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RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT

FOR DECEMBER 1969

A. F. Rupp

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ORNL-TM-2828

Contract No. W-7405-eng-26

Isotopes Development Center

RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR DECEMBER 1969

A. F. Rupp

JANUARY 1970

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Oak Ridge, Tennessee
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RADIOISOTOPE PRODUCTION AND MATERIALS DEVELOPMENT

NEUTRON PRODUCTS - 08-01-01

A. Biomedical Radioisotopes

1. Thulium-171

Nothing to report.

2. Phosphorus-33

The purpose of this project is to develop methods of preparing hundred-millicurie quantities of carrier-free phosphorus-33 containing $\leq 5\%$ phosphorus-32. This radioisotope of phosphorus has a lower energy beta and a longer half-life than phosphorus-32, which makes it useful for autoradiography work in biochemistry, agriculture, and metallurgy.

Two methods of preparing phosphorus-33 are being evaluated. One method is the irradiation of sulfur-33 enriched to greater than 92% sulfur-33 in a fast neutron flux to prepare the radioisotope by the $^{33}S(n,p)^{33}P$ reaction. The other method of preparation is the fast neutron irradiation of chlorine-36 to prepare phosphorus-33 by the $^{36}Cl(n,\alpha)^{33}P$ reaction. Highly enriched targets are necessary to reduce the phosphorus-32 content of the phosphorus-33 produced from either of the targets.

a. Phosphorus-33 Preparation from Enriched Sulfur-33

The enriched 92% sulfur-33 target material (potassium sulfate, anhydrous) contained chemical impurities that were introducing scandium-46 and chromium-51 into the phosphorus-33 product. These impurities were probably introduced during previous chemical processing of the target for recovery of phosphorus-33. Work in December was devoted to purifying the $K_2{}^{33}SO_4$. Assay of the material has not been completed.

The yield of phosphorus-33 from the target containing 96 mg sulfur-33 (92% enriched) has been approximately 230 to 250 mCi of phosphorus-33 per irradiation. Additional 92% sulfur-33 is being prepared as potassium sulfate to increase the yield of phosphorus-33 per batch to ≥ 300 mCi.

b. Phosphorus-33 Preparation from Enriched Chlorine

The purpose of this work is to prepare highly enriched chlorine-36 for target material for future phosphorus-33 production by the $^{36}Cl(n,\alpha)^{33}P$ method and to supplement current phosphorus-33 needs now normally supplied by the $^{33}S(n,p)^{33}P$ method.

A 4.7-g target of $K^{36}Cl$ was removed from the HFIR to recover the phosphorus-33 activities and determine the specific activity of the chlorine-36. There were 1580 mCi of phosphorus-33 and 63 mCi of phosphorus-32 recovered from the target; the phosphorus-33 will be used

to fill current needs. The target material is being purified at present and assays are not complete on the isotope abundance of the chlorine-36. Future plans are to put the $K^{36}Cl$ target back in the HFIR for further enrichment of the chlorine-36.

3. 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 has very limited use in humans because of its high-energy (1.52 MeV) gamma-ray emission and its relatively short half-life (12.4 hr). Potassium-43, with a half-life of 22.4 hr and principal gamma-ray emissions of 0.373 and 0.617 MeV, is more suitable for metabolic and clinical studies. 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.

A 6 mCi potassium-43 preparation was shipped to Johns Hopkins for evaluation under our cooperative program. The potassium-43 was separated from the calcium-43 target material (63.6% enrichment) by sorption on a Bio-Rad AG 50W-X8 resin and step-wise elution with 0.2 and 0.5 M HCl. Complete separation from sodium-24 contamination was also achieved.

The first 100 ml, which was eluted with 0.2 M HCl, was analyzed and found to contain 0.48 mCi of iridium-192. Based on the irradiation parameters for the target, this quantity of iridium-192 implies that approximately 380 mg of natural iridium was present initially. Work is still in progress to define the origin of the iridium-192 contamination. The platinum crucible used to calcine the recovered calcium target is suspect, although activation analysis of a piece of the crucible yielded a value of only 28 ppm iridium. A sample of $CaCO_3$ was calcined in the crucible and analyzed for iridium by activation analysis along with an uncalcined sample. The sample heated in the crucible contained 33 ppm of iridium and no platinum compared with 0.4 ppm of iridium for the unheated sample of CaCO , indicating that iridium is selectively leached from the crucible. If this is the case leaching of iridium from a crucible should decrease with use.

Another interesting aspect to the problem of long-lived contaminants is a study made on the residue of a potassium preparation shipped to MIT in June and returned for analysis. The recent analysis showed chromium-51 as the principal contaminant with lesser quantities of iridium-192 and cesium-134. Other minor gamma activities identified were antimony-124, cesium-137, scandium-46, rubidium-86, and cobalt-60. The target

used for this product had not been recovered in a platinum crucible, unless this was done originally in recovering the enriched stable isotope. The trace of antimony activity, which has been observed in decayed samples of recent products, is derived from the quartz irradiation ampules.

Measurements of the beta spectrum are in progress to elucidate the decay scheme of potassium-43. Work is still in progress to determine the relative efficiency of the detector for the assignment of relative intensities to the gamma rays reported last month. The decay of a sample of potassium-43 was studied for approximately ten half-lives and the value of the half-life obtained from following the decay of the two prominent gamma transitions, 373 and 617 keV, is 22.44 and 22.46 hr, respectively. Additional work will be done to define the standard error.

4. Rubidium-84

The 0.51-MeV annihilation radiations of rubidium-84 are used in diagnostic studies of heart muscle. The objectives of this work are to prepare rubidium-84 by fast neutron irradiation of enriched strontium-84 [$^{84}Sr(n,p)^{84}Rb$ reaction] and to characterize the rubidium-84 product. Rubidium-83, which has gamma energies near 0.51 MeV, is an undesirable radionuclide impurity in rubidium-84 products.

A 190-mg SrO target (75% isotopic enrichment of strontium-84) was irradiated for 46 days at a fast neutron flux of 2.6×10^{14} n/cm^2.sec.

The rubidium activities were separated from the enriched strontium-84 target by precipitation of the strontium with fuming nitric acid and purified by ion exchange. The purified rubidium fraction was assayed for rubidium-83, rubidium-84, and rubidium-86 with a Ge(Li) detector and a 4096 channel gamma spectrometer. The yields of rubidium-84 and rubidium-86 were confirmed by counting on a calibrated NaI(Tl) crystal and 400 channel gamma spectrometer. The yields of rubidium activities from the strontium-84 target were 0.45 mCi of rubidium-83, 1.81 mCi of rubidium-84, and 8.3 mCi of rubidium-86. This radioisotope preparation was unsuitable for use in heart muscle studies because of the high rubidium-83 content.

The yield of rubidium-84 was much lower than the anticipated[1] approximately 14 mCi. The disagreement between the rubidium-84 found and that expected (based on short irradiations at lower neutron fluxes) has not been explained.

The rubidium-83 results from the $^{84}Sr(n,2n)^{83}Sr \rightarrow {}^{83}Rb$ reaction. Little can be done to improve the $^{83}Rb/^{84}Rb$ ratio, since both are produced by fast neutron reactions.

The rubidium-86 content results from two successive neutron captures by the strontium-84 target material: $^{84}Sr(n,\alpha)^{85,85m}Sr \rightarrow {}^{85}Rb$ and $^{85}Rb(n,\alpha)^{86}Rb$. The yield of rubidium-86 from the target is in fair agreement with the computed yield using known cross sections for these reactions.

Additional irradiations will be made to explain the anomalous results found between this experiment and previous work.[1]

[1] H. B. Hupf and H. A. O'Brien, Jr., The Reactor Production of ^{84}Rb from Strontium, Intern. J. Appl. Radiation Isotopes 18: 417-20 (1967).

B. Research Radioisotopes

1. 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 days; $E_{\beta\text{-av}}$ = 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.

The thermal conductivities were measured for vacuum hot-pressed specimens of Tm_2O_3, Yb_2O_3, and a mole-ratio mixture of 0.46 Yb_2O_3-0.54 Tm_2O_3. The mixed-oxide pellet was prepared by calcining an oxalate precipitate prepared by homogeneous precipitation from aqueous solution. The purpose of making measurements on ytterbia, the decay product of $^{170}Tm_2O_3$, is to show that the thermal conductivity of thulia-ytterbia mixtures is a linear function of the concentrations. The physical characteristics of the specimens are given in Table 1.

Table 1. Physical Characteristics of Tm_2O_3, Yb_2O_3, and Mole-Ratio Mixture

	Height, in.	Diameter, in.	Weight, g	Density, g/cm^3	% Theoretical Density
Tm_2O_3	0.503	1.000	56.32	8.71	98.0
Yb_2O_3	0.480	0.995	53.43	8.76	95.5
0.46 Yb_2O_3-0.54 Tm_2O_3	0.520	0.998	55.42	8.30	92.0

The thermal conductivities (λ) were measured in the range 400-900°C with a thermal comparator using Pyroceram 9606 as a thermal conductivity standard. The preliminary measurements, corrected to theoretical density by means of the Maxwell equation

$$\lambda_{TD} = \lambda_{meas} \left\{ \frac{2 + P}{2 - 2P} \right\} ,$$

where P is the porosity of the pellet, are shown in Table 2.

Table 2. Thermal Conductivities of Tm_2O_3, Yb_2O_3, and Mole-Ratio Mixture

Tm_2O_3		Yb_2O_3		0.46 Yb_2O_3-0.54 Tm_2O_3	
Temperature, °C	λ, W/cm.°C	Temperature, °C	λ, W/cm.°C	Temperature, °C	λ, W/cm.°C
422	0.0373	441	0.0330	441	0.0353
539	0.0349	549	0.0310	548	0.0331
647	0.0328	629	0.0295	650	0.0317
721	0.0316	741	0.0276	743	0.0294
813	0.0289	887	0.0256	868	0.0274

These preliminary data are measurements on uninstrumented pellets and are subject to systematic errors of calibration. Work in January will be devoted to making measurements with an instrumented, 1-in.-high ytterbia pellet to verify the calibration.

The seawater leach-rate experiment was terminated when the Naval Radiological Defense Laboratory was closed. D. Kubosi, who is now located at the Naval Ordnance Laboratory, stated that the data were available, but there has been no response from the Space Electric Power Office to a request for funding of a final report.

The thulia wafer specimens for future compatibility experiments will be under irradiation during January.

D. Miscellaneous

1. HFIR Target Testing

During the calendar year 1969 a total of eight stringers for long-term irradiations in the beryllium reflector and 146 rabbits for short-term irradiations in the hydraulic tube facility of the HFIR were leak tested prior to irradiation to insure the safety and success of the experiment.

Of these 154 targets tested, a total of 23 were rejected, giving a rejection rate of 14.9%. During calendar year 1968 the rejection rate was 19.5%. It is interesting to note that 70% of the failures during 1969 were detected by the visual ethylene-glycol test and only 30% by the pressure test. A similar distribution was noted in 1968, when 65% of failures were found by the ethylene-glycol test and 35% by the pressure test. These data would seem to indicate that most faults in preparing targets are gross and easily detectable by visual means and that only a few are small enough to pass the visual test and be detected by the pressure test.

2. Terbium-157

Nothing to report.

E. Supporting Technology

1. Radioisotope Characterization

The continuing program of keeping aware of properties of radioisotopes (half-lives, decay schemes, production cross sections) has resulted in useful outputs to the scientific and technological community. American Nuclear Society personnel used our data on properties of radioisotopes and licensing requirements in the Radioisotope Directory (Nuclear News, December 1969), which is intended to supersede the Isotope Index. ASTM Committee E-10 requested a copy of the Isotope User's Guide for each of its approximately 120 members to be used as a source of standardized data.

2. Radioisotope Special Analysis and Quality Control

No unusual problems in assay of radioisotope products have been encountered recently. In connection with the survey on status and needs for standard radioactive materials, supported by the National Academy of Sciences, discussions took place here and in Cincinnati with members of ASTM and with hospital and public health personnel. ASTM Committee E-10 on radioisotopes and radiation effects agreed to poll its members on whether or not the National Bureau of Standards should expand its program in radioactivity standards. Gathering of information by use of a questionnaire is largely completed, and a panel of specialists in various fields is being formed to recommend further investigation and to draw conclusions and make recommendations. Professor L. R. Zumwalt of North Carolina State University will serve as chairman of the panel, and other members include Dr. G. Friedlander of Brookhaven National Laboratory, Dr. J. H. Harley of the AEC Health and Safety Laboratory, and Dr. E. Saenger of Cincinnati General Hospital and the University of Cincinnati. Arrangements for two additional members are pending. With respect to the status of standards, it had seemed reasonable to think that few inaccurate calibrations are presently being made, but responses from competent workers show that groups of standards from two well known commercial suppliers have recently been seriously in error.

A problem related to assay (i.e., disintegration-rate measurement) is the measurement of external radiation from a source. A few months ago the calibration of devices for gamma radiation from americium-241 was described; standards of that nuclide were obtained from IAEA. A new problem is quantitation of bremsstrahlung from titanium tritide sources; no comparable standard for such a low-energy continuous spectrum is available. As in the case of americium-241, the usual survey instruments are not calibrated for the energy range, nor are they sufficiently sensitive. It was recommended that a source of vanadium-49, which emits approximately 20% 4.5-keV X rays, be used as a standard. The specific gamma-ray constant is approximately 0.86 mR/hr at 1 meter from a 1-mCi point source. If a second standard is needed, iron-55 would be appropriate and has the virtue of direct traceability to NBS and IAEA primary standards.

A significant quality-control study was initiated in an attempt to explain the deformations of cesium-137 sources reported last month. The usual analyses were performed: cesium-137 assay, chloride, sulfate, carbon, rubidium, and spectrographic examination. Because NH_4Cl_6 is isomorphous with CsCl, and presumably would vaporize on heating, possibly causing swelling of CsCl sources, ammonium determinations were made. Microscopic examinations and determinations of water-insoluble solids were also done. Carbon determinations were done on the solids. Only the total carbon value was anomalous; attempts to determine the form of the carbon were not successful. Therefore the analytical program did not furnish an explanation of the deformations, which were avoided by changes in operating procedures.

CYCLOTRON PRODUCTS - 08-01-02

A. Biomedical Radioisotopes

1. Rubidium-83

The chief objectives of this work are to supply rubidium-83 for medical and research applications under cooperative programs and to develop the technology for preparing a high-purity product acceptable for human use. Rubidium-83 is prepared by cyclotron bombardment of strontium-84 by the following reactions:

$$^{84}Sr(p,2n)^{83}Y \xrightarrow{EC,\ \beta^+} {}^{83}Sr \xrightarrow{EC,\ \beta^+} {}^{83}Rb\ ,$$

$$^{84}Sr(p,pn)^{83}Sr \xrightarrow{EC,\ \beta^+} {}^{83}Rb\ ,$$

$$^{84}Sr(p,2p)^{83}Rb\ .$$

Rubidium-83 has a half-life of 83 days and principal gamma emissions of 0.521 (100%), 0.530 (66%), and 0.553 (36%) MeV.

A request was received from our cooperative participant at the University of Utah for rubidium-83 to be used in muscular dystrophy studies. A 133-mg sample of 75.6% isotopically enriched $^{84}SrCO_3$ was irradiated in the ORNL 86-Inch Cyclotron and has been set aside to allow decay of the strontium-83 to rubidium-83 before processing.

A new cation exchange procedure for the separation of rubidium (I) from strontium (II) using Bio-Rad AG 50-X8 resin has been investigated and found satisfactory for this product. This new procedure was selected because it offers the advantage of using dilute HCl (0.5 M) rather than an aqueous NH_4Cl solution as the eluting agent for rubidium (I), thereby freeing the final rubidium-83 product from excessive amounts of ammonium salts which are difficult to remove completely. The actual processing and analysis of this product run will be performed during January.

2. Gallium-67

The objectives of this program are twofold: 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.

Two gallium-67 production runs were processed and shipped during December. The total gallium-67 production for the last quarter of 1969 is given in Table 3.

Table 3. Experimental Gallium-67 Preparations Made During October-December 1969

Month	Number of Process Runs	Number of Shipments	Gallium-67 Shipped, mCi[a]
October	2	2	308
November	2	5	212
December	2	5	225
Total	6	12	745

[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 naturally abundant zinc electrodeposited on copper was tested as a prelude for the fabrication and testing of an isotopically enriched zinc-68-on-copper target. The target, which was bombarded with an average beam current of 230 μA, showed signs of melting in the center. Preliminary data obtained to date indicate that a target fabricated from 99.3% enriched zinc-68 may offer an improvement in yield (in mCi/hr) of approximately 2-1/2 times that obtained with the natural zinc tubing target currently being used.

3. Indium-111

The objectives of this program are twofold: to elucidate, define, and optimize pertinent production parameters (e.g., 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 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.

The use of this product in humans makes product characterization and quality control very important and necessary items. In an attempt to completely characterize the chemical composition of the initial indium-111 products being produced by the HBr-isopropyl ether extraction procedure currently being investigated, an aliquot of the purified product was analyzed by spark-source mass spectrometry. The analytical data from the first indium-111 product separated by the HBr-isopropyl ether extractions is now being computer processed.

The radionuclidic purity of this product was determined by gamma-ray spectrometry using a Ge(Li) detector and a 4096 channel pulse height analyzer. Spectra taken approximately 2 days and 35 days after irradiation showed only the radioactive isotopic impurities, indium-110m (4.9 hr) in the first sample and indium-114m (50 days) in the second. The indium-114m-114 content of the indium-111 product was estimated to be 0.02% of the indium-111 one half-life (2.81 days) after end of bombardment. One of our cooperative participants estimates that this amount of indium-114m-114 impurity will result in approximately 8% additional dose to the patient. Based on present experience, they believe that the "radionuclidic impurities pose negligible additional risk to patients."

Recovery of the enriched cadmium-111 target material used for indium-111 production by the $^{111}Cd(p,n)^{111}In$ reaction has been investigated. The procedure consists basically of precipitation of $^{111}Cd(OH)_2$ with NaOH, followed by repeated aqueous washes to remove excess base, and finally calcination at 425°C to the oxide. Initial recovery results indicate that $\geq 93\%$ of the original target material can be easily and economically recovered. This value includes both processing and assay sample losses.

Another development run for indium-111 has been scheduled for early January to gain additional operating experience for the HBr-isopropyl ether extraction process and to check the reproducibility of the pertinent production parameters (e.g., chemical yield, production rate and yield, chemical and radionuclidic purity of the product, and target recovery). Preliminary consideration will also be given different types of cyclotron targets for indium-111 production by the $^{112}Cd(p,2n)^{111}In$ reaction. Since the cross sections for the (p,2n) reaction on cadmium-112 is approximately twice that for the (p,n) reaction on cadmium-111, it makes this alternative look promising for higher production rates if the target fabrication problems can be overcome.

4. Neodymium-140

The purpose of this work is to develop the technology for preparing neodymium-140 and to supply experimental products through medical

cooperatives for evaluation as diagnostic scanning agents. Neodymium-140 (3.3 days) decays by electron capture to praseodymium-140 (3.39 min) which subsequently decays by positron emission to stable cerium-140. The principal photons useful for scanning studies are the praseodymium and neodymium X rays and the annihilation photons of the positron. The objectives are to determine the yield of activity that can be produced from the $^{141}Pr(p,2n)^{140}Nd$ reaction, to develop the separation procedures necessary to isolate carrier-free neodymium-140 from the approximately 200-mg praseodymium target, and to characterize the neodymium-140 product.

Two cyclotron irradiations have been made. The yields of neodymium-140 from the 2-hr irradiation of the 200-mg praseodymium-141 targets were 30.0 and 37.0 mCi from total beam currents of 172.3 and 175.5 μA, respectively.

The chemical separation of carrier-free neodymium-140 and the 200-mg praseodymium target was completed in 3 hr by use of high-pressure ion exchange. The rare earths were sorbed on a 50-cm by 6-mm column of 20- to 35-μm Dowex 50 type resin. Separation was achieved by elution at 1000 psi and 70°C with 0.3 M α-hydroxyisobutyric acid (pH 4.4). There was no detectable praseodymium in the neodymium-140 product (limit of detection = 13 μg/ml). The product was free of other radionuclides.

Information feedback from the cooperatives indicates neodymium-140 has excellent possibilities as a general scanning agent. By proper chemical treatment it can be made to go to the bone, brain, or kidneys of experimental animals. These applications for localization of tumors in organs by positron scanning may be extended to human patients soon.

Future work will include supplying experimental products to the cooperatives and making the radioisotope available to more medical investigators. Some experimental work will be directed toward increasing the neodymium-140 yield by target redesign.

FISSION PRODUCTS - 08-01-03

A. Krypton-85 Enrichment

Low level units AB, B, C, and D (see Fig. 1) and the intermediate level unit CD had run through a normal operating cycle and had reached essential equilibrium this month. On December 15 the depleted gas from the ends of all units was removed and stored, and the product material from units AB, B, and C (a total of 3.3 liters or 432 Ci) was transferred into unit CD for a high-level run. Normal fission-product krypton with approximately 3.4% mass 85 content was used to replace the enriched product and depleted material removed from the units, bringing the gas pressures in all units up to approximately 3 psig.

All six units should now operate through a normal 60-day cycle. At the completion of the cycle, gas transfers will again be made to keep

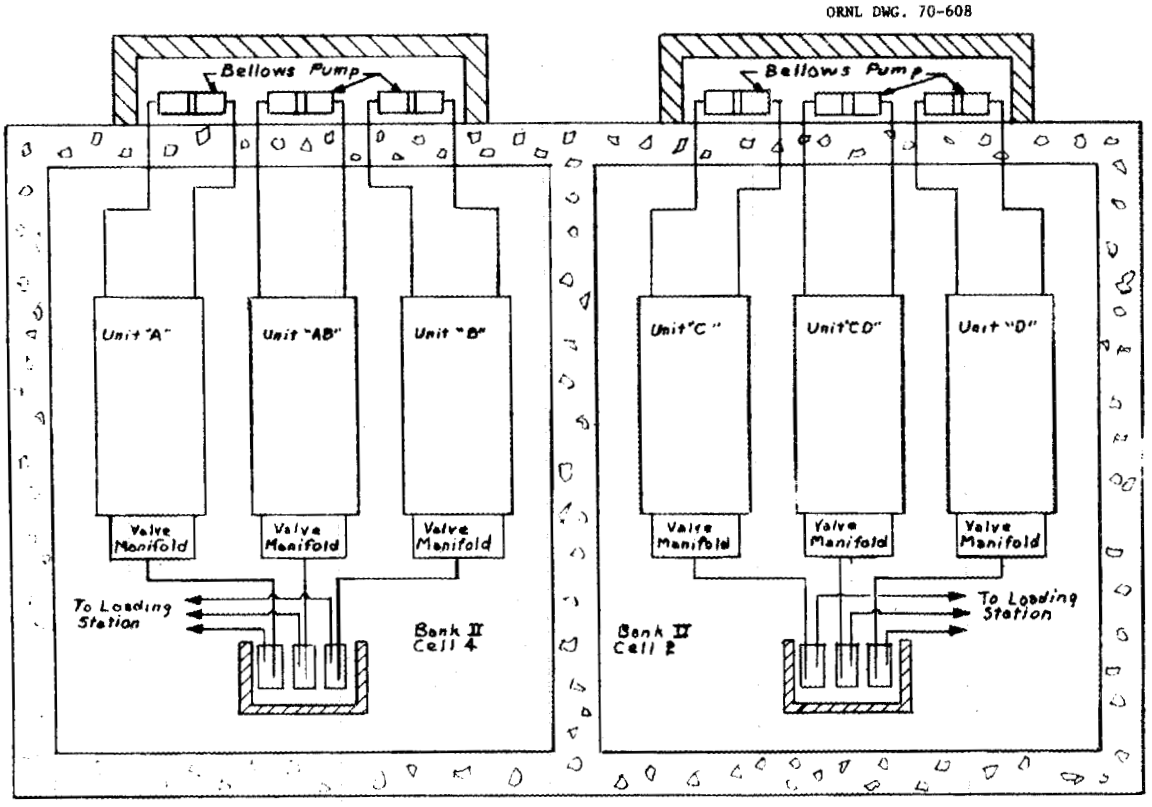

Fig. 1. Schematic Arrangement of Krypton-85 Columns.

the high-level units rising in product concentrations and the low-level units operating to provide pre-enriched material for feed to the high-level operation.

The total curie activity contained in each unit, the elapsed operating time on the current run, and the count rate at each end of the center product section are 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
A	515	36	3,500
AB	1071	6	12,000
B	843	6	11,300
C	1044	6	10,500
CD	1906	60	15,500
D	1150	36	13,000

In addition to the material in the operating units there are 605 Ci of normal (3.4%) feed material in a 40-liter shielded transfer tank and 518 Ci of depleted (<1%) material in the storage cell, making a total of 7652 Ci of krypton-85 stored or in process.

B. Cesium-137 Process and Product Technology

1. Fission-Product Process Study

The purpose of this project is the study and development of economical processes for recovery, purification, and conversion of fission products (cesium-137 in particular) obtained from reactor fuel reprocessing operations. The current objectives are to prepare the conceptual flow diagrams for the three cesium-137 recovery processes proposed, to estimate the number of process functional units, and to initiate studies on the equilibrium phase behavior of the CsCl-KCl system.

Preliminary flow diagrams have been prepared for three processes outlined in the October-November report. Each process uses a different eluant to remove the cesium compound loaded on the inorganic ion exchanger. Process 1 uses hydrochloric acid as the eluant, process 2 uses nitric acid, and process 3 uses a mixture of $(NH_4)_2CO_3$ and NH_4OH.

An attempt has been made to estimate the number of functional units (N in the equation presented in IDC Newsletter for July 1969, ORNL CF-69-8-13) from the preliminary flow diagram for each of the three processes. A functional unit may be defined as a group of equipment and accessories that are required to perform one significant process function. Based on this definition, the estimated number of functional units is given in Table 4.

Table 4. Functional Units for Three Processes

Equipment with Accessory	Number of Functional Units
Process 1 (HCl as Eluant)	
Shielded Transfer Tank and Feed Storage	1
Evaporating Crystallizer	1
Centrifuge	1
Dryer or Calciner	1
Total	4
Process 2 (HNO_3 as Eluant)	
Shielded Transfer Tank and Feed Storage	1
Evaporator	1
Crystallizer	1
Centrifuge	1
Chlorinator	1
Dryer or Calciner	1
Total	6
Process 3 [$(NH_4)_2CO_3$-NH_4OH as Eluant]	
Shielded Transfer Tank and Feed Storage	1
Primary Evaporator	1
Chlorinator	1
Evaporating Crystallizer	1
Centrifuge	1
Calciner	1
Total	6

It should be noted that both processes 2 and 3 have six functional units. The number of functional units alone is insufficient for the economic evaluation of different processes. However, the fact that the $(NH_4)_2CO_3$-NH_4OH solution would be less corrosive than HNO_3 and that the former is a more efficient eluant than the latter (see Table 2, page 5 of IDC Newsletter for May 1969, ORNL CF-69-6-14) may imply that process 3 is economically more attractive than process 2.

A study on the equilibrium phase behavior of the CsCl-KCl system has been initiated to investigate the peculiar behavior of the CsCl-KCl system reported.[2] This behavior refers to the possible absence of the solid phase transition of CsCl containing a small amount of KCl. The transition temperature of pure CsCl is 469°C. A brief literature survey has been conducted to search for any published data for the system. The only data of some significance found thus far are those reported in two technical papers.[3,4] The first one deals with the crystallization surface (temperatures of appearance of the first crystals) for the Cs, K, Na || Cl system, while the the second one presents the phase diagram for the CsCl-KCl system with some indication of the presence of more than one solid phase.

In January the major emphasis will be devoted to studies on the equilibrium phase behavior of the CsCl-KCl system. The work on development of a generalized semiempirical method for evaluating various conceptual designs will be continued.

2. Cesium Chloride Slab Sources

A method was developed for repairing stainless steel CsCl powder storage cans that had been punctured for gas sample analysis.

A test powder can filled with NaCl was punctured four times using the gas sampling puncture equipment. These holes were plugged with stainless steel wire leaving approximately 3/16 in. of excess wire protruding. The wire was fused with the powder can by using two separate pulses from a Heliarc welding torch. The repaired test powder cans that were leak tested under vacuum while immersed in ethylene glycol were tight. The test can was sectioned through the welds and weld penetration was equal or greater than the original can wall thickness of 0.065 in. At present the powder can is being examined by the Source Safety Testing group to determine if it meets the USA DOT requirements for a "special form" container.

[2] H. H. Van Tuyl et al., Interim Report on Low-Cost Cesium Radiation Sources from Hanford Waste Management Program, Battelle Northwest Laboratory, BNWL-CC-2245 (July 1969), p. 5.

[3] I. I. Il'yasov and A. G. Bergman, Melting Points in the Cs, K, Na || Cl Ternary System, Russian J. of Inorg. Chem. 7(3): 355-6 (1962).

[4] S. Żemczużny and F. Rambach, Schmelzen der Alkalichloride, Z. Anorg. Chem. 65: 418-20 (1910).

RADIATION SOURCE DEVELOPMENT -- 08-01-04

A. Strontium-90 Silicate Sources

1. Matrix Studies

Nothing to report.

2. Encapsulation

Nothing to report.

3. Characterization

Test solutions and two pounds of inactive strontium silicate minibeads have been prepared for the characterization studies.

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

A technique to produce curie quantities of elemental carbon-14 was needed to satify two requirements: a stable carbon-14 compound, such as MoC, is needed for the development of carbon-14 sources and for elemental carbon-14. A library search revealed two techniques that appeared to be applicable. Rutherford[5] has produced elemental carbon-13 from CO using hydrogen reduction on an iron catalyst at a reaction temperature of 600°C. Yields of greater than 90% were obtained with impurity levels of less than 1%. Ruderman and Wu[6] have produced elemental carbon-14 by heating barium carbonate-^{14}C in the presence of various metals (Na, K, Mg, Li, Mn) in a sealed quartz tube. Rutherford's technique had three disadvantages: the iron catalyst was difficult to prepare, the hydrogen reduction coupled with curie quantities of carbon-14 was considered to be potentially hazardous if an explosion occurred, and iron catalyst contamination of the elemental carbon could interfere with the use of the final product. Ruderman's direct conversion technique appeared to be more applicable to our particular problem; however, their study was exploratory in nature and the highest yield they obtained was 60%. Since neither technique fulfilled our requirements, a technique which utilizes concepts from both procedures was developed. In the new procedure magnesium metal is used to reduce $^{14}CO_2$ obtained by acid decomposition of $Ba^{14}CO_3$. The $^{14}CO_2$ gas is circulated through a quartz tube containing a boat of magnesium metal heated to 750°C in an electric tube furnace. The first run of high specific activity (85%) ^{14}C-barium carbonate that was processed to elemental carbon had a greater than 90% yield.

C. Alpha Source Fabrication Development

Nothing to report.

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[5]William M. Rutherford, Preparation of Elemental Carbon-13 in High Yield From $^{13}CO_2$, J. Am. Chem. Soc. 88: 179 (1966).
[6]I. W. Ruderman and C. S. Wu, Preparation of Carbon-14 from $Ba^{14}CO_3$-^{14}C, AECD-2412 (1948).

D. Sources for Medical Application

1. Blood Flow Patterns Using Radioactive Minibeads

A study of blood flow patterns in animals at the Cardiovascular Research Institute in California requires radioactive minibeads in the 10- to 20-μm-dia size range. The experimental procedure consists of injecting small quantities of radioactivity in the form of small spheres into the animal's blood stream and then following the path of the spheres with appropriate radiation detection equipment. The spheres deposit in the arterioles as a function of blood flow rate to the various parts of the body and deviation from normal blood flow can be observed by scanning. Strontium-90 silicate will not necessarily be used; but normal strontium silicate is being used as a carrier for other radionuclides, since the fabrication techniques for this material have been developed.

The procedure consisted of injecting strontium silicate fines (screened through a 325-mesh screen) into an air stream (2 liter/min) produced by applying a vacuum to the bottom of the sphere-forming furnace. The minibeads were collected on glass wool placed in the lower end of the tube; essentially all of the feed was converted to spheres with an overall recovery of 95%.

2. Sources for Low-Level Beta Irradiation of Animal Skins

A proposal for the fabrication of four low-level sources (two sources 1 to 2 rads/hr and two sources 10 to 15 rads/hr) was submitted in answer to an inquiry for sources to be used for low-level beta irradition of the skin of animals in an experimental program conducted by the National Aeronautics and Space Administration. Two designs were submitted: a square source capsule (8 by 8 cm) with a 0.005-in.-thick stainless steel window and an active plate made of strontium-90 silicate minibeads encapsulated in an aluminum matrix and a flexible source that could be used to follow the contour of the body of the animal.

The design of a high-pressure gamma process irradiator is approximately 80% complete.

RADIOISOTOPE SOURCE SAFETY TESTING - 08-01-04

A. Special Form Prototype Testing

Nothing to report.

B. Testing Sources with Manufactured Leaks

Nothing to report.

C. Surveillance of Regulations and Cask Evaluations

Up-to-date information on radioisotope regulations is maintained by review of the Federal Register. Comments were forwarded to AEC-ORO regarding USA DOT Federal Highway Administration's Notice No. 69-17, Safe Loading of Motor Vehicles. It was pointed out that while the proposed changes to regulations pertaining to the securing of cargoes being transported by motor vehicles will not present major problems to the transport of large radioactive material containers, minor word changes in the proposed regulations would simplify their utility as applied to radioactive material packaging. It was suggested that large quantity radioactive material and fissile packaging be included in the list of exempted articles, since the safety standards already in effect for these packagings are much more stringent than the proposed regulations.

Large quantity radioactive material packages are evaluated prior to application for AEC certification and petitions for USA DOT approval. The SNAP-7D Thermoelectric Generator and the UKAEA Fissile Package GB:0504A/GB:TCC5 were evaluated to determine compliance with the safety standards of AEC Manual Chapter 0529. The SNAP-7D device is a 224,000-Ci strontium-90 powered thermoelectric generator the U. S. Navy used to power a floating meteorological station in the Gulf of Mexico. This generator is scheduled to be returned to ORNL for inspection and storage. The UKAEA Fissile Package is a British Type B, Class I fissile shipper similar to the design listed in IAEA transport regulations. This package will be used to transport kilogram quantities of $^{233}UO_2(NO_3)_2$ solution from ORNL to a Euratom site.

A certification that the SNAP-7E Thermoelectric Generator meets the requirements of AECM-0529 was received from AEC-ORO. A petition for a special permit was prepared in the format of and including the information required by 49 CFR 170. This petition has been sent to the Hazardous Materials Regulations Board of the Department of Transportation.

The structural and thermal analysis of the light-weight drum package for transuranium radionuclides is continuing.

New designs of shipping containers are tested to determine their effectiveness during transport. Testing of shielding effectiveness and thermal dissipation has been delayed, since the 6000 lb of lithium hydroxide monohydrate to be used in the shielding test has not been received.

D. Source Leak Test Guide

A guide for leak testing encapsulated radioactive sources is being edited for publication.

E. Sealed Source Testing

Nothing to report.

F. Unsealed Source Testing

Testing of commercial tritium foils was continued. Since previous testing did not cover the temperature span between 25 and 100°C, additional tests were made at 44°C. The foils were cut into eight equal area sections and one of these sections was tested at the stated temperature. The foils were purged with dry air for 16 hr prior to testing to minimize differences due to humidity. In each test a section of foil was placed in a quartz tube which had been preheated to the desired temperature; a dry air purge was provided to sweep the released tritium into the recovery train to obtain a 2-hr sample. The tritium losses from the 1-, 4-, 8-, and 15-Ci/in.^{2} foils were 0.000126, 0.0000126, 0.0000088, and 0.0000099%, respectively. Most of the losses were in the form of tritium oxide.

G. Examination of Commercial Sources

A 600-mCi krypton-85 source which had leaked in use has been examined to determine the location and nature of the leak hole. This source had been used in a gaging operation for approximately 1.5 years. The radioactive gas was singly encapsulated in a monel cylinder having a 0.002-in.-thick window at one end. The seals were made using a silver solder. The source was located in a holder and was held in place by a coil spring. The leak was found to be a 3/8-in. separation of the silver solder seal holding the window to the source capsule. A visual inspection at a magnification of 30X did not reveal the reason for the failure of the seal, but it was probably due to a weak spot in the braze area.

Thermal cycling might be a problem for this type of source design, since it was noted that each time the microscope light was turned on, the window expanded and made a pinging noise. This same noise occurred when the window was allowed to cool. Repeated flexing of the window in thermal cycling might cause fatigue failure of the seal.

Small patches of a brown-colored foreign material were visible on the window of the source. In some cases the brown material was easily removed, revealing clean metal beneath, in others pit holes were found. None of these pit holes were observed to extend through the window.

Publication: R. G. Niemeyer, Containment Integrity of ^{226}Ra and ^{241}Am Foils Employed in Smoke Detectors, ORNL TM-2684 (Oct. 21, 1969).

TECHNOLOGY UTILIZATION - 08-01-05

A. Information Center

The number of accessioned items available for searching by keywords has now reached 16 thousand, and there is virtually no indexing backlog. New acquisitions are being indexed and accessioned as soon as received.

During December, 89 requests for information were received plus requests for more than 300 IIC reports and reprints. A list of reviews in progress is given below:

Title	Author(s)	Status, % complete
Cement-Polymer Combinations	Martha Gerrard, P. S. Baker, R. E. Greene	75
DID Research and Developments - 1969	Isotopes Information Center Staff	85
Flow Measurement	R. H. Lafferty, Jr.	5
Indium-113m: Preparation and Uses	Martha Gerrard	10
Iodine-125	P. S. Baker, Martha Gerrard	25
Isotopic Methods of Examination and Authentication in Art and Archaeology	F. J. Miller, E. V. Sayre, B. Keisch	90
List of AEC Radioisotope Customers with Summary of Radioisotope Shipments	Ruth Curl, compiler	At Printer
Oceanography[a]	Isotopes Information Center Staff	90
Patent Review - Process Radiation Development	R. E. Greene, P. S. Baker, Helen Warren	80
Potato Sprout Inhibition by Radiation	F. E. McKinney	15
Radiation vs Glass	D. N. Hess	20
Radioisotopes in Automatic Control Systems	R. H. Lafferty, Jr.	-
Radioisotopes in the Pharmaceutical Industry	P. S. Baker, Martha Gerrard	5
Radioisotopes in the Steel Industry[b]	Republic Steel Corporation	10
Radioisotopes in the Textile Industry	F. J. Miller	90
Radioisotopes in X-Ray Fluorescence Analysis	R. H. Lafferty, Jr.	5
Selected Abstracts of World Literature on Production and Industrial Uses of Radioisotopes, 4th Quarter 1969	Martha Gerrard, P. S. Baker	10
Self-Diffusion in Liquids	F. J. Miller	75
Snow Gaging and Water Management	James L. Smith	-
Strontium-90	Roberta Shor, R. H. Lafferty, Jr., P. S. Baker	85
Technetium-99m: Preparation and Uses	Martha Gerrard, P. S. Baker	25
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

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

B. Isotopes and Radiation Technology

Galley proof for Isotopes and Radiation Technology 7(3) was checked and returned to the publisher (DTIE), and manuscript for 7(4) was completed.

In order to acquaint heart specialists with the current work being sponsored by the AEC and NHI on nuclear-powered heart-assist devices, a special "Heart" issue of Isotopes and Radiation Technology 7(2) was mailed to 2500 members of the American Heart Association and the American College of Cardiology. The entire issue is devoted to a consideration of these devices and includes descriptions of several isotope-powered heart circulatory devices, discussions of the preparation of candidate power fuels, a brief mention of the cardiac pacemaker, and a report on in-phantom dosimetry measurements.

C. Publications

P. S. Baker, Robert H. Lafferty, Jr., Martha Gerrard, R. E. Greene, F. E. McKinney, F. J. Miller, and D. N. Hess (Eds.) and A. F. Rupp (Adviser), Isotopes and Radiation Technology 7(2) (Winter 1969).

P. S. Baker, "Nuclear Data," pp. 3-63 in Handbook of Radioactive Nuclides, Part I, ed. by Yen Wang, The Chemical Rubber Company, Cleveland, Ohio, 1969.

P. S. Baker, "Radionuclides for Industrial Applications," pp. 503-70 in Handbook of Radioactive Nuclides, Part VII, ed. by Yen Wang, The Chemical Rubber Company, Cleveland, Ohio, 1969.

Martha Gerrard, Milk Preservation by Ionizing Radiation: Literature Review, ORNL-IIC-26 (November 1969).

Martha Gerrard and P. S. Baker, Selected Abstracts of World Literature on Production and Industrial Uses of Radioisotopes, ORNL-IIC-22 (Part 3) (December 1969).

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 - 03-03-02

A. Oceanographic Systems Study

Preparations for the Oceanside, California, experiment in February are continuing. The new core injection device has been tested in the laboratory and will be ready for the field test at Oceanside.

A stationary detector has been assembled and preliminary testing has been accomplished. This detector will be used to continuously monitor a core injection. The detector is being operated with 2000 ft of electromechanical cable. Preparations for calibrating this detector with a gold-198 tagged sand core are nearly complete and the calibration will be accomplished by the end of January.

Several interpolation techniques can be applied to the raw data obtained during past surveys. Since the actual surveyed areas represent only 1% of the total study area, it is assumed that an interpolation technique is required to accurately describe the transport system. Since the required degree of interpolation is not known, several theories are being tested. The rate of movement of the perimeter defining the area containing the radioactive tagged sand can now be calculated directly from the raw data, linear interpolation in the x and y direction, linear interpolation in the x or y direction, cubic spline[7] interpolation in the x and y direction, and cubic spline interpolation in the x or y direction. By applying these techniques to the same data, the most suitable technique can be determined. This information can be factored into the data acquisition techniques which may ultimately simplify data acquisition and data treatment. Interpolation is also a necessary step in providing quantitative data and a radiation material balance. The immediate goal is to process all of the previously collected data and to present the results in topical reports. Studies for simplification of the survey system controls are continuing.

B. National Security

Nothing to report.

RADIATION PROCESSING APPLICATIONS DEVELOPMENT

ORGANIC SYNTHESIS WITH ISOTOPIC SOURCES - 08-04-01

A. Woolen Mill Effluent

The results from the dieldrin analysis has been received from Southeast Water Laboratory. They are shown in Table 5.

Table 5. Results from Dieldrin Analysis of Irradiated Woolen Mill Effluent Samples

Volume, ml	Oxygen Pressure, psi	Time Irradiated, min	Total Dose, R	Percent Light Transmission	Dieldrin, ppb
			Yellow-Gold Dye Vat Solution		
Control				8.2[a]	350
100	1600			9.3[a]	53
100	Atmospheric	12	1.36×10^6	63[a]	12
100	1600	6	6.8×10^5	98[a]	41
100	1600	12	1.36×10^6	Not measured	11
100	1600	12	1.36×10^6	Not measured	11
			Bright Orange Dye Vat Solution		
Control				5.1[b]	850
100	1600			55.1[b]	96
100	Atmospheric	12	1.36×10^6	98[b]	61
100	1600	10	1.13×10^6	99[b]	7
100	1600	12	1.36×10^6	Not measured	97
100	1600	12	1.36×10^6	Not measured	97
			Total Woolen Mill Waste		
Control				22[c]	450
100	2000			18[c]	60
100	Atmospheric	71	8×10^6	29[c]	10
100	2000	71	8×10^6	38[c]	21
100	2000	71	8×10^6	38[c]	21

[a]At 412 mμ maximum. [b]At 410 mμ maximum. [c]At 400 mμ maximum.

[7]J. L. Walsh, J. H. Ahlberg, and E. N. Nilson, Best Approximation Properties of the Spline Fit, J. of Mathematics and Mechanics 11(2): 225-34 (1962).

B. Radiolytic Hydrolysis of Glycidol to Glycerin

Synthetic glycerin is being produced at a rate of 250 million lb per year. In 1968 FMC started synthetic glycerin production by a peracetic acid-glycidol process at its new Bayport, Texas, plant which is a 40 million lb per year unit.

Since the epoxide function of glycidol can be hydrolyzed it was felt that gamma radiolysis could be utilized in such a process. Irradiation of glycidol in different solvent systems was investigated and a 50% water-50% acetic acid mixture was the most efficient. This result was probably due to the increased solubility of oxygen in acetic acid which could be used to form the hydroxyl radicals that are essential for the conversion (hydrolysis) of glycidol to glycerin. An unknown product, probably an acid or an ester, was also produced in smaller amounts. Mass spectrometric analysis is being conducted on the unknown product. The initial concentration of glycerin immediately after irradiation (1.12×10^7 R) showed a dependence on the volume of solvent used.

A study was made to compare the rate of conversion between identical solutions of glycidol in 50% water-50% acetic acid. In one case the solution was maintained at room temperature for 50 hr without exposure to radiation. In the other the solution was irradiated for 1 hr at 1.12×10^7 R/hr and then maintained at room temperature for 50 hr. The experiment was repeated with both samples under 1500 psig of oxygen. The results are shown in Table 6.

Table 6. Rate of Conversion to Glycerin of Identical[a] Irradiated and Nonirradiated Solutions of Glycidol and 50% Water-50% Acetic Acid

Time, hr	Nonirradiated Sample		Irradiated Sample[b]	
	Glycerin, %	Unknown, %	Glycerin, %	Unknown, %
		No Pressure		
2	8.9	7.6	14.3	11.3
3	12.0	8.1	21.2	12.8
4	14.3	8.1	23.0	12.5
5	15.8	8.9	25.7	13.7
6	21.7	9.5	30.2	14.1
7	22.9	11.1	30.9	14.2
24	62.7	15.7	68.9	19.1
50	77.2	20.1	77.8	20.9
		1500 psig of Oxygen		
1			30.7	12.3
2	12.1	9.2	32.6	12.7
4	14.3	9.2	36.2	13.3
5	18.3	9.8	40.9	13.7
6	21.1	10.3	43.1	14.8
7	26.8	11.3	c	c
24	60.8	17.2	c	c
48	77.4	19.8	c	c

[a] 5 ml of glycidol and 10 ml of 50% water-50% acetic acid.
[b] Dose - 1.12×10^7 R/hr for 1 hr.
[c] No measurements made due to weekend.

There was not much difference in the rate of conversion when 2.73×10^6 R/hr for 1 hr was used and there was no advantage in increasing the total dose. No additional studies are planned.

The efforts in January will be concentrated on allyl alcohol and its conversion to glycerin and perhaps on long chain hydrocarbon conversion to fatty acids.

RADIOISOTOPE SALES

Requests for quotations were received from Sanders Nuclear Corporation for an approximately 30,000-Ci strontium-90 heat source and from Lovelace Foundation for 5- to 10-Ci quantities of strontium-89 and yttrium-91. An order was received from Japan (Mitsubishi) for 5001 Ci of promethium as Pm_2O_3.

Shipments made during December included 6,100 Ci of cesium-137 to Lockheed-Georgia Nuclear Laboratory (material in the form of sources; the activity was billed in previous fiscal years), 12 Ci of molybdenum-99 to Australian Atomic Energy Commission, 105,882 Ci of strontium-90 to Isotopes, Inc. (material in the form of sources; the activity was billed in previous fiscal years), 15,000 Ci of tritium to Great Britain (Radiochemical Centre), and 30,900 Ci of strontium-90 to Aerojet General Corporation.

A list of outstanding large orders is shown below:

Isotope	Amount, Ci	Customer
Cesium-137	360,000	Brookhaven National Laboratory[a]
Cesium-137	~8,000	Lockheed-Georgia Nuclear Laboratory[a]
Cesium-137	~180,000	American Nuclear Corporation[a]
Cesium-137	~207,000	U. S. Army, Natick Laboratories[a]
Cesium-137	205,000	Industrial Reactor Laboratories, Inc.
Cesium-137	1,180,000	Radiation Machinery Corporation
Cesium-137	~194,000	Atomic Energy of Canada Limited
Cesium-137	200,001	General Nuclear, Inc.
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	~5,000	Miscellaneous orders
Strontium-90	300,000	CEA, France
Strontium-90	~532,000	Isotopes, Inc.[a]
Strontium-90	~105,000	Isotopes, Inc.[a]
Strontium-90	~41,000	Brookhaven National Laboratory
Strontium-90	4,951	Radiation Machinery Corporation
Strontium-90	~2,000	Miscellaneous orders
Promethium-147	5,100	Reactor Centrum Nederland
Promethium-147	5,000	Mitsubishi Corporation (Japan)
Carbon-14	1,445 mCi	Catholic University

[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.

Tables 7 and 8 give the ORNL and non-ORNL cyclotron customers and runs for December 1969.

Table 7. ORNL Cyclotron Customers and Runs for December 1969

Customer	Product	Target	Time, hr:min Beam	Misc	Total	Total Charges
Isotopes	Neodymium-140	Praseodymium-141	2:00	1:05	3:05	$ 302.92
	Rubidium-83	Strontium-84	2:00	1:15	3:15	308.75
	Gallium-67	Zinc	2:00	:30	2:30	255.04
Physics	Cobalt-61	Nickel-64	1:15	1:15	2:30	247.50
	Cobalt-61	Nickel-64	1:15	1:15	2:30	247.50
ORAU	Gallium-67	Zinc	2:25	1:15	3:40	343.85
	Gallium-67	Zinc	1:00	:30	1:30	160.04
Total			11:55	7:05	19:00	$1865.60

Table 8. Non-ORNL Cyclotron Customers and Runs for December 1969

Customer	Product	Target	Time, hr:min Beam	Misc	Total	Total Charges
Idaho Nuclear Corporation	Rhenium-183, Rhenium-184	Tungsten	4:00	1:15	5:15	$ 820.10
New England Nuclear Corporation[a]	Yttrium-87	Strontium-87	3:30	1:15	4:45	750.10
	Rubidium-84	Krypton	1:30	:25	1:55	341.67
	Yttrium-87	Strontium-87	3:30	1:15	4:45	750.10
	Gold-195	Platinum	14:00	1:15	15:15	2706.77[b]
Huntington Hospital	Rubidium-84	Krypton	8:00	:50	8:50	1383.32
International Chemical & Nuclear Corporation[a]	Gold-195	Platinum	5:00	1:15	6:15	1446.77[b]
Stock	Rubidium-84	Krypton	2:00		2:00	
Totals			41:30	7:30	49:00	$8198.83

[a]Cobalt-57 runs to International Chemical & Nuclear Corporation and New England Nuclear Corporation from last month have been shipped for a total of $17,461.78.
[b]Purchased but not shipped.

The radioisotope sales proceeds and shipments for the first five months of fiscal years 1969 and 1970 are given in Table 9.

Table 9. Radioisotope Sales and Shipments

Item	7-1-68 thru 11-30-68	7-1-69 thru 11-30-69
Inventory items	$213,539	$229,248
Major products	43,330	48,773
Radioisotope services	83,916	71,951
Cyclotron irradiations	64,606	45,549
Miscellaneous processed materials	28,403	37,096
Packing and shipping	33,902	30,425
Total Radioisotope Sales	$467,696	$463,042
Total Radioisotope Shipments	1,163	1,011

RADIOISOTOPE PRODUCTION

CESIUM-137 PROCESSING

No cesium-137 processing was done this month. One shipment of 58,000 Ci of cesium-137, in 145 units, was transferred for source fabrication.

STRONTIUM-90 PROCESSING

Two $^{90}SrTiO_3$ preparations were made using our new potassium carbonate precipitation procedure. Material balances and process details for the two runs are shown below:

	Batch No. 10SR-69	Batch No. 11SR-69
Feed Analysis, Ci		
Based on feed storage analysis	69,900	46,200
Based on individual batch analysis	58,900	42,500
TiO_2 Addition, g	949	663
pH Adjustment, liters		
3 M $KHCO_3$	0.5	0.9
5 M K_2CO_3	5.0	5.0
Product - $^{90}SrTiO_3$		
Weight, g	1,696	1,477
Activity concentration (by calorimetry), Ci/g	28.3	31.7
Uncalcined product, Ci	7,500	0
Total activity, Ci	55,600	46,800
Waste		
Volume, liters	78	68
Strontium-90 content, Ci	<10	100
Total Strontium-90 Activity Balance, Ci		
Feed	58,900	42,500
Product	55,600	46,800
Waste	0	100
Percent accounted for	94.4	110.3
Combined Material Balance, Ci		
Feed	101,400	
Accounted for	102,500	
Percent accounted for	101	

The apparent discrepancies in the two runs probably resulted from a holdup of product in the system during the first batch. The general handling aspects of the new process are definitely superior to those of the ammonium carbonate process and product quality is comparable.

MAINTENANCE

Decontamination of three manipulator cells used for cesium-137 processing and general operations is underway. All waste and disposable equipment has been removed from these cells and remote decontamination was started.

The top shield from a shielded service area was removed in preparation for the installation of a filter/dehumidifier in the off-gas system.

The piping in the unloading station for feed shipping casks is being revised for use with the new HAPO-type cesium-137 cask; this work is approximately 70% complete. A dummy cask head has been fabricated and will be used to check out this revised piping as well as to familiarize operating personnel with the new cask connection procedures.

One of the two large-volume chemical makeup systems was removed except for the solution addition lines to process vessels. Two glove boxes will be installed in the area formerly occupied by these makeup tanks. The glove box system will be used for preparation of encapsulation materials, inspections, and similar work.

An existing large-volume gas storage system was tested and repiped to provide an argon supply system for several uses in FPDL. This revision will eliminate the handling of large numbers of gas cylinders. The nitric acid storage system was also reworked so that the main storage tank can be filled from a tank truck by gravity flow. This revision provides the ability to receive the less-expensive bulk acid and eliminates the problems associated with storage, handling, and pumping of nitric acid in small containers.

There were nine manipulator pulls during the month; four were for mechanical repairs and five for boot replacement. Two loads of contaminated waste were delivered to the Solid Waste Disposal Area. Disposal of solid waste is now on a current basis. A large-volume shielded waste container has been fabricated and is undergoing tests. This container has less shielding than the presently used waste containers but will accept a 55-gal drum (vs approximately 15 gal total volume for the heavily shielded containers). With proper segregation and precleaning of contaminated waste, the number of waste transfers should be reduced significantly by the use of this new container.

SOURCE FABRICATION

A. Cesium-137

Fabrication of BNL-strip $^{137}CsCl$ sources continued on a routine basis. Twenty-six of these units were fabricated and tested; these are now on shelf-test preparatory to shipment. This cycle included 11 fully loaded sources and 15 patterned sources (five different types). Only one reject, a leaking outer capsule, was experienced during this cycle. To date 59 rectangular sources have been fabricated and tested by the new procedures.

Welding of a batch of cylindrical 400-Ci $^{137}CsCl$ fueled sources was started. This order is for 210 units of which 84 have been completed and are on shelf-test for Radiation Machinery Corporation.

Three short-form BNL-strip sources containing 2,000 Ci each were shipped to Lockheed-Georgia Company.

B. Strontium-90

One 720-W $^{90}SrTiO_3$ source (106,000 Ci) was loaded into a generator and shipped to Isotopes, Inc. The SNAP-23 source (166,000 Ci) was loaded into an experimental generator and shipped to Westinghouse.

SHORT-LIVED FISSION PRODUCT OPERATIONS

Nothing to report.

SPECIAL FORM MATERIALS

Four hot-pressed $^{90}SrTiO_3$ pellets were canned in individual customer-supplied stainless steel containers which were welded, tested, and loaded into two casks for shipment. Calorimetric assay of the completed units showed that all were identical in heat output - 52.5 W each. These pellets were made from $^{90}SrTiO_3$ prepared by the potassium carbonate procedure. The average power density of the pellets was 0.93 W/cm^3. No significant differences in pellet characteristics were observed in these pellets as compared to similar pellets made from $^{90}SrTiO_3$ prepared by the ammonium carbonate process.

OPERATIONAL SUMMARY

	December 1969		1969 Totals	
	Number	Amount, Ci	Number	Amount, Ci
Cesium-137				
HAPO shipments received	0	0	0	0
Product batches prepared	0	0	20	231,000
Sources fabricated	26	112,000	322	647,000
Special form containers	0	0	0	0
Strontium-90				
HAPO shipments received	0	0	0	0
Product batches prepared	2	102,500	11	409,500
Sources fabricated	0	0	0	0
Special form containers	4	30,900	9	32,900

PROCESS STATUS

Physical Inventory

Item	Cesium-137,[a] Ci	Strontium-90,[b] Ci
In-process materials	244,500	1,693,600
Product powder	433,800	930,000
Sources in fabrication	60,200	232,900
Completed sources	401,600	404,400[c]
Total	1,140,100	3,260,900

[a]Includes 835,000 Ci of billed material.
[b]Includes 420,000 Ci of billed material.
[c]Includes stored sources (SNAPs, Sentinel, etc.).

MISCELLANEOUS RADIOISOTOPE PROCESSING

Short-Lived Fission Products			Processed Units			Service Irradiations	
Radioisotope	No. of Batches	Amount, Ci	Radioisotope	No. of Batches	Amount, mCi	Type	No. of Irradiations
Xenon-133	2	1,200	Indium-114	1	1,285	HFIR monitor	1
Iodine-131	2	160	Copper-67	2	28	ORR monitor	1
Ruthenium-103	1	22	Calcium-47	1	12	Platinium-196	3
			Barium-131	1	9	Y_2O_3	1
			Iodine-131 (elemental)	1	200	^{67}ZnO	1
						Gold	1
						SiO_2	1
Total	5	1,382		6	1,534		9

A recovery run of cerium-144 is in progress at the Short-Lived Fission Product cells. The feed material for this run consists of retained fission-product cuts from the recovery of other isotopes. A sample of this feed material indicates that approximately 40 Ci of cerium-144 is present. This product will be purified for small quantity shipments to customers requiring processed material. Requirements for greater than 50-Ci quantities of aged cerium-144 cannot be met with this method of processing.

ADMINISTRATIVE SECTION

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

Table 10. Travel by IDC Personnel

Site Visited	Purpose of Visit
Knoxville, Tennessee	Attend training session on use of MT/ST
Oak Ridge, Tennessee	Attend ORAU Medical Division symposium

Table 11. Visitors to IDC

Visitors (affiliation)	Purpose of Visit
Atlantic Richfield Hanford Corporation	Discuss manipulator cell design and equipment
National Bureau of Standards	Discuss reactor and isotopes problems
Accelerator and Radiation Source Review Committee	Inspect cyclotron facilities

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