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RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR APRIL 1971

A. F. Rupp

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Isotopes Development Center

RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR APRIL 1971

A. F. Rupp

Work Sponsored by
AEC Division of Isotopes Development

MAY 1971

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RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR APRIL 1971

A. F. Rupp

RADIOISOTOPE PRODUCTION AND MATERIALS DEVELOPMENT

REACTOR-PRODUCED ISOTOPES - 08-01-01

A. Biomedical Radioisotopes

1. Phosphorus-33

The purpose of this project is to develop methods of preparing hundred-millicurie quantities of carrier-free phosphorus-33 containing ≤5% phosphorus-32. The two methods of producing phosphorus-33 being evaluated are based on the irradiation of highly enriched targets of sulfur-33 (>92 at. %) or chlorine-36 (approximately 63 at. %) in a fast neutron flux. Phosphorus-33 (25.2 days; 0.248-MeV β^-_{max}) has both a longer half-life and a lower energy beta than phosphorus-32 (14.3 days; 1.709-MeV β^-_{max}), which makes it advantageous for autoradiography, longer ecological and agricultural experiments than with phosphorus-32, synthesis of tagged complex organo-phosphorus compounds, and double labeling experiments.

A target of 1.74 g enriched (67%) $K^{36}Cl$ was irradiated for two reactor cycles. After two weeks' decay the target was processed to give 1 Ci phosphorus-33 containing 3% phosphorus-32 as a final product. The only gamma impurity detected was 0.7 μCi/ml of chromium-51 activity. The phosphorus-33 product was analyzed by spark source mass spectrometry for elemental content. The elements detected and the amounts are shown below:

Element	Amount (μg/ml)	Element	Amount (μg/ml)
Aluminum	3	Sodium	20
Boron	0.4	Nickel	0.2
Barium	1.5	Phosphorus-31	0.6
Bromine	0.02	Lead	0.8
Calcium	16	Silicon	4
Chromium	0.4	Strontium	0.06
Copper	0.6	Titanium	1.5
Iron	3	Zinc	5
Potassium	1	Sulfur	10
Magnesium	4	Phosphorus-33	0.17
Manganese	0.1		

One interesting feature of the above analysis is the detection of phosphorus-33 atoms well above the sulfur-33 background. The activity concentration of phosphorus-33 was measured as 26 mCi/ml, equivalent to 0.17 μg/ml. Since the 31 mass is unambiguously assigned to phosphorus, the analysis specifies the isotopic abundance of phosphorus-33 atoms in the phosphorus product as ∿20%.

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-43, with a half-life of 22.4 hr and gamma-ray emissions of 0.373 and 0.617 MeV, is potentially useful for metabolic and clinical studies of blood flow, rejection of transplanted organs, and kidney function because the dose rate would permit multiple doses.

One batch of potassium-43 was made. Data for the run are summarized in Tables 1 and 2.

Table 1. Yield of Potassium-43 from Calcium-43 Target

	Target 332
Weight ^{43}CaO (∿50%),[a] mg	242
Irradiation time, hr	65
Product delivery date	3/31/71
Total product at 8:00 AM on delivery date, mCi	
Potassium-43	15.1
Potassium-42	1.9 (12.3%)

[a]Recovered February 1971. Isotopic analysis: ^{40}Ca, 21.9 at. %; ^{41}Ca, <0.001 at. %; ^{42}Ca, 8.31 at. %; ^{43}Ca, 50.7 at %; ^{44}Ca, 19.0 at. %; ^{45}Ca, ≤0.01 at. %; ^{46}Ca, <0.001 at. %; and ^{48}Ca, 0.057 at. %.

Spark source mass spectrographic analysis of target (wt ppm): Al (500); B (30); Ba (10); Cd (80); Cr (60); Ca (50); Fe (300); K (500); Mn (80); P (30); Pb (100); Si (>5,000); S (>10,000); Ce (50); and Yb (350).

Table 2. Radionuclides Found in Dissolver Sample by Gamma Scanning

Radionuclide	Half-life	Activity at Discharge[a] (mCi)	Activity Concentration at Discharge (μCi per mg of target)
Potassium-43	22.5 h	37.7	156
Potassium-42	12.4 h	8.8	36
Sodium-24	15.0 h	7.4	31
Manganese-56	2.58 h	186	770
Strontium-87m	2.83 h	21	87
Calcium-47	4.53 d	0.55	2.3
Scandium-46	83.8 d	4.5	19
Scandium-47	3.35 d	obscured by gold-199	
Europium-152m	9.3 h	15	62
Europium-156	15.2 d	0.6	2.5
Samarium-153	46.8 h	4	17
Cadmium-115	53.5 h	3	12
Lanthanum-140	40.2 h	1.3	5.4
Ytterbium-169	32 d	0.9	4
Ytterbium-175	101 h	220	910
Cerium-143	33 h	0.2	0.8
Gold-199	3.15 d	3	12
Zinc-69m	13.8 h	0.5	2

[a]Total target.

The abnormal radiocontaminants in the dissolver solution, which correspond with the spectrographic analysis in Table 1, resulted from a mishap during the last target recovery. The major portion of the target was spilled in the hood and had to be recovered by vacuuming into a suction flask. The target was dissolved in nitric acid and then recovered by the usual oxalate precipitation which also carried the rare-earth contaminants. The recovery was ∿90%, but it was noted that the calcined powder had a much greater density than usual and resulted in a greater target loading for the same size ampule than was previously possible. The spectrographic analysis points to sulfate as the chief component difference and suggests that $CaSO_4$ could offer potential for greater target loading. This incident points out the effectiveness of the chemical procedure because the cation column removes the contaminants, and none were detected in the potassium-43 product. The elution of the target calcium will also remove most of the rare-earth contaminants.

Shipments were made under medical cooperative programs to Johns Hopkins Medical Institution, Peter Bent Brigham Hospital, and V. A. Hospital Durham.

B. Exploratory Development of Products and Techniques

1. Gadolinium-153

Gadolinium-153 (241 days) is an important radionuclide because it decays by electron capture to yield predominantly 100-keV photons. This energy range is useful in backscatter and transmission gaging, as well as having applications in other fields. Highly enriched transmutation product gadolinium-152 is prepared by chemically separating it from neutron burned-out europium control plates from HFIR. The >90% abundant gadolinium-152 is not available from calutron separations and can only be obtained at reasonable cost as a by-product of the HFIR reactor operations at the present time.

Work is continuing on the purification of the gadolinium recovered from a HFIR control rod. A sample of this material will be converted to Gd_2O_3 and irradiated to check the gadolinium-153 yield.

C. Reactor Products Pilot Production (Production and Inventory Accounts)

Processed Units		Service Irradiations	
Radioisotope	Amount (mCi)	Type	Number
Calcium-47	24	Platinum-196	3
Palladium-109	1,260	U_3O_8	1
Copper-67	12		
Scandium-46	17,900		
Silver-111	288		

D. Special Sources

1. Cobalt-60

A program to produce cobalt-60 with a specific activity of greater than 1000 Ci/g was started in June 1969. This source material will be used for making improved radiographs of high density materials. A sample containing approximately 12 grams of cobalt-59 was neutron irradiated to produce cobalt-60 and reduce the cobalt-59 content to less than 5%. Nickel, which is a decay product of cobalt-60, will be removed to obtain a specific activity of 1000 Ci/g.

The cobalt metal target will be dissolved, and the nickel contaminant will be removed by ion exchange. The cobalt-60 will be reduced to elemental cobalt by electrodeposition and reshaped to the desired form by hydraulic pressing. The metal pellets will then be encapsulated in stainless steel capsules.

E. The Rare-Earth Radionuclides

It is becoming increasingly apparent that various insoluble compounds of rare earths (e.g., sulfides) in the form of colloids may be extremely useful in mechanically outlining obstructions due to cancerous tissue in the capillary and lymphatic systems of the body. The wide span of half-lives and gamma radiations available in this class of radionuclides which have essentially identical chemical properties allows a wide choice.

Many of these nuclides exhibit Mössbauer properties again with a wide range of resonance cross section, natural width, minimum observable width, and recoil energy.

The rare earths occupy a region of the chart of the nuclides where the spherical nuclei change to permanently deformed nuclei, with abrupt changes in nuclear structure properties. This transition region is particularly attractive for comparison of the observed nuclear properties to theoretical predictions.

As part of a continuing program of preparing high-purity rare-earth radionuclides we have investigated the preparation of the following Mössbauer radionuclides:

Nuclide	Half-Life	Mode of Preparation	Mössbauer Nuclide	Mössbauer Line (keV)	Ref.
^{156}Eu	15.2 days	$^{154}Sm(n,\gamma)^{155}Sm \xrightarrow{\beta^-} {}^{155}Eu(n,\gamma)^{156}Eu$	^{156}Gd	89.0	1
^{156}Tb	5.4 days	$^{156}Gd(p,n)^{156}Tb$	^{156}Gd	89.0	2,3
^{160}Tb	72.4 days	$^{159}Tb(n,\gamma)^{160}Tb$	^{160}Dy	86.8	4
^{166m}Ho	1200 years	$^{165}Ho(n,\gamma)^{166m}Ho$	^{166}Er	80.6	5
^{149}Eu	93 days	$^{149}Sm(p,n)^{149}Eu$	^{149}Sm	22	6

[1]A. F. Kluk, Ph.D Thesis, Vanderbilt University, 1971.
[2]D. J. McMillan, J. H. Hamilton, M. Mladjenovic, J. C. Manthuruthil, and J. J. Pinajian, *Z. Phys.* **241**, 103-116 (1971).
[3]D. J. McMillan, J. H. Hamilton, and J. J. Pinajian, *Phys. Rev.* (in press).
[4]M. A. Ludington, J. J. Reidy, M. L. Wiedenbeck, D. J. McMillan, J. H. Hamilton, and J. J. Pinajian, *Nucl. Phys.* **A119**, 398 (1968).
[5]J. C. Manthuruthil, H. J. Hennecke, and J. J. Pinajian, L-Subshell Conversion Line Intensity Ratios for the 82.45 Pure M1 Transition in Ho^{166}, *Radioactivity in Nuclear Spectroscopy — Techniques and Applications*, Gordon & Breech, New York, 1970 (in press).
[6]O. K. Harling, C. A. Ventrice, and J. J. Pinajian, *Phys. Rev.* **132**, 807 (1963).

For each of these radionuclides we have examined the production parameters, separation chemistry, purity, and characterization of the nuclide. The unique opportunity offered by the preparation of relatively large quantities of extremely high-purity radionuclides has presented us with a number of very interesting nuclides (from the nuclear structure viewpoint). We have

exploited this fact by carrying on cooperative programs with the Chemistry Division, Physics Division, and Nuclear Data Group of the Oak Ridge National Laboratory as well as carefully selected universities. Of the near spherical nuclides, 54.5-day europium-148 and 120-day gadolinium-151 have been investigated;[7,8] of the slightly deformed, 242-day gadolinium-153 is being investigated; and of the deformed nuclei, 5.1-day terbium-156 and 15.2-day europium-156 have been examined.[9,10]

1. Gadolinium-151

Carrier-free 120-day gadolinium-151 offers a rare-earth radionuclide decaying by electron capture (EC) to europium-151 and emitting gamma rays (predominantly 153.57, 174.65, and 243.22 keV) well suited for collimation. Hence, it has been suggested for use in long-term animal studies. The 21.6-keV gamma ray of europium-151 has been used in Mössbauer studies.

The principal modes of production of gadolinium-151 have been through the use of accelerators using the $^{151}Eu(p,n)^{151}Gd$ reaction and the $^{151}Eu(d,2n)^{151}Gd$ reaction, as well as spallation reactions initiated by protons on erbium and tantalum.

Although gadolinium-151 has been extensively investigated for the past decade because of its nuclear structure (gadolinium-151 is in the transition region, i.e., the region in which the nuclei are changing from the spherical to the deformed), comparatively little is known of the cross sections of the reactions used to produce it or even the production rate. Recently Bullock[11] reported the yield using 17.5-MeV protons having an upper limit of 72.5 μCi/mA.hr, while a yield of 5.1 mCi/mA.hr was obtained with 16-MeV deuterons.

A sample of 590 mg $^{151}Eu_2O_3$, isotopically enriched to 96.67% in europium-151, was irradiated with 15-18 MeV protons using the capsule target facility of the ORNL 86-Inch Cyclotron which employed a 189-μA beam current. After irradiation the $^{151}Eu_2O_3$ was dissolved in HNO_3, precipitated with NH_4OH, filtered, and redissolved in HCl. The rare earths were purified by passage through a Dowex 1 anion exchange column. The purified rare earth after additional treatment was then sorbed onto a Dowex 50W, 20-40 μm cation exchanger operating at 80°C and 120 psi. Elution with 0.25 *M* α-hydroxyisobutyric acid effectively separated the gadolinium-151 from the target material. The gadolinium-151 was separated from the butyrate by passing the solution through a Dowex 50W cation exchanger in the chloride form. An overall yield of about 85% was realized, and the total time of separation was less than 4 hr.

The yield based on the 153.57-keV photon (6.9%) and the 243.22-keV photon (6.2%) was 129 μCi/hr (±20%), which is equivalent to 680 μCi/mA.hr using a capsule-type target.

---

[7] J. W. Ford, A. V. Ramayya, and J. J. Pinajian, *Nucl. Phys.* A146, 397 (1970).
[8] V. Lakshminarayana, D. R. Dunn, S. M. Brahmavar, J. H. Hamilton, and J. J. Pinajian, *Bull. Am. Phys. Soc.* 16, 538 (1971).
[9] McMillan, *Z. Phys.* 241, 103-16 (1971).
[10] McMillan, *Phys. Rev.* (in press).
[11] R. J. Bullock, N. R. Large, I. L. Jenkins, A. G. Wain, P. Glentworth, and D. A. Newton, *J. Inorg. Nucl. Chem.* 31, 1929 (1969).

Figure 1 shows the gamma-ray spectrum taken with a 30-cm^3 Ge(Li) detector. The principal impurity was 242-day gadolinium-153 (~11%) produced by the concomitant $^{153}Eu(p,n)^{153}Gd$ reaction. This suggests, of course, that the cross section for the reaction is large and that it would be a good method of producing carrier-free gadolinium-153. Tracer quantities of 6.2-year europium-150 and 14-year europium-152, <0.05% and <0.13% respectively, were also observed.

The proposed decay scheme, published earlier,[12] is shown in Fig. 2. In the published work, only the relative gamma-ray intensities using the 153.57-keV lines as 100 were cited. If one assumes pure M1 transitions for the 21.54-, 196.46-, 260.39-, and 286.00-keV transitions, then by using the total internal conversion coefficients given by Hager and Seltzer,[13] one can estimate the absolute intensities of the gamma rays. The estimate of error in such a calculation is $^{+10\%}_{-50\%}$. These results are shown in Table 3. During the coming period, we will examine the gamma-ray spectrum of the 242-day gadolinium-153.

Table 3. Gamma-Ray Energies and Intensities From the Decay of Gadolinium-151 to Europium-151

Gamma-Ray Energy (keV)	Intensity[a,b] (%)
21.54	3.2
106.51	0.09
110.53	0.008
153.57	6.9
174.65	3.3
196.46	0.03
239.00	0.16
243.22	6.2
260.39	0.06
284.98[c]	—
286.00	0.09
306.52[c]	—
307.43	1.1
328.23	0.09
332.12	0.01
353.51	0.14

[a]Intensities are based on assumption of pure M1 transition for 21.54-, 196.46-, 260.39-, and 286.00-keV transitions. Total internal conversion coefficients calculated from the work of Hager and Seltzer.

[b]Estimate of error $^{+10\%}_{-50\%}$.

[c]Deduced energy, predicted by Coulomb excitation, studied. Photon is not observed.

[12]Ford, *Nucl. Phys.* A146, 397 (1970).
[13]R. S. Hager and E. C. Seltzer, *Nucl. Data* A4(1 and 2), 1 (1968).

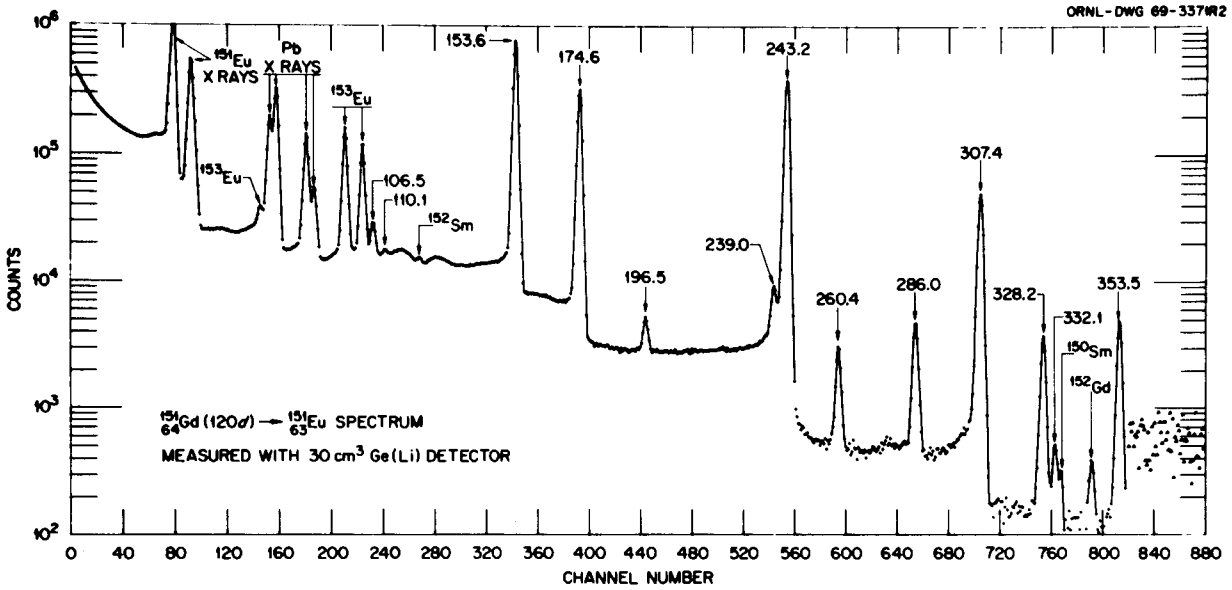

Fig. 1. Gamma-Ray Spectrum of Gadolinium-151 30-cm^3 Ge(Li) Detector.

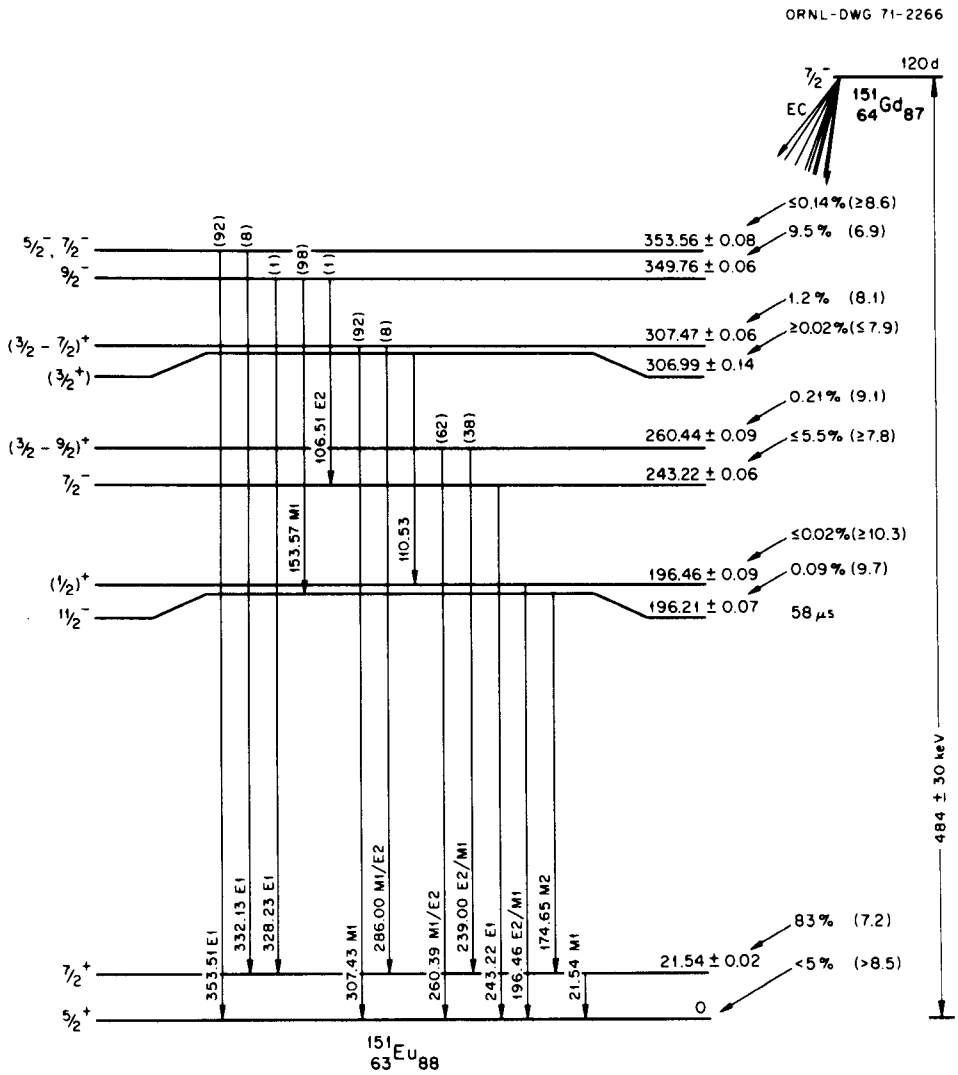

Fig. 2. Decay Scheme of 120-day Gadolinium-151.

2. Terbium-156

The terbium radioisotopes offer a wide spectrum of Mössbauer isotopes:

Nuclide	Half-life	Mössbauer Transition Energy (keV)
Terbium-161	7.0 days	25.7, 74.6
Terbium-160	72.4 days	86.8
Terbium-158	150 years	79.5
Terbium-156	5.1 days	89.0
Terbium-155	5.4 days	60.0, 86.5

We originally worked with 72.4-day terbium-160 to develop our separation technique and determine yields because of its convenient (for these purposes) half-life. The production rate was found to be 21.1 Ci per 100 mg $^{159}Tb_2O_3$ based on the irradiation of 71 mg of the oxide in the removable beryllium reflector region for a full cycle of 23.62 MWd. This work has been reported in a 1967 newsletter[14] and has been published.[15] The original separation work was based on the use of the standard α-hydroxyisobutyric acid/Dowex 50W cation exchanger system. The introduction of the use of ultrafine resin size (in the micron region) combined with high pressure (about 500 psi)[16] provided a much more rapid technique. This modification was introduced in the study of the preparation of 5.1-day terbium-156, which was produced in the ORNL 86-Inch Cyclotron by the $^{156}Gd(p,n)^{156}Tb$ reaction. Figure 3 shows a typical elution curve for this type of system

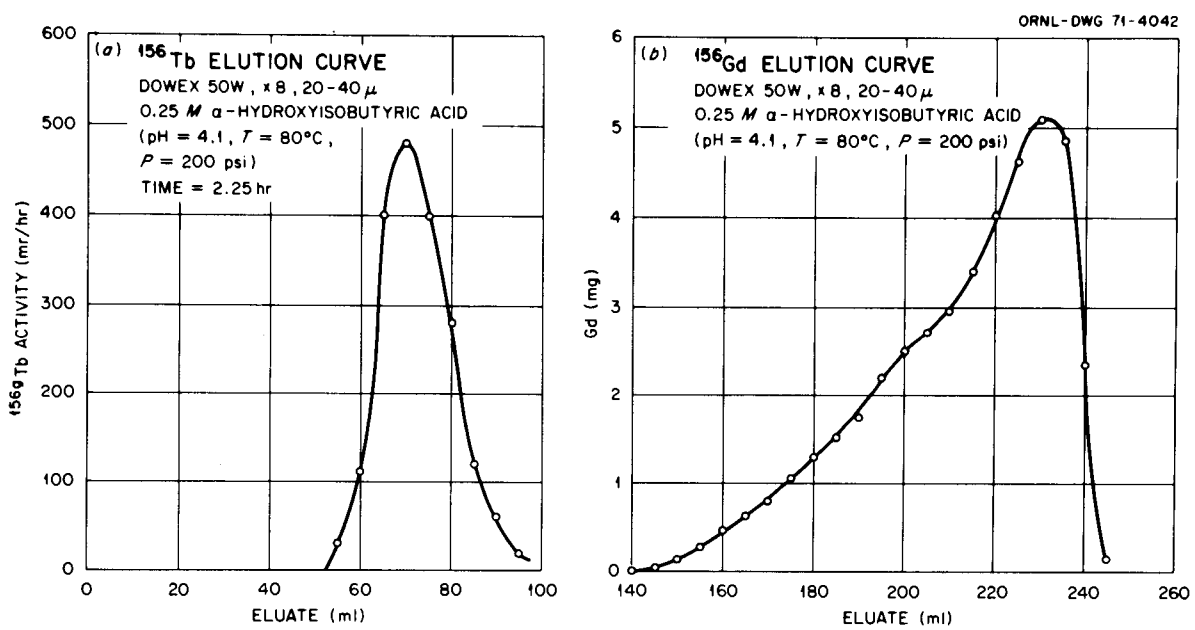

Fig. 3. Elution of Carrier-Free Terbium-156 and 50 mg Gadolinium-156.

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[14]A. R. Rupp, *Isotopes Development Center Newsletter for April 1967*, ORNL-CF-67-5-23, Oak Ridge National Laboratory.
[15]Ludington, *Nucl. Phys.* A119, 398 (1968).
[16]D. O. Campbell and S. R. Buxton, *Ind. Eng. Chem. Process Design Develop.* 9, 89 (1970).

and demonstrates the clear-cut advantages of the high-pressure system. An additional modification is the use of the cation exchanger for the removal of the butyrate complex from the rare earth. The eluate obtained from the isobutyric acid elution is made 0.25 *M* in HCl and sorbed onto Dowex 50W. The butyrate is washed off with 0.5 to 1.5 *M* HCl, and the terbium-156 is stripped off with 6 *M* HCl. Figure 4 shows such an elution for terbium-156 complex. The total time for the process chemistry using these modifications has been reduced from >20 hr to ∿4 hr. The production technique, chemical separation procedures, and yield information have been reported.[17] A yield of ∿900 μCi/hr was realized using the window target facility.

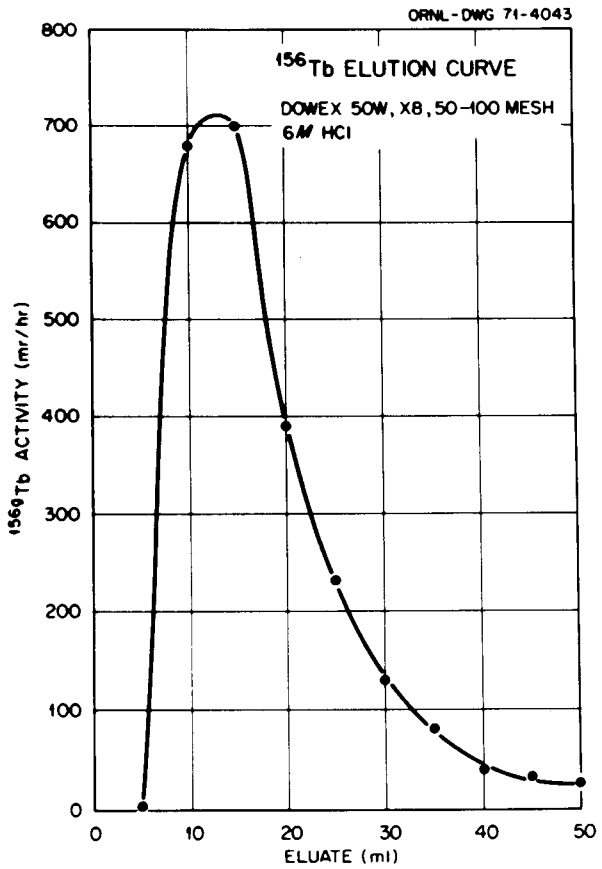

Fig. 4. Elution of Carrier-Free Terbium-156.

These high-purity products of terbium-156 were analyzed for radiation characteristics. Since terbium-156 is a deformed nucleus, it is of particular interest to nuclear structure groups. In cooperation with Vanderbilt University, gamma-ray singles and coincidence measurements were carried out with Ge(Li) detectors. A total of 102 transitions were observed, of which 52 were previously unreported. Of these, 78 were placed in a decay scheme. This work has been submitted for publication.[18] In addition, the conversion electron spectrum was measured with an iron, double-focusing spectrometer, and the K conversion coefficient was obtained for 34 of the transitions.[19]

F. Angular Correlation

It is essential for both production yield information and for dose calculations to know the absolute number of gamma rays emitted per disintegration, as well as the degree of the internal conversion of these gamma rays. Ordinarily, in gamma-ray spectroscopy one obtains only the relative intensities of the gamma rays.

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[17]A. F. Rupp, *Isotopes Development Center Newsletter for April 1968*, ORNL-CF-68-5-16, Oak Ridge National Laboratory.
[18]McMillan, *Phys. Rev.* (in press).
[19]McMillan, *Z, Phys.* 241, 103 (1971).

In many cases, and this is particularly true in the rare-earth region, only the relative intensities of the gamma rays are known and very little, if any, is known of either the multipolarity of the radiation or the spin and parity of the levels. The multipolarity describes the character of the radiation — whether the radiation field is electric (E) or magnetic (M) in nature and how much angular momentum is carried off (1, 2, 3 . . . units) by the gamma ray. Knowledge of the multipolarity allows one to calculate the degree of internal conversion of the gamma rays. If the transition is of mixed polarity, for example, partly M1 and partly E2, then these calculations become even more dependent on the knowledge of the degree of admixture.

By the use of angular correlation measurements (i.e., the measurements of the intensity of gamma rays as a function of angle), we can obtain information which allows us to determine the multipolarities of the gamma rays and spins of the excited levels. The principle of this counting technique is shown in Fig. 5.

Once the multipolarities of the gamma rays and the spins of the levels are known, theoretical conversion coefficient tables[20] give the conversion coefficients, and from this one can calculate the absolute transition rates. This, in effect, is an alternate way of obtaining information on the electron intensities as well.

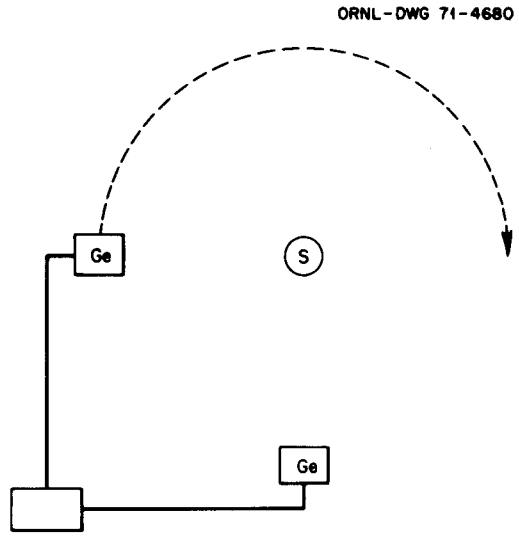
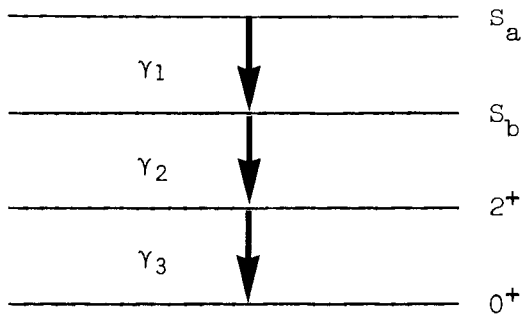


Fig. 5. Simplified Schematic of Ge(Li)-Ge(Li) Coincidence System.

A typical cascade of three gamma rays as shown below can be considered:



[20]See for example R. S. Hager and E. C. Seltzer, *Nucl. Data* A4, 1-235 (1968).

Known are the spins and parity of the ground state (0^+) and the first excited state (2^+), the multipolarity of the $2^+ \rightarrow 0^+$ transition, and, of course, the energies of γ_1, γ_2, and γ_3; unknown are the spins and parity of the second excited state S_b and the third or highest excited state S_a as well as the multipolarities of the $S_a \rightarrow S_b$ transition and the $S_b \rightarrow 2^+$ transition. The stationary detector, usually a Ge(Li) detector, is gated on γ_3. The movable detector system, usually NaI(Tl) for convenience in mobility, sees γ_2 only when γ_3 is seen by the stationary detector. With multiparameter equipment we can also measure γ_1 at the same time; thus, we have a γ_3-γ_2 correlation and γ_3-γ_1 correlation. In actual practice, for example, by counting at three angles you obtain from this angular correlation experiment the experimental coefficients of the parameters A_2 and A_4 for each cascade. From the γ_3-γ_2 cascade data we obtain the spin of the second excited level (S_b) and the multipolarity of γ_2, and from the γ_3-γ_1 cascade we obtain the spin of the third excited level (S_a) and the multipolarity of γ_1.

In practice the simple schematic diagram of Fig. 5 may be a Ge(Li)-Ge(Li) two parameter coincidence system as shown in Fig. 6.

Although convenient tables[21] for consecutive cascades have been available, similar tables for (1-3) cascades have not been available. We have generated a set of 52 graphs for several of the most useful cases of spins and multipolarities for the (1-3) cascades.[22]

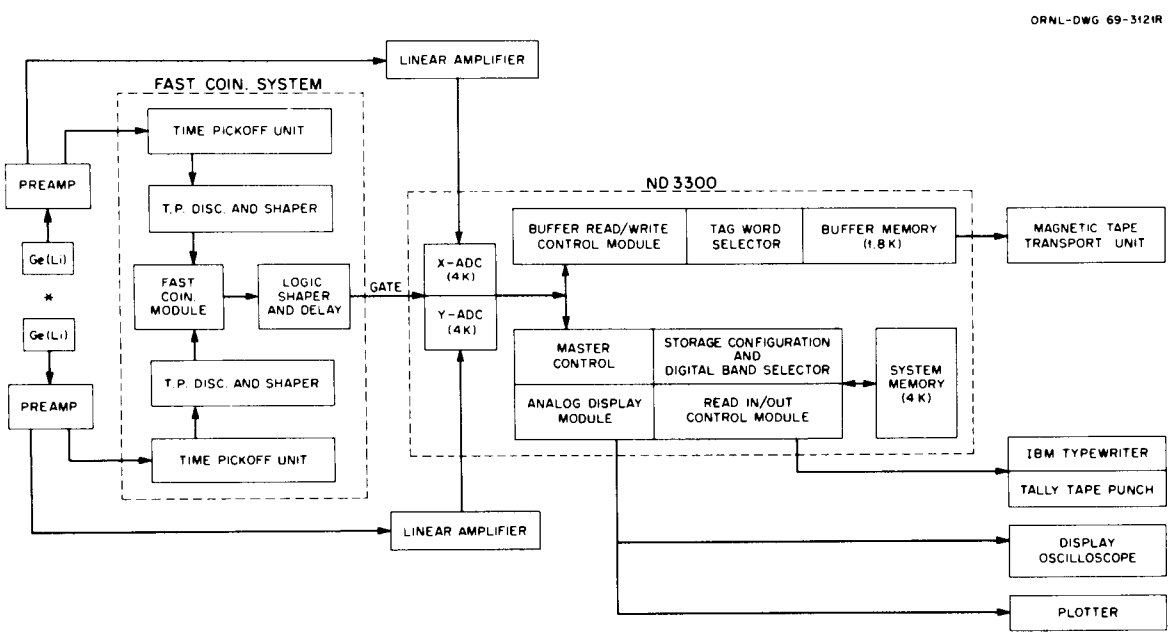


Fig. 6. Ge(Li)-Ge(Li) Two Parameter Coincidence System.

[21]H. W. Taylor, T. McPherson, B. Singh, and F. S. Prato, *Tables of Directional Correlation Coefficients A_2 and A_4 for Gamma-Gamma Cascades*, University of Toronto, 1970.

[22]A. V. Ramayya, J. H. Hamilton, P. E. Little, E. Collins, and J. J. Pinajian, *Graphs for the Analysis of (1-3) Gamma-Gamma Directional Correlations*, ORNL-4619, Oak Ridge National Laboratory (April 1971).

G. Carbon-14

All of the Be_3N_2 targets have been removed from the Hanford reactor, and two carbon-14 production runs have been completed. Radiochemical analysis of the product from the first run indicates a carbon-14 isotopic composition of 96.9%. This product contains 28.3 Ci and is the highest carbon-14 enrichment that has been produced at ORNL and, as far as we know, anywhere else. Analysis on the second production run has not been completed.

ACCELERATOR-PRODUCED ISOTOPES - 08-01-02

A. Biomedical Radioisotopes

1. Gallium-67

The objectives of this program are to determine the optimal target configuration for gallium-67 (78.2 hr) production by the $^{68}Zn(p,2n)^{67}Ga$ reaction 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.

Gallium-67 decays by electron capture with the emission of four main gamma rays of 93, 184, 296, and 388 keV with intensities of 40, 23, 20, and 8%, respectively.

Weekly production of gallium-67 has been continued with five batches being prepared and shipped to customers. A summary of the production experience for the third quarter of FY 1971 is shown in Table 4. All data are for high-purity natural zinc tube targets (0.375-in. OD by 0.020-in. wall thickness) bombarded with 22-MeV protons. The average beam current was intentionally reduced during some irradiations in order to prolong target life and allow maximum yield. By irradiating this tube target design at lower beam currents (e.g., 250 μA), the target life can be extended to greater than 6 hr, and the anticipated yield per target increased to approximately 650 mCi at end of bombardment. The gallium-67 thick-target production rate under the present cyclotron operating conditions seems to be approximately 10-15% lower than previously cited.[23] This parameter will be watched carefully to determine whether there is an unexpected relationship between beam current and production rate which is being affected by machine operating conditions.

Additional target development has been undertaken on a zinc flat plate target design. In order to reduce the metal thickness from the surface

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[23]A. F. Rupp, *Radioisotope Program (8000) Progress Report for September 1970*, ORNL-TM-3183, Oak Ridge National Laboratory, p. 11.

Table 4. ORNL Gallium-67 Production Experience for Third Quarter of FY 1971

Run No.	Av Beam Current (μA)	Innage (hr)	Integrated Beam (μA-hr)	^{67}Ga Cyclotron Yield[a] (mCi)	Integrated ^{67}Ga Production Rate[a] (mCi/mA-hr)	^{66}Ga[b] (%)	Gallium Chemical Yield (%)
68-67-64A	301	2.58[c]	777	347	447	0.4	94.7
68-67-64B	323	2.58[c]	835	383	458		
68-67-65	315	1.67[c]	525	237	451	3.0	101
68-67-66A	322	1.00[c]	322	170	512	0.5	95.0
68-67-66B	308	2.58[c]	743	351	473		
68-67-67A	318	2.00[c]	636	291	458	1.8	86.0
68-67-67B	318	2.17[c]	690	283	410		
68-67-68A	311	2.08[c]	646	282	436	1.8	96.1
68-67-68B	312	2.50[c]	781	318	406		
68-67-69A	308	2.58[c]	795	348	437	0.4	93.4
68-67-69B	306	2.00[c]	612	289	472		
68-67-70A	307	1.17[c]	359	Target not processed			
68-67-70B	309	6.98	2159	878	407	3.1	94.9
68-68-72	302	2.00[c]	604	279	462	4.3	92.7
68-67-73A	297	1.42[c]	421	170	405	2.0	73.4
68-67-73B	314	0.92[c]	288	191	663		
68-67-74	239	2.95[c]	705	319	453	1.0	89.9
68-67-75	245	5.00	1223	491	402	0.7	89.8
68-67-76	259	6.42	1663	758	456	0.7	89.5

[a]Values calculated as of end of bombardment.
[b]Time reference: 8:00 AM on day after shipment.
[c]Irradiation terminated by target failure.

to the cooling water and improve heat flow, a zinc flat plate was fabricated with an additional 0.072 in. of zinc milled from the proton beam target area. This flat plate was irradiated for 5 hr at an average beam current of 304 μA (integrated beam = 1522 μA-hr) and a beam energy of 22 MeV. The beam area was chemically etched with HCl, and the dissolver solution was recovered for analysis. More than 1 Ci of gallium-67 had been produced in this zinc flat plate at end of bombardment at a thick-target production rate greater than 660 mCi/mA-hr. The gallium-67 production rate was not determined in this equipment due to scheduling delays resulting from high target radiation levels and routine gallium-67 processing requirements. Chemical etching of the gallium-67 activity from the flat zinc plate would not appear to be feasible for routine production due to the time and the in-cell equipment required. Additional development work will be done on this point in the future; the current required level of production can be obtained from the tube target design.

Preliminary procedures have been developed and tested for the final product finishing of the gallium-67 citrate preparation to be supplied ORAU as part of the project of Cooperative Group to Study Localization of Radiopharmaceuticals. One hundred millicuries of gallium-67 (reference time = 1200 on the day following shipment from ORNL) will be supplied to ORAU as the citrate and packaged in individual units (10 mCi/unit) for sterilization and

certification. The composition of the finished product will be: gallium-67 = 1.25 mCi/ml at reference time; gallium-66 $\leq$ 0.06 mCi/ml at reference time; sodium citrate = 3.5 mg/ml; sodium chloride = 8.2 mg/ml; and benzyl alcohol = 8.2 mg/ml. Additional product finishing development will be undertaken in conjunction with ORAU during May.

2. Indium-111

The objectives of this program have been to define and optimize the pertinent production parameters for the economical production of indium-111 of satisfactory quality for radiopharmaceutical manufacture. The current objectives are to obtain practical operating experience of the procedures developed by pilot production and to provide this radionuclide to interested customers as a special research material — in batch lots — for their evaluation. Indium-111 has been suggested for such studies as spinal-cerebral cisternography, aerosol lung studies, delayed brain scanning, visualization of the lymphatic system, metabolic studies of indium-labeled macroaggregates and colloids, and tumor localization. Indium-111 has gamma emissions of 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.

One batch of indium-111, approximately 79 mCi total at end of one-day decay allowance, was prepared and portions were supplied to interested clinicians as a special research material. A rough draft of our accumulated production and processing experience has been prepared and will be submitted for publication soon.

3. Rubidium-84

The objectives of this program are to improve the technology for the production of higher purity rubidium-84 by use of an enriched krypton-84 target. Rubidium-84 is usually produced by proton bombardment of natural krypton targets, but this product is unsuitable for positron camera scanning due to appreciable amounts of rubidium-83 produced simultaneously. The use of rubidium-84 for myocardial scanning with a positron camera has increased the demand for a higher purity product.

One rubidium-84 production run using an enriched krypton-84 target was made, and the product was supplied to customers. Additional enriched krypton-84 has been obtained and blended with the older target material.[24] The preliminary production parameters and product composition of the newer target material are compared with those of the older material in Table 5. It can be seen that this new target material offers a $^{84}Rb/^{83}Rb$ activity ratio more than four times larger than a product prepared from naturally abundant krypton and almost twice as large as that obtained with the older

[24]Rupp, ORNL-TM-3183, p. 12.

target. It should be noted that the rubidium-86 is also greatly reduced from that obtained from a natural target. The inventory volume of the new target material is approximately 2-1/2 times that of the volume of the older target and should be sufficient for continued routine production.

Table 5. Rubidium-84 Production Parameters for Various Enriched Krypton-84 Targets

Target	Production Rate (mCi/mA-hr)	Product Composition (% activity)			$^{84}Rb/^{83}Rb$ Activity Ratio
		^{83}Rb	^{84}Rb	^{86}Rb	
Natural	∿78[a]	9.7	68	22	7.0
"Old" ^{84}Kr	121	4.8	87.8	7.4	18.3
"New" ^{84}Kr	∿128	2.9	88.9	8.2	30.7

[a]Estimated.

4. Medically Useful Radioisotopes of Bismuth

The objectives of this program are to evaluate the pertinent production parameters and to provide bismuth-206 for evaluation as a diagnostic radionuclide in medicine. Bismuth-206 has been used to delineate brain tumors in patients with known brain tumors which are poorly visualized by more common radionuclides. Even though very favorable tumor-to-background activity ratio has been obtained by earlier workers, the lack of an appropriate scanning system suitable for the high-energy emissions of this nuclide has retarded the evaluation of the isotope. A cooperative program has been established with Vanderbilt Medical School for the evaluation of bismuth-206 as a diagnostic radionuclide using their high-energy photon scanning system.

The purpose of this program is to prepare radioisotopes of bismuth of high radiochemical purity in high yield, establish a definitive decay scheme, measure the production yield, prepare a suitable radiopharmaceutical product, and test the product.

The examination of bismuth-206 by groups in Germany (Mundinger), Netherlands (van der Werff), and England (Matthews) has demonstrated that this isotope is better for brain tumor localization than other nuclides such as ^{43}K, ^{84}Rb, ^{52}Mn, ^{74}As, ^{110}Ag, ^{68}Ga, ^{65}Zn, ^{95}Zr, ^{131}I, and various compounds labelled with ^{131}I, ^{110}Ag, and ^{51}Cr. Bismuth-206 has a very high tumor/brain concentration ratio and has, as well, a low concentration in muscle and blood. Other bismuth radioisotopes are also potentially very useful because of the availability of half-lives varying from 28 years for bismuth-207 to 1.8 hr for bismuth-201.

a. Bismuth-206

An experimental production procedure has been devised and the preliminary testing begun for the large-scale preparation of bismuth-206 via the $^{207}Pb(p,2n)^{206}Bi$ reaction. Three natural lead targets of varying lead thicknesses have been prepared by electrodepositing lead from a tetrafluoroboric acid solution onto a flattened copper tube substrate. The thickest of these targets (about 0.0035-in. lead thickness) was irradiated in the ORNL 86-Inch Cyclotron for 1 hr with 22-MeV protons at an average beam current of 132 μA. Visual inspection of the target showed no signs of heat dissipation problems, indicating that future irradiations of this target design can be performed at higher beam currents. The lead was removed from the copper tube by electrostripping in 1 *M* HNO_3 solution. The proposed chemical processing involves basically the precipitation of $PbCl_2$ to remove the major amount of lead, followed by cation-exchange chromatography to finally separate the bismuth-206(III) from traces of lead(II) and copper(II). Preliminary experiments indicate that this production approach could be used with an enriched lead-207 target (>90% lead-207) to produce >50 mCi bismuth-206 per hour of cyclotron time with <2% bismuth-205 contamination at end of bombardment (bismuth-206 prepared from natural lead contained approximately 35% bismuth-205). Work will be continued on the development of this production method using enriched lead-207 so that high purity bismuth-206 may be distributed for applications evaluation.

Earlier we reported[25] on the production rate of bismuth-206, the separation chemistry, and the characterization of the bismuth-206 decay in terms of the gamma rays observed, giving gamma-ray energies and intensities. From these data a decay scheme was proposed which incorporates a number of changes. For example, the 107.2-keV gamma ray cited by Lederer, Hollander, and Perlman[26] is no longer assigned to the decay of bismuth-206.

Production rate and dose rate information require, however, the knowledge of the degree of internal conversion of the gamma rays — i.e., the internal conversion coefficient. We mentioned in the December newsletter[25] the anticipated correlation of our work with that of the Japanese. This correlation with the Japanese work is being pursued by J. C. Manthuruthil (Aerospace Research Laboratory, WPAFB). M. Fujioka (Department of Applied Physics, Tokyo Institute of Technology) has supplied us with unpublished internal conversion coefficient data — i.e., conversion electron intensities.

An example is given to show how this correlation is made. The 803.10-keV line is the most prominent line in the decay[27] — being a factor of ∿1.5 more intense than the 881.01-keV gamma ray and ∿2.5 more intense than the

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[25]A. F. Rupp, *Radioisotope Program (8000) Progress Report for December 1970*, ORNL-TM-3282, Oak Ridge National Laboratory, pp. 13-16.
[26]C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes*, 6th ed., Wiley, New York, 1967.
[27]A. F. Rupp, *Radioisotope Program (8000) Progress Report for February 1971*, ORNL-TM-3341, Oak Ridge National Laboratory, pp. 6-10.

516.18- and 537.45-keV gamma rays. It is emitted during the transition from the first excited state (the 803.10-keV level), a $2^+ \rightarrow 0^+$ transition. Such transitions, according to nuclear systematics, are pure E2 transitions. Hence, one knows the theoretical value for the conversion coefficients, i.e., α_K, etc. Recalling that the (electron intensity)/(gamma-ray intensities) = $e/\gamma = \alpha$, we now have a means of correlating the Japanese electron conversion data to our gamma-ray intensity data, since we know what α_K should be and have experimental values for electron and gamma-ray intensities. By this means we are able to obtain many experimental internal conversion coefficients of the mixed transitions which would not be possible from theoretical considerations unless one knew the mixing ratios. This has resulted in the determination of the multipolarities of many of the transitions which in turn resulted in unique spin assignments of some levels and limitation assignments for other levels. These data are now being interpreted by Manthuruthil on the basis of shell model calculations in order to prepare our paper for publication.

b. Bismuth-205

The December newsletter[28] reported work on 6.24-day bismuth-206 which involved the use of a mass separator for the isolation of a high-purity fraction of bismuth-206 activity. In the course of that work, it was pointed out that we were able to obtain high-purity samples of 15.3-day bismuth-205 activity, enabling us to make progress on the examination of bismuth-205.

An additional sample of bismuth-205 was prepared by irradiating radiogenic lead (obtained from the National Lead Company) having an isotopic mass analysis of <0.10% lead-204, 88.59% lead-206, 8.36% lead-207, and 3.05% lead-208. A 0.029-in. lead foil was irradiated for 4 hr with ~18.5-MeV protons using a 26.7-μA beam current. At this incident energy bismuth-205 is formed primarily by the reaction: $^{206}Pb(p,2n)^{205}Bi$ (Q = -11.57 MeV). In addition, varying amounts of 11.8-hr bismuth-203, 11.2-hr bismuth-204, and 6.24-day bismuth-206 plus a negligible amount of 28-year bismuth-207 are formed. By allowing the sample to "cool" ~100 days only the 15.3-day bismuth-205 with faint traces of the 6.24-day bismuth-206 was seen. The lead strip was dissolved in warm nitric acid. The solution was evaporated down to a few milliliters, and the lead precipitated out of chilled fuming nitric acid (~90%). Approximately 10 mg iron(III) carrier was added and the bismuth-205 coprecipitated with $Fe(OH)_3$. After the precipitate was washed, it was dissolved in a minimum amount of hydrochloric acid and reprecipitated. This process was repeated and the bismuth activity then extracted into dithizone solution, and final counting samples were prepared as described earlier.[28]

The decay scheme of bismuth-205 as given by Lederer, Hollander, and Perlman shows 29 gamma rays (out of 38 gamma rays observed) placed in 17 levels of lead-205. Again, as in the case of bismuth-206, assay for yield determination and dose consideration is dependent on an accurate knowledge of the decay scheme and on the knowledge of the degree of internal conversion

[28]Rupp, ORNL-TM-3282.

of the photons. The cooperative program among the Isotopes Development Center (ORNL), the Aerospace Research Laboratory (Wright-Patterson Air Force Base), the Lawrence Radiation Laboratory (Livermore), and the Physics Department of Vanderbilt University was extended to include the examination of our bismuth-205 product. As before, Compton suppression work was done at Livermore (Fig. 7), multiparameter coincidence work at Vanderbilt, high energy gamma-ray spectroscopy at Wright-Patterson, and production, chemical separation, and singles low energy gamma-ray spectroscopy at Oak Ridge National Laboratory.

Figures 8a and 8b show the decay scheme: 116 gamma rays placed (out of 134 gamma rays) in 25 levels of lead-205. Included in the figure are the relative gamma-ray intensities based on the 703.45-keV gamma ray taken as 100,000.

B. Accelerator Pilot Production (Production and Inventory Accounts)

Table 6 gives the April 1971 accelerator irradiations for ORNL and non-ORNL customers.

Table 6. Accelerator Irradiations and Runs for April 1971

Product	No. of Runs	Time (hr:min)			Total Charges
		Beam	Misc.	Total	
		ORNL Programs			
Gallium-67	5	17:40	5:25	23:05	$2,418.41
Cobalt-61	3	6:45	3:40	10:25	1,019.57
Indium-111	1	1:30	0:50	2:20	253.06
Bismuth-206	1	1:00	0:55	1:55	220.75
Total		26:55	10:50	37:45	$3,911.79
		Non-ORNL Programs			
Yttrium-87	3	8:15	3:45	12:00	$1,860.30
Rubidium-84	1	7:05	1:50	8:55	2,196.65
Total		15:20	5:35	20:55	$4,056.95

C. Angular Correlation

See Angular Correlation Section under Reactor Products.

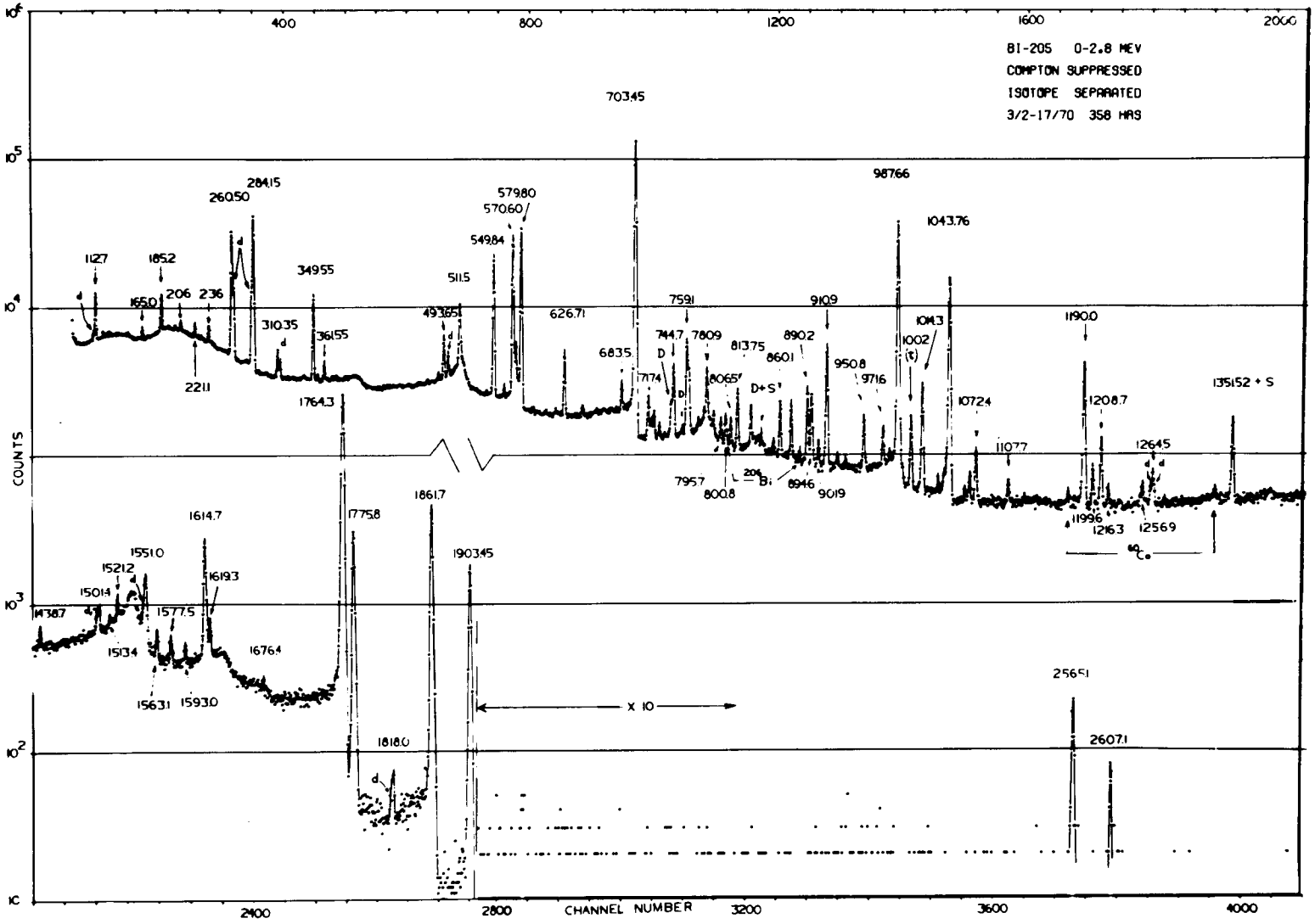

Fig. 7. Compton-Suppressed Spectrum of Bismuth-205.

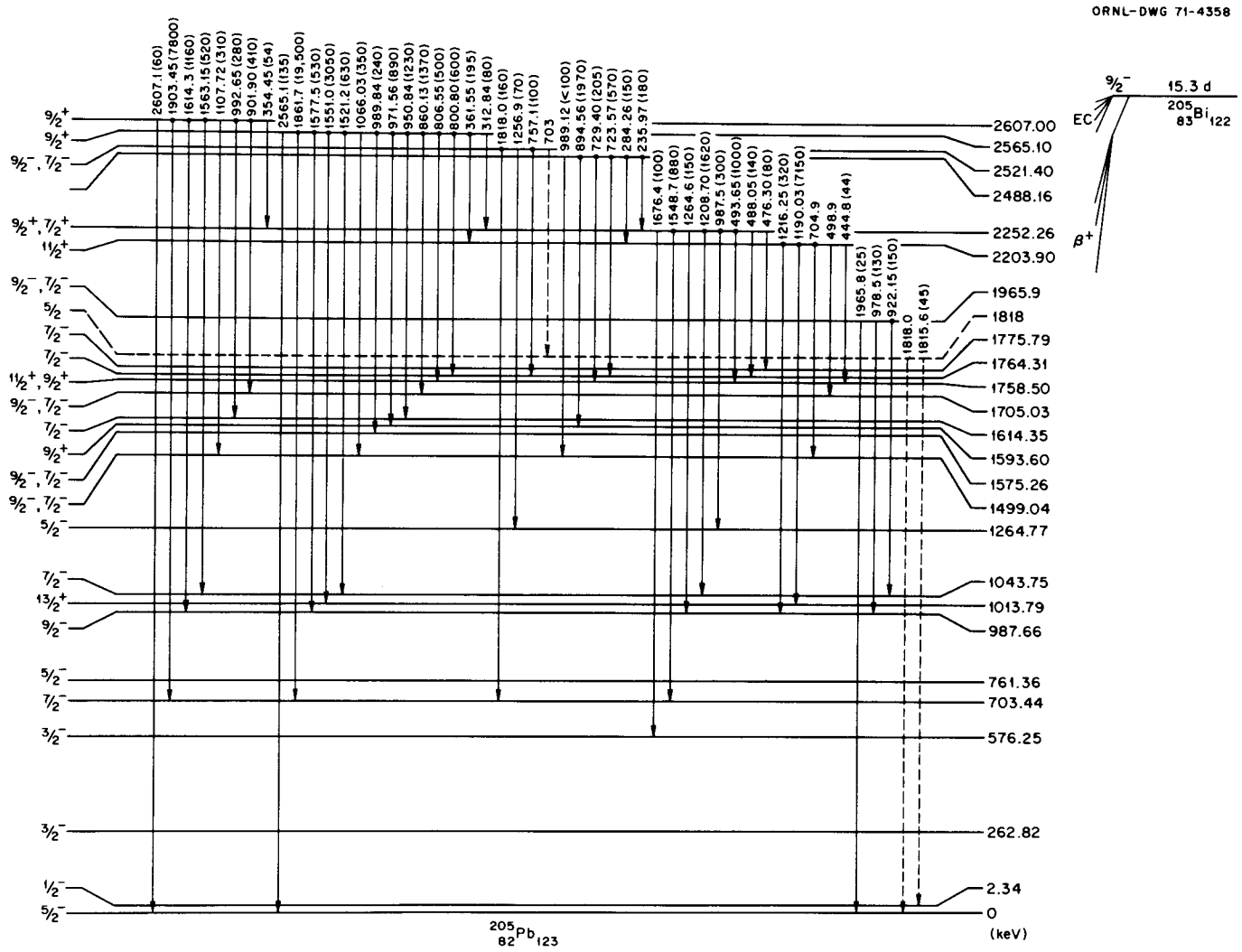

Fig. 8a. Decay Scheme of Bismuth-206.

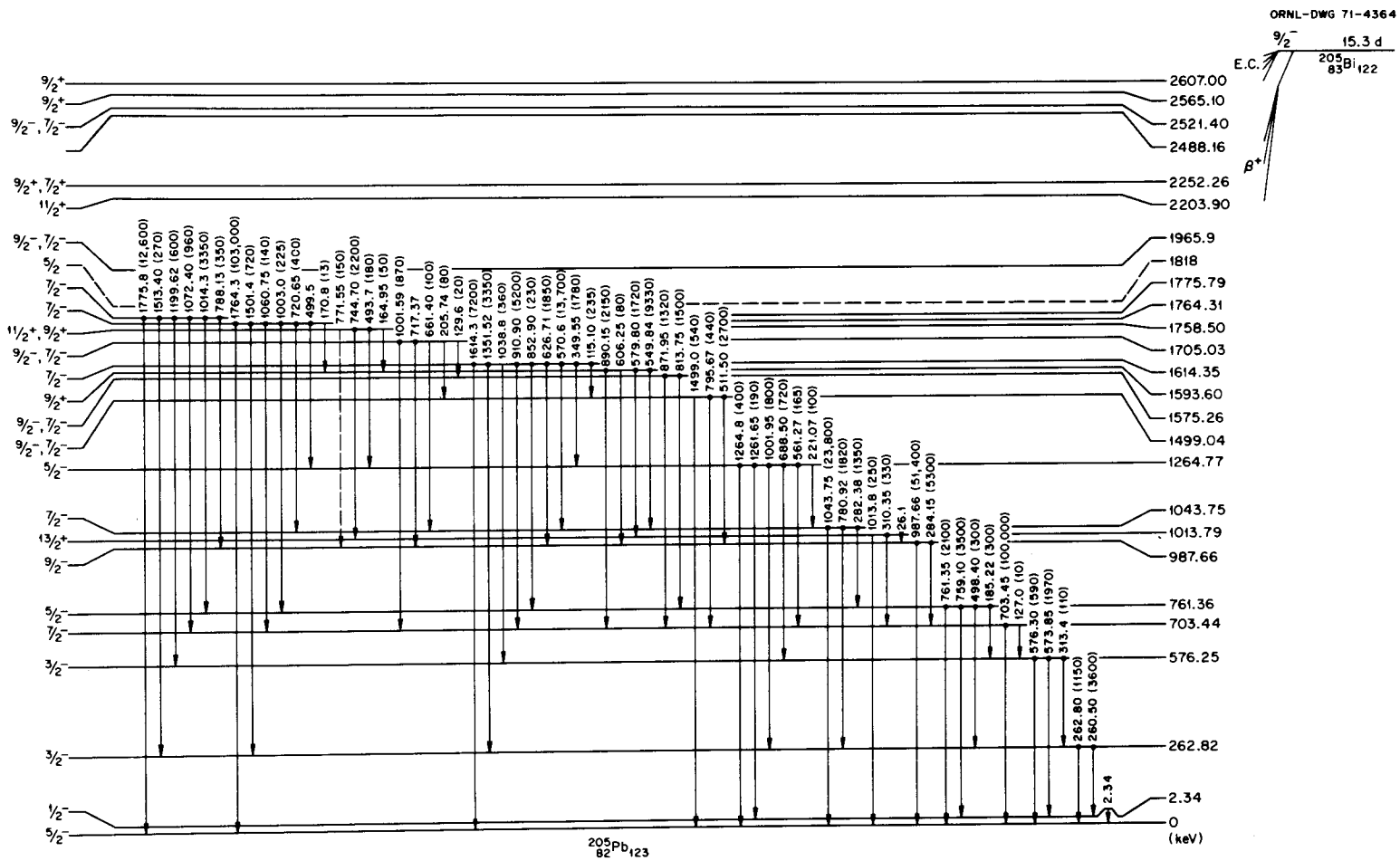

Fig. 8b. Decay Scheme of Bismuth-206.

FISSION PRODUCTS - 08-01-03

A. Krypton-85 Enrichment

Trouble developed in the liquid nitrogen trap system, and some of the transfer valves on units AB and B stuck. The cell containing units A, AB, and B (see Fig. 9) will be shut down for repairs.

The depleted ends were removed from units C and D and were replaced with normal 3.7% feed.

Unit	Activity in Unit (Ci)	Time Since Last Product Removal (days)	Count Rate in Product Section (counts/min) Mar 1971	Apr 1971
C	1784	110	6,650	6,250
CD	2309	360	16,100	17,850
D	1647	110	6,400	6,000

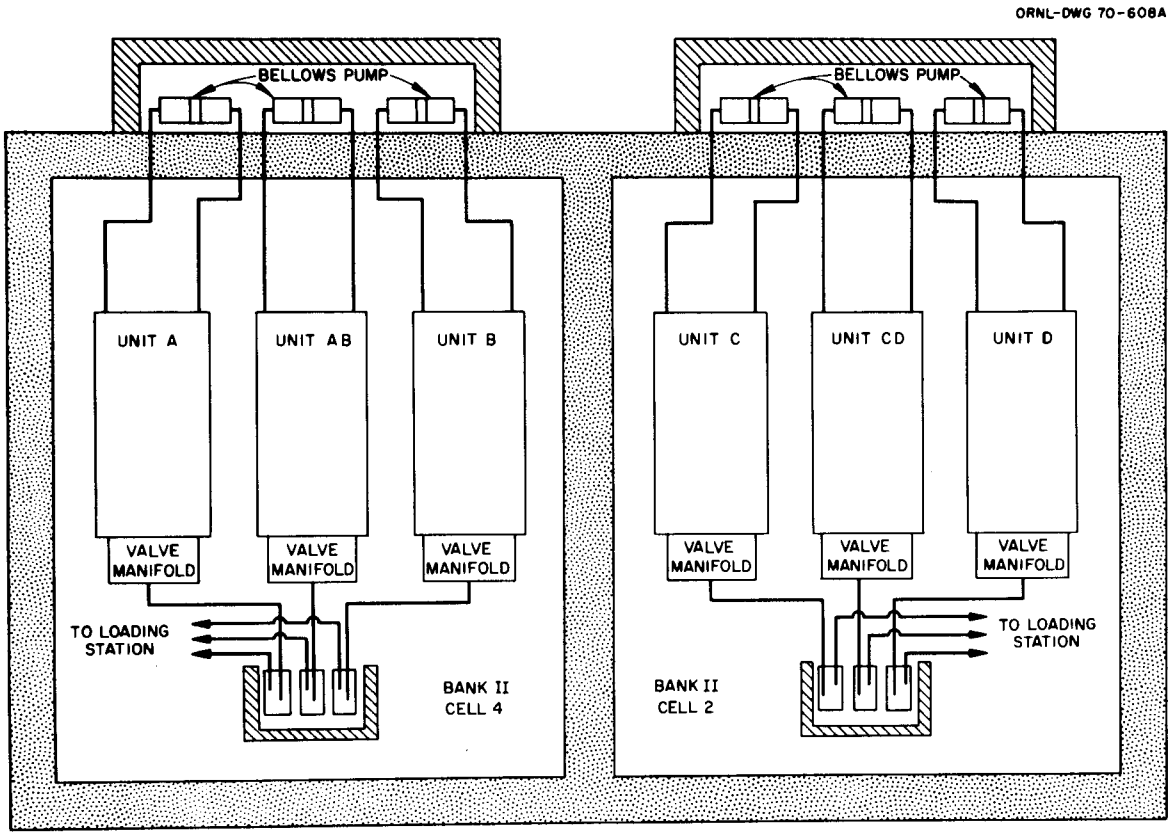

Fig. 9. Schematic Arrangement of Krypton-85 Columns.

B. Cesium-137 Gamma Source Development

1. Cesium Source Form Development

Cesium-137 chloride has been the compound of choice for radiation sources in applications of moderate temperature conditions. In many respects cesium chloride is the ideal compound; the cesium weight per unit volume is high, the radiation resistance of the CsCl is excellent, the compatibility with stainless steel at ambient temperatures is excellent, and its preparation is straightforward. However, the projected conditions of use of cesium-137 gamma sources are increasingly severe with respect to temperature, and some applications indicate the need for a cesium source material which has low solubility. In view of these projected conditions of use, the testing of $^{137}CsCl$ at elevated temperatures is being done and the development of a low solubility, temperature-resistant source form of cesium is being studied.

a. Alternative Cesium-137 Source Form Development

Two alternative methods for preparation of cesium tantalates have been studied. The first one is based on the molten $CsOH$-Ta_2O_5 reaction in a vessel provided with a reflux condenser. The reaction was carried out under reflux to regulate release of water vapor produced in the reaction:

$$2CsOH(melt) + Ta_2O_5(s) \rightarrow 2CsTaO_3(s)\downarrow + H_2O(g)\uparrow \qquad (1)$$

In the preparation of cesium niobates described in previous reports, it was found that the reaction similar to the one above proceeded rather slowly at low temperatures (below ~250°C). On the other hand, in the high temperature range, the process becomes dominated by the decomposition of CsOH with losses of an excessive amount of cesium according to:

$$2CsOH \rightarrow Cs_2O + H_2O \qquad (2)$$

Since reaction (1) is similar to that in the formation of cesium niobates, a regulated release of water vapor would not only favor reaction (1) but also minimize cesium losses due to reaction (2) at high temperatures. A low-speed agitator was also used in some molten $CsOH$-Ta_2O_5 reaction experiments to promote mass transfer in the reaction system.

The basis for the second method is the slurry reaction between Ta_2O_5 and Cs_2CO_3 (instead of CsOH) as in

$$Ta_2O_5(s) + xH_2O(\ell) \rightarrow Ta_2O_5 \cdot xH_2O \rightarrow H_2Ta_2O_6 \cdot (x-1)H_2O(aq) \qquad (3)$$

$$Cs_2CO_3(aq) + H_2Ta_2O_6 \cdot (x-1)H_2O(aq) \rightarrow 2CsTaO_3(s)\downarrow + xH_2O(\ell) + CO_2(g)\uparrow \qquad (4)$$

The reaction was carried out at the boiling point under reflux with mechanical agitation. The products from both methods were heated to ~200°C to drive off moisture at the conclusion of the reactions and then calcined at 750°C.

In aqueous solutions, tantalum compounds are known to exhibit even more complex behavior than niobium compounds. Accordingly, some difficulties have been encountered in the analysis of tantalum by the x-ray absorption edge method that requires the sample to be in aqueous solution. Currently under development is an x-ray fluorescence technique which is capable of the direct analysis for tantalum in solid samples. Summarized in Table 7 are the atomic ratios of Ta/Cs for various products from the $CsOH-Ta_2O_5$ slurry reaction and the molten $CsOH-Ta_2O_5$ reaction as determined by the x-ray absorption edge method. Also appearing in Table 7 are weight losses in drying and calcination steps as well as the solubility in sea water. Similar data for products from the Cs_2CO_3-CsOH reaction are not yet available.

Table 7. Chemical Composition, Solubility, and Weight Losses for Various Cesium Tantalate Products

Sample No.[a]	Drying or Calcining Condition	Weight % of Sea Water Insolubles	% Weight Losses Per Step[b]	Ta/Cs Atomic Ratio of Product
		$CsOH-Ta_2O_5$ Slurry Reaction Product		
A-1	As dried	-	2.2	1.20
B-1	in air	-	2.0	1.24
A-2	750°C, in	54.9	2.2	1.11
B-2	nitrogen	86.3	2.6	1.12
A-3	750°C, in	54.8	4.1	-
B-3	air	87.9	3.8	1.12
		Molten $CsOH-Ta_2O_5$ Reaction Product		
C-1	As dried	69.1	2.4	0.62
D-1	in air	74.8	-	0.60
C-2	750°C, in	74.5	6.7	0.74
D-2	nitrogen	74.6	1.2	0.73
C-3	750°C, in	75.0	7.2	-
D-3	air	75.6	1.4	0.80

[a]The slurry reaction time for sample number B's was twice that for sample number A's. Sample numbers D-1, -2, and -3 were reacted at higher temperatures (up to ∿650°C) than C-1, -2, and -3 (up to ∿350°C).

[b]Weight percent based on the sample weight at the beginning of each step.

Table 7 demonstrates that the changes in both chemical compositions (Ta/Cs ratio) and weight losses of the product from the slurry reaction with the reaction time are almost negligible. The composition changes only slightly upon calcination. However, the amount of insolubles is increased considerably (from 55% to 86-88%) by the extended slurry reaction time. The product weight losses after calcination were appreciably smaller in the nitrogen atmosphere (2.2-2.6%) than in air (3.8-4.1%). In the molten $CsOH-Ta_2O_5$ reaction, the reaction temperature has very little influence on the chemical composition of the product. On the other hand, the weight losses during calcination are reduced remarkably (from 6.7-7.2% to 1.2-1.4%) when the reaction is carried out at high temperatures. The amount of insolubles obtained after calcination is practically unaffected by the reaction temperature. It is to be noted that provision of the reflux condenser in the molten salt reaction resulted in an improvement in regard to the amount of insolubles as well as the weight losses (in comparison with similar molten $CsOH-Nb_2O_5$ reactions without a reflux condenser reported previously).

Some of the samples listed in Table 7 exhibited a considerably higher solubility (as much as approximately 5-6 times) in distilled water than in sea water. This phenomenon can be explained as follows:

1. Distilled water may tend to hydrolyze cesium tantalates producing fine colloidal particles which go through the filter. This is based on the observation that some filtrates appear turbid immediately after filtration, and the turbidity increases upon standing in air. The suspensions eventually precipitate after some time, presumably resulting from the reaction with CO_2 gas in air.

2. Hydrolysis of cesium tantalates mentioned above may also result in formation of soluble tantalates (low Ta/Cs ratio).

3. In sea water, the presence of strong electrolytes, such as NaCl, probably reduced the tendency to form colloidal particles.

In Table 8, the estimated volumetric specific activity (Ci/cm^3) of potential cesium-137 tantalate gamma source compounds is compared with that of other cesium-137 compounds. It is seen that the cesium tantalate with the Ta/Cs ratio of 1 has somewhat higher activity (77.4 Ci/cm^3) than the equivalent cesium niobates (74.4 Ci/cm^3). However, the reverse appears to be true for the tantalate with the Ta/Cs ratio of 2 as compared with the equivalent niobates. Also to be considered in comparing the volumetric specific activity for different source compounds is the density of the compound which would exert some influence on the efficiency of an irradiator.

b. Improvement and Characterization of $^{137}CsCl$ Source Form

Presented in Table 9 are the values of coefficient of linear thermal expansion computed from the data obtained by the Brinkman dilatometer discussed previously.[29] According to Table 9, the expansion coefficient varies

[29]Rupp, ORNL-TM-3341, pp. 15-16.

Table 8. Volumetric Specific Activity of Various Potential Cesium-137 Source Compounds

	Density[a] (g/cm^3)	Total Cesium Content (wt %)	Volumetric Specific Activity (Ci/cm^3)[c]
$CsTaO_3$	6.6	37.1	77.4
$CsTa_2O_{5.5}$	7.1[b]	23.1	52.0
$CsNbO_3$	4.8	48.5	74.4
$CsNb_2O_{5.5}$	5.5[b]	33.0	57.5
CsCl	4.0	78.8	100.4

[a]Assume 0% voids.
[b]Estimated value.
[c]Assume an isotopic purity of 36.5% cesium-137, the balance being mostly cesium-135.

Table 9. Coefficient of Linear Thermal Expansion for Pure CsCl and KCl-CsCl Mixtures

Heating Cycle No.	Temperature Range (°C)	Coefficient of Linear Thermal Expansion[a] [in./(in.·°C) x 10^5]
	Pure CsCl	
1	150-300	5.49
	300-480	9.09
	480-550	239
2	150-400	5.15
	475-550	111
3	150-400	4.32
	485-550	103
	2 mole % (or 0.9 wt %) KCl-CsCl (Molten)	
1	100-250	3.52
	350-400	12.5
	450-500	79.8
2	150-400	3.83
	450-500	54.7
3	100-350	4.15
	450-500	55.0

[a]Thermal expansion characteristics in the temperature ranges not indicated are rather complex in nature.

greatly depending upon whether the temperature range is below or above the solid phase transition point. For example, for pure CsCl, the coefficient above the transition point (464°C) is approximately 22-44 times as high as that below the transition. An addition of a small amount of KCl (e.g., 0.9 wt %) reduces this difference drastically, but there still is a dramatic change (13- to 23-fold) in the expansion coefficient as the material is heated through the transition temperature range. The presence of KCl also tends to reduce the absolute value of the coefficient considerably. In both cases, the behavior of the material (cylindrical pellet) in the first cycle is rather complex, presumably due to the thermal equilibration (annealing) effect.

C. Cesium-137 Pilot Production (Production and Inventory Accounts)

1. Processing and Process Status

No cesium-137 processing was done this month. Some rerouting of stored solutions was done to prepare the cesium-137 process system for a complete line check. The current cesium-137 process status is as follows:

Item	Cesium-137 (Ci)
In-process material	559,000
^{137}CsCl product	18,600
Sources in fabrication	100,000
Completed sources awaiting shipment	46,500

2. Operational Summary

Item	April 1971		FY 1971	
	Number	Amount (Ci)	Number	Amount (Ci)
HAPO shipments received	0	0	1	388,200
Product batches prepared	0	0	0	0
Sources fabricated	0	0	62	43,400
Special form containers loaded	0	0	17	198,800
Sources shipped	0	0	283	163,300
Special form containers shipped	9	101,800	21	199,800

3. Current Orders

Current orders for cesium-137 as sources or bulk powder are shown below:

Customer	Amount (Ci)	Estimated Shipping Date
J. L. Shephard and Associates	1,350	May 1971
Atomic Energy of Canada Limited	1,500	May 1971
CEA, France	98,417	June 1971
American Hoechst Corporation	5	June 1971
Lockheed-Georgia Company[a]	~35,000	June 1971
Brookhaven National Laboratory	203,000	July 1971
Total	339,272	

[a]The cesium-137 activity to be supplied by Lockheed-Georgia Company.

An order is on hand for Atomic Energy of Canada Limited for ∿154,300 Ci of cesium-137 as bulk powder or sources to be scheduled and shipped as released by the customer.

D. Strontium-90 Pilot Production (Production and Inventory Accounts)

1. Processing and Process Status

Seven batches of $^{90}Sr_2TiO_4$ with a total yield of 273,000 Ci were prepared. Nearly all of this was used, along with some older inventory material, in fabrication of pellets for sources. Twenty-four pellets were pressed on the induction-heated hot press. There were five pellet failures, three resulting from pellets sticking in the dies and two that broke during ejection. The hot press was also used to calcine all of the $^{90}Sr_2TiO_4$ processed. Considerable deterioration of the inner graphite sleeve of the hot-press furnace has occurred during its 42 in-cell cycles. The 20 good pellets were used to fuel a total of 12 strontium-90 units, four for Teledyne Isotopes, two for the U. S. Navy, and six for Gulf Energy and Environmental Systems. The loadings are summarized below.

Customer	No. of Units	No. of Pellets	Total Watts	Total Curies Strontium-90
Teledyne	4	12	2,904	427,000
U. S. Navy	2	2	451	66,300
Gulf Energy and Environmental Systems	6	6	318	46,800
			3,673	540,100

Plans for May include preparation of a batch of ^{90}SrO, fabrication of some test ^{90}SrO pellets, and preparation of one batch of $^{90}Sr_2TiO_4$. The current process status for strontium-90 is as follows:

Item	Strontium-90 (Ci)
In-process material	667,000
Strontium-90 products	263,000
Sources in fabrication	749,000
Returned SNAP sources	295,000
Completed sources awaiting shipment	19,500
	1,993,500

2. Operational Summary

Item	April 1971 Number	April 1971 Amount (Ci)	FY 1971 Number	FY 1971 Amount (Ci)
HAPO shipments received	0	0	0	0
Product batches prepared	7	273,000	18	618,000
Sources completed	0	0	0	0
Special form containers loaded	0	0	0	0
Shipments to customers	1	25	30	796,700

3. Current Orders

Current orders for strontium-90 as sources or bulk powder are as follows:

Customer	Amount (Ci)	Estimated Shipping Date
U. S. Navy	208,000	a
U. S. Navy	424,000	a
Gulf Energy and Environmental Systems	~47,000	May 1971
Total	679,000	

[a]Further work and subsequent shipment are pending receipt of further instructions from customer.

An order is on hand for approximately 20,000 Ci as bulk powder to be scheduled and shipped when released by Teledyne Isotopes.

4. Source Fabrication

Four $^{90}Sr_2TiO_4$ source liners were fabricated. These sources contained an average of 52,000 Ci strontium-90 each (342 W). Problems involving the deformation of the liners after fueling are currently under investigation. Shipment of these source liners has been delayed pending the outcome of these investigations. An increase of the diameter of up to 0.060 in. has been measured.

Four Sentinel-25 sources, each containing approximately 106,000 Ci (723 W), are being stored (unwelded) due to welding problems. Test pieces furnished by the customer (Teledyne) would not yield sufficient weld penetration to allow the welding of the loaded sources. At the request of the customer the remaining test pieces were returned, and the customer will do the weld development and furnish a welding procedure that will assure proper penetration.

Three capsules and two liners for the SNAP-21 sources for the Navy have been dimensionally inspected and found suitable. Two additional liners are on hand, but they are 0.010 in. too large in diameter, which allows only 0.010-in. clearance with the capsules. The bottoms have been welded into the three capsules and are being ultrasonically inspected by the Metals and Ceramics Division.

E. Strontium Pellet Vacuum Hot Press

Five inactive $SrTiO_3$ pellets were pressed to obtain data applicable to the in-cell pressing of strontium-90 compounds. Data obtained from these pellets are listed in Table 10.

Some of the factors affecting the final pellets are (1) the effect of system pressure on die body temperature, (2) the effect of redesigning the

Table 10. Data from $SrTiO_3$ Pressed Pellets

Pellet No.	Prepressed Height (in.)	Final Height (in.)	Bottom Punch Height (in.)	Ram Pressure Applied During Cool-Down	Ratio Prepressed Height / Final Height	Ram Travel (in.)	Average Die Body Temperature (°C)	Power Supply Settings	$SrTiO_3$ Weight (g)	$SrTiO_3$ Density (g/cm^3)	System Vacuum [(-) in Hg]
1	3.938	1.772	1.5	Yes	2.2	2.166	1280-1320	35 V; 290 A	716	4.1	26
2	3.0	1.303	3.0	No	2.3	1.697	1330-1390	36 V; 275 A	599	4.7	29+
3	4.5	1.655	2.0	No	2.7	2.845	1380-1420	35 V; 260 A	735	4.5	29+
4	4.38	3.22	2.0	No	1.4	1.16	1350-1390	32 V; 250 A	1331	4.5	29+
5	3.38	1.95	1.5	No.	1.73	1.43	1360-1390	36 V; 260 A	655.2	3.3	29+

die rest to elevate the die body into a higher temperature zone, (3) the effect of maintaining ram pressure during the cool down cycle, and (4) the use of powdered unsintered $SrTiO_3$ compared with crushed, sintered $SrTiO_3$ as starting material.

A discussion of the five pellets follows.

1. The first pellet had a density of 4.1 g/cm^3. The bottom 1/3 in. of the pellet failed to sinter. Three factors which would account for the nonsintering are: (a) the electrode feed-throughs to the heater were located about 0.3 in. higher than the design specified, which raised the heater in relation to the die body and created a cool zone at the bottom of the pellet, (b) the vacuum pump used on the system was unable to reduce the system pressure below 26-1/2 in. mercury vacuum [At this pressure the multilayer molybdenum reflector insulation (which surrounds the heater) is not as efficient as at lower pressures. The maximum die body temperature was 50-140°C below that which was achieved with a lower system pressure.], and (c) a 1-1/2-in.-high bottom punch was used. This in conjunction with the raised heater position placed the bottom portion of the pellet in a lower temperature zone.

   A ram pressure of 4000 psig was maintained on this pellet until the die body temperature dropped to 1000°C. The pellet was cracked on the upper surface, possibly as a result of continuation of the ram pressure.

2. The second pellet had the highest density of the five pellets pressed so far, 4.7 g/cm^3, and was almost completely sintered. In making this pellet, a 3-in.-high bottom punch was used which placed the pellet into the high temperature zone and a vacuum pump capable of reducing the system pressure to greater than 29.9 in. mercury vacuum was used. The multilayer reflector insulation is much more efficient at this lower pressure as is evidenced by the higher die body temperature (i.e., 1360°C compared to 1300°C) in this test.

The ram pressure was removed from the pellet as soon as the heater power was cut off, and the pellet was not cracked on its upper surface. All subsequent pellets were allowed to cool without ram pressure being applied during cool down.

3. The third pellet had a density of 4.5 g/cm^3. The only change made between pressing pellet 2 and this pellet was the use of a 2-in.-high bottom punch instead of a 3-in.-high bottom punch. The smaller bottom punch allowed an additional inch of prepressed powder to be placed in the die body and produced a larger final pellet (1.66 in. high compared to 1.31 in. for pellet 2). The lower density resulted from the bottom portion of the pellet not being sintered. The nonsintering may have resulted from the pellet not being high enough in the hot zone or additional heat loss through the bottom punch or a combination of these conditions. Both of these conditions could have been the result of using the smaller-in-height bottom punch. Additional tests will be conducted to resolve this question.

4. The fourth pellet had a density of 4.47 g/cm^3. This pellet was made from coarsely crushed, old, sintered pellets; while the first three pellets were made using fresh unsintered $SrTiO_3$ in powder form.

   A 2-in.-bottom punch was used and, although the die body temperature was essentially the same as the other runs, the bottom third of the pellet was crumbly with little cohesion.

   The inside surface of the die body was pitted badly enough after this run to require honing in the shops. After the previous three runs, all that was required was to pass 600-grit paper over the inside surface to remove residual graphite. The top and bottom punches were also heavily scored.

5. The fifth pellet was made using a revised die rest, which raised the die body into a higher temperature region. Since the 7-in.-high die body was still in the shops being honed, a 6-in. die body was used. The power supply began to malfunction while this pellet was being pressed, necessitating two shutdowns due to blown fuses. After the first shutdown, the system had to be cooled and pressurized so that the heater could be realigned (a short circuit developed during the cool-down period). The second shut-down involved letting the die body cool down while the fuse was being replaced. A third blown fuse made it impractical to continue the run. Although the die body temperature was in the 1300 to 1390°C range for over an hour with a ram pressure of 4000 psi, very little sintering took place. The density of the pellet was only 3.33 g/cm^3.

1. Surface Temperature Measurement

A contact pyrometer was used to measure surface temperatures at critical places (near O-ring locations) on the hot press. Since this press is made entirely of stainless steel (except the TZM transmitter rod) and has a different cooling system than the previous hot press, it was necessary to determine if any temperatures were outside the operating range of the Viton O-rings. The measured temperatures are listed in Table 11.

Table 11. Surface Temperature Measurements
(Die Body Temperature — 1390°C)

Location	Temperature (°C)
Sight glass flange	80
Bottom bellows flange	<50
Upper and lower housing flanges	95
Side wall surface	145
TZM transmitter rod flange	225

The temperature limitations of Viton for continuous operation are listed in Table 12.

Table 12. Continuous Service Limits of Viton O-Rings

Length of Service (hr)	Recommended Maximum Temperature
>3000	450°F (232°C)
1000	500°F (260°C)
240	550°F (288°C)
48	600°F (216°C)

The only O-ring location that approaches the recommended maximum temperature is the one which seals the TZM transmitter rod. To date there has been no evidence of leakage from any of the O-rings.

2. Proposed Tests

It has been planned to make some SrO pellets, but the $(NH_4)_2CO_3$ used in the preparation of the powder was needed at FPDL. When the material is available a batch of SrO will be prepared.

The power supply that malfunctioned during the pressing of pellet 5 is being replaced. The revised die rest can then be evaluated by pressing additional $SrTiO_3$ pellets ($SrTiO_3$ powder is on hand).

F. Beta Source Development

1. Strontium-89 Sources for Ames Research Center

Eleven strontium-89 beta radiation sources were designed and fabricated. Radiation measurements were made on each source to determine the radiation dose rate and the homogeneity of the radiation from the sources. Of the 11 sources, nine met the desired radiation specifications of homogeneity and radiation dose rate and these were shipped to NASA at Moffet Field, California.

Beta sources were needed to irradiate a volume in which insects were to be grown and studied for radiation effects. The cycle for the development of the insects from egg to larva to insect was short (∿3 days); therefore, the irradiations were designed to be given over 72-hr periods. The irradiations were to be given at three dose rates — 800, 1200, and 1600 rads — in 72 hr. The irradiation study was to begin on April 22, 1971, and a second experiment on May 22, 1971; therefore, the source design and dose measurements considered the decay of the radioactivity.

Strontium-89 was chosen because its half-life (50.5 d) is sufficiently long. Since the beta radiation energy (1.46 MeV) is high enough to penetrate the matrix in which the insects are grown and it is monoenergetic, the design of the source and the calculation of dose rates are simplified. Fission product strontium-89 was used rather than the radioactivity produced by neutron irradiation of enriched strontium-88.

The fission product strontium-89 has a higher specific activity than that produced by neutron irradiation of target material. The radioactive contaminant of fission product strontium-89 is strontium-90 and is usually less than 1%. The high specific activity and the absence of gamma radiation would reduce the amount of bremsstrahlung and gamma radiation in the source. The beta energy is absorbed or degraded less, and a more homogeneous source is obtained.

The source was made with an active area larger than needed. When it was placed in equipment for irradiating the insects, a 1/8-in. strip around the perimeter was shielded. This region normally contains a greater concentration of radioactivity.

Filter paper 1-1/4 by 1-1/4 in. was bonded to 1-1/4 x 1-1/4 x 0.020-in.-thick aluminum pieces. The required quantity of strontium-89 solution (in a volume sufficient to wet the paper) was added dropwise to the filter paper. After the source was dried under a heat lamp, it was placed in a Lucite holder and a 0.006-in. aluminum window was placed over the radioactive area and bonded with epoxy to the Lucite source holder. Twenty microcuries of strontium-89 in the source would produce approximately 11.0 rads/hr at a distance of 1.0 mm from the center of the source.

Radiographs were made of the source before and after encapsulation to qualitatively measure the homogeneity of the source.

2. Technetium-99

Surface dose rate measurements using the extrapolation chamber were made to determine source output. Previous measurements made to establish input proved to be very valuable in that only two sources out of 11 fabricated failed to meet the specifications. The vibrating reed electrometer and electronic recorder combination was calibrated by the Instrumentation and Controls Division, Instrument Calibration Group, to ensure the accuracy of the current measurements. The diameter of each of the extrapolation chamber collector electrodes was measured by the Fabrication Department mechanical inspector with a tool-maker's microscope. A previously measured

2-cm-diam technetium-99 source was carefully remeasured using two 10-mm collector electrodes (nominal diameter) and one 3-mm collector electrode. This standardized source will be used in the future for checking the instrument while it is at ORNL.

The surface dose rate measurements made with the extrapolation chamber and the required values are shown below. Sources 1 and 6 were discarded because they did not fall within the limits of the specifications.

Source No.	Dose Rate (rads/hr) Measured	Required
1	11.2	12.33
2	19.8	18.51
3	25.5	24.69
4	12.5	12.33
5	19.2	18.51
6	28.3	24.69
6A	24.22	24.36
1A	11.62	12.17
7	16.4	17.1
8	23.9	25.7
9	33.8	34.3

Source uniformity measurements were made using the extrapolation chamber equipped with a 3-mm-diam collector electrode. These readings were taken on a grid with 5-mm center-to-center spacings, producing 25 readings for each source. The percentage variation based on the average of the high and low reading for the various sources is given below.

Source No.	Percentage Variation
1A	±8.8
2	±10.3
3	±9.5
4	±9.1
5	±9.4
6A	±9.4
7	±6.8
8	±6.9
9	±10.6

The standard deviation for No. 7 (the source with the least percentage variation) was 3.5% and for No. 9 (the source with the greatest percentage variation) was 2.2%. The extrapolation chamber readings were normalized by taking the arithmetic mean of each source as 100% and converting the readings to percentage values. These values were then used as a basis for calculation of the standard deviation.

G. Promethium-147 Source Fabrication, Powder Shipments, and Current Orders

Current orders for promethium as bulk powder are shown below:

Customer	Amount (Ci)	Estimated Shipping Date
McDonnell Douglas Astronautics	50,000	May 1971[a]
Atomic Energy of Canada Limited	1,000	June 1971
Minnesota Mining & Manufacturing Co.	2,500	June 1971
Radiochemical Centre, England	1,000	June 1971
Total	54,500	

[a]To be shipped from Richland, Washington.

H. Short-Lived Fission Products Pilot Production (Production and Inventory Accounts)

Isotope	Number of Batches	Amount (Ci)
Xenon-133	2	∿800
Iodine-131	1	38

APPLICATIONS AND TECHNOLOGY SUPPORT - 08-01-04

A. Radioisotope Characterization, Quality Control, and Standards

1. Radioisotope Special Analysis and Quality Control

Attention this month centered on radioactivity standards, especially the ACS symposium on the subject and the subsequent meeting of the National Research Council subcommittee. Attendees included representatives of such organizations as ASTM, AIF, EPA, AECL, Euratom, FDA, NBS, and RCC. A local paper, "A Survey of Needs for Radioactivity Standards," was presented by S. A. Reynolds. The NRC subcommittee reviewed the report of the standards panel, of which over 600 copies have now been distributed, and considered the preparation of a calibration manual and establishment of an information office. Representatives from the American Nuclear Society Information Center on Nuclear Standards proposed possible collaboration in information activities related to radioactivity standards. On behalf of the NRC subcommittee and panel, a letter was published in *Science*[30] commenting on activities of NRC, NBS, and ACS.

[30]S. A. Reynolds, *Science* 171, 955 (1971).

B. Radiation Processing (Not Supported by 08 Program; WFO Account)

1. Radiolytic Oxidation of Dye Waste Solution

Pollution of rivers by dye waste effluent from textile mills and dye chemical manufacturers is a serious problem. Past experimental work has shown that radiolytic oxidation of the waste solutions may be a practical method of destroying certain dyes and chemicals. The purpose of this investigation is to study and develop a method of radiolytic oxidation. The method developed must be economically feasible. A cobalt-60 gamma radiation source is being used to irradiate samples under a high pressure oxygen atmosphere. The solutions include both individual dyes and mixtures of dyes. This work is being supported by the American Association of Textile Chemists and Colorists (AATCC) and the Environmental Protection Agency.

a. Neolan Yellow BG Dye Effluent

The radiolytic oxidation of Neolan Yellow BG (an azo metalized dye) dye effluent was investigated. This dye effluent was prepared as if it would be used as a textile dye for wool. It contained 3 g of dye per liter and 67 g of H_2SO_4 per liter of effluent. Samples of the effluent under 1500 psi oxygen were irradiated in the cobalt-60 gamma irradiator at doses of 10^6 R and 5×10^6 R. Table 13 shows the results of the experiments. The color of the dye effluent changed from a dark orange to a light yellow at 5×10^6 R.

Table 13. Transmission and COD of Neolan Yellow BG Dye Effluent Samples

	Transmission (%)			COD
	400 mμ	500 mμ	600 mμ	(mg/liter)
No exposure	1	1	36	920
10^6 R	1	6	83	800
5×10^6 R	36	69	91	570

b. Intracolor Intralite Fast Brilliant Blue 2GLL Dye Solution

The radiolytic oxidation of Intracolor Intralite Fast Brilliant Blue 2GLL dye solution was investigated. This direct dye solution was furnished as a standard dye by arrangements with the American Association of Textile Chemists and Colorists. The dye solution contained 0.44 g dye, 4.44 g tetrasodium pyrophosphate, 0.25 g sodium sulfate, and 0.21 g intravon G (a wetting agent) per gallon of solution. Samples of this solution under 1500 psi oxygen were irradiated at doses of 10^6 R and 5×10^6 R in the

cobalt-60 gamma irradiator. The solution changed from a dark blue to a light blue at 10^6 R exposure and to a clear solution at 5×10^6 R exposure. The results of the irradiation are shown in Table 14.

Table 14. Transmission and COD of Intracolor Intralite Fast Brilliant Blue 2GLL Dye Solution

	Transmission (%)			COD
	500 mμ	580 mμ	650 mμ	(mg/liter)
No exposure	33	1	17	182
10^6 R	84	69	77	102
5×10^6 R	97	98	98	65

c. Intracolor Intrazone Black 2BN Dye Solution

The radiolytic oxidation of Intracolor Intrazone Black 2BN (a disazo dye) dye solution was investigated. This dye was furnished as a standard dye by arrangement with the American Association of Textile Chemists and Colorists. The dye solution contained 0.34 g dye, 21.04 g sodium sulfate, 2.10 g ammonium sulfate, 0.084 g intravon G, and acetic acid to adjust the solution to a pH of 5.5 to 6.0 in a gallon of solution. Samples of this dye solution under 1500 psi oxygen were irradiated at doses of 10^6 R and 5×10^6 R in the cobalt-60 irradiator. The solution changed from a dark purple to clear with the 5×10^6 R exposure. The sample which had received 10^6 R contained a black precipitate after the irradiation. This black precipitate was filtered out of the solution before transmission and COD measurements were made. The results of the irradiation are shown in Table 15. The COD of the filtered solution was lower than either the starting solution or the solution which received 5×10^6 R exposure. The radiolytic oxidation was effective in lowering the COD and color of this dye solution.

Table 15. Transmission and COD of Intracolor Intrazone Black 2BN Dye Solution

	Transmission (%)			COD
	500 mμ	560 mμ	700 mμ	(mg/liter)
No exposure	11	3	47	104
10^6 R	93[a]	89[a]	96[a]	30[a]
5×10^6 R	99	96	100	71

[a]Black precipitate was filtered from the sample before analysis.

2. Radiolytic Oxidation of Dye Waste Solutions with Dynamic System

Development work has been concentrated on the high oxygen pressure radiolytic oxidation of objectionable chemicals in waste effluents. A small dynamic demonstration system has been developed by which it is possible to pump a waste effluent through a holdup reservoir located in a cobalt-60 gamma irradiator. This sytem has been operated at 0-1500 psi of oxygen, which may be needed to enhance the radiolytic oxidation of waste organic chemicals. The pumping speed and the size of the holdup reservoir may be adjusted to give the desired radiation dose to the organic chemicals in the effluent. Since it would be desirable to use a smaller radiation source and faster pumping speeds, the dyes are held in the holdup reservoir by adsorbing them on charcoal located there. The dyes should remain on the charcoal until destroyed by radiation.

The dynamic system consists of a pump, a compressor, a gamma radiation source, a holdup reservoir, and the connecting pressure tubing. A Lapp pump is used to pump the solution through a holdup reservoir, which is a 5-in.-diam spiral coil of 1/8-in.-diam stainless steel pipe containing the charcoal. This holdup reservoir is located in the gamma irradiator. The oxygen is compressed to 1500 psi and put into the feed from the Lapp pump to the irradiator. Hence the feed solution is saturated with oxygen at 1500 psi before it is pumped through the irradiator.

A preliminary experiment was performed to determine the capacity of the charcoal for benzyl cyanine 6B dye. The dye effluent was 0.09 g/liter of the dye in water. The pH of the solution was near 7. About 15 g of charcoal was placed in a column and the dye effluent was passed through at 50 ml/hr. The breakthrough occurred at 1.4 liters. The results, normalized to 150 g of charcoal, are shown in Table 16.

The dynamic system was used to check the effect of radiation alone on the benzyl cyanine 6B dye when adsorbed on charcoal. The dye solution was pumped at 2 liters/hr through the charcoal reservoir which was located in a 10^7 R/hr field. The breakthrough occurred at about 25 liters of dye effluent. It appears that the capacity of the charcoal was raised from 14 to 25 liters when placed in a radiation source.

The dynamic system was used to measure the effect of radiolytic oxidation (at 1500 psi oxygen) on benzyl cyanine 6B dye adsorbed on charcoal. The solution was pumped at 2 liters/hr through a duplicate reservoir filled with 150 g of charcoal. The analytical results of the experiment are shown in Table 16. No breakthrough was found at 108 liters, at which point the experiment was terminated. When the oxygen was cut off, breakthrough occurred but disappeared again when the oxygen at 1500 psi was applied to the system. It may be concluded from this experiment that charcoal can effectively hold up benzyl cyanine 6B dye while it is destroyed by radiation when the oxygen pressure on the system is 1500 psi.

The dynamic system seems to be an effective system in a gamma radiation field to radiolytically oxidize benzyl cyanine 6B dye when the dye is adsorbed by charcoal located in a radiation field. It appears necessary

to have the oxygen in equilibrium with the solution at high pressures (1500 psi). Charcoal appears to adsorb the dye until it is destroyed by radiation.

Table 16. Analytical Data for Radiolytic Oxidation of Benzyl Cyanine 6B Dye

	No Exposure Atmospheric	10^7 R Field Atmospheric	10^7 R Field 1500 psi O_2
Weight of Charcoal (g)	150[a]	150	150
Flow Rate (liters/hr)	0.50[a]	2.00	2.00
Capacity Charcoal (liters)	14[a]	25	b
Original Transmission at 550 mμ (%)	2.5	2.5	2.5
Transmission (%) at 550 mμ after			
13 liters[a]	95		
16 liters[a]	65		
24 liters		91	
27 liters		82	
108 liters			96.5
Original COD (mg/liter)	59	59	59
COD (mg/liter) after			
13 liters[a]	1.1		
16 liters[a]	6.0		
24 liters		16.8	
27 liters		19.0	
58 liters			19.3
108 liters			13.0

[a]Results normalized to 150 g charcoal.
[b]No breakthrough at 108 liters.

C. Radioisotope Safety

1. Tritium Foils

A study to determine tritium losses from foils used in certain types of gas chromatograph detectors under various conditions of use is in progress. Tests are being conducted to determine the effect of various foil cleaning procedures on tritium losses. The tests consist of immersing a foil in a solvent and ultrasonically cleaning it for 30 min. The solvent is then analyzed for tritium content. Solvents being used are 5% alcoholic KOH, heptane, hexane, and toluene. After testing, the foils are rinsed with methanol and allowed to dry. Each foil will be cleaned several times in

a particular solvent and then be subjected to a standard temperature test to determine if the cleaning procedure affects the tritium loss under known test conditions.

TECHNOLOGY UTILIZATION - 08-01-05

A. Information Center

In April, 133 requests for information were filled with dispatch of 225 items or documents — 141 (including 50 flyers) were already prepared at the Center. Four sales letters were translated, and the NASA cooperative translation project was completed. A list of reviews in progress is shown below.

Title	Author(s)	Status (% Complete)
Iodine-125	P. S. Baker and Martha Gerrard	35
Patent Literature on Process Radiation and Irradiator Design, Part 1. U. S. Patents 1950 Through 1968	R. E. Greene, Helen S. Warren, and P. S. Baker	At reproduction
Potato Sprout Inhibition by Radiation, Part 2	F. E. McKinney	50
Selected Abstracts of World Literature on Production and Industrial Uses of Radioisotopes, Part 1 of 1971	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	At reproduction
Technetium-99m: Preparation and Uses	Martha Gerrard and P. S. Baker	65
Radioisotopes in the Textile Industry	F. J. Miller	90
Grain Disinfestation — A Worldwide Review	F. E. McKinney	20
Presowing Irradiation of Seed	Russian book translated and edited by Martha Gerrard	Final typing being proofed
Iodine-131 Production Methods	French report being translated by Martha Gerrard	References being checked
Radioelectrochemistry	Helen P. Raaen	1st draft completed
Nuclear Methods in Study of Environment	Helen P. Raaen	Draft 50% completed
Fruit and Vegetable Bibliography (revision)	F. E. McKinney	Final copy being proofed

B. Isotopes and Radiation Technology

Isotopes and Radiation Technology 8(4) galley proofing was completed, manuscript for 9(1) was completed, and writing and editing for 9(2) were started.

C. Publications

Martha Gerrard and P. S. Baker, *Selected Abstracts of World Literature on Production and Industrial Uses of Radioisotopes*, ORNL-IIC-30, Part 4 (April 1971).

P. S. Baker, A. F. Rupp, and Associates, *Isotopes and Radiation Technology* 8(3) (Summer 1971).

V. I. Katko, Study of the Extraction and Shaping of Ion Beams in Plasma Sources, *Kernenergie* 10, 85-90 (1967), translated by Martha Gerrard and J. J. Pinajian from Russian, ORNL-tr-2462.

RADIOISOTOPE APPLICATIONS DEVELOPMENT

SYSTEMS ENGINEERING APPLICATIONS - 08-03-02

A. Oceanographic System Study

Radionuclides appear to be useful tools to study sand transport phenomena and, although many experiments have been conducted to demonstrate their effectiveness as tracers, little is known concerning whether or not the dynamic systems in which they are used can be tagged well enough to achieve quantitative data leading to an understanding of basic mechanisms. The Radioisotope Sand Tracing (RIST) study has progressed through equipment development to the point where important system tagging parameters can be studied.

A RIST field experiment was conducted at Wrightsville Beach and Masonboro Inlet, North Carolina, from March 1 to March 11, 1971. A total of 40 Ci (at shipping time) of gold-198-199 was used as the tagging material on the sand. Four injections were made as line sources or point sources.

The PDP-8/I computer system was used on board the survey vehicle for the first time, and the system performed with no machine malfunctions. Real-time plotting was almost accomplished by having the machine read the punched paper tape generated by the regular RIST Data Collection System. With this technique, data plotting was behind real-time a minimum of 50 linear feet. Plans to completely interface the computer system into the data collection system are proceeding. This will yield real-time data processing and data plotting and eliminate the problems of attending to and reading the paper tape.

Onboard plotting proved valuable to the operators and some off-line data processing was accomplished in generating interpolated radiation intensity contour maps. Data processing has been continued since the computer system returned to Oak Ridge National Laboratory. The data report on this field test is 50% complete, and data reduction is 75% complete.

All the equipment shipped to Masonboro except the computer system and one detector assembly has been shipped to Point Mugu, California, for the next field experiment, tentatively scheduled for June 1971.

RADIOISOTOPE SALES

A request for quotation was received from Sanders Nuclear Corporation for four strontium-90 pellets containing 50 W and encapsulated into liners.

Shipments made during the month include 101,584 Ci of cesium-137 to CEA, France; 100 Ci of krypton-85 to Japan Radioisotope Association; and 5,000 Ci of tritium to New England Nuclear Corporation. Other shipments include 139.96 Ci of xenon-133, 319 Ci of krypton-85, and 7,210 Ci of tritium.

The radioisotope sales proceeds and shipments for the first nine months of FY 1970 and FY 1971 are given in Table 17.

Table 17. Radioisotope Sales and Shipments

Item	7-1-69 thru 3-31-70	7-1-70 thru 3-31-71
Inventory items	$ 478,828	$447,232
Major products	83,570	96,103
Radioisotope services	238,329	221,249
Cyclotron irradiations	99,957	93,153
Miscellaneous processed materials	59,529	49,823
Packing and shipping	56,600	63,917
Total	$1,016,813	$971,477
Number of radioisotope shipments	1,925	2,403

ADMINISTRATIVE

Visitors to the IDC and travel by IDC personnel are given in Tables 18 and 19.

Table 18. Visitors to IDC

Affiliation	Purpose of Visit
Atomic Power Authority Bombay, India	Production of radioisotopes in power reactors
Vanderbilt University	Obtain isotopes information
CEA, Saclay	Tour IIC and discuss radioisotope production
Gesellschaft fur Kernforschung M.B.H., Karlsruhe, West Germany	Discuss curium-244 source fabrication
Union Miniere, Brussels	Discuss radioisotope production

Table 18. continued

Affiliation	Purpose of Visit
General Electric Company	Discuss strontium
Central Bureau of Nuclear Measurements, Euratom	Discuss radioisotope standards
American Nuclear Society Information Center on Nuclear Standards	Discuss possible collaboration in information activities related to radioactivity standards
Container Corporation	Discuss processing paper effluent
Owens-Illinois Paper Company	Discuss processing paper effluent

Table 19. Travel of IDC Personnel

Site Visited	Purpose
Vanderbilt University Nashville, Tennessee	Discuss proposed holmium-166m experiment and final calculations for bismuth-205 decay scheme
New York	Attend American National Standards Institute on Package Leak Testing
Augusta, Georgia	Attend ANS meeting on Neutron Sources and Applications
Dallas, Texas	Attend SINB Seminar on Innovative Applications of Radiation
Nashville, Tennessee	Conduct experiment at Vanderbilt
Aero-Jet General Sacramento, California	Discuss strontium source contract

PUBLICATIONS

Journals

V. Lakshminarayana, D. R. Dunn, S. M. Brahmavar, J. H. Hamilton, and J. J. Pinajian, ^{148}Eu Decay, *Bull. Am. Phys. Soc.* 16, 538 (1971).

D. C. Camp, V. Ananthakrishnan, A. Ramayya, J. Hamilton, J. Manthuruthil, and J. Pinajian, The Decay Scheme of ^{205}Bi, *Bull. Am. Phys. Soc.* 16, 492 (1971).

Reports

A. V. Ramayya, J. H. Hamilton, P. E. Little, E. Collins, and J. J. Pinajian, *Graphs for the Analysis of (1-3) Gamma-Gamma Directional Correlations*, ORNL-4619, Oak Ridge National Laboratory (April 1971).

S. A. Reynolds, *A Survey of Needs for Radioactivity Standards*, ORNL-TM-3239, Oak Ridge National Laboratory (September 1970).

A. F. Rupp, *Radioisotope Program (8000) Progress Report for March 1971*, ORNL-TM-3380, Oak Ridge National Laboratory.

Eugene Lamb, *Isotopic Power Fuels Monthly Status Report for March 1971*, ORNL-CF-71-4-29, Oak Ridge National Laboratory.

Translation

K. E. Scheer and W. Maier-Borst, On the Preparation of ^{99m}Tc for Medicinal Purposes, *Nucl. Med.* 3, 214-17 (1963), translated from German by J. J. Pinajian, ORNL-OLS-17/71.

Papers Presented

D. C. Camp, V. Ananthakrishnan, A. V. Ramayya, J. H. Hamilton, J. Manthuruthil, and J. J. Pinajian, The Decay of ^{205}Bi, presented at *1971 Spring Meeting of the American Physical Society, Washington, D. C., April 26-29, 1971.*

V. Lakshminarayana, D. R. Dunn, S. M. Brahmavar, J. H. Hamilton, and J. J. Pinajian, ^{148}Eu Decay, presented at *1971 Spring Meeting of the American Physical Society, Washington, D. C. April 26-29, 1971.*

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