid dosimeters are now commercially available.

Several types of calorimeters of various materials (carbon, aluminum, copper, etc.) have been fabricated for calibrating the response of thin-film dosimeters for industrial radiation-process users and radiation-chemistry experimenters. These calorimeters can be used either in air or in vacuum and in high-intensity pulsed or continuous radiation fields. The first draft of an ASTM standard method of test for thin-film dosimeters has been written. A simple thin-film dosimeter package has been devised, matching the geometry and response of biological test samples (radiation-resistant bacteria) for precise radiation-sterilization calibrations.

A calibration system has been developed for dose and dose-rate studies of thin dosimeters, specifically calorimeters and dye films, that are exposed to monoenergetic-electron and to high-intensity, pulsed electron beams. Basically, a measurement is made of energy deposition by a perpendicularly incident electron beam in a thin layer of a homogeneous medium. The primary dose-measuring device is a thin aluminum calorimeter. Comparisons of the response of the system with electron-transport calculations by Berger and Seltzer have shown good agreement for incident monoenergetic beam energies of 0.5 to 1.0 MeV.

Several old and new dosimeter systems were evaluated for their response characteristics (energy dependence, dose-rate dependence, temperature dependence, response range, and reproducibility). These included clear Perspex, dyed Perspex, blue cellophane, several commercial dye films and papers, cellulose acetate, cellulose acetate butyrate, polycarbonate, and thermoluminescence systems.

The holographic interferometer, previously developed for three-dimensional dose-distribution studies of pulsed broad-spectrum electron beams, has now been used with monoenergetic electron beams. Current inves-

tigations show that this device can be used to determine the average spectrum of a beam during a single pulse. A two-beam interferometer has also been built to measure energy imparted during the irradiation of a water absorber. This device can be used for doses as low as 10 krads and can be used with either pulsed or continuous beams. The latter instrument was also used as an accurate nontactile thickness gage, measuring to about a wavelength of light.

Two high-resolution scanning photometers were designed and built for use in evaluating dose-distribution images in radiochromic films. One of these instruments is a scanning microdensitometer for use with transparencies, and the other is a scanning microspectrophotometer for use with both reflecting and transparent mediums.

Plan for Next Period

Project terminated

Reports Published

- W L McLaughlin, E K Hussmann, H Eisenlohr, and L Chalkley, A Chemical Dosimeter for Monitoring Gamma-Radiation Doses of 1 – 100 krad, *Int J Appl Radiat Isotop*, 22, 135-40 (1971)
- M Rosenstein, *Electron Depth-Dose Distribution Measurements in Polystyrene*, Doctoral Dissertation, University of Maryland, 1971
- W L McLaughlin, M Rosenstein, E K Hussmann, and J J Lantz, Jr., Scanning Densitometer for Continuous Recording of Spectral Transmission Density at Low Spatial Contrast, *J Res Nat Bur Stand, Sect C*, 75, 23-7 (1971)
- M Rosenstein, W L McLaughlin, and J Silverman, Energy Deposition of Electron Beams in Insulating Materials, in *Conf Proc Electron and Ion Beam Science and Technology, Fourth International Conference*, Los Angeles, CA, May 1970, pp 591-604, R Bakish (Ed.), The Electrochemical Society, 1970
- E K Hussmann and W L McLaughlin, Dose-Distribution Measurement of High-Intensity Pulsed Radiation by Means of Holographic Interferometry, *Radiat Res*, 47, 1-14 (1971)
- M Rosenstein, H Eisen, and J Silverman, Electron Depth-Dose Distribution Measurements in Polystyrene as a Function of Angle and Thickness, *Trans Amer Nucl Soc*, 14, 397-98 (1971)
- H Eisen, *Electron Depth-Dose Distribution Measurements in Metals and Two-Material Slabs*, Doctoral Dissertation, University of Maryland, 1971

- M J Berger, Distribution of Absorbed Dose Around Point Sources of Electrons and Beta Particles in Water and Other Media, *J Nucl Med*, 12, MIRD Suppl No 5 7-23 (March 1971)
- M J Berger, Energy Depositions by Low-Energy Electrons Delta-Ray Effects in Track Structure and Micro-dosimetric Event-Size Spectra, *Proceedings of 3rd International Symposium in Microdosimetry* Stresa, Italy, October 18-22, 1971
- M J Berger and S M Seltzer, Absorption of Electron Energy in Multilayer Targets, *Trans Amer Nucl Soc*, 14 887-88 (1971)

Estimated Date of Final Report

Open literature publications

HIGH INTENSITY RADIATION DEVELOPMENT LABORATORY OPERATIONS*

Objective of Work

- The objective of this project is to use the appropriately staffed, equipped, and maintained facilities at the High Intensity Radiation Development Laboratory (HIRDL) in preparing sources, loading and modifying portable irradiators, intermittently or continuously irradiating large and small samples, providing reference gamma fields for dosimetry standardization, and developing radiation-engineering techniques and standards

Progress and Results

At present the HIRDL inventory includes 1.9×10^3 kCi of Co-60, 66 kCi of Cs-137, and 38 kCi of Sr-90. Approximately 300 kCi of the Co-60 is being used in the Gamma Facility. Operations in the Gamma Facility totalled 35,667 hours of irradiation time given to 1,473 samples in 632 separate irradiations. Dose rates presently available in this facility range from 3×10^3 R/hr to 8.5×10^6 R/hr.

In a continuing program to upgrade the capability of the irradiation cell, a new elevator has been installed in the irradiation-cell pool to decrease the time necessary to modify loading the source plaque for its various tasks. A stepwise programmer has also been installed on the General Mills Model 300 manipulator so that a plane 4.5 ft high and 10 ft long can be automatically scanned. In-cell support irradiations ranged from pep-

perment cuttings for the BNL Biology Department to monomer-impregnated concrete pipe.

Preparation-cell usage included inspecting and loading 96 sources for Pakistan, reclaiming 26 radioactive Co-alloy tensile specimens for use in corrosion studies, upgrading the activity of the Biology Dept's Gamma Field Irradiator, inspecting Natick sources destined for Penn State, loading and unloading of various sources for the round-robin calibration, and repairing a small Cs-137 irradiator for the Biology Dept.

Maintenance tasks have continued, with the Model-8 manipulators requiring extensive care. A recheck of the absolute filters in the cell ventilation system was made using an aerosol smoke and dust photometric method. The system was 99.9% effective in removal of the test aerosol.

Plan for Next Period

Near-capacity operation of the preparation and irradiation cells will be required to accomplish the research, development, and service tasks scheduled. No major facility changes or additions are contemplated.

Reports Published

None

Estimated Date of Final Report

Continuing project

RADIATION SOURCE DEVELOPMENT, TESTING, AND EVALUATION*

Objective of Work

The objectives of this project are to develop safe, versatile, and economic standard gamma and beta sources and to devise appropriate testing, inspection, and calculation procedures.

Progress and Results

The automated pool-air-cycling corrosion apparatus was completed, and a trial run showed satisfactory functioning. An attempt to load about 0.6 MCi of Co-60 demonstrated the need of modification of the active source holder because of thermal expansion effects. Long-term automated cycling tests were initiated on high (~ 400 Ci/g) and normal (~ 80 Ci/g)

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*J I Wagner, Brookhaven National Laboratory, Upton NY 11973, Contract AT(30-2)-GEN-16

intensity cobalt sources and on inactive dummy sources, which are adjacent to and irradiated by active cobalt and cesium sources. The inactive sources, which have experienced the same corrosion conditions as the active sources, can thus be examined in the laboratory without radiation hazard.

Testing of potential cladding materials and corrosion mechanisms continued at a low level. Internally heated dummy sources partially immersed in a very dilute aqueous solution (saturated with Pyrex) showed extremely rapid build-up of solids at the source surface/liquid/vapor interface at a solution temperature of 90°C.

Analytical calculations and experimental measurements have begun in a study of the relative characteristics of irradiators containing Cs-137 and Cs-134 sources. The purpose of this work is (1) to compare analytical and experimental results obtained with pure Cs-137 and Cs-134 sources as well as with sources containing a known mixture of the two isotopes; and (2) to compare relative benefits of the two isotopes in thick- and thin-target irradiations in two-pass and

multipass irradiator systems and in thin-rectangular and thick-cylindrical source-element geometries.

The following progress has been made:

1. Two sets of sources were designed (fabricated by ORNL): one set was of the typical BNL flat type ($\frac{1}{4}$ in. thick) and the other set was a thick (1.0 in. diameter) cylindrical type. Each set contained three sources: one with pure Cs-137, one with pure Cs-134, and a third with half of each. Each source activity was less than 2 Ci.
2. An automatic programmed x-y-z positioner was designed, constructed, and installed for use in the HIRDL irradiation cell (see figure) and made use of the existing heavy-duty General Mills arm in that cell.

A thorough review of the technical and economic aspects of, and future prospects for, separated fission product radioisotopes was prepared by the Division of Isotopes Development, using AEC staff and contractor input. Participation by BNL involved primarily the

Programmed movement of GM arm automatically exposes instrumented targets to low level cesium sources in dose distribution experiments.

preparation of economic and application comparisons of existing and projected Co-60, Cs-137, and Cs-134/Cs-137 sources

Source standardization activities are discussed under the "General Engineering and Technical Assistance," reported below

Plan for Next Period

Experimental comparisons of low-activity Cs-137 and Cs-134 sources will be completed. Accelerated source corrosion testing will continue. Response techniques to maximum-credible-accident situations involving cesium sources, e.g., source rupture in a pool, will be developed. Strontium source studies will be accelerated.

Reports Published

J. Wagner and A. Oltmann, BNL Cobalt Radiation Sources, *Trans. Amer. Nucl. Soc.*, 14, 498-99 (1971)

Estimated Date of Final Report

Continuing project

DOSIMETRY AND DOSIMETER DEVELOPMENT*

Objective of Work

The objectives of this project are to devise, improve, and evaluate dosimetry methods applicable to large-scale irradiators, to consult and render assistance to USAEC contractors, and to cooperate with AEC contractors, personnel of other government agencies, industrialists, and representatives of educational and private institutions in the dissemination and exchange of information and techniques.

Progress and Results

The procedure for use of the ferrous sulfate-cupric sulfate dosimeter has undergone several revisions and has finally passed the ASTM balloting and been accepted for publication as a standard procedure. The ceric sulfate dosimetry procedure has, after several revisions, passed both ASTM Dosimetry Section balloting and Subcommittee D/20 balloting. The procedure is presently being reviewed by an ASTM editorial committee before final publication as an ASTM standard procedure. The new procedure for the Fricke dosimeter

has passed Dosimetry Section balloting and, after several revisions, has been submitted for balloting in Subcommittee D/20.

The long-term studies of solutions of radiochromic dye (RCD) cyanide powders have been completed. It was found that these solutions can be used successfully for accurate measurement of absorbed dose in the range from 5 to 500 krad. Solutions may be stored for six months or more if they are protected from ultraviolet light. Numerous solvents for the dye powders were tested — ethoxy ethanol, ethoxy methanol, dimethyl sulfoxide, and isopropanol, among others. For the work at this laboratory the isopropanol was found to be best suited with respect to dose-range reproducibility, accuracy, availability, and safety. Work continues on supported and unsupported RCD films as well as on impregnated papers.

Tests on a new 10-mm-thick ultraviolet-transmitting Lucite dosimeter manufactured in Argentina show it to be similar to the Perspex-HX dosimeter. The nonlinear usable total-dose range of this system was found to be slightly better than that of Perspex-HX (1 to 5 Mrads). The dosimeter bleaches thermally at a rate that is temperature as well as total-dose dependent.

Studies were made to test the effects of monomolecular coatings on Perspex-HX dosimeters. The four coatings used were applied by exposing the dosimeters to ammonia plasma, carbon dioxide, moist air, and helium. The purpose of the coatings was to inhibit oxygen diffusion into the dosimeters, which is related to postirradiation bleaching. No long-term improvements were noted for any of the coatings.

A large volume of standard ferric ion solution was prepared and standardized for BNL by a commercial analytical laboratory. Samples of this solution are being sent to three commercial laboratories and three institutions (government laboratories and/or universities) for cross calibration. Upon completion of standardization procedures, the solution will be made available to other institutions and commercial companies for spectrophotometer calibration.

Further calibration of the HIRDL Gamma Facility continued in this period, and several new tubes were added and calibrated. Previous calibrations were rechecked. Calibrations were made on all portable irradiator units passing through HIRDL, including one Brookhaven Portable Cesium Development Irradiator (BPCDI) and the Portable Cesium Irradiator (PCI).

An IAEA Panel of Consultants was assembled in Vienna in April, 1971, for the purpose of drafting an IAEA Food Irradiator Calibration Manual. The first rough draft of BNL's contribution to this document has

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been completed. This publication will replace the previously scheduled AEC Food Irradiator Calibration Manual.

The Fifth Dosimetry Workshop was held in February 1971, in Goleta, CA, with EG&G as host. The three main topics discussed were Radiation Dosimetry in Radiation Processing, Dosimetry in Medical Irradiation, and Dosimetry in Industrial Machine Irradiation. The Workshop was attended by about 80 people from 50 institutions, approximately one-half being from commercial companies.

Plan for Next Period

Arrangements are underway for the Sixth Dosimetry Workshop planned for June 1972. The host will be the Atomic Research Establishment at Risoe, Denmark. The theme will be Dosimetry in Radiation Research, and it is expected that dosimetry standards for medical products and food dosimetry will receive considerable discussion.

Cooperation with the IAEA and with individual foreign laboratories on international dosimetry standards and irradiator calibration will continue, as will involvement with ASTM on dosimetry procedures. Beta dosimetry, delayed this year, will be emphasized next year.

Reports Published

- F. X. Rizzo, L. Galanter, and K. Krishnamurthy, *Tabulated Dose Uniformity Ratio and Minimum Dose Data for Gamma Irradiator Design*, USAEC Report BNL-50145 (Rev.), December 1970.
- F. X. Rizzo, L. Galanter, and K. Krishnamurthy, *Tabulated Dose Distribution Data for Gamma Irradiator Design*, USAEC Report BNL-50147 (Rev.), December 1970.
- F. X. Rizzo, L. Galanter, and K. Krishnamurthy, *Tabulated Dose Distribution Data for Gamma Irradiator Design. I. Cobalt-60 Square Plaque Sources*, *Int. J. Radiat. Eng.*, 1(1): 15-48 (July 1971).

Estimated Date of Final Report

Continuing project.

PORTABLE IRRADIATOR SUPPORT*

Objective of Work

The objective of this project is to ensure the operational capability of new and reassigned pool-type

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Installing the tank for a Mark IV Research Irradiator in Lyallpur, Pakistan.

and portable research irradiators owned or financed by the USAEC. Design, construction, and operation consultation concerning irradiator operation, e.g., source handling, and irradiator elements — source elevators, conveyors, instrumentation, etc. — will be provided to foreign or domestic installations of AEC-DID interest.

Progress and Results

Photographs received from Pakistan in early 1971 showed the tank and tower for the Mark IV pool-type Research Irradiator being placed in the ground prior to construction of the building to house the unit (see figure). When later photographs showed the building in the final stage of construction, the final consignment was shipped including the isotopic radiation sources (25,000 Ci of Co-60). BNL has only one fully approved isotope shipping container; since that cask is in periodic

use for routine domestic shipments, it seemed unwise to release it for the several-month round trip to Pakistan. One of the old ETR casks** (previously used for overseas shipments but currently nonapproved) was set aside for the shipment. An added advantage of its use is that the ETR cask may be left with the irradiator in Pakistan for source storage in the unlikely event of tank leakage. The Pakistani AEC concurred in this decision and, after the understandable delays in gaining formal approval from all concerned parties, a Department of Transportation Special Permit was issued for the shipment.

Final installation of the irradiator by a BNL engineer will occur in early 1972. This will be followed by a visit of a dosimetry expert to assist the Pakistani researchers in calibrating the unit.

Shipboard Irradiator II, after its short-term use in Louisiana and subsequent shipment to Korea, was installed in that country by a BNL technician. Repair of some minor damage suffered in use in Louisiana and in shipment was necessary before the unit could be made operational. Of particular concern was the fraying observed in the hoist cable. The cable was replaced with one of finer wire construction which should reduce the possibility of a recurrence of this problem.

After thorough testing of the unit and instruction of personnel by the technician, the irradiator was turned over to the Korean operators.

When the irradiator was being packaged in Louisiana, the radiation-monitoring equipment seemed faulty. Rather than risking the sending of defective instruments to Korea, they were returned to BNL, and replacements were sent by air freight to Korea.

During routine operation of Shipboard Irradiator III on loan to the University of Nebraska, the hoist cable failed. A new cable, retained at BNL for emergencies, was sent by air freight to the University, where it was installed with the assistance of a BNL engineer returning from a meeting in California. The new cable was also of finer wire construction such as that installed in Korea. The unit was examined carefully for additional damage but was found to be in excellent condition. After cleaning, lubrication, and some minor adjustments, the irradiator was returned to operation.

Plan for Next Period

Installation of the Mark IV Pool Type Research Unit in Pakistan will be completed.

Transfer of the Brookhaven Portable Cesium Development Irradiator (BPCDI) No. 1 from Argentina to Brazil is planned. The Indian AEC has suggested the possibility of moving BPCDI No. 2 from Trombay to a coastal location.

The Portable Cesium Irradiator (PCI) may be transferred from its current California location to Louisiana.

Reports Published

None

Estimated Date of Final Report

Continuing project

GENERAL ENGINEERING AND TECHNICAL ASSISTANCE*

Objective of Work

The objective of this project is to maintain the technical cognizance required by the programs conducted at the High Intensity Radiation Development Laboratory (HIRDL) or involved in HIRDL consultations. Activities include the planning and designing of advanced irradiators, maintenance of source standards, assisting of off-site investigators, upgrading of on-site facilities to meet changing service requirements, and continued examining of the extent, nature, and control of undesirable side effects of radiation processing.

Progress and Results

Technical assistance requests from commercial organizations, universities, and government or government-sponsored facilities continued at a high level, most involved dosimetry, calibration, and shielding problems. Irradiator design activity was slight.

Source calibration and standardization activities were transferred to this group during this period. Source calibration at BNL consists of standardization of a few sources by calorimetry, then using these standard sources as references to assay the remainder of the inventory. Both the calorimeter, used for standardization, and the source assayer have been in use at HIRDL for many years. Recent concern over the existence of a truly adiabatic system in the calorimeter has led to extensive modification and testing. With these changes,

**The ETR casks were previously used to ship cobalt sources irradiated in the Experimental Test Reactor (ETR).

*J. H. Cusack, Brookhaven National Laboratory, Upton, NY 11973, Contract AT(30-2)-GEN-16

coupled with theoretical calculations, it has been determined that the system remains adiabatic over much more adverse conditions than exist during normal operation

Previous data collected from the source assayer relating the activity of an unknown source to that of a standardized source were taken from a moving chart paper. This technique, and the use of instrumentation with a high rate of drift, led in some cases to gross errors. This problem has been corrected with the design and fabrication of highly stable instrumentation containing a digital readout

A major portion of the effort relating to radiation-source standardization at BNL is in conjunction with an interfacility cross-calibration program, the purpose of which is to set up industry-wide (and, possibly, international) standards. The present participants in this program are General Electric Co., Neutron Products, Inc., Atomic Energy of Canada Ltd., Commissariat à l'Energie Atomique (Saclay, France), the Radiochemical Centre (Harwell, England), and Brookhaven National Laboratory

An IAEA-sponsored Consultants Meeting on Source Standardization was held in Baden, Switzerland, in August 1971. This meeting, an outgrowth of the BNL source-standardization efforts, was attended by English, French, German, Swiss, Canadian, and US (BNL) representatives. Major emphasis in the resulting consultants report was on source-specification requirements, recommendations for future work stressed isodose plotting and spectrum-distribution determinations by experimental and Monte Carlo calculation techniques

Plan for Next Period

Technical-assistance requests are expected to continue. A growing interest in the development, production, and use of cesium sources is especially apparent.

Investigation of new irradiator concepts will continue.

Source standardization activities will continue in cooperation with the IAEA and with commercial organizations.

Reports Published

None

Estimated Date of Final Report

Continuing project

FIELD DOSIMETRY STANDARDIZATION*

Objective of Work

The objective of this work is to ensure the accuracy and reproducibility of all gamma dose-measurement data generated under AEC sponsorship or in support of AEC food and process irradiation interests.

Progress and Results

Dosimetry and/or calibration procedures performed by personnel at Michigan State University, Bulk Grain Irradiator facility, University of Hawaii (research irradiator), Hawaii Development Irradiator (HDI) facility, University of Washington, and Bureau of Commercial Fisheries at Seattle and Gloucester were reviewed in depth.

The dosimetry section of the strawberry petition was expanded to include the irradiations performed at the University of California in the Portable Cesium Irradiator (PCI) and the Mark II Research Irradiator.

Reviews of the dosimetry sections of the marine products petition and the petition for papayas are in progress.

Technical assistance in dosimetry, dose calculation, and dose standardization was given to several companies and universities. Among the recipients were Gamma Processing Corporation, Radiation Technology, Inc., Tampa Bay Engineering, University of Massachusetts, New York State Atomic and Space Authority, Korean Government, Medical Department, BNL, AME, Inc., Radiation International, Inc., Brazilian AEC, Argentine AEC, Plastics Division, Allied Chemical Corp., U.S. Veterans Administration, Neutron Products, Inc., NUMEC, and the Radiation Division of BNL.

Plan for Next Period

Brookhaven National Laboratory will continue to review reports by AEC contractors prior to their publication. Dosimetry updating or recalibration will be conducted at AEC facilities upon request.

Final drafts of the strawberry and marine-products FDA petitions will be completed. All other FDA petitions will undergo in-depth reviews and evaluation before submission.

*F. X. RIZZO, Brookhaven National Laboratory, Upton, NY 11973, Contract AT(30-2)-GEN-16

Reports Published

None

Estimated Date of Final Report

Continuing project

**PATENT REVIEW – PROCESS
RADIATION DEVELOPMENT*****Objective of Work**

The objective of this search by the Isotopes Information Center (IIC) is to review, categorize, abstract, and compile U S , British, and Canadian patent literature regarding process radiation to provide backup information for the DID Process Radiation Staff. In addition, other foreign patents are being included as spinoff from the Center's retrieval system.

The audience for this review will include all potential manufacturers of materials produced by irradiation as well as manufacturers of radiation-processing equipment. This review should provide those manufacturers with information concerning processes or equipment that have been developed and thus reduce the possibility of patent infringement. The review should also be valuable to both manufacturers and research and development institutions to help direct the effort of new work in the field. If kept current, this report should be a valuable index to the growth of the radiation-processing industries.

Progress and Results

During the last year Part 1 was completed. It contains information on more than 700 U S patents for the years 1950 through 1968 as well as a partial listing of the 1969 and 1970 patents.

*R E Greene, P S Baker and H S Warren, Oak Ridge National Laboratory, P O Box X, Oak Ridge, TN 37830, Contract W-7405-eng-26

The subject index, although not exhaustive, contains approximately one thousand terms taken from compounds given in the title, from typical starting materials and end products, and from basic processes (e g , synthesis, polymerization). An analysis of the findings, from which trends may be depicted, is included.

Patent cards were reproduced as an appendix to the full report and were prepared for all in-scope patents. Each card contains patent number, country, assignment date, application date, priority date, category (process or irradiator design), title, assignee, patentee, code class, international code class (where applicable), and an abstract when available and not prohibited by copyright laws. When no abstract was available, the U S Patent Gazette description was used as a substitute. When such sources were still inadequate, pertinent information on the radiation application was reproduced from the patent itself.

An AEC-generated announcement of the availability of the document was made on August 12, 1971.

Plan for Next Period

Indexing of Part 2, British and Canadian patents, is following the same procedure as that for Part 1, and subject and assignee indexes will be prepared. Patent cards will be compiled and listed numerically in an appendix to the report. The basic report will include findings and graphs showing growth and trends.

A 1969 and 1970 supplement to Part 1 is in preparation.

Reports Published

R E Greene, H S Warren, and P S Baker, *Patent Literature on Process Radiation and Irradiator Design Part I United States Patents 1950 Through 1968*, USAEC Report ORNL-IIC-24 June 1971.

Estimated Date of Final Report

Publication of Part 2 is expected in late 1972.

RADIATION PRESERVATION OF FOODS

Astronaut Buzz Aldrin demonstrates for the camera how to make a sandwich with irradiated bread. Both irradiated bread and bread from irradiated flour have been tested satisfactorily on several Apollo flights. (Photo courtesy of Manned Spacecraft Center, National Aeronautics and Space Administration, Houston, Texas.)

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Radiation Preservation of Foods

The objective of the Radiation Preservation of Foods Program is to develop the process of low-dose radiation treatment of food (pasteurization, disinfestation, and control of pathogens) to the point of commercialization. Technical feasibility and cost-benefit studies have identified East Coast finfish, papayas (and mangoes), strawberries, shrimp, and mushrooms as being the most promising products of all those studied, for these, a concerted research and development effort is to continue in order to bring the irradiation process to the point of commercialization. The majority of the work to be performed must necessarily be a direct input toward the preparation and presentation of Irradiated Food Petitions to the Food and Drug Administration (FDA). Since radiation is classified as a food additive, all the requirements imposed by the FDA for chemical-additive petitions are also imposed on radiation processing.

During 1969, the program was reduced because of budgetary limitations, and only that work necessary to complete the accumulation of data for the strawberry and papaya petitions has been continued. There is also some work continuing on East Coast finfish, but this effort has been reduced until advanced microbiological studies are completed and the AEC has reviewed all the data. The AEC has completed a 2-year animal feeding study on irradiated strawberries and is preparing a petition to the FDA requesting that a regulation be issued allowing unlimited human consumption of this

product. A second animal-feeding study with rats and mice on irradiated papayas has been completed. A reproduction study with dogs is scheduled for completion in early 1972, and the petition for papayas should be submitted to FDA approximately 6 to 8 months later.

The AEC is also completing a technical feasibility study on the use of irradiation in conjunction with centralized retail cutting of fresh red meats. At the present time, any decision to conduct 2-year animal-feeding studies will await the completion of the U.S. Army's feeding study on radiation sterilized beef, which is scheduled for completion within the next 3 years.

The AEC responsibility for this program is divided between the Division of Isotopes Development (DID) and the Division of Biology and Medicine (DBM), with DID responsible for overall program direction, the technology and feasibility of the process, and the selection of items in the program. Once the technology has been established, DBM initiates and conducts the necessary wholesomeness and safety studies designed to demonstrate that consumption of the irradiated food product constitutes no public health hazard. A list of DBM contracts in this field may be obtained by contacting that division directly.

Projects concerned with design and construction of irradiators for the food program and with dosimetry in all irradiators in the program are also reported in the Radiation Processing section of these summaries.

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MARINE PRODUCTS DEVELOPMENT IRRADIATOR PROGRAM*

Objective of Work

The primary objective of this program is to conduct radiation research on a semicommercial scale to develop data that will demonstrate the feasibility of extending the shelf life of irradiated fresh seafoods. The program also provides irradiation services for other Federal, state, and municipal agencies or private industry.

Progress and Results

The procedure developed to attain the minimum effective dose is as follows: (a) purchase commercially produced haddock fillets wrapped in conventional cellophane units in 10-lb shipping containers, (b) irradiate them to doses of 50 to 250 krad at increments of 50 krad, (c) store samples of the controls and of each of the five dose levels at 33°F, 42°F, and 46°F, and (d) analyze the wrapped cellophane packages of fillets at appropriate storage intervals for total bacterial count and subject duplicate samples to sensory (odor) evaluation.

Haddock caught and processed in January, April, and June showed that the minimum effective dose level as determined by bacterial counts and odor rejection fell between 100 and 250 krad. Further testing is necessary to establish more precisely the minimum effective dose.

In cooperation with private industry, a study of the effects of ionizing radiations at different rates is being made. The MPDI radioactive source imparts energy at a level of about 10^2 rads/sec in contrast with the ultrahigh rate of 10^{13} rads/sec imparted by machine-generated pulsed electrons. The effects of ionizing radiation from different sources and at different dose rates have been compared before but not to the magnitude of dose-rate difference in the current study.

Generally, there is good agreement between the two systems with respect to bactericidal effect against *Bacillus pumilus* and *Streptococcus faecalis*. There is a difference indicated between the two systems of ionizing radiation with respect to material damage mechanisms in complex molecular structures. Adverse chemical effects seem to be more pronounced when caused by ionizing radiation of low dose rate (10^2 rads/sec) than by an ultrahigh (10^{13} rads/sec) rate. Materials tested so far include insulin, plasma, enzymes, fibrino-

gen, and detergents. A report on the damage due to rate effects will be published.

Plan for Next Period

The plan for the next period is (1) to perform at least two more simulated shipping studies to establish the minimum effective dose and carry out actual shipping studies and (2) to continue to provide irradiation services and maintain the MPDI at peak efficiency.

Reports Published

- L. J. Ronsivalli, J. D. Kaylor, E. J. Murphy, R. J. Learson, and M. S. Schwartz, Studies in Petition-Oriented Aspects of Radiation Pasteurization of Fishery Products, in *Preservation of Fish by Irradiation*, Report of a Panel, Vienna, Dec 15-19, 1969, IAEA, Vienna, pp 1-11, November 1970 (STI/PUB/196).
- J. D. Kaylor, E. J. Murphy, J. H. Green, and W. S. Hill, *Marine Products Development Irradiator Program Irradiated Fresh Shrimp Shipping Studies*, USAEC Report TID-25543, October 1970.

Estimated Date of Final Report

Continuing project

RADIATION PASTEURIZATION OF FRESH MEATS AND POULTRY*

Objective of Work

The overall objective of this work is to provide a technology by which centralized cutting and packaging of fresh meats and poultry may be achieved. The technology will (1) control microbial spoilage by use of ionizing radiation plus conventional refrigeration, (2) control pigment and lipid oxidation principally through effective atmosphere control and packaging as well as possible additives such as ascorbic acid, and (3) minimize fluid exudation with drip-controlling agents. Controlling these deteriorative processes through such an integrated procedure will make feasible the complete preparation of fresh red-meat consumer cuts and poultry at a central location to provide sufficient product life to permit distribution to retail outlets under refrigeration.

*J. D. Kaylor, National Marine Fisheries Service, Atlantic Fishery Products Technology Center, Emerson Ave., Gloucester, MA 01930, Contract AT(49-11)-3005

*W. M. Urbain, Department of Food Science and Human Nutrition, Michigan State University, East Lansing, MI 48823, Contract AT(11-1)-1689

Progress and Results

The procedure developed through laboratory-scale tests to attain the objective is as follows: (a) treating the meat with condensed phosphates (e.g., tripolyphosphate) to control liquid exudation and to maintain normal red color; (b) packaging each cut in oxygen-permeable film, grouping of a number of such packaged cuts, and placing them in a bulk container which is sealed under vacuum; (c) irradiating the bulk-packaged product at 100 to 200 krad with gamma radiation; (d) distributing and storing under refrigeration; and (e) opening the bulk container at the retail outlet, at which time the normal red color develops and the product has a normal display-case life (3 days).

An attempt to obtain a picture of the microbial outgrowth patterns of beef handled according to the proposed procedure is in progress, which involves examination of the beef for (a) numbers of organisms present (both mesophilic and psychrophilic) and (b) coliform, *Clostridium perfringens*, Staphylococcus (coagulase positive), and salmonella microorganisms.

Knowledge of the outgrowth pattern is important for characterizing the sensory spoilage which ultimately occurs and also for assessing any health hazard which might develop.

The primary purpose of the phosphate treatment is to control "drip" (weepage of liquid) from the meat cut. Two methods of incorporation of the phosphate have been studied: dipping or spraying a water solution and injecting through hollow-needle pumping devices. On the basis of similar phosphate additions to certain meats, limits of 0.5% added phosphate and 3.0% total weight gain (including solvent water) have been set.

In the final step of the handling procedure, the meats are exposed to air to develop and maintain the normal red color. Chemical analyses have determined that the oxygen of the air oxidizes the fat present in the meat. This is true also of meats as they are now handled. The addition of ascorbic acid prevents this oxidation. The level of fat oxidation, however, is very low and efforts to correlate taste panel evaluations with chemical analyses have not established that a flavor change is caused by this oxidation.

Oxygen must be removed from the primary package to secure meat color stability. Design of a packaging system to accomplish this is being undertaken.

Plan for Next Period

Future work will be aimed at completing the microbiological studies, determining the best method of phosphate addition, and designing an effective pack-

aging system. Meat handling procedures prior to cutting also will be investigated.

Reports Published

W. M. Urbain, G. G. Giddings, and W. W. Ballantyne, *Radiation Pasteurization of Fresh Meats and Poultry*, Annual Report, USAEC Report COO-1689-6, 1971.

Estimated Date of Final Report

Continuing project.

A STUDY OF CONTROL OF PUBLIC HEALTH PROBLEMS USING IRRADIATION*

Objective of Work

This project was undertaken to determine the impact of irradiation on microorganisms of public health significance in ground red meat, the behavior of the residual microflora during simulated commercial handling of ground red meat, and the usefulness of commonly accepted evaluation systems for public health safety in meat products.

Progress and Results

Most of the exploration of usefulness was based on a low dose application (68 krad), with recognition that the ultimate dose level would depend on many factors. Various dose levels were applied when individual factors such as the resistance of a specific species of microorganism were to be studied. Pathogens and indicator organisms were studied to determine the relative sensitivity of representative species. Ground red meat products were chosen because their sanitation is one of the major challenges of the industry and because the quality of these products seems to be a plateau of minimal tolerance by regulatory agencies for public health protection.

Previous data indicated that ground red meat commercially distributed is near the threshold of microbial and detectable organoleptic spoilage. Shelf life beyond the point of retail sales is only 1 or 2 days. Even as improved by irradiation, this product does not fit into the ultimate goal for maximum public health protection, which would consist of central processing, packaging, and irradiation. Irradiation would make central

*R. Burt Maxcy, Department of Food Science and Technology, University of Nebraska, Lincoln, NB 68503, Contract AT(11-1)-2038.

Comparative counts and LD₅₀ values calculated from mortality figures of chick embryos

Treatment	MPN per ml	Plate count per ml			LD ₅₀ (Number of cells)
		PCA	BGA	SSA	
Unirradiated	2.0×10^9	1.0×10^9	9.6×10^8	3.2×10^8	0.5
Irradiated	5.7×10^5	6.4×10^5	3.5×10^5	1.6×10^4	1.3

processing and wide distribution possible through extension of shelf life. Furthermore, even the low dose of 68 krad reduces the pathogenic and toxin-producing organisms by 90 to 99%, thus providing a public motivation for acceptance of the irradiated product.

During the last year, basic data have been obtained on which the generalizations in the preceding paragraph are based. The applicability of the commonly accepted methods for determining salmonellas, coliforms, and other members of the Enterobacteriaceae was determined. Cell injury affects the number of microorganisms recovering on selective media, and the reduced recovery after irradiation is of the same general magnitude found when other methods of cell injury are involved.

Relative infectivity of irradiated and unirradiated salmonella cells was determined using the egg yolk sac challenge technique. In addition, counts were made with most probable number (MPN) technique and plate count on plate-count agar (PCA), brilliant green agar (BGA), and salmonella-shigella agar (SSA). Data (Table) indicate that irradiated cells are less capable of initiating growth even in the highly favorable environment of an egg yolk sac. The fear of increased infectivity from salmonellas after irradiation is therefore unfounded and will remain so until some unforeseen data to the contrary can be presented.

Since irradiation naturally alters the microflora, the nature and significance of the residual microorganisms were studied. A major share of the residual microorganisms belongs to the *Moraxella* group, which includes *Moraxella*, *Acinetobacter*, and perhaps closely related genera. Certain of the species were extremely radio-resistant. Exploratory work on the source and significance of these bacteria indicated that they were widespread in the various inputs to ground beef. In ground beef irradiated at 68 krad, the surviving members of the *Moraxella* group grew slower than other types of microflora both at 5°C and at 25°C. Thus, irradiation does not add to the significance of these organisms.

Work on the Enterobacteriaceae indicated that there are a great number of these contaminants present in ground meat. The diversity of isolates indicated that many of the microorganisms presently associated either directly or indirectly with gastro-intestinal disorder may have their origin in ground meat products. There are many diseases caused by food-borne microorganisms that might be controlled or eliminated by a reduction in the number of microorganisms in products taken home.

Plan for Next Period

Fundamental studies of radiation pasteurization of ground red meat will be continued, with particular emphasis on higher radiation doses at the pasteurization level.

Reports Published

- N. P. Tiwar and R. B. Maxcy, Impact of Low Doses of Gamma Radiation and Storage on the Microflora of Ground Red Meat, *J. Food Sci.*, 36: 833-34 (1971).
 R. B. Maxcy, N. P. Tiwar, and C. C. Anagnostis, *Study of Control of Public Health Problems Using Irradiation, Annual Report, May 1, 1969-April 30, 1970*, USAEC Report COO-2038-2, 1971.

Estimated Date of Final Report

Continuing project

RADIATION TECHNOLOGY IN CONJUNCTION WITH POST-HARVEST PROCEDURES AS A MEANS OF EXTENDING THE SHELF LIFE OF FRUITS AND VEGETABLES*

Objective of Work

The overall objective of this program is to demonstrate the use of radiation in controlling post-harvest

*E. C. Maxie, University of California, Davis, CA 95616
 Contract AT(11-1) 34, P 80

Percent Marketable Shasta Strawberries After Irradiation and Shipment

Shipment method	Destination	Date shipped	Dose krad	Percent marketable fruit		
				Before shipment	On arrival	After 3 days of simulated storage
Truck	Minneapolis	7/16/70	0	98 92	94 93	90 31
			150		91 97	91 46
			235		91 52	90 92
Truck	Atlanta	7/31/70	0	98 54	93 66	86 67
			150		95 75	93 19
			235		94 80	90 05
Truck	Atlanta	8/26/70	0	98 67	85 26	87 59
			150		88 47	86 45
			235		90 96	86 60
Truck	Atlanta	9/17/70	0	96 27	85 01	45 63
			150		91 17	81 83
			235		92 70	86 97
Truck	Atlanta	10/1/70	0	98 40	94 43	84 49
			150		95 58	92 99
			235		94 78	89 80
Surface ship	Honolulu	8/16/70	0	97 96	49 98	19 66
			150		92 64	80 34
			235		89 89	84 18
Surface ship	Honolulu	8/21/70	0	98 66	95 51	85 26
			150		97 10	87 50
			235		94 97	85 60

disease of selected fruits and vegetables. Work performed in the terminal year of the contract was aimed at determining the effectiveness of irradiation in controlling the post-harvest incidence of decay in California strawberries after commercial shipping, storage, and handling.

Progress and Results

During the 1970 California harvest season, seven test shipments of irradiated strawberries were made, using near-commercial conditions to determine the effectiveness of irradiation in controlling the incidence of decay after commercial handling. Preliminary results were reported last year. The irradiation levels used were 150 and 235 krad at the point of maximum absorbed dose, with a uniformity ratio of 1.35.

Both irradiation and the physical operation of the pilot plant irradiator tended to contribute (although not significantly) to the slightly higher amount of fruit that suffered from mechanical injury as a result of transportation, however, the market acceptability of the fruit was judged not to be affected.

The Portable Cesium Irradiator (PCI) used for the 1970 studies did less mechanical injury to the fruits than did the Mobile Gamma Irradiator (MGI) used for the preliminary 1969 studies.

These tests (Table) showed irradiation to be significantly effective in controlling decay during and after shipments of long duration such as the marine shipment to Honolulu. Transcontinental shipments via refrigerated trucks showed less effect, although decay control was consistently better with the irradiated samples.

Irradiation to a maximum dose of 150 krad (111 krad minimum) has a slight but consistent superiority to controlling decay over the maximum dose of 235 krad (173 krad minimum).

Plan for Next Period

This contract terminated on June 1, 1971.

Reports Published

E. C. Maxie, *Radiation Technology in Conjunction with Post Harvest Procedures as a Means of Extending the Shelf Life of Fruits and Vegetables*, Annual Report, USAEC Report UCD-34P80-9, 1971.

Estimated Date of Final Report

This project has been terminated.

HAWAII FOOD IRRADIATION PROGRAM: STUDY OF DOSIMETRY, TOLERANCE, AND SHELF LIFE EXTENSION RELATED TO DISINFESTATION OF FRUITS AND VEGETABLES*

Objective of Work

The primary objective of this project is to develop data in support of a petition to the Food and Drug Administration (FDA) for approval of radiation treatment and early commercialization of fruit and vegetable disinfestation by ionizing radiation. Papaya is the primary product under study because of the concentration, organization, and growth of the industry. Hawaii has a vital interest in radiation disinfestation of papayas and other fruits and vegetables. The State's exports require development of effective and economic treatments for products subject to quarantine measures.

*A M Dollar, Department of Agriculture, State of Hawaii, Contract AT(26-1)-374

Present treatments are batch processes that impose delays prior to packaging and shipment. Radiation treatment not only effectively controls insects subject to quarantine but also is a continuous process applicable to packaged products, allowing efficient handling.

Progress and Results

Treatment of papaya at an absorbed dose of more than 21 krad provides probit 9 assurance that no fruit flies will emerge to create a hazard at the destination. Comprehensive studies have demonstrated clearly that papaya and other fruits and vegetables can tolerate absorbed doses in excess of those required to achieve disinfestation. Extensive data collected for papaya handled under commercial-scale conditions supported earlier University of Hawaii research studies showing that there were no adverse changes in appearance, color, aroma, texture, or nutritional values during the market life of papaya treated at absorbed doses of less than 150 krad. These data have been evaluated and are being prepared for FDA petition action.

Optimum disinfestation treatment of papaya by ionizing radiation requires continuous flow of fruit from receipt at the processing plant until ready for

Papaya decay control. (Saturated vapor/hot water treatment)

shipment. For treatment of the quantity of fruit on a continuous basis (now a batch system), a hot water treatment that can be carried out in 30 sec or less must be adopted. The conventional hot water dip treatment at $118^{\circ} \pm 2^{\circ}\text{F}$ for 20 min was compared with high-temperature, short-time treatments with a continuous system that consisted of a pump, heater, and spray unit recycling only 30 gallons of water. After hot water treatment at the desired temperature, fruits were irradiated at a minimum absorbed dose of 25 krad and then stored under normal commercial conditions.

Fruit decay is reduced two-fold by treatment at $145^{\circ} \pm 5^{\circ}\text{F}$ for 20 sec when compared to conventional treatment. Another two-fold improvement in marketing life is obtained by combining the treatment with a disinfection dose of 25 krad. Treatment at 145°F for 25 sec increases the risk of chill injury (surface scald) when fruits are stored at $50^{\circ} \pm 5^{\circ}\text{F}$. The longer treatment delays color development by 1 day when fruits are stored at $75^{\circ} \pm 2^{\circ}\text{F}$. Time-temperature tolerance curves were determined for the range of 100 to 150°F (see figure).

Plan for Next Period

The petition will be completed and submitted to FDA once biological safety evaluations are finished. Commercial-scale marketing evaluations will be undertaken once FDA approval can be obtained. The facility can treat commercial quantities on a cost recovery basis while developing a suitable commercial facility near the production center of Hawaii.

Reports Published

A. M. Dollar, M. Hanaoka, G. A. McClish, E. K. Akamine, J. H. Moy, E. Ross, and K. Ohinata, *Hawaii Food Irradiation Program*, USAEC Report NVO-374-17, Vols. 1 & 2, 1970.

Estimated Date of Final Report

Continuing project.

LOAN OF 50,000 CURIES OF Co-60 FOR USE IN RADIATION FACILITY FOR RESEARCH IN FOODS*

Objective of Work

The aim of this project is to make available, on the campus of the University of Massachusetts, a 50,000-Ci

Co-60 radiation facility to be used for research on the preservation and public health aspects of food and food products. Four areas will be investigated: (1) effect of a simultaneous heating and radiation process to inactivate *Salmonella senftenberg* in egg, (2) stability of food lipids to radiation, (3) degradation of carbohydrate systems by radiation, and (4) sensitivity of psychrophilic spoilage microorganisms in seafoods to radiation. In addition, arrangements are being made to make the facility available to the entire University and to Amherst, Smith, Mt. Holyoke, and Hampshire Colleges.

Progress and Results

Installation of the radiation facility began in January, 1971, and the Co-60 was delivered on April 1. Preliminary dosimetry and in-service training of operating personnel by the AEC and the facility contractor have been completed. The facility is on a standby basis until final system security and monitoring measures can be completed.

Plan for Next Period

Research will be initiated when final monitoring and security measures are installed.

Reports Published

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