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RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR MARCH 1970

A. F. Rupp

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ORNL-TM-2965

Contract No. W-7405-eng-26

Isotopes Development Center

RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR MARCH 1970

A. F. Rupp

APRIL 1970

OAK RIDGE NATIONAL LABORATORY
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RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR MARCH 1970

A. F. Rupp

RADIOISOTOPE PRODUCTION AND MATERIALS DEVELOPMENT

NEUTRON PRODUCTS - 08-01-01

A. Biomedical Radioisotopes

1. Thulium-171

Nothing to report.

2. Potassium-43

The objectives of this project are: to prepare potassium-43 by the $^{43}Ca(n,p)^{43}K$ reaction, using isotopically enriched ^{43}CaO targets, in quantities sufficient for medical and biological experiments; to define a method for separating potassium-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-42 has been used, primarily in animals, to locate tumors, to tag red blood cells, and to study blood flow, but it has very limited use in humans because of its high beta energy (3.53 MeV), its high-energy (1.52 MeV) gamma ray, and its relatively short half-life (12.4 hr). Potassium-43, with a half-life of 22.4 hr and gamma-ray emissions of 0.373 and 0.617 MeV, is more suitable for metabolic and clinical studies, because the lower dose rate would permit multiple doses. It has been suggested as a possible tool for studying blood flow through the heart and for diagnosing myocardial infarctions or immune rejection of transplanted organs. It has also proved capable of diffusing through a kidney with greatly reduced renal function, providing a good image of this organ.

A 90.1-mg target of enriched ^{43}CaO (~62%) was irradiated 65 hr in a fast flux of approximately 10^{15} n/cm^2.sec; the potassium-43 process yield was 10.7 mCi. An ion-exchange process was carried out using Bio-Rad AG 50W-X8 (100-200 mesh) with pneumatic over-pressure to increase the flow rate to 3 ml/min, or 7.8 ml/min.cm^2. The column was a commercial Bio-Rex 0.7-cm glass column with the reservoir flange modified to take a ground glass ball joint for easy in-cell manipulation. Air pressure was supplied from a portable precision regulator assembly equipped with an automatic solenoid cutoff valve which closes in the event of a fitting failure. Tests with this system prove that flow rates of up to 16 ml/min can be attained using 28 psi even with 200- to 400-mesh resin. The flow rate is a linear function of the pressure which can be controlled precisely with the regulator. Improved temperature control was achieved by wrapping the column with a foam neoprene insulation. The eluting solution is heated before it is placed in the reservoir and then it is maintained above 80°C by use of a heat lamp and an aluminum reflector on the opposite side of the reservoir. The column was operated at approximately 80°C.

In addition to a significant shortening of the process time, the separation between the sodium-24 contaminant (resulting from activation of sodium in the target) and the potassium-43 was increased. The column was eluted with 100 ml of 0.2 N HCl followed by approximately 100 ml of 0.5 N HCl. The sodium-24 was eluted between 34-39 column volumes (130-150 ml), including the first 100 ml of 0.2 N HCl. The potassium-43 product was collected between 45-53 column volumes (170-200 ml). The eluant was collected in 10-ml fractions and the activities in each were examined with a Ge(Li) detector. Platinum and gold-199 activities were found in each fraction indicating that these contaminants, which originate from the calcining of the target in platinum, are in colloidal states. Iridium-192 originating from the same source is also observed, but it is apparently washed out of the column before the sodium-24 is eluted.

Nine millicuries of the product was shipped to Johns Hopkins under our cooperative program. Two patients and a dog were scanned with excellent results. The dog had been studied in an earlier experiment[1] and, by comparison with the earlier scans, it was concluded that the myocardial infarct which had been visualized in the earlier scan had healed. An autopsy will be done to confirm this. The results of this work will be reported in a paper which has been accepted for the July 1970 national meeting of the Society of Nuclear Medicine.

The beta spectrum of potassium-43 is being studied with a beta spectrometer using data taken with a source prepared from the most recent product. The next experiment will test the use of finer (200-400) mesh resin and faster column flow rates.

3. Phosphorus-33

Nothing to report.

4. Rubidium-84

The purpose of this project is to determine the yield and purity of rubidium-84 prepared by fast neutron irradiation of enriched strontium-84 by the $^{84}Sr(n,p)^{84}Rb$ reaction.

Experimental targets were irradiated in fluxes of approximately 10^{14} and 10^{15} n/cm^2.sec for various periods of time. The experimental results are shown in Table 1.

Table 1. Rubidium Yield from Neutron Irradiation of Strontium-84

Flux, n/cm^2.sec			Irradiation	Yield of Rb, μCi/mg ^{84}Sr		
Thermal	Resonance[a]	Fast	Time, days	^{83}Rb	^{84}Rb	^{86}Rb
2.5×10^{14}	1.6×10^{13}	1.2×10^{14}	5	0.048	7.3	0.093
			10	0.13	16.6	0.43
2.6×10^{15}	1×10^{14}	8×10^{14}	5	0.28	33.8	4.83
			10	0.33	43.5	12.7
7.5×10^{14}	6×10^{13}	2.5×10^{14}	23	0.45	16.2	47.2
			46	0.40	19.2	68.8

[a]Resonance flux is for gold not cadmium.

[1]A. F. Rupp, Radioisotope Program (8000) Progress Report for January 1970, ORNL-TM-2876, Oak Ridge National Laboratory, p. 1.

The increase in rubidium-84 yield was not proportional to the increase in the fast neutron flux, which may indicate burnout of the rubidium-84 by a thermal or epithermal (n,γ) reaction. The rubidium-86 impurity increased rapidly with the higher neutron fluxes and longer irradiation times. The increase in the rubidium-86 is due to the growth and subsequent activation of rubidium-85 resulting from the decay of strontium-85m and strontium-85. The rubidium-83 impurity content, usually from 0.7-2.4% of the rubidium-84 activity, is an undesirable contaminant due to its 0.52- to 0.53-MeV gammas which interfere with the use of the rubidium-84 positron annihilation photons. The rubidium-83 is produced by the $^{84}Sr(n,2n)^{83}Sr \rightarrow {}^{83}Rb$ reaction. The present work on variables in rubidium-84 production as influenced by neutron flux and irradiation time is completed.

B. Research Radioisotopes

1. Chlorine-36

Nothing to report.

2. Lanthanum-137

Nothing to report.

C. Isotopic Power

1. Thulium-170

The objectives of this research are to provide sufficient data on thulium-170, which has been proposed as an isotopic power source for short-duration missions, to permit reasonable assessment of its potential application, and to permit preliminary engineering design of power sources. The economics for reactor production of thulium-170 (128.5 day; E_{β^-av} = 0.32 MeV) from natural thulium-169 are favorable and the sesquioxide offers a promising fuel compound, having a practical specific power of approximately 2 W/g and a power density of approximately 16 W/cm^3. The study will include measurements of thermal conductivity and thermal diffusivity, leach rates for safety analysis, high-temperature compatibility with suitable containment metals, vapor pressure, and fuel form densification.

Two tungsten Knudsen cells of different capacities have been designed for measurement of the vapor pressure of Tm_2O_3 and Tm_2O_3-Yb_2O_3 mixtures up to approximately 2300°C and are being fabricated.

The preliminary thermal conductivity data reported in December 1969 (ref. 2) have been checked and additional measurements were made on an instrumented Yb_2O_3 pellet. The revised data corrected to theoretical density are shown in Table 2. The thermal conductivities, λ (W/cm.°K), have been fit in the least squares sense as a linear function of the absolute temperature, °K. The equations for Yb_2O_3, Tm_2O_3, and 0.46 Yb_2O_3-0.54 Tm_2O_3, respectively, are:

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[2]A. F. Rupp, Radioisotope Program (8000) Progress Report for December 1969, ORNL-TM-2828, Oak Ridge National Laboratory.

$$1/\lambda = 0.02350T + 8.783 \quad (1)$$

$$1/\lambda = 0.02363T + 9.174 \quad (2)$$

$$1/\lambda = 0.02154T + 13.524 \quad (3)$$

Table 3 lists the evaluation of $1/\lambda$ at various temperatures along with the 95% confidence limits.

The $^{170}Tm_2O_3$ wafers for compatibility studies were discharged from the reactor on March 5. Work will begin on opening irradiation slugs and assembling compatibility specimen couples during April.

Table 2. Thermal Conductivities of Tm_2O_3, Yb_2O_3, and 0.46 Yb_2O_3-0.54 Tm_2O_3 Mole Ratio Mixture at Theoretical Density

Tm_2O_3		Yb_2O_3		0.46 Yb_2O_3-0.54 Tm_2O_3	
Temp, °C	λ, W/cm.°C	Temp, °C	λ, W/cm.°C	Temp, °C	λ, W/cm.°C
422	0.0389	411	0.0452	441	0.0344
539	0.0348	526	0.0419	548	0.0320
647	0.0328	637	0.0395	650	0.0307
721	0.0312	653	0.0381	743	0.0282
813	0.0282	753	0.0367	868	0.0261
		866	0.0346		
		441	0.0337		
		547	0.0306		
		625	0.0287		
		741	0.0266		
		887	0.0242		
		422	0.0396		
		527	0.0365		
		633	0.0327		
		773	0.0282		
		932	0.0260		

Table 3. Thermal Resistance ($1/\lambda$) and 95% Confidence Limits for Tm_2O_3, Yb_2O_3, and 0.46 Yb_2O_3-0.54 Tm_2O_3

Temp, °C	Tm_2O_3		Yb_2O_3		0.46 Yb_2O_3-0.54 Tm_2O_3	
	$1/\lambda$, cm.°C/W	95% Confidence Limit, cm.°C/W	$1/\lambda$, cm.°C/W	95% Confidence Limit, cm.°C/W	$1/\lambda$, cm.°C/W	95% Confidence Limit, cm.°C/W
200	20.35	± 2.87	19.84	± 6.29	23.72	± 3.23
400	25.07	± 1.70	24.54	± 3.91	28.02	± 1.99
500	27.44	± 1.20	26.89	± 2.89	30.17	± 1.44
600	29.80	± 0.89	29.24	± 2.21	32.32	± 0.97
700	32.16	± 0.99	31.59	± 2.22	34.48	± 0.97
800	34.52	± 1.40	33.94	± 2.92	36.63	± 1.44
1000	39.25	± 2.53	38.64	± 5.11	40.94	± 2.60
1200	43.98	± 3.75	43.34	± 7.59	45.25	± 3.89
1400	48.70	± 5.00	48.04	±10.16	49.56	± 5.22

D. Miscellaneous

1. HFIR Target Testing

Nothing to report.

E. Supporting Technology

1. Radioisotope Characterization

Half-lives of radionuclides are important not only in correcting for decay, but in calculating radioactivity when assays are in terms of weights and mass analyses. Examples of nuclides where this is particularly important are the transuranic elements, notably plutonium, americium, and curium.

A review was made of a tabulation of half-lives and other properties of transuranic elements contained in a new report.[3] Agreement of the listed selections with ours was satisfactory except for plutonium-241, for which our most recent estimate is 14.5 ± 0.5 years as compared to the reference value of 14.98 ± 0.33 years. Information was obtained from workers engaged in determining the half-life of plutonium-241 (among others) by calorimetry and by mass spectrometry; their current tentative values are 14.5 and 14.6 years, respectively. A similar review is being made of another new report,[4] which lists properties of selected nuclides of mass numbers 13-144.

2. Radioisotope Special Analysis and Quality Control

The survey of uses and needs for standard radioactive materials being made for the National Research Council continues as planned. Interviews with producers and users of standards are now being emphasized. Travel during March included the St. Louis-San Francisco Bay area, and April travel will be to points in the southeast and the Washington-New York-Boston section. Increasing attention is being given to the effect of quality of standards on regulatory decisions.

For local quality control, standards of several radionuclides have been ordered from the National Bureau of Standards and commercial suppliers.

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[3] W. D. Burch, et al., Semiannual Report of Production Status and Plans for Period Ending June 30, 1969, Transuranium Processing Plant, ORNL-4447, Oak Ridge National Laboratory.

[4] M. E. Meek and R. S. Gilbert, Summary of Gamma and Beta Energy and Intensity Data, NEDO-12037, General Electric Company (January 1970).

CYCLOTRON PRODUCTS - 08-01-02

A. Biomedical Radioisotopes

1. Rubidium-83

Nothing to report.

2. Rubidium-84

Nothing to report.

3. Gallium-67

The objectives of this program are to determine the optimal target configuration for gallium-67 (78 hr) production in acceptable purity and quantity and to provide gallium-67 for clinical applications research and development. Interest in this isotope has been spurred by evidence, obtained by the Medical Division of Oak Ridge Associated Universities (ORAU), of a high uptake of carrier-free gallium-67 by lymphoid tumors in both animals and humans. This interesting discovery was made during the evaluation of gallium-67, produced in the ORNL 86-Inch Cyclotron by the $^{68}Zn(p,2n)^{67}Ga$ reaction, as a bone-scanning agent for a patient with Hodgkin's disease.

Three gallium-67 preparations were made and shipped during March, and seven smaller orders for gallium-67 were filled for clinicians interested in evaluating the potential of this radionuclide for their applications. The total gallium-67 production for the first quarter of 1970 is shown in Table 4.

Table 4. Experimental Gallium-67 Preparations Made During the Quarter Ending March 31, 1970

Month	Number of Process Runs	Number of Shipments	Amount[a] of ^{67}Ga Shipped, mCi
January	2	7	247
February	2	7	336
March	2	6	266
Total	6	20	849

[a]Data calculated as of 8:00 AM on the day following shipment to be consistent with the ORNL decay allowance policy.

Work has been continued on cyclotron target development. A target of natural zinc electrodeposited on copper was tested during a production run as a prelude for the fabrication and testing of an isotopically enriched zinc-68-on-copper target. This target was similar to one tested

earlier[5] except that it was annealed longer at 390°C in an attempt to improve the Zn-Cu interface bonding and to increase the heat flow. The target was bombarded with 22-MeV protons at two different beam currents, first for 3 hr with a 235-μA beam and then for 25 min with a 265-μA beam. When the target was removed from the cyclotron, it showed signs of extensive melting, and appreciable amounts of loose radioactive gallium contamination were encountered. Even though this target offers the possibility of much larger scale production with less gallium-66 contamination in the product, the extensive loss of target integrity would seem to rule out this target configuration.

4. Indium-111

The objectives of this program are twofold: (1) to elucidate, define and optimize pertinent production parameters (cyclotron target material, cyclotron beam energy and current conditions, adverse side reactions leading to undesirable isotopic impurities, target processing and product purification procedures, and target recovery, if necessary) and (2) to make sufficient material available to cooperative participants for their evaluation of indium-111 as a diagnostic radionuclide in medicine. Such applications would include spinal-cerebral cisternography, aerosol lung studies, delayed brain scanning, visualization of the lymphatic system, metabolic studies of indium-labeled macroaggregates and colloids, and possible applications for tumor localizations. Indium-111 has both a favorable decay scheme [gamma emissions of 173 keV (89%) and 247 keV (94%)] ideally suited for external detection and an optimal half-life (2.81 days) for labeling and distribution studies which must be carried out over 24 hr or longer. The use of indium in such lengthy studies is currently impossible with the indium-113m (1.73 hr) isotope now in general use.

Additional operating experience on the HBr-isopropyl ether extraction purification process for indium-111 was obtained from a development run during March. Four shipments of indium-111, totaling approximately 95 mCi, were supplied to our medical cooperative participants for their application and evaluation. Due to unexpected and unexplained events during cyclotron operation, both the cyclotron production rate and yield were approximately 15% lower than for past irradiations. Possible explanations of this behavior might be (1) a misaligned capsule such that the beam was not hitting the ^{111}CdO target squarely or (2) wandering of the proton beam off the target area during irradiation. Chemical yield, chemical and radionuclidic purity of the product, target recovery, and iron decontamination were all acceptable and would seem feasible for routine indium-111 production. The complete chemical processing procedure which will be extensively tested during future development runs is given below in moderate detail.

Indium-111 is produced in the ORNL 86-Inch Cyclotron by 22-MeV proton bombardment of ^{111}CdO contained in an internal beam capsule target.[6] After irradiation the targets are removed from the cyclotron and transferred to a high level cell for processing. The target material (^{111}CdO) is prepared by high temperature calcination (>400°C) of $^{111}Cd(OH)_2$ and should, therefore, be free of pyrogens. The outside of the target is cleaned with freon before being opened in a high level cell. The aluminum capsule target is opened and the ^{111}CdO transferred to a 250-ml beaker.

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[5]A. F. Rupp, ORNL-TM-2828, p. 8.

[6]J. A. Martin and F. L. Green, Cyclotron Target for the Irradiation of Chemical Compounds, Nucl. Sci. Eng. 1: 185 (1956).

The aluminum target is then leached with approximately 3 M HNO_3 to dissolve any target material that may remain in the aluminum capsule. After the dissolver solution is sampled for production yield data, it is evaporated to dryness. The solid nitrates are converted to bromides by repeated evaporations from 6 M HBr solution. After conversion of the sample to the bromides, it is dissolved in 6 M HBr and transferred to a clean, dry separatory funnel. The sample is then extracted two times into purified isopropyl ether. The ether is purified by passing it through a column containing molecular sieve in the upper portion to remove alcohols and alumina in the lower portion to remove any peroxides that might have formed during storage. This purification scheme has been suggested by members of the ORNL Chemistry Division as an easy, safe, and convenient method for removing peroxides from ethers. The purified ether is required to improve the solvent extraction yields which have been noted (literature) to fluctuate due to impurities in the ether, presumably peroxides. The combined organic phases are washed two times with 6 M HBr. Then the indium is "stripped" from the organic phase by two treatments with 8 M HCl (HCl prepared from reagent grade HCl and sterile water). The combined strip solutions are washed two times with purified isopropyl ether to reduce the iron content in the final product. The product is evaporated to dryness and then taken up with 0.05-0.10 M HCl (dilute HCl prepared with sterile water). The product is finally filtered through a 0.45-μm Millipore filter into a stock solution bottle. The final volume of the product is made up to the desired quantity with 0.05 M HCl, which is also filtered through the Millipore filter.

Chemical spot tests for Cd(II) and Fe(III) are employed for process quality control. Practical operating limits of detection for Cd(II) and Fe(III) of <20 μg/ml and <5 μg/ml, respectively, are employed for this product.

Fabrication and testing of an alternative target design for the production of indium-111 by the $^{112}Cd(p,2n)^{111}In$ reaction were completed during March. The new design employs cadmium metal rather than the oxide. Initial test models were fabricated using natural cadmium to avoid committing a large inventory of isotopically enriched cadmium-112 until the design is shown to be feasible.

Initital results were not encouraging; the target failed at a beam current of 140 μA. This failure was apparently due to localized heating at the top of the capsule where the proton beam is the most intense. This initial target was designed with 0.035-in. inner wall thickness so that the inside tube would not collapse during the swaging operation. Consideration will be given to a design with reduced wall thickness (approximately 0.020-in. total thickness of aluminum) in an attempt to reduce the temperature gradient and to improve heat removal. Such a design will probably require the use of a mandrel during the swaging operation to avoid crushing the target.

B. Miscellaneous

As part of a continuing effort to assist cooperative participants to fill specialized requirements for specific radionuclides not usually available, miscellaneous, short-term tasks are occasionally undertaken. The demand or potential usefulness of these radionuclides is not considered adequate to justify extensive effort or a continuing development program, but the services performed are required to meet program objectives.

1. Promethium-143

Promethium-143 (13.59 days) was requested by Battelle-Northwest for use in inhalation studies. A 174-mg sample of $^{144}Sm_2O_3$ (95.1% isotopically enriched) was fabricated into a window-type target[7] for production of high radionuclidic purity promethium-143 by the following path:

$$^{144}Sm(p,2n)^{143}Eu \xrightarrow{\beta^+,\ EC} {}^{143}Sm \xrightarrow{\beta^+,\ EC} {}^{143}Pm\ .$$

The $^{144}Sm_2O_3$ target was irradiated with 22-MeV protons for 6.3 hr at an average beam current of 86.8 μA (integrated beam = 547 μA-hr). The irradiated sample was set aside for one week to allow decay of short-lived activities before processing. The sample was dissolved in dilute nitric acid and sampled for production rate and yield data. The promethium-143 product was separated from large amounts of europium-146-149 activities [formed by (p,n) reactions on isotopic impurities in the original samarium target] and from macro amounts of target material by high pressure cation exchange chromatography using 0.15 M α-hydroxyisobutyrate (pH = 4.7) as the eluting agent. The carrier-free promethium-143 product was "carried" on $Fe(OH)_3$ to separate it from the α-hydroxyisobutyrate and then separated from iron by anion exchange chromatography in 6 M HCl solution. The final product was evaporated to dryness and found to be free of solids (the only method used to ensure separation of the product from the target which formed no long-lived samarium radioisotopes under these irradiation conditions).

A preliminary thick-target production rate and a cyclotron yield of 0.44 μCi/μA-hr and 38 μCi/hr, respectively, were determined for this irradiation configuration. The radionuclidic purity of the product was estimated to be greater than 99.8% with only a trace of promethium-144 [arising from the (p,n) reaction on neodymium impurities present in the target material] detected. A chemical spot test for iron indicated less than 5 μg of Fe(III) per ml in the final product.

FISSION PRODUCTS - 08-01-03

A. Krypton-85 Enrichment

Three lots of product material were removed from the cascades and were transferred to the Sales Department. These lots consisted of 136 Ci of 19.9% krypton-85, 299 Ci of 21.0% krypton-85, and 407 Ci of 26.4% krypton-85.

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[7] J. J. Pinajian and T. A. Butler, ORNL 86-Inch Cyclotron Facility for Isotope Production, Isotopes and Radiation Technology 1(2): 137 (1963).

Internal transfers of product and reloadings with normal feed material were made in columns A, AB, B, and CD (see Fig. 1), and the present status of each cascade is shown below. The count rate indicates the level of enrichment.

Unit	Activity in Unit, Ci	Time Since Last Product Removal, days	Count Rate in Product Section, counts/min February 1970	March 1970
A	760	125	5,400	4,300
AB	849	5	8,200	3,600
B	706	5	7,500	2,750
C	1,044	95	2,600	3,900
CD	1,613	6	13,550	5,550
D	1,186	43	5,150	7,300

Column A does not appear to be building up in a normal manner, and a defective bellows pump is suspected; this is now being investigated.

A 40-liter cylinder of feed material was received containing 1860 Ci of 3.3% krypton-85.

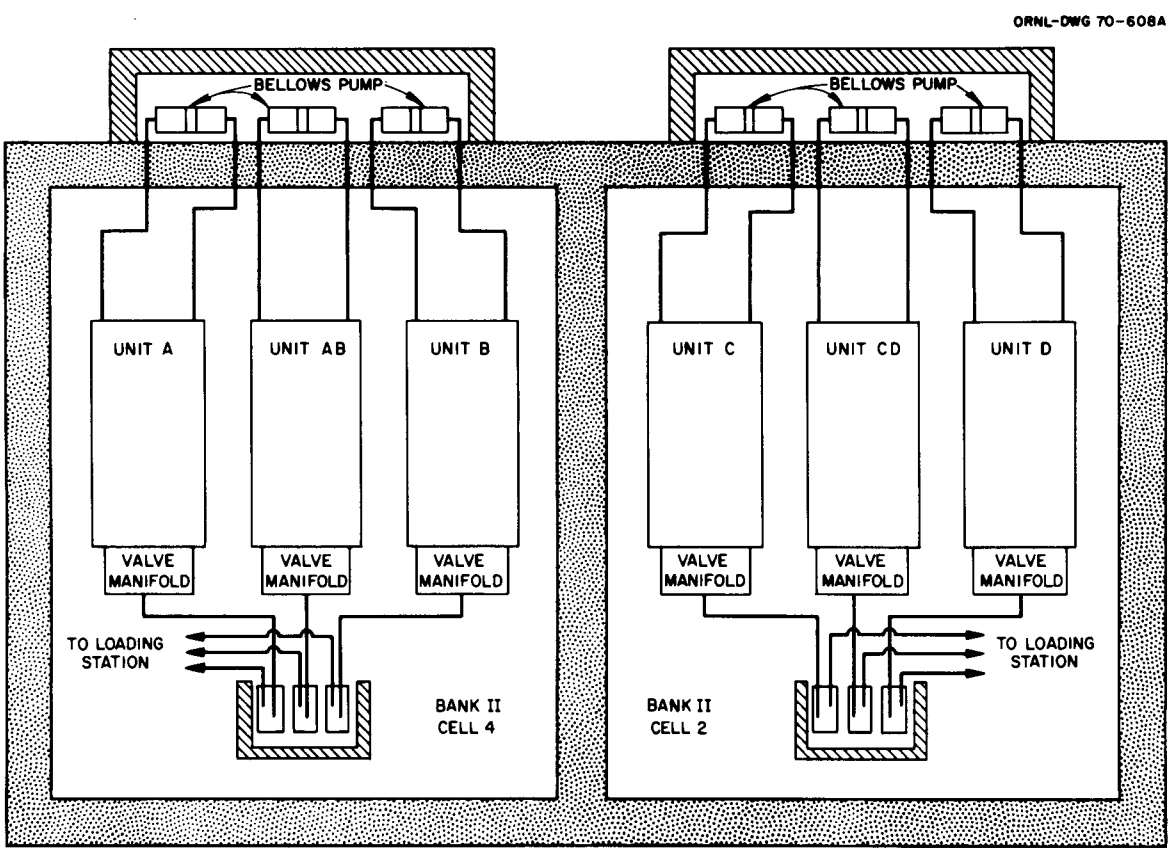

Fig. 1. Schematic Arrangement of Krypton-85 Columns.

B. Cesium-137 Process and Product Technology

1. Fission Product Process Study

The purpose of this project is the study and development of economic processes for recovery, purification, and conversion of fission products (cesium-137 in particular) obtained from reactor fuel reprocessing operations.

The current objectives are: (1) to develop a simple and efficient method for preparing homogeneous CsCl-KCl mixtures of various compositions, (2) to obtain preliminary differential thermal analysis (DTA) thermograms for selected mixtures, and (3) to study the cesium-137 process economics. The study on the CsCl-KCl system has been undertaken primarily to investigate whether the solid phase transition of CsCl may be eliminated in the presence of a small amount of KCl.

Homogeneity of a mixed sample is especially important in the DTA studies in view of the extremely small size of the sample. Accordingly, a study has been carried out to develop a simple and efficient method for preparation of homogeneous CsCl-KCl solid mixtures from pure crystalline material of the individual compounds.

Four selected compositions (in terms of the KCl content) of the mixtures included 2, 5, 50, and 95 mole % KCl. The preweighed mixture was put into a small vial containing a couple of glass beads and mounted on a miniature laboratory shaker, where it was blended for a time period of 1, 5, or 10 min. Samples from various mixtures have been submitted for chemical analyses to evaluate the efficiency of mixing. The results are not yet available.

In the ^{137}CsCl product obtained from the chemical processing of fission products, the impurity KCl is present, presumably as a phase distinct from ^{137}CsCl because of the large difference in the solubility of the two salts. The method of blending described above would produce a mixture similar to this type of product.

Thermograms were obtained for the CsCl-KCl mixtures of four different compositions that had been blended for 10 min. At least three separate runs were made for each mixture. The first run is mainly for the purpose of forming a molten mixture. For mixtures containing less than 50 mole % KCl (or higher than 50 mole % CsCl), the thermogram from the first run was similar to that for pure CsCl; that is, it always exhibited both solid-solid and solid-liquid transitions at temperatures which varied with the composition of the mixture. However, subsequent runs using the same mixture that has been previously molten resulted in thermograms in which no solid-solid transition was observed. The preliminary results on characteristic transition temperatures of such CsCl-KCl mixtures are summarized in Table 5.

The data in Table 5 indicate that addition of approximately 1 wt % (or approximately 2.2 mole %) of KCl to pure CsCl would eliminate the solid-solid transition. As the KCl content is increased to approximately 31 wt % (or 50 mole %), two transition peaks appear upon freezing. As expected, further increase in the KCl content to approximately 90 wt % (or 95 mole %) raises both the melting and freezing peak temperatures considerably.

Table 5. Characteristics of DTA Thermograms for Various CsCl-KCl Mixtures[a]

KCl Content of Mixture		Transition Peak Temperature,[b] °C			Solid-Solid Transition Temperature, °C
			Freezing		
Weight %	Mole %	Melting	First Peak	Second Peak	
0[c]	0[c]	639-645	634-640	None	431-437
0.98	2.18	636-644	633-637	None	None
2.36	5.16	633-635	626-633	None	None
30.83	50.15	610-611	622-623	603-604	None
89.34	94.98	752-757	748-753	None	None

[a]Mixtures that have been previously molten.
[b]The transition peak temperature varied with the heating rate and other factors.
[c]From thermogram for pure CsCl.

A modification of the Zevnik and Buchanan method is expressed by the following equation for the fixed capital cost (F):

$$F = f_c NCP^n (I/\bar{I}) \qquad (4)$$

where

C = a constant to be determined for a given range of production capacity,

f_c = complexity factor = 2×10^m,

m = f_t (process temperature factor) + f_p (process pressure factor) + f_a (process alloy factor),

P = production capacity, Ci/year,

n = factor derived from a statistical correlation between F and P,

N = number of functional units,

I = current value of construction cost index, and

$\bar{I}$ = construction cost index for a reference F versus P correlation.

In this equation, evaluation of parameters, C, n, and $\bar{I}$ would require a systematic analysis of extensive cost data for fission products processes which are not available thus far.

However, since the three processes under consideration for the production of $^{137}CsCl$ are similar in nature, the values of C, n, and $\bar{I}$ may be assumed to be approximately the same for all the processes. Also, the production capacity, P, may be specified and the current construction cost index, I,

can be estimated. In this way, the fixed capital costs, F, for the three processes may be compared through estimated values of the complexity factor, f_c, and the number of functional units, N, alone. Grouping all the terms on the right hand side of Eq. (4) (except f_c and N) as K,

$$F = f_c NK \ . \qquad (5)$$

Assuming that the three processes under consideration are to be operated under essentially atmospheric pressure, the process pressure factor $f_p = 0$.

The preliminary results shown in Table 6 are based on the values of f_a, f_t, and N estimated in earlier reports.[8-10]

Table 6. Comparison of Fixed Capital Costs for Various Processes for Production of $^{137}CsCl$

Process	N	f_a	f_t	f_c	F
1 HCl as eluant	4	0.4	0.06	5.76	23.04 K[a]
2 HNO_3 as eluant	6	0.3	0.06	4.59	27.52 K[a]
			0.11	5.14	30.86 K[a]
3 $(NH_4)_2CO_3$-NH_4OH as eluant	6	0.2	0.07	3.73	22.36 K[a]

[a]$K = CP^n(I/\bar{I})$.

Since K consists of parameters that are common to all three processes, a tentative conclusion drawn from Table 6 is that the fixed capital cost for Process 3 is slightly lower than Process 1. Process 2 seems to be the least economical from the standpoint of the fixed capital cost.

Studies on the phase behavior of the CsCl-KCl system will continue. The emphasis will be placed on determination of the lowest KCl content that can eliminate the solid-solid transition of the mixture. The process economics study will continue as time permits.

2. Entrapped Air in Cold-Pressed CsCl Pellets

Some swelling of cold-pressed $^{137}CsCl$ pellets has been observed when the pellets are subjected to thermal anneals at temperatures of about 400°C. It is well known that varying amounts of air are entrapped in powders which are formed into solid bodies by cold pressing at ambient pressures. This study was undertaken to determine the extent of entrapped air.

Five CsCl pellets were pressed at 120,000 psi in a 7-mm-dia die. The average density of the pellets was 3.93 g/cm^3 compared with a theoretical density of 3.99 g/cm^3. The pellets were submitted for entrapped air analysis.

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[8]A. F. Rupp, ORNL-TM-2828, p. 12.
[9]A. F. Rupp, ORNL-TM-2876, p. 11.
[10]A. F. Rupp, Radioisotope Program (8000) Progress Report for February 1970, ORNL-TM-2910, Oak Ridge National Laboratory.

In the analysis, the pellets were placed in a chamber and the chamber evacuated for 5-10 min. The chamber was sealed, the pellets were melted, and the released air was measured. The air released from each pellet, the pellet weights, and the calculated air pressures are shown in Table 7. It is assumed that the entrapped air is evenly distributed over the entire void space in the pellet.

No conclusion can be made at present as to the significance of the data relative to the swelling of $^{137}CsCl$ pellets.

Table 7. Data on Entrapped Air in Pressed CsCl Pellets

Pellet No.	Air, cm^3/g of CsCl	Weight of Pellet, g	Air Pressure in Pellet, psi
1	0.069	1.506	269
2	0.044	1.508	172
3	0.036	1.511	141
4	0.043	1.507	167
5	0.041	1.506	160

C. Absorption of Xenon-133 in Isotonic Sodium Chloride Solutions

Studies to develop techniques which make use of standard gas ampules and efficiently dissolve xenon-133 in sterile isotonic sodium chloride solutions are being made.

In some of the experiments, dry sodium chloride was sealed with the xenon-133, and degassed saline solution was admitted to the ampule upon breaking the glass break-seal tip. The stem of the ampule was then broken and the solution forced out of the ampule and into a bottle by air or saline solution via a hypodermic needle and a syringe. Care was taken to exclude air in contact with the solution containing xenon so that the xenon would remain in the solution for a longer time.

Other experiments used the same procedure and degassed distilled water to dissolve the xenon-133. The solution was made isotonic by the addition of sodium chloride solution.

The conclusions of the experiments were that either method efficiently dissolved the xenon-133. Cooling in an ice water bath to below room temperature did not affect further dissolution of the xenon. Greater than 90% of the xenon present in the ampule was dissolved in the solutions (approximately 10 mCi/ml).

RADIATION SOURCE DEVELOPMENT - 08-01-04

A. Strontium-90 Silicate Sources

1. Matrix Studies

Laboratory tests have been performed to determine the feasibility of producing a graphite matrix for encapsulating the strontium silicate minibeads. The minibeads were held at a temperature of 1050°C in a graphite reaction tube and contacted with propane gas. This produced a soft nonadherent coating which was difficult to control. Dense tough coatings using this process are deposited at temperatures between 1200 and 1300°C. It was necessary to operate at 1050°C because strontium silicate begins to soften above this point. Another approach is being investigated in which the minibeads will be mixed in a graphite powder containing a binder that will be extruded. The final step will be to decompose the binder at elevated temperatures.

Attrition tests to determine the feasibility of using strontium silicate minibeads in a fluid bed reactor have been completed using inactive minibeads. The results of a test in which toluene was used as the fluidizing medium show that after approximately 70 hr of operation the attrition rate had dropped to an undetectable level. This was for approximately 30 g of minibeads with a bed expansion of 300%. Additional tests involving radioactive minibeads will be necessary to properly define this effect.

2. Encapsulation

Nothing to report.

3. Characterization

Nothing to report.

B. Fabrication of Carbon-14 and Technetium-99 Radiographic Sources

Nothing to report.

C. Alpha Source Fabrication Development

Nothing to report.

D. Low-Energy X-Ray Studies

To develop sources useful in secondary X-ray fluorescent analysis, tests are being made using various source capsule geometries.

The system which is used for the detection of low-energy X rays and gammas is a Thermal Measurement Corporation analyzer with a Model 451 pre-amplifier and a lithium-drifted silicon crystal. The system was calibrated using sources with known gamma energies and X rays which were produced as secondary K alpha and K beta energies.

This lithium-drifted silicon detector and analyzer can resolve the low-energy peaks with a resolution of 350 to 400 eV. Environmental conditions (high temperature, vibration, fluctuations in power, or power failure) which occur will produce detrimental effects upon the resolution of these low-energy radiation photopeaks. A periodic check of the change in resolution will be made in order to reduce errors in analyses.

Rock samples were examined by the technique of double fluorescent X-ray excitation (i.e., americium-241 60-keV excitation of molybdenum which excites those X-ray energies below molybdenum K X-ray energy). In addition to calcium and iron, traces of arsenic and strontium were found.

E. Absorption of Scandium-46 Gamma Radiation in Concrete

Auditing the thickness of concrete in new highway construction now requires coring at various locations. The State of Pennsylvania is now considering the use of small sources imbedded at the ground-concrete interface at the time the concrete is poured. Changes in the ratio of gamma peaks provide a thickness measurement.

The change in the ratio of the two gamma peaks in scandium-46 was observed in varied thicknesses (1.875 to 8.500 in.) of concrete. A source containing approximately 10 μCi of scandium-46 covered with 4-mil aluminum foil was used in the experiment. A 3 by 3 in. NaI crystal coupled to a 512-channel analyzer was used to observe the number of counts in the 889- and 1120-keV photopeaks. The peak ratio was observed to decrease as the concrete thickness increased.

RADIOISOTOPE SOURCE SAFETY TESTING - 08-01-04

A. Special Form Prototype Testing

Prototype sources are subjected to the tests prescribed for "special form material." On the basis of these tests, sources of similar design may be judged to pass the requirements for "special form material." This is a continuing program.

A 130-Ci ^{137}CsCl source, doubly encapsulated in type 316L stainless steel, met the requirements for USAEC "special form material" on the basis of the temperature test (heating in air to a temperature of 800°C and remaining at that temperature for a period of 10 min) and on test results obtained from sources of similar design. The overall dimensions of the source were 1.95-in. length and 0.49-in. dia. The inner capsule contained an aluminum cup to prevent the source material from entering the free void space during normal conditions. At elevated temperatures the aluminum cup quickly loses its strength and collapses (melting point approximately 660°C).

B. Testing Sources With Manufactured Leaks

Nothing to report.

C. Surveillance of Regulations and Cask Evaluations

Nothing to report.

D. Source Leak Test Guide

Nothing to report.

E. Sealed Source Testing

Sealed sources are tested to evaluate their ability to contain the radioactive material under normal and abnormal conditions.

A 0.25-in.-thick by 2.38-in.-dia gamma source containing 20 mCi of $^{241}AmO_2$ was tested to determine if it could meet the test requirements for the ORNL I-A classification. The source consisted of a disk of aluminum with a concentric groove cut to contain the active material. The source was covered with a 0.010-in.-thick aluminum cover sealed to the disk with epoxy. The vibration test was not made because a tester was not yet available, and the source was too large for our shear test equipment. The source passed all other tests except the impact test in which it was only moderately damaged, but the seal was broken. It is unlikely that this source would be subjected to an impact of 20 ft-lb in actual use. This is equivalent to the impact received if the source were to be dropped from a height of 188.6 ft onto a steel plate. The following is a summary of the tests performed:

Class A Operating Temperature Test - The source was immersed in an ice bath (32°F) for 24 hr and then heated in air at 212°F for 24 hr. No leaks were found.

Class A Thermal Shock Test - The source was heated at 212°F for 1 hr, plunged into an ice water bath (32°F), and then placed in a container of dry ice (-70°F) for 15 min. No leaks were found.

Class A Maximum Temperature Test - The source was heated at 400°F for 1 hr, then cooled to room temperature. No leaks were found.

Class I External Pressure Test - The source was placed in a pressure vessel and held for 15 min at 30 psig. No leaks were found.

Class I Crushing Force Test - The source was placed on edge in a press and a 200-lb load was applied for 15 min. No leaks were found.

Class I Puncture Resistance Test - The source was placed on the 1/8-in.-dia pin so that the impact was on the lid covering the groove which contained the source material. A 50-g weight was dropped from a height of 3 ft, striking the opposite side of the source. No leaks were found.

Class I Impact Resistance Test - The source was placed with its edge resting on the impact pad and a 5-lb weight was dropped on it from a height of 4 ft. The capsule was moderately deformed by this test and failed to pass the leak test.

F. Unsealed Source Testing

Nothing to report.

TECHNOLOGY UTILIZATION - 08-01-05

A. Information Center

The Isotopes Information Center now has 18,000 accessioned items. The backlog has been cleared and documents are being indexed and accession cards made immediately upon receipt. Punching of the Termatrex cards has started again, and another 1600 items have been included on the computer tapes. During March, 220 IIC reports (including a few "Understanding the Atom" booklets) and reprints were sent out in answer to queries, and 48 requests that required specific answers were filled. The documents requested included the Customer List and the worldwide isotope-use abstract compilations. Twelve letters were translated for Isotopes Sales, and 500 pages of a Russian machine translation were edited in a cooperative project with NASA. The edited translation of the Russian concrete-polymer book was sent to DTIE for typesetting. A list of reviews in process (for answering future queries) is given below:

Title	Author(s)	Status, % Complete
Cement-Polymer Combinations	V. I. Solomatov, translated from Russian and edited by Martha Gerrard, P. S. Baker, and R. E. Greene	At printers
DID Research and Developments - 1969	Isotopes Information Center Staff	90
Indium-113m: Preparation and Uses	Martha Gerrard	10
Flow Measurement	R. H. Lafferty, Jr.	5
Iodine-125	P. S. Baker and Martha Gerrard	25
Isotopic Methods of Examination and Authentication in Art and Archaeology	F. J. Miller, E. V. Sayre, and B. Keisch	95
Oceanography[a]	Isotopes Information Center Staff	90
Patent Review - Process Radiation Development	R. E. Greene, P. S. Baker and Helen Warren	85
Potato Sprout Inhibition by Radiation	F. E. McKinney	35
Radiation vs Glass[b]	D. N. Hess	20
Radioisotopes in the Pharmaceutical Industry	P. S. Baker and Martha Gerrard	5
Radioisotopes in the Steel Industry[c]	Republic Steel Corporation	10
Radioisotopes in X-Ray Fluorescence Analysis	R. H. Lafferty, Jr.	5
Selected Abstracts of World Literature on Production and Industrial Uses of Radioisotopes	Martha Gerrard and P. S. Baker	10
Self-Diffusion in Liquids	F. J. Miller	75
Strontium-90	Roberta Shor, R. H. Lafferty, Jr., and P. S. Baker	85
Technetium-99m: Preparation and Uses	Martha Gerrard and P. S. Baker	30
Radioisotopes in the Textile Industry	F. J. Miller	90
Use of Radioisotopes in Sedimentology	V. Romanovsky, translated and edited by Martha Gerrard	10
Wheat Disinfestation - A Worldwide Review	F. E. McKinney	5
Wood Plastics	R. E. Greene	75
Yttrium-90	Martha Gerrard	10
Permuted Isotopes and Radiation Technology Indexes	Helen Raaen	10

[a]Draft completed; being held at DID.
[b]Indefinite.
[c]Will be in sections; first section now in hand.

B. Isotopes and Radiation Technology

Galley proofing for Isotopes and Radiation Technology 7(4) was completed, as was most of the manuscript for 8(1).

RADIOISOTOPE APPLICATIONS DEVELOPMENT

BASIC TECHNOLOGY DEVELOPMENT - 08-03-01

A. Liquid Scintillation Development

1. Alpha Radiation Damage in Phosphors

Nothing to report.

2. Cerenkov Counting

Nothing to report.

SYSTEMS ENGINEERING APPLICATIONS - 08-03-02

A. Oceanographic Systems Study

One of the major goals of the RIST program is to develop techniques to determine volume of sediment transport. To accomplish this, several tasks must be completed: 1) development of a new injection technique, 2) improvement of stationary detector designs, and 3) development of computer programs to analyze the data.

The detector assembly that had been equipped with the electromechanical cable has been disassembled. It has been determined that the failure of this equipment was caused by salt water leaking into a section of the tongue where the wires are spliced. This section was potted with a silicone material which did not bond with the teflon-coated wires. We will attempt to etch the teflon to obtain bonding or will install water seals on the tube that contains the splices.

The Oceanside, California, test data are being treated and contours will be available in approximately three weeks.

The computer programs for RIST are still in the development stage due to the lack of a good working theoretical model of sediment transport. Any mathematical model that reasonably fits the physical phenomenon must be empirically derived. The collected data are processed, studied, and reprocessed until the data reductions reasonably fit the field experiment observations. When there are not enough field data to define some parameters, the collected data help to design the next field experiment for pursuit of these parameters.

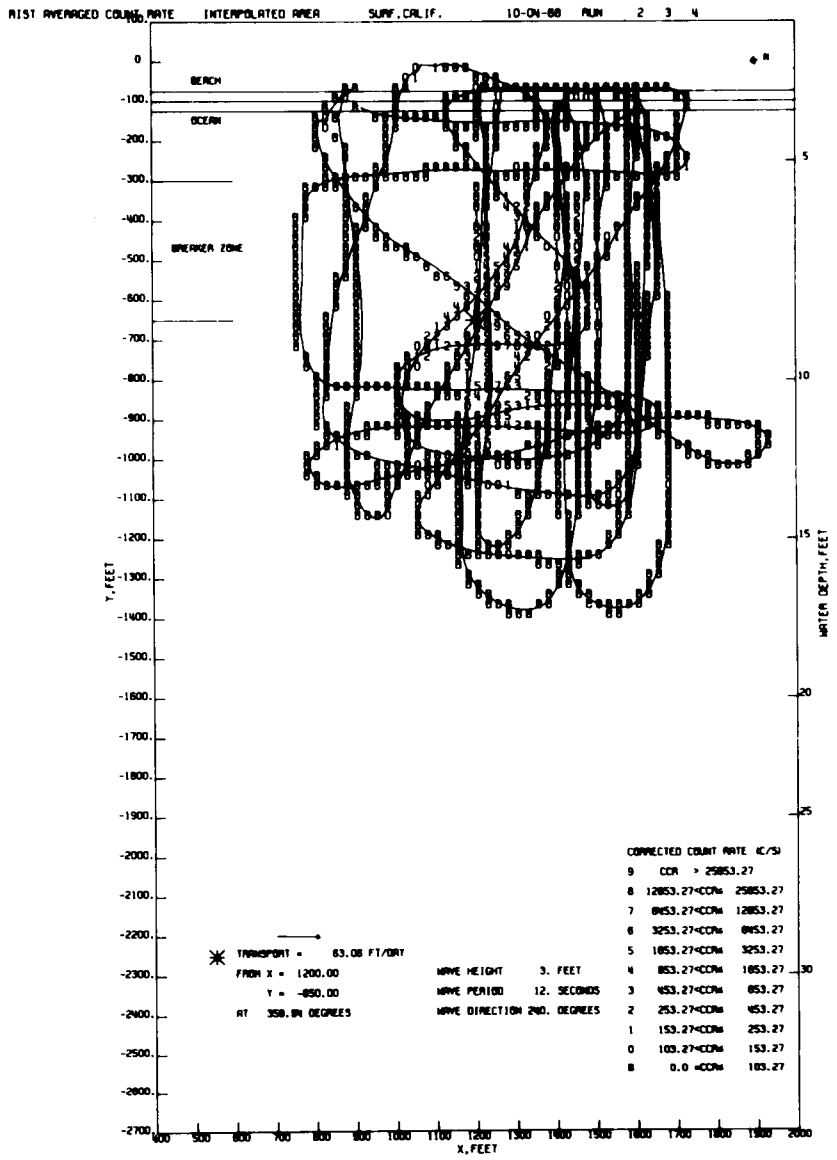

Fig. 2. Radiation Data Along Detector Track.

Figure 2 displays the results of one computer program. The data input of this program comes from the first processing program[11] which places the count rates (corrected for decay back to time of injection and for background) on an x-y grid. In this program the x-y grid is divided into 25-ft squares, and all count rates within any one square are averaged and a one digit number representing this average count rate is assigned to that square. The figure also displays the track line of the detector as a solid line. In an attempt to calculate a linear velocity of sand particles in

[11] H. R. Brashear et al., Computer Plotting of Data from the Mobile Amphibious Detection System Used in Radioactive Sand Tracer Studies, ORNL-TM-2212, Oak Ridge National Laboratory (July 1968).

transport, all count rates above background (B) are processed using the following equation:

$$T_{(x)} = \Sigma\ RX/R_T t\ ,$$

where

R = averaged radiation count rate,
X = distance from injection in the x direction,
R_T = sum of all count rates above B,
t = time from tagged sand insertion,
$T_{(x)}$ = x component of the transport vector.

A similar procedure is carried out in the y direction and a resultant vector established from these two vectors.

Figure 3 is the result of the linear interpolation portion of the program. The interpolation is done only in the x direction which coincides with the

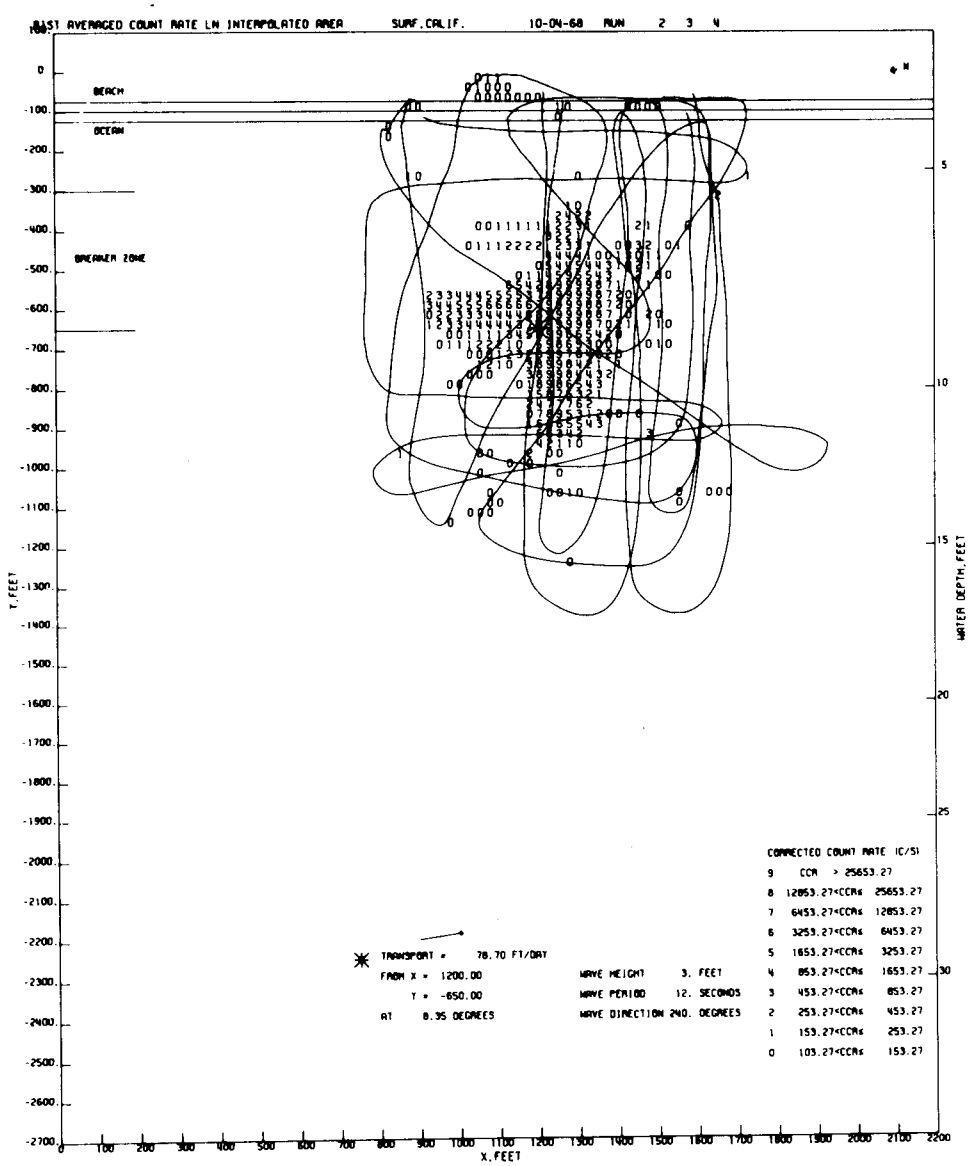

Fig. 3. Detector Track and Significant Points of Radiation.

transport direction (358°) calculated in Fig. 2. Interpolation is done between averaged count rates only and no extrapolation or weighing factors are used. The apparent linear velocity is calculated again to give a better estimate of the actual linear surface rate. For clarity, the background values have been omitted from the plot.

Figure 4 displays the contouring portion of the program. The count rates obtained from the linear interpolation are plotted as contours.

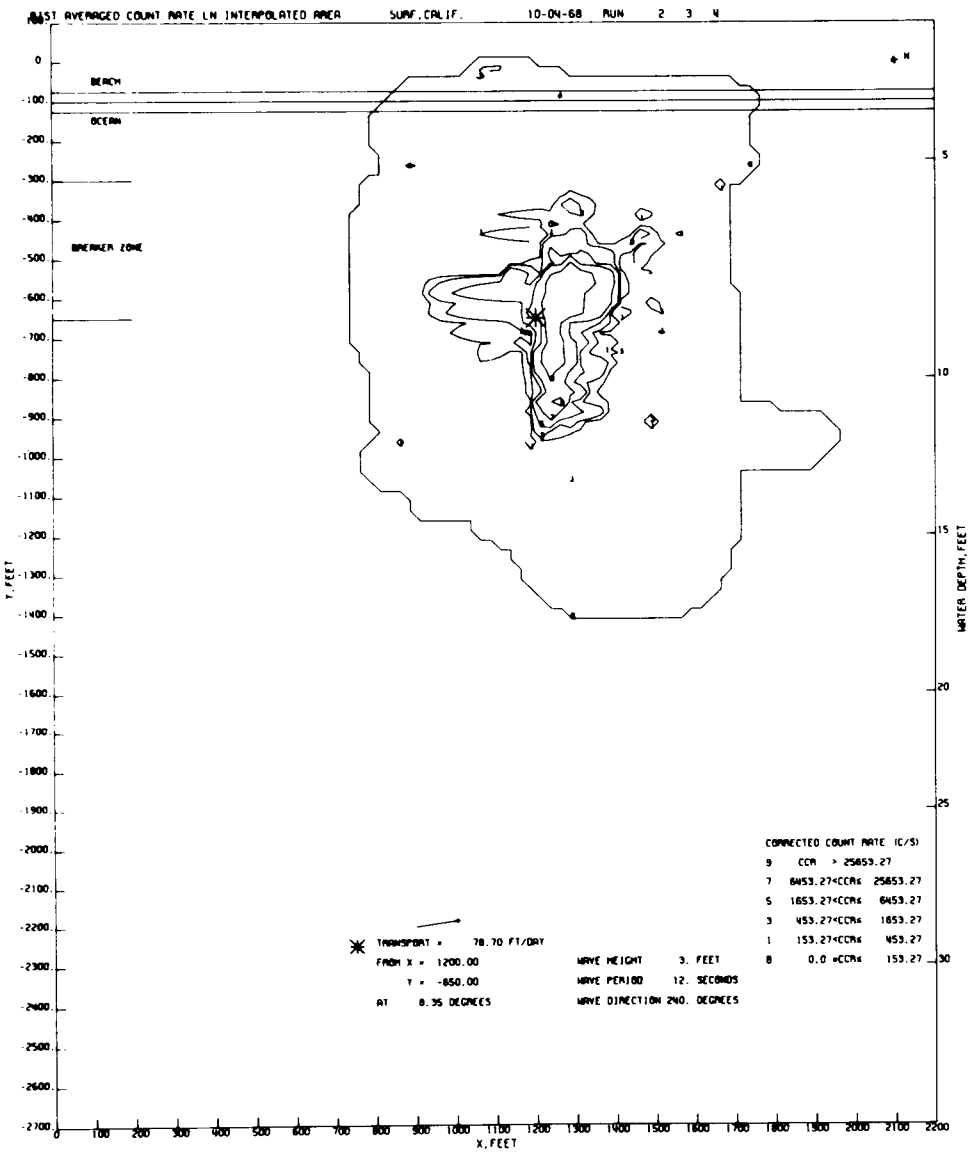

Fig. 4. Total Survey Area and Transport Contour.

RADIATION PROCESSING APPLICATIONS

ORGANIC SYNTHESIS WITH ISOTOPIC SOURCES - 08-04-01

A. Straight Chain Fatty Acids

The radiolytic oxidation of long chain hydrocarbons is fairly complex; therefore, shorter chain hydrocarbons are being used to study the mode of oxidation.

The gamma radiolytic high pressure oxidation of 2-hexene was continued. As stated previously[12] we felt 2-hexene was oxidizing at the double bond. Butyric acid, butyraldehyde, acetic acid, acetaldehyde, and water have been identified by retention time on gas-liquid chromatography and the structures have been confirmed by infrared spectroscopy and mass spectroscopy. We have had difficulty in trapping sufficient amounts of samples from the gas-liquid chromatograph in order to do infrared spectroscopy and mass spectroscopy on the remaining components of our irradiated samples. Longer irradiations of 2-hexene are planned so that greater amounts of the unidentified components can be obtained. Work on 2-hexene should be completed in April.

The study of the radiolytic oxidation of 2-hexene was undertaken to aid in understanding the radiolytic oxidation of tall oil on which we will begin work in April.

RADIOISOTOPE SALES

An inquiry was received from Allgemeine Elektricitats-Gesellschaft, Germany, for up to 2,000,000 Ci of cesium-137. An inquiry was received from International Nutronics for 200,000 Ci of cesium-137.

Notable shipments made during March include 120,000 Ci of strontium-90 to CEA, France; 20 mCi of chlorine-36 to Radiochemical Centre, England; 2,000 Ci of cesium-137 to University of North Carolina; 1,300 Ci of cesium-137 to J. L. Shephard and Associates; and 45,000 Ci of tritium to Radiochemical Centre, England. Other shipments made were 166,810 mCi of krypton-85, 169,825 mCi of xenon-133, 97,655 Ci of tritium, and 2 Ci of enriched 20-25% krypton-85.

A listing of outstanding large orders is shown on page 24.

The radioisotope sales and proceeds and shipments for the first eight months of FY 1969 and FY 1970 are given in Table 8.

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[12] A. F. Rupp, ORNL-TM-2910, p. 20.

Isotope	Amount, Ci	Customer
Cesium-137	~203,000	Brookhaven National Laboratory
Cesium-137	~180,000	American Nuclear Corporation[a]
Cesium-137	~80,000	Lockheed Georgia Company[a]
Cesium-137	~207,000	U. S. Army Natick Laboratories[a]
Cesium-137	205,000	Industrial Reactor Laboratories, Inc.
Cesium-137	120,000	Radiation Machinery Corporation
Cesium-137	~190,000	Atomic Energy of Canada Limited
Cesium-137	40,000	Radiation Resources, Inc.
Cesium-137	20,000	Radiochemical Centre
Cesium-137	200,001	CEA, France
Cesium-137	200,001	Radiation Resources, Inc.
Cesium-137	~3,000	Miscellaneous orders
TOTAL	1,648,002	
Strontium-90	~532,000	Isotopes, Inc.
Strontium-90	~105,000	Isotopes, Inc.[a]
Strontium-90	~41,000	Brookhaven National Laboratory
Strontium-90	~556,982	U. S. Navy
Strontium-90	~3,000	Miscellaneous orders
TOTAL	1,237,982	
Promethium-147	5,100	Reactor Centrum Nederland
Promethium-147	5,001	Mitsubishi Corporation (Japan)
Promethium-147	150,001	McDonnell Douglas Astronautics Company[b]
TOTAL	160,102	
Carbon-14	1,445 mCi	Catholic University
Tritium	30,000	Radiochemical Centre, England
Cobalt-60	6,200	Puerto Rico Nuclear Center

[a]Activity has been billed in previous fiscal years. Material is now being fabricated into sources or has already been fabricated into sources and is stored for future requests for shipments.
[b]Material is to be shipped from Richland, Washington.

Table 8. Radioisotope Sales and Shipments

Item	7-1-68 thru 2-28-69	7-1-69 thru 2-28-70
Inventory items	$449,310	$411,318
Major products	66,423	72,537
Radioisotope services	132,701	153,163
Cyclotron irradiations	105,304	94,512
Miscellaneous processed materials	45,922	57,944
Packing and shipping	55,803	48,960
Total Radioisotope Sales	$855,463	$838,434
Total Radioisotope Shipments	1,862	1,665

Table 9 gives the March 1970 cyclotron irradiations and runs for ORNL and non-ORNL customers.

Table 9. Cyclotron Irradiations and Runs for March 1970

Product	No. of Runs	Time, hr:min Beam	Misc.	Total	Total Charges
ORNL Research Programs					
Cobalt-56	1	4:00	1:10	5:10	$ 616.54[a]
Cobalt-61	2	3:00	1:30	4:30	447.50
Gallium-67	2	4:55	2:20	7:15	758.95
Indium-111	1	2:00	1:05	3:05	292.92
Total		13:55	6:05	20:00	$2,115.91
Non-ORNL Research Programs					
Bismuth-205	1	1:00	1:15	2:15	$ 335.50
Rubidium-84	1	9:30	1:15	10:45	1,724.99
Yttrium-87	1	2:00	1:15	3:15	540.10
Total		12:30	3:45	16:15	$2,600.59

[a]Purchased but not shipped.

RADIOISOTOPE PRODUCTION

CESIUM-137 PROCESSING

No cesium-137 processing was done this month.

STRONTIUM-90 PROCESSING

Two strontium-90 product preparations were made in March. The first, Run No. 4Sr70, was a $^{90}SrCO_3$ precipitation which yielded 37,000 Ci of finished product, and the second run, No. 5Sr70, was a standard $^{90}Sr_2TiO_4$ precipitation which yielded 49,000 Ci of product. A total of 86,000 Ci of strontium-90 product was prepared during March. These products were made by using the NH_4OH-$(NH_4)_2CO_3$ precipitation procedure. The analytical data and activity balance calculations for five runs that have been made during CY 1970 are shown below. A total of 225,000 Ci of strontium-90 product powder has been prepared this calendar year.

	Batch Number				
	1Sr70	2Sr70	3Sr70	4Sr70	5Sr70
Feed Analysis,[a] Ci	42,800	47,500	44,300	37,800	50,300[b]
TiO_2 Addition, g	650	753	903	None	361
Product					
Weight, g	1,501	1,525	1,678	757	1,231
Specific activity, Ci/g	30.7	28.5	29.7	49.1	39.7
$CO_3^{=}$, wt %	0.15	0.05	0.21	-	-[c]
Total product, Ci	46,100	43,500	49,900	37,200	48,900
Waste					
Volume, liters	90	92	113	95	107
Strontium-90 content, Ci	130	60	335	70	55
Activity Balance, Ci					
Feed	42,800	47,500	44,300	37,800	50,300
Product	46,100	43,500	49,900	37,200	48,900
Waste	130	60	335	70	55
Combined Activity Balance, Ci					
Feed	222,700				
Accounted for	226,200				
Percent accounted for	101.6				

[a]Based on individual batch analysis.
[b]Based on P-23 feed analysis.
[c]Analytical results are not available.

GENERAL OPERATIONS

Three high temperature compatibility tests on curium-244 oxide sources and strontium-90 titanate sources are in progress. These tests are being performed as part of the compatibility studies being conducted for the isotopic power programs.

A failure in the cooling water service lines to the induction-heated hot press in one of the cells has required the removal of the furnace. The cell will be decontaminated to a personnel entry level. Modifications will be made during this period to provide this cell with inert atmosphere capability. Entry will also be required for the installation of electrical leads and services to the new resistance-heated hot press. It is planned to initiate maintenance work in April.

The shielded Drum Waste Carrier was put into operation during March. A total of six waste loadouts of debris from the manipulator cell bank was made in the Drum Waste Carrier. No difficulties have been experienced in the loading or unloading of the carrier containing a sealed 55-gal drum. The contents of the drum are preliminarily decontaminated and bagged in plastic; each bag is probed to determine its radiation level. After installation of the lid on the drum, the drum is decontaminated to a smear level of less than 1 mr/hr and remotely loaded into the Drum Waste Carrier. The loaded Drum Waste Carrier is decontaminated to smear tolerance and transferred to the burial ground where the drum is lowered into an auger hole for permanent burial.

Waste compaction studies were initiated to determine the compaction ratio of general manipulator cell waste. The one test conducted indicates that with a pressure of 6.0 psig a compaction ratio of 2.3:1 can be obtained for mixed waste. The waste consisted of incremental pressing of materials such as 1) bagged manipulator boots, gloves, glassware; 2) styrofoam, tin cans, paper towels; 3) glassware, tygon, plastic; and 4) bagged wet wipes. It is planned to conduct tests with similar groupings at 10 and 15 psig.

The HEPA filters in both the 120 and 220 banks of the hot off-gas filtration system were changed out during March. The filters had become plugged with NH_4NO_3 during the processing of strontium-90.

MAINTENANCE

The installation of the hot off-gas dehumidifier system has been completed. The water service lines and the associated instrumentation are currently being installed. The off-gas scrubber pumps have been removed and the piping is being revamped in a pump cell. It is planned to decontaminate this cell, install a stainless steel floor pan, and install new scrubber pumps. The dehumidifier-scrubber maintenance program will be completed in April. This system will reduce the plugging problems associated with the off-gas HEPA filters.

SOURCE FABRICATION

A. Cesium-137

Fabrication of miscellaneous $^{137}CsCl$ sources continued. The sources completed this month are listed below.

Customer	No. of Sources	Cesium-137, Ci Per Source	Cesium-137, Ci Total
Radiation Resources, Inc.	100	400	40,000
Atomic Energy of Canada Limited[a]	2	1,100	2,200
Atomic Energy of Canada Limited[a]	1	1,000	1,000
J. L. Shepherd and Associates[a]	10	130	1,300
University of North Carolina[a]	1	2,000	2,000
Robbin Laboratories[a]	1	16	16
AEC-New York Operations Office[a]	1	4	4
American Nuclear Corporation	2	500	1,000
American Nuclear Corporation	1	20	20
Total	119		47,540

[a]Shipped (others in storage pending shipment).

Miscellaneous source fabrication will continue during April. There are 40 more sources on order, representing 11 different designs. The pellets for these sources have been fabricated, and some of the liners have been completed.

All of the sources fabricated in March received the recently revised quality assurance tests including heat tests at 400°C plus redundant dimensional and leak checks. "Special form" sources will continue to be heat tested at 400°C, but we are considering limiting heat testing of teletherapy and similar sources to 250°C because their requirements of high pellet density and minimum clearance can produce stresses in the source capsule due to the normal expansion of CsCl in heating to 400°C.

In most miscellaneous source runs, it is uncommon for as many as ten units to be of a given design, and normally there are only one or two. This complicates the scheduling of operations and necessitates frequent set-up changes to accommodate the various source sizes, types of loading, weld procedures, etc. The problems of in-cell inventories for a large assortment of sources are obvious. The order from Radiation Resources, Inc. for 100 identical 400-Ci sources presented an unusual opportunity for the scheduling of operations for a number sources large enough to represent something approaching "mass production." The procedures used for this run of 100 units are outlined below, and the differences between this and runs which include an assortment of designs are noted.

The source fabrication facility consists of three manipulator cells, which will be referred to as Cells A, B, and C below. Cell A is the powder handling and source loading cell; Cell B is used for welding and leak testing; and Cell C is used for furnace testing, source decontamination, and final checking. The 100 sources were fabricated as follows:

1. A set of special tools was fabricated specifically for use with the 100 sources. Since tool and equipment fabrication is expensive, it is not usually economical to provide special tools for each and every source design. However, when the cost can be applied to 100 units, such fabrication becomes feasible.

2. The work area in Cell A was cleared of all equipment not associated with loading of these sources. Normally, there will be several sizes of die bodies, gages, storage pans, etc. in the area.

3. The 100 liners (inner capsules) were loaded and capped in one continuous operation. Elimination of such items as frequent resetting of weights and changes in loading fixtures was definitely time saving.

4. The liners were transferred to Cell B and welded in one continuous operation. Continuous welding of identical items is even more of a time advantage than is continuous loading since resetting of the remote welder is eliminated. Also, test welds are reduced since the "post" weld of one series becomes the "pre" weld of the next. Test welds are normally made before and after each series of ten or fewer identical fueled welds.

5. After the liners had been leak tested and furnace tested, they were cleaned to low contamination levels. This was a definite departure

from normal procedures. In most cases, since there will be sources at several stages of fabrication, the most cleaning that a given liner gets is a rinse before being inserted into its capsule.

6. Cell C was decontaminated to smear tolerance. New, smear-clean equipment was set up and the dried liners were loaded into their capsules. The caps were inserted into the capsules with clean tools. In this case a holding block was used to retain the capsule while the cap was inserted by means of a hydraulic jack. This minimized the handling of capsules and also protected the weld areas.

7. Cell B was cleaned before capsule welding was started. The capsules were then welded and leak tested in one continuous operation.

8. Before the final furnace test on the sources, they were decontaminated to shipping tolerance. This required very little effort since the capsules had been well protected from contamination during welding. Cell C was cleaned to smear tolerance and a new test furnace was used. An additional advantage to this pre-cleaning is that significant leaks which might occur during furnace testing will be detected by contamination checks after furnace testing. None were detected in this case.

The total cycle of 100 units, from beginning of powder baking to the time at which the finished sources were put on shelf test, required 32 shifts of slightly more than two weeks of elapsed time. By contrast, the fabrication of the other 19 sources of individually different designs made this month required essentially the same amount of time.

At the request of the AEC, six representative units from a batch of 46 rectangular $^{137}CsCl$ sources prepared for the U. S. Army Natick Laboratories were given a long-term furnace test. All 46 sources had been furnace tested 4 hr at 400°C during their fabrication. Since the highest temperature which these sources are expected to reach during use will be during the shipment of the irradiator from Lockheed, Georgia, to Natick, the customer wanted a test to duplicate both temperature and time conditions. The calculated maximum temperature is 250°C and the shipping time is about ten days. The six test sources were heated to 375°C, and this temperature was maintained (±5°C) for two weeks. The sources were dimensionally checked and leak tested. No dimensional changes or leaks were detected.

B. Strontium-90

Nothing to report.

C. Miscellaneous Sources

Sources fabricated and shipped during March 1970 include a 10-mCi carbon-14 target, a 200-Ci promethium-147 source, and 16 strontium-90 pellets.

SPECIAL FORM MATERIALS

One shipment of 120,000 Ci of $^{90}SrTiO_3$ in eight identically loaded shipping cans was made to France (CEA).

OPERATIONAL SUMMARY

	March 1970		FY 1970	
	Number	Amount, Ci	Number	Amount, Ci
Cesium-137				
HAPO shipments received	0	0	0	0
Product batches prepared	0	0	0	0
Sources fabricated	119	47,500	304	352,900
Special form containers	0	0	25	7,700
Strontium-90				
HAPO shipments received	0	0	0	0
Product batches prepared	2	86,000	16	646,000
Sources fabricated	0	0	0	0
Special form containers	0	0	40	332,900

PROCESS STATUS

Physical Inventory

Item	Cesium-137, Ci	Strontium-90, Ci
In-process materials	238,500	954,000
Products in storage	382,500	942,000
Sources in fabrication	8,000	0
Completed sources	387,500	528,000
Total	1,016,500	2,424,000

MISCELLANEOUS RADIOISOTOPE PROCESSING

Short-Lived Fission Products		Processed Units		Service Irradiations	
Radioisotope	Total, Ci	Radioisotope	Total, Ci	Type	Number
Xenon-133	1,200	Copper-67	12	Platinum-196	4
Iodine-131	40	Calcium-47	17	Y_2O_3	1
Mixed fission products	0.1	Palladium-109	3,461	Fe-Ni	1
		Osmium-191	353	Platinum-190	1
				Cerium rod	1
Total	1,240.1	Total	3,843	Ruthenium-97 sponge	1
				Total	9

The charcoal traps in the hot off-gas and cell ventilation system of the Short-Lived Fission Products Facility were changed this month. Work on cerium-144 has been temporarily suspended.

ADMINISTRATIVE SECTION

Travel by IDC personnel and visitors to the IDC are given in Tables 10 and 11.

Table 10. Travel by IDC Personnel

Site Visited	Purpose of Visit
Vanderbilt University, Nashville	Discuss radiation sources for medical use
Washington, D. C.	Attend meeting of National Academy of Sciences Subcommittee
Miami Beach, Florida	Attend 2nd Annual Nuclear Medicine Seminar
Smithville, Tennessee	Examine display site

Table 11. Visitors to IDC

Visitor Affiliation	Purpose of Visit
Battelle-Northwest, Richland	Tour Isotopes Information Center
OECD/ENEA, Paris, France	Tour Isotopes Information Center
General Nuclear	Americium-241 sources
Industrial Nucleonics	Carbon-14 sources
Ministry of Public Works, Venezuela	Sediment tracing
Corps of Engineers	Sediment tracing
Catholic University	Carbon-14 targets
Isotopes Inc. and U. S. Navy, Ft. Belvoir	Strontium-90 sources, design of new compact heat source, isotope kilowatt study, and SrO data

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63. T. D. Chikalla, PNL, Richland, Washington
64. D. F. Cope, AEC Site Representative, ORNL
65. D. C. Davis, AEC, Oak Ridge, Tennessee
66. J. C. Dempsey, AEC, Washington, D. C.
67. W. K. Eister, AEC, Washington, D. C.
68. E. E. Fowler, AEC, Washington, D. C.
69. J. D. Goldstein, AEC, Washington, D. C.
70. A. Gottschalk, Argonne Cancer Research Hospital, Chicago, Illinois
71. F. D. Haines, AEC, Washington, D. C.
72. J. W. Irvine, MIT (Consultant)
73. J. Lawrence, Lawrence Radiation Laboratory, Berkeley, California
74. J. E. Machurek, AEC, Washington, D. C.
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