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REVIEW OF RADIOISOTOPES PROGRAM, 1967

J. H. Gillette

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

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ISOTOPES DIVISION

REVIEW OF RADIOISOTOPES PROGRAM, 1967

J. H. Gillette

Compiled by

Ingrid B. Lane

Contributions from

P. S. Baker
T. A. Butler
F. N. Case
R. A. Robinson
R. W. Schaich
H. F. Stringfield

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OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

SUMMARY

During the past year, new and improved radioisotope preparations were provided to medical research groups through cooperative programs for medical evaluation. The radioisotope preparations included ^{69m}Zn, ^{43}K, ^{92m}Nb, ^{33}P, ^{67}Cu, ^{95m}Tc, and ^{123}I. The feasibility and economics of producing ^{171}Tm as an isotopic power heat source for use in an artificial heart were investigated. The high neutron flux capabilities of the HFIR (used to study neutron production of radioisotopes) were determined in the beryllium reflector and the hydraulic tube positions.

The thermal neutron cross section and resonance integral of ^{140}Ba were determined to be 1.57 ± 0.3 and 13.6 ± 1.4 barns, respectively, and the effective cross section of ^{204}Tl was determined to be 23.1 barns.

Thermal diffusion enrichment of the ^{85}Kr in the fission-product krypton provided 240 Ci of 13.97% ^{85}Kr and 370 Ci of 27.62% ^{85}Kr.

Field tests with ^{133}Xe-tagged sand were carried out at three locations to evaluate equipment and to develop more effective procedures for use in studying the direction, the velocity, and the volume of sand movement.

Beta, low energy gamma, secondary X-ray, and neutron sources were developed, fabricated, and characterized for varied applications, including the following. The feasibility of producing ^{153}Gd in sufficient quantity and purity to be used for the Mars Atmosphere Density Sensor source was determined, and alternative radioisotope sources were investigated. An experimental blood irradiator was designed and fabricated using ^{90}Sr. An ^{55}Fe source was prepared for use in an X-ray diffractometer. Two types of radiographic cameras were designed and fabricated: one for use with ^{147}Pm and ^{169}Yb and the other for use with ^{241}Am and ^{57}Co. A process was developed for producing high intensity beta sources consisting of stainless steel capsules containing strontium silicate spheres embedded in an aluminum matrix.

The reliability of radioactive source capsules used to contain radioactive materials and shipping containers used to transport radioactive materials was determined under conditions of normal use as well as in the event of an accident. An applications rating guide was developed to determine the required source classification for the intended application of a source and the expected environmental conditions experienced during its operating life. Tests were begun to determine if certain selected ORNL source capsules meet the USAEC requirements for "special form material." Test capsules made of 304L and 316L stainless steel, Inconel, and Hastelloy C were exposed to cesium chloride at 1300°C to determine their compatibility with cesium chloride under conditions which may exist in the event of a fire. A heat transfer study was performed on a Sugarman shipping cask in combination with the wood fire and impact shield in which it will be shipped.

The activities of the Isotopes Information Center included writing state-of-the-art reviews and production and applications brochures; publishing the quarterly Technical Progress Review, Isotopes and Radiation Technology; establishing a continuing and up-to-date collection of indexed material on the production and applications of isotopes; providing technical and editorial assistance to DID; and answering inquiries concerning the production and applications of isotopes.

The Fission Products Development Laboratory received 1,040,000 Ci of ^{137}Cs feed. Approximately 315,000 Ci of ^{137}CsCl and ~230,000 Ci of ^{90}Sr were pressed into pellets. Radioisotope sales income showed a decrease in 1967 as compared with sales in 1966. Numerous articles in journals were published as well as reports, and many papers were presented at scientific and technical meetings.

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ISOTOPES PRODUCTION AND SALES

REVIEW OF RADIOISOTOPES PROGRAM, 1967

J. H. Gillette

PREPARATION AND APPLICATION OF RADIOISOTOPE RESEARCH MATERIALS

Techniques and procedures were developed for preparing and characterizing radioisotopes (that were previously unavailable) for use as research materials in the medical, biological, and physical sciences. Neutron-produced radioisotopes were obtained by use of the Oak Ridge Research Reactor (ORR) and the High Flux Isotopes Reactor (HFIR), and neutron-deficient radioisotopes were produced by proton irradiation in the ORNL 86-Inch Cyclotron.

Through neutron-irradiation studies the following objectives are accomplished: (1) examination of the principal modes of neutron interaction [(n,γ), (n,p), and (n,α)] with natural abundance or enriched isotope targets, (2) development of appropriate chemical or physical processes to recover and purify the desired radioisotope, (3) characterization of the product purity and determination of unknown or uncertain nuclear properties such as half-lives and decay schemes, and (4) establishment of cooperative programs with research groups to promote the beneficial application of radioisotopes.

Cyclotron-produced radioisotopes which decay by positron (β^+) emission and/or electron capture are associated with X-ray emission and annihilation radiation (0.511 Mev), that are easily detected but have minimal radiation effects. These products are principally used in medical diagnosis, Mössbauer applications, and metallurgical studies, as well as several research applications. In addition, these radioisotopes are of special interest to internal conversion spectroscopists and researchers for defining details of nuclear structure and behavior. Information derived from the production techniques and characterizations of these cyclotron-produced radioisotopes is of use to medical centers and other institutions in developing the utility of local cyclotrons.

HFIR Irradiation Facilities

The high neutron flux capabilities of the HFIR were used to study the neutron production of radioisotopes. The HFIR is a light-water-cooled and water-moderated research reactor, fueled with ~9.4 kg of highly enriched ^{235}U, which operates at a power level of 100 Mw_{th}.[1,2] The reactor core (Fig. 1) consists of a series of concentric fuel regions, each ~2 ft high. A 5-in.-dia hole in the center of the core serves as a flux trap. A hydraulic tube facility is located in the center of this region for experimental irradiations of short duration at high neutron flux. From the center line of this facility, the distance to the nearest fuel region is ~2.75 in. The fuel region is surrounded by a concentric ring of beryllium reflector ~1 ft thick. In the reflector region are 38 vertical experimental facilities. The ones closest to the fuel region are located in the removable portion of the beryllium reflector and are ~2 in. from the nearest fuel.

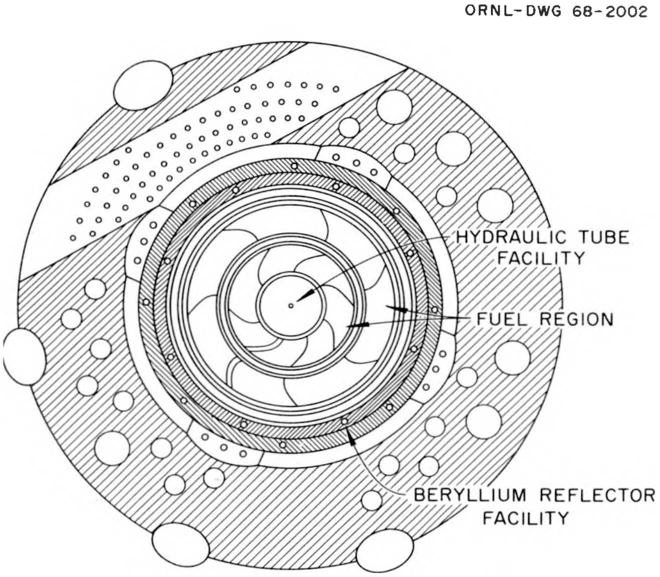


Fig. 1. HFIR Core and Selected Irradiation Facilities.

At the higher fluxes available in the HFIR, nuclear reactions such as (n,p) and (n,α), which are impractical at lower fluxes, are now proving to be feasible for radioisotope preparation. Target holders were designed, tested, and approved for utilization in the removable beryllium facility adjacent to the cylindrical fuel pieces and in the hydraulic tube access facility along the central axis of the flux-trap region.

The thermal and the fission neutron fluxes were measured in the beryllium reflector and the hydraulic tube of the HFIR (Table 1). In the beryllium reflector position, the two stainless steel monitors (Nos. 0 and 1) were located ~0.5 in. above and below the reactor center line, while No. 2 was 2.5 in. above the center line. In the hydraulic tube, the top six positions were monitored. The thermal neutron flux was determined by measuring the amount of ^{60}Co produced by the ^{59}Co(n,γ)^{60}Co reaction. The thermal flux is based on the 2200-m/sec cross section of 37 barns for the ^{59}Co(n,γ)^{60}Co reaction and is not corrected for the resonance neutron contribution. The fission neutron flux was determined by measuring the amount of ^{54}Mn produced by the ^{54}Fe(n,p)^{54}Mn threshold reaction. A fission neutron cross section of 0.061 barn was used for this reaction, and a virgin fission neutron spectrum was assumed to be present in the hydraulic tube.

Table 1. Thermal and Fission Neutron Fluxes in the HFIR

Hydraulic tube position	Monitor No.	Flux, neutrons/cm^2.sec	
		^{59}Co(n,γ)^{60}Co	^{54}Fe(n,p)^{54}Mn
	Beryllium Reflector		
	0	1.30×10^{15}	2.64×10^{14}
	1	1.28×10^{15}	2.89×10^{14}
	2	1.09×10^{15}	2.17×10^{14}
	Hydraulic Tube		
1	1	–	2.92×10^{14}
	2	–	3.50×10^{14}
2	1	–	5.37×10^{14}
	2	–	5.95×10^{14}
3	1	–	7.10×10^{14}
	2	–	7.76×10^{14}
4	6-67-2	2.46×10^{15}	8.27×10^{14}
5	1	2.79×10^{15}	9.07×10^{14}
	2	2.71×10^{15}	8.83×10^{14}
	3	2.66×10^{15}	8.27×10^{14}
	4	2.72×10^{15}	8.82×10^{14}
	5	2.79×10^{15}	8.76×10^{14}
	6	2.72×10^{15}	8.39×10^{14}
6	6-67-3	2.42×10^{15}	8.84×10^{14}

Preparation of Radioisotopes for Medical Applications

Several cooperative programs were established with medical research institutions throughout the country. Since radioisotopes are playing an increasingly important part in the diagnosis and treatment of disease, new radioisotope products were prepared for medical evaluation. Often these new preparations are of a low general demand, difficult to prepare, or not economically attractive to commercial suppliers. By supplying these previously unavailable radionuclides to medical research groups through these cooperative programs, the production and separation variables and the utility of a specific nuclide can be studied concurrently.

Thulium-171

The feasibility and economics of producing ^{171}Tm as an isotopic heat source for use in an artificial heart were investigated. The minimal intrinsic radiations of ^{171}Tm (1.9-y beta emitter) make it a highly desirable candidate heat source for biological applications if it can be produced sufficiently free of ^{170}Tm contamination.

To be competitive with alternative heat-source candidates (principally ^{238}Pu and ^{147}Pm) for the artificial heart, the ^{170}Tm/^{171}Tm ratio must be reduced to the order of 10^{-3} to 10^{-5}. Both technical and economic factors of three methods for producing ^{171}Tm by neutron irradiation of (1) natural thulium, (2) enriched ^{170}Er, and (3) natural erbium and the product refinement necessary to reduce the quantity of ^{170}Tm were investigated in detail. Methods of product refinement include natural decay, electromagnetic separation, and high neutron flux burnout of ^{170}Tm.

It was concluded that ^{171}Tm with the purity required for the artificial heart program would cost ~\$10,000 per thermal watt; therefore, it is not competitive with other candidates.

Zinc-69m

Zinc-69m (half-life, 14 h) has been used by medical investigators at Walter Reed General Hospital to perform *in vivo* and *in vitro* scintillation scans of human prostate glands. Studies to determine if there is a difference in uptake between normal and abnormal tissue are in progress. Apparently, no tissue stores zinc preferentially, and the value of ^{69m}Zn as a prostate scanning agent will depend upon its incorporation into a complex ion or molecule which will allow accumulation of zinc in the gland.

Production of ^{69m}Zn in the ORR was studied by two nuclear reactions using isotopically enriched targets. The fission neutron cross section for the ^{69}Ga(n,p)^{69m}Zn reaction was determined to be 0.50 ± 0.14 mb. The zinc product was separated from the gallium target by solvent extraction. Production of ^{69m}Zn by the ^{68}Zn(n,γ)^{69m}Zn reaction produced an average yield of 2.22 mCi of ^{69m}Zn per mg of zinc target.

Potassium-43

A number of medical investigators have expressed interest in the availability of 22-h ^{43}K for use in studying potassium metabolism, localizing malignant breast tumors, and tagging red blood cells. The use of commercially available ^{42}K is limited by its short half-life for these types of investigation.

Two targets of 63.6% enriched ^{43}Ca as $CaCO_3$ were irradiated in the ORR to produce ^{43}K by the ^{43}Ca(n,p)^{43}K reaction. The potassium was separated from the target by ion exchange. An average yield of ~39 μCi/mg of calcium was obtained from which the average production cross section was computed to be 1.5 mb.

Niobium-92m

The reactor production and chemical separation of 10.2-d ^{92m}Nb from natural and enriched molybdenum targets are being studied. Niobium may be of use as a tumor detecting agent in nuclear medicine.

Irradiation of two targets of natural abundance molybdenum (3.26 mg of ^{92}Mo) and two targets of 98.27% isotopically enriched molybdenum (22.4 mg of ^{92}Mo) in the hydraulic tube facilities of the HFIR and ORR, respectively, produced an average of 0.497 and 1.68 mCi of ^{92m}Nb, respectively. The fission neutron cross section for the ^{92}Mo(n,p)^{92m}Nb reaction averaged 4.92 mb.

Procedures for separating trace amounts of niobium from molybdenum targets without the use of HF, which is undesirable for medical products, have been investigated. Niobium was eluted from a column of alumina (previously treated with 0.1 N HNO_3) with 0.1 N HCl while molybdenum remained on the column. Niobium was also separated from alkaline solutions containing molybdenum by adsorption on $Fe(OH)_3$. At pH values of <7.5, molybdenum is not carried by the $Fe(OH)_3$ precipitate.

Phosphorus-33

The availability of millicurie quantities of ^{33}P offers many opportunities for research, especially in the medical and biochemical sciences. In comparison with ^{32}P, ^{33}P has a longer half-life (25 vs 14.3 d), a lower maximum beta energy (~0.25 vs ~1.7 Mev), and, consequently, less bremsstrahlung production. Specific uses for ^{33}P include autoradiography, study of relative effects of recoil energy of sulfur produced by decay of ^{32}P and ^{33}P, long-term ecological and agricultural studies, synthesis of tagged complex organophosphorus compounds, and double labeling of phosphorus in complex molecules such as nucleic acids.

Millicurie quantities of ^{33}P, relatively free of ^{32}P, were produced in high fast neutron flux positions of the ORR and HFIR by two nuclear reactions: ^{33}S(n,p)^{33}P and ^{36}Cl(n,α)^{33}P. Isotopically enriched ^{33}S and ^{36}Cl were used as reactor targets. Chemical procedures for separation of carrier-free phosphorus activities from elemental sulfur and potassium chloride were developed.

Copper-67

As a radioisotopic tracer of copper, 12.9-h ^{64}Cu is generally used because it can be easily produced from stable ^{63}Cu by the (n,γ) reaction. However, in many applications its usefulness is severely limited by its short half-life. Copper-67 (61 h), on the other hand, has a half-life about five times longer than that of ^{64}Cu and is the longest-lived radioisotope of copper. In a cooperative research effort with Mayo Clinic, ^{67}Cu is being used to study the metabolism of copper in patients with Wilson's disease and in their relatives to detect a possible inherited tendency for this disease. Tentative results show that extrapolations of the metabolic behavior of copper based on short-lived ^{64}Cu are incorrect and must be obtained using ^{67}Cu.[3]

Copper-67 can be produced in the ORR and the HFIR in millicurie quantities after a 2-week irradiation of enriched ^{67}Zn targets. After separation of the copper on an anion-exchange column, the enriched zinc is recovered for reirradiation.

Technetium-95m

While performing experiments to devise a method for dissolving irradiated molybdenum metal targets (used in preparation of ^{95m}Tc), it was discovered that ordinary, unstabilized 3 to 30% H_2O_2 will dissolve molybdenum metal. The resulting yellow solution is stable at room temperature in concentrations below 17.8 mg of molybdenum per ml. At higher concentrations a yellow precipitate appears which is readily soluble in dilute ammonium or sodium hydroxide. The reactions involved are probably complex; however, an overall reaction may be represented by

$$Mo + 3H_2O_2 \rightarrow H_2MoO_4 + 2H_2O \ .$$

The rate of dissolution of molybdenum foil and powder was determined for various aqueous dilutions of H_2O_2 and for concentrated and diluted HCl and HNO_3 (Table 2).

Table 2. Dissolution of Molybdenum at 27°C

Solvent	Dissolution rate, mg/hr	
	Mo foil[a]	Mo powder[b]
30% H_2O_2	300	Violent reaction
15% H_2O_2	150	Violent reaction
7.5% H_2O_2	100	Vigorous reaction
3% H_2O_2	Negligible	Rapid
12 $\underline{M}$ HCl	Negligible	Negligible
3 $\underline{M}$ HCl	Negligible	Negligible
15 $\underline{M}$ HNO_3	Negligible	Negligible
3 $\underline{M}$ HNO_3	Negligible[c]	Negligible[c]

[a] 1 x 1 x 0.01 in.

[b] 100 mesh.

[c] HNO_3 slowly converted the molybdenum to an insoluble salt.

A 30% H_2O_2 solution is a convenient reagent for dissolving irradiated molybdenum metal foil or wire targets, and a 3% H_2O_2 solution is convenient for powdered molybdenum samples. The method has the advantage of providing a target solution which is free of extraneous anions (i.e., chlorides, nitrates) and is often more suitable for subsequent chemical processing.

Technetium-95m (60 d, 0.200-Mev gamma) is of interest to researchers studying the metabolism of technetium and technetium compounds in the body. Currently, 6-h ^{99m}Tc is the only commercially available radioactive technetium; it is rather short-lived for biological and compound tagging studies.

The production of ^{95m}Tc in the ORNL 86-Inch Cyclotron from natural molybdenum targets by two simultaneous reactions, ^{95}Mo(p,n)^{95m}Tc and

$^{96}Mo(p,2n)^{95m}Tc$, was investigated. Two 1/4- by 6-in. molybdenum tube targets bombarded at an average beam current of 265 μa/hr produced an average of ~3 mCi of ^{95m}Tc per hr. The tubes were dissolved in 15% H_2O_2, and the technetium activities were extracted from the aqueous phase (adjusted to 2 N NaOH) with 2,4-dimethyl pyridine. The technetium activities were then back-extracted into water by adding benzene to the organic phase.[4,5] Technetium-95m was isolated after a 40-day period to allow the ^{96}Tc activity to decay.

Iodine-123

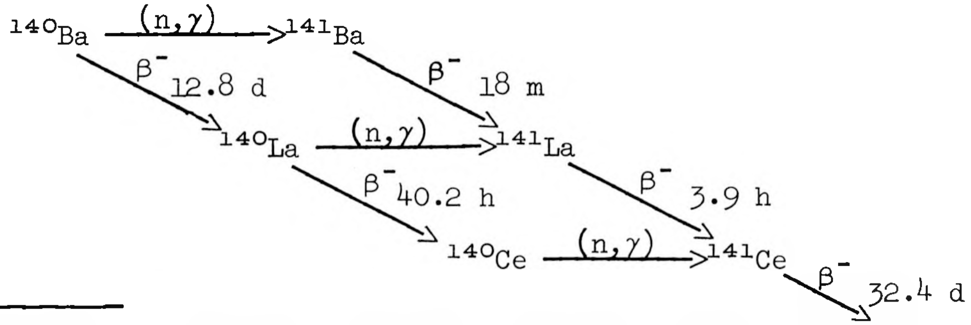
Iodine-123 (13 h), produced in the ORNL 86-Inch Cyclotron by the $^{123}Te(p,n)^{123}I$ reaction from ~80% isotopically enriched ^{123}Te, is being supplied to medical institutions through a special exemption granted by the U. S. Atomic Energy Commission. The production yields obtained from 16 runs indicate an average ^{123}I yield of 0.476 mCi per hr per mg of target. Based on these results, ~225 mCi of ^{123}I per 190-μa beam hour can be produced, compared with ~80 mCi per beam hour reported previously. During these runs, the target processing time was reduced from ~4-1/2 to ~2-1/3 hr. The valuable ^{123}Te target material has been recovered from each run and subsequently reused. The recovery yields have averaged ~96% of the starting material.

Cross-Section Determinations

Thermal Neutron Cross Section and Resonance Integral of Barium-140*

Due to the high fission yield of the 140 mass chain and the moderately long half-life of 12.8-d ^{140}Ba, the thermal neutron cross section and the resonance integral of this nuclide were obtained to determine the effect of ^{140}Ba on the neutron economy of nuclear reactors. Measurements were made with and without 40-mil-thick cadmium neutron filters, which have an effective cutoff energy of 0.54 ev. The thermal neutron cross section and resonance integral of ^{140}Ba were determined to be 1.57 ± 0.3 and 13.6 ± 1.4 barns, respectively. These values imply only minor neutron losses in the reactor due to neutron absorption by ^{140}Ba.

Each target consisted of 10 mCi of fission-product ^{140}Ba, which was purified by anion exchange using 0.01 M $(NH_4)_2CO_3$ to remove lanthanum, cerium, and uranium. After a 24-hr irradiation in a high flux position of the ORR, the ^{141}Ce fraction produced by the reaction series



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*Work performed in cooperation with the Chemistry and the Analytical Chemistry Divisions of ORNL.

was isolated from the ^{140}Ba target, and both were assayed using NaI gamma-ray spectrometry. The determination of ^{141}Ce required careful chemical separation since the amount of ^{141}Ce produced was five orders of magnitude less than that of ^{140}Ba. During the 24-hr irradiation, ~10% of the ^{141}Ce was formed from ^{140}La by neutron capture and <0.5% from ^{140}Ce by neutron capture.

The neutron fluxes were measured with 0.151% cobalt alloys using a 2200-m/sec value of 37 barns and a resonance integral of 75 barns for cobalt. The thermal neutron flux represents the Maxwellian component of the spectrum, and the resonance flux represents the epithermal flux per unit lethargy.

Effective Cross Section of Thallium-204*

The sufficiently long half-life (3.8 y) and pure beta emission of ^{204}Tl enhance its usefulness as a heat source for the generation of electrical power. Thallium-204 can be prepared from ^{203}Tl by neutron capture, and an activity concentration of ~35 Ci/g has been obtained (the specific activity of pure ^{204}Tl is 461 Ci/g). The burnup cross section of ^{204}Tl was determined in order to optimize irradiation conditions to obtain the highest possible specific activity.

The thermal neutron cross section and resonance integral of ^{204}Tl can be determined by irradiating pure ^{204}Tl [prepared by the ^{204}Pb(n,p)^{204}Tl or ^{204}Hg(p,n)^{204}Tl reactions] and measuring the amount of ^{205}Tl by mass spectrometry or by irradiating pure ^{203}Tl and measuring the ^{205}Tl by mass spectrometry. The first method is limited by the low yields of ^{204}Tl produced by the (n,p) and the (p,n) reactions. If ^{203}Tl is irradiated, the target must contain $\leq$1000 ppm ^{205}Tl to accurately measure the increase of ^{205}Tl after irradiation. The use of available calutron-enriched ^{203}Tl was precluded since the initial amount of ^{205}Tl in this material is too high.

Thallium-203 target material (5 mg) was obtained by chemically separating ^{203}Tl from four old ^{203}Hg product solutions. Four ~300-μg samples were inserted into the beryllium reflector of the HFIR and irradiated for ~45 days. Paired cobalt and niobium monitors were also inserted to measure the neutron flux. Gamma-ray analysis of the irradiated ^{203}Tl targets failed to detect any traces of ^{203}Hg. Mass analyses of the irradiated ^{203}Tl targets and the effective cross sections for ^{203}Tl and ^{204}Tl which were determined are given in Table 3.

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*Work performed in cooperation with the Chemistry and the Analytical Chemistry Divisions of ORNL.

Table 3. Mass Analyses of Irradiated Thallium Samples[a] and Effective Reactor Cross Sections of ^{203}Tl and ^{204}Tl

Composition, %			Thermal flux, neutrons/cm^2.sec	Effective cross section, barns	
^{203}Tl	^{204}Tl	^{205}Tl		^{203}Tl	^{204}Tl
93.78 ± 0.05	5.81 ± 0.04	0.410 ± 0.020	10.8 x 10^{14}	16.0	23.3
93.85 ± 0.02	5.76 ± 0.02	0.393 ± 0.008	10.2 x 10^{14}	16.7	24.0
96.98 ± 0.04	2.81 ± 0.03	0.211 ± 0.016	5.58 x 10^{14}	14.7	—
97.83 ± 0.02	2.03 ± 0.02	0.134 ± 0.007	3.80 x 10^{14}	15.0	22.1

[a] Initial composition: 99.900 ± 0.002% ^{203}Tl, 0.100 ± 0.002% ^{205}Tl, and <10 ppm ^{204}Tl.

KRYPTON-85 ENRICHMENT BY THERMAL DIFFUSION

A thermal diffusion pilot plant for the enrichment of ^{85}Kr from fission-product krypton was operated to gain experience and to develop design criteria for a full-scale plant. The pilot plant consists of six water-cooled tube bundles with calrod heaters in the center of each tube. Small bellows pumps circulate the gas through the tube bundles.

At the beginning of the year all six thermal diffusion column bundles were emptied and shut down when water leaks occurred in two separate bundles within a day of each other. The system was not started again until all of the columns had been retubed. During the period required to make these changes, a water treatment system was installed to improve the quality of the cooling water. A system to help avoid the loss of krypton when a calrod fails and perforates a tube was also designed. The column bundles were divided into sections that can be rapidly isolated from each other by automatic valves when sensing instruments detect a calrod failure or a sudden change in column pressure.

One cascade of three columns of the ^{85}Kr thermal diffusion isotope enrichment system was loaded with ~1500 Ci of 3.8% ^{85}Kr and operation began on June 17, 1967. Product fractions were removed from the three columns after 67 days of operation; the ^{85}Kr isotopic abundance ranged from 8.7 to 11.5%, which is below the minimum expected value of 15%. The product from the three columns and 7.8% ^{85}Kr from a previous run were put into the center section of one column for further processing. The other five columns were loaded with normal feed (~3.5% ^{85}Kr). Product fractions were removed in December; 240 Ci had been enriched to 13.97% and 370 Ci had been enriched to 27.62%.

RADIOISOTOPE SAND TRANSPORT STUDY

A cooperative program to study the movement of sand around the Point Conception (California) area was initiated in 1966. The two major participants are the Isotopes Development Center (AEC-DID) and the U. S. Army Coastal Engineering Research Center; other participants include the Department of Navy, Department of Air Force, State of California, and National Aeronautics and Space Administration. The experimental program consists of releasing a quantity of ^{133}Xe–tagged sand in a specified area and surveying this area with a detector system to determine the direction, the volume, and the velocity of sand movement. The study should provide data to predict the movement of materials such as highly toxic fuel contamination in seawater (resulting from a missile launching abort), possible sites for harbors that will not be subject to excessive sand blocking, and locations for the construction of recreational beaches. The potential use of the detector system can also be seen for underwater prospecting for radioactive materials.

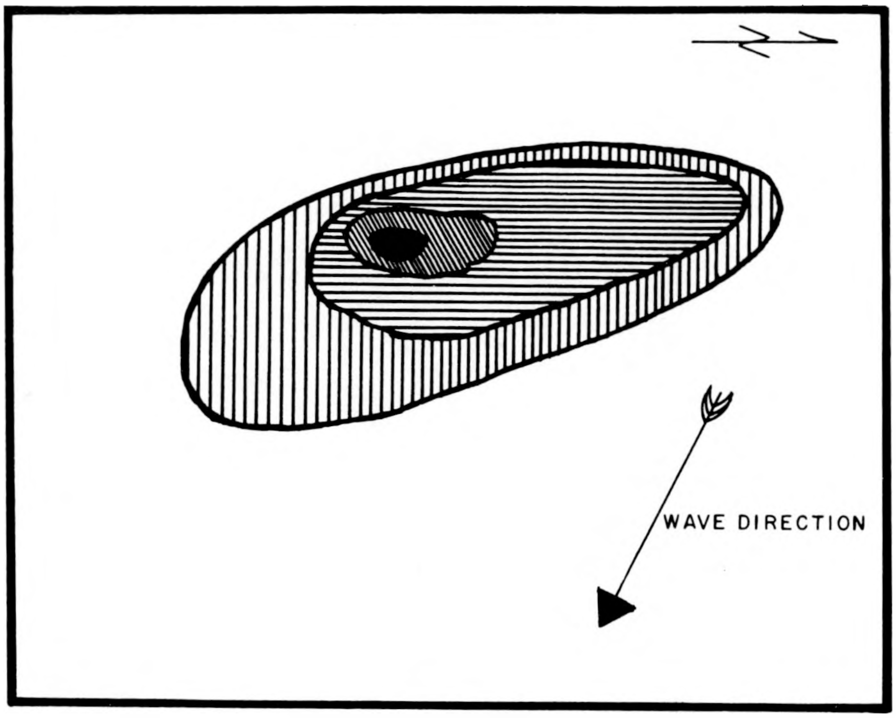
During the past year, field tests were carried out at three locations to evaluate equipment and to develop more effective procedures: Cape Kennedy, Florida (in April); Surf, California (in June); and Point Conception, California (in December). The ocean-floor radiation detector was tested at Cape Kennedy by placing a liter of sand tagged with 15 mCi of ^{133}Xe in a 3-ft-dia area at a depth of 30 ft, ~1/2 mile off the coast. Rapid movement of the sand was confirmed by the absence of radioactivity at the point of injection <5 min after the sand was placed on the ocean floor. After 30 min, the sand had dispersed in an area ~100 yd on either side of the injection point. The system was able to detect sand particles containing as little as 0.1 μCi of ^{133}Xe per cm^3 of sand.

A successful field test was conducted in June at Surf (near Vandenberg Air Force Base). The sand (~200,000 g) tagged with 2 Ci of ^{133}Xe was placed by scuba divers at a depth of ~30 ft. The operation of the survey system was excellent; the system operated in high surf, in waves up to 12 ft, and in areas where large rocks were present. The direction of sand movement was determined and followed for 2 days. The sand was traced for ~1-1/2 miles, and spectra obtained confirmed ^{133}Xe as the radioisotope in the sand.

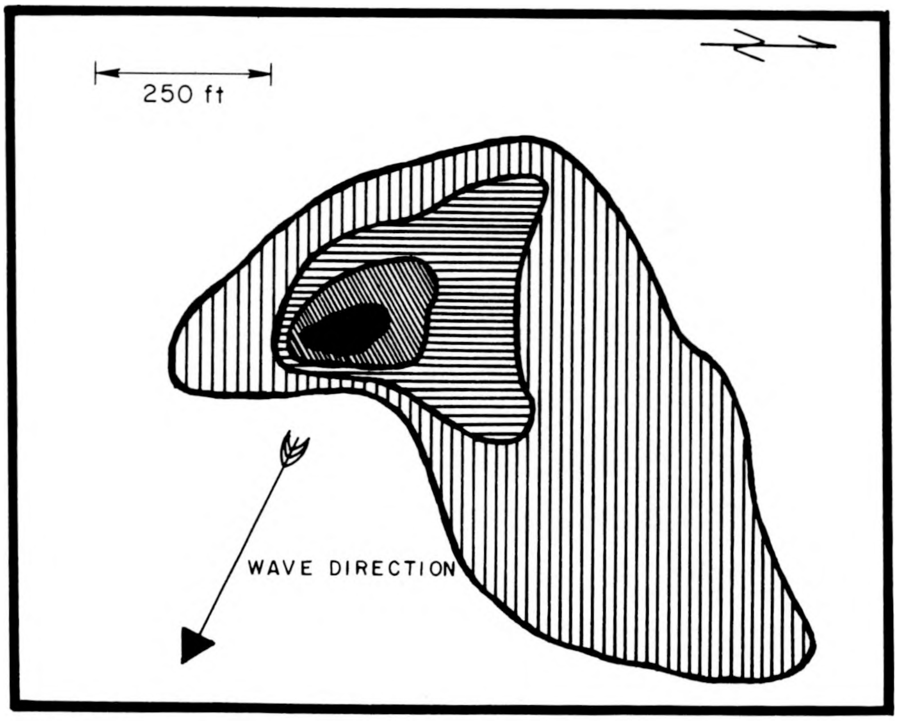
Data obtained during the June transport study were evaluated, and contour maps of sand movement were drawn (Fig. 2). Although the data used for Fig. 2 have not been corrected for decay, the type of contour that is being developed can be seen. During the 2 days after dispersion, the sand changed from a northern to a southern movement along the shore which appears to be the direction of total sand movement in the Surf area.

Results from the field test indicated that a technique for correlating data and that a shelter (Fig. 3) to protect the electronic instruments from the weather and the sea were needed. The detection system was modified to automatically correlate information that is required for

ORNL DWG. 67-8504



1 DAY AFTER RELEASE OF ^{133}Xe TAGGED SAND



2 DAYS AFTER RELEASE OF ^{133}Xe TAGGED SAND

Fig. 2. Change in Direction of Xenon-133 Tagged Sand.

data analysis. It monitors time, boat position (digital radar system), and radiation counts every 10 sec.

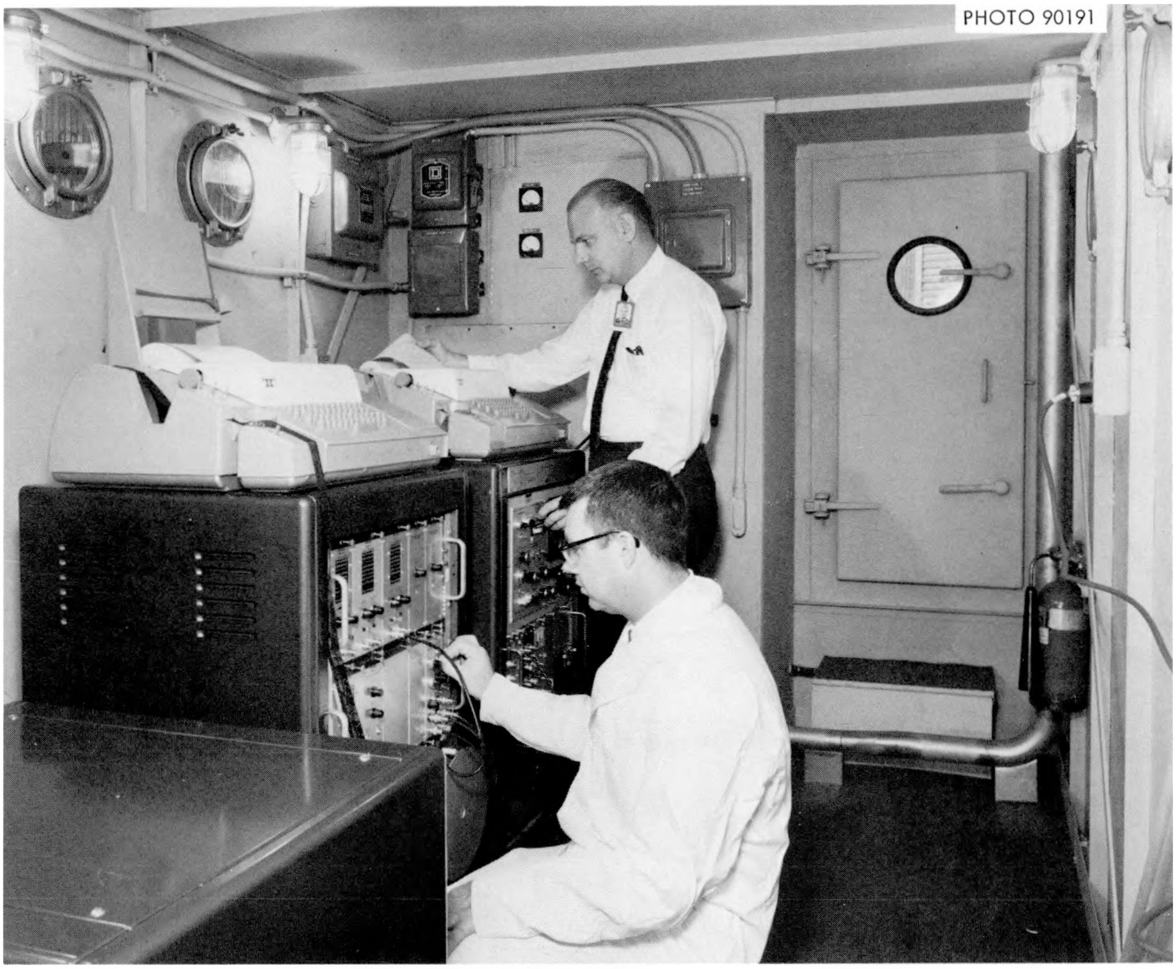


Fig. 3. Instrument Cabin for Vehicle Used to Tow Mobile Detector.

The third test was conducted at Point Conception. Injection was made at three sites: north of the point, on the point, and south of the point. Although these areas were surveyed for ~2 weeks, very little radiation data were collected. Contrary to results obtained at Surf, the sand appeared to undergo appreciable burial or essentially no transport. The same result would be obtained by no transport as by burial since it is practically impossible to place the detector assembly directly on the injection point. Future tests will attempt to answer these questions.

RADIOISOTOPE SOURCE DEVELOPMENT

During the past year, Radioisotope Source Development has included the development, fabrication, and characterization of beta, low-energy gamma, secondary X-ray, gamma, and neutron sources for varied applications. Assistance has been provided to other governmental agencies and industry to meet particular needs. Specific projects include the development of ^{153}Gd production methods, design and fabrication of an extracorporeal blood irradiator, preparation of an ^{55}Fe source for use in X-ray diffraction, design and fabrication of sources for use in portable radiographic cameras, and development of techniques for the fabrication and encapsulation of ^{90}Sr silicate into forms suitable for source applications.

Production Study of Gadolinium-153 for Use in the Mars Atmosphere Density Sensor

The feasibility of producing ^{153}Gd in sufficient quantity and purity to be used for the Mars Atmosphere Density Sensor source was determined, and alternative radioisotope sources were investigated. A gamma-ray output of 7.4×10^{11} photons/sec at Mars is required; the source activity requirement for ^{153}Gd is 150 Ci. Europium-155 and ^{57}Co are possible substitutes for ^{153}Gd. The order of production feasibility is (1) ^{153}Gd from natural europium, (2) ^{155}Eu from enriched ^{154}Sm, (3) ^{57}Co (cyclotron produced), and (4) ^{153}Gd from enriched ^{152}Gd.

Production of ^{153}Gd from Enriched ^{152}Gd

Although pure ^{153}Gd has a specific activity of 3500 Ci/g, calculations indicate that the maximum specific activity attainable in a flux of 3×10^{14} neutrons/cm^2.sec is 78 Ci of ^{153}Gd per g of ^{152}Gd. Since both the cross-section ratio (^{152}Gd:^{153}Gd) and the resonance integral ratio are of the order of 10^{-2}, no significant advantage results from use of a flux that is differently moderated. Therefore, other reactors will not produce a significantly higher specific activity than that obtained in the ORR.

Three 330-mg targets containing 13.5% ^{152}Gd mixed with aluminum to decrease the effects of flux depression were irradiated in the ORR for 37, 56, and 93 days. The size of these targets is comparable with those of targets which would be used in production runs in order to obtain a realistic estimate of actual production yields. The presence of ^{155}Gd and ^{157}Gd, each having large thermal cross sections, results in severe flux depression during the early stages of irradiation. Computed and experimental yields are given in Table 4.

Table 4. Yield of Gadolinium-153 from 13.5% Enriched Gadolinium-152

Irradiation time, days	Flux, neutrons/cm^2.sec	Yield of ^{153}Gd, Ci/g		
		per Gd	per ^{152}Gd	Computed
37	2.48×10^{14}	7.45	55	54
56	2.88×10^{14}	3.56	26	35
93	2.67×10^{14}	2.42	18	21

All three irradiation times exceeded the optimum. Irradiation for 15 to 20 days should yield at least 65 Ci of ^{153}Gd per g of ^{152}Gd.

The samples irradiated for 56 and 93 days contained ^{156}Eu and ^{160}Tb produced by the $^{156}\mathrm{Gd}(n,p)^{156}\mathrm{Eu}$ and $^{158}\mathrm{Gd}(n,\gamma)^{159}\mathrm{Gd}\xrightarrow[18\ h]{\beta^-}{}^{159}\mathrm{Tb}(n,\gamma)^{160}\mathrm{Tb}$ reactions, respectively. In the 93-day irradiation 0.1 Ci of ^{160}Tb per Ci of ^{153}Gd was produced. The computed ^{160}Tb:^{153}Gd ratio for a 20-day irradiation of 13.5% ^{152}Gd at 2×10^{14} neutrons/cm^2.sec is 2.3×10^{-3}. Use of more highly enriched targets would lower the impurity level somewhat but chemical purification would still be necessary.

The required output of the Mars Atmosphere Density Sensor source is 20 gamma Ci or 7.4×10^{11} photons/sec at Mars, and the proposed area[6] is 10 cm^2. For this source, the specific activity is critically important in the range of 20 to 50 Ci/g. At lower specific activity, self-absorption makes it impossible to attain the required output; at higher specific activity self-absorption will be slight.

The specific activity of ^{153}Gd produced from 13.5% ^{152}Gd will not be high enough for use in the Mars Atmosphere Density Sensor. A limited quantity of more highly enriched ^{152}Gd is available from which it may be possible to produce 130 Ci of ^{153}Gd with a specific activity of 23 Ci/g. The output of the resultant source would be only 15 gamma Ci at Mars. No allowance has been made for losses during chemical processing or source fabrication. Therefore, production of ^{153}Gd from enriched ^{152}Gd is not feasible for this application.

Alternative Production Method

Gadolinium-153 can also be produced by neutron bombardment of ^{151}Eu by the reaction sequence

$$^{151}\mathrm{Eu}(n,\gamma)^{152}\mathrm{Eu}\xrightarrow[9.3\ h]{\beta^-}{}^{152}\mathrm{Gd}(n,\gamma)^{153}\mathrm{Gd}\ .$$

The computed maximum yield in a 2×10^{14} neutron/cm^2.sec flux is 17.6 Ci per g of ^{151}Eu. If natural europium (47.82% ^{151}Eu) is used, this corresponds to 8.5 Ci of ^{153}Gd per g of target. Irradiation of natural europium also produces 12.4-y ^{152}Eu and 16-y ^{154}Eu which must be separated from gadolinium. After separation, the computed specific activity of ^{153}Gd is 78 Ci per g of gadolinium.

Alternative Nuclides

In cooperation with Research Triangle Institute, ORNL reviewed possible substitutes for ^{153}Gd in the Mars Atmosphere Density Sensor. Two alternative isotopes were found: ^{155}Eu and ^{57}Co.

Europium-155 (1.8 y) can be made by irradiation of ^{154}Sm by the reaction

$$^{154}\mathrm{Sm}(n,\gamma)^{155}\mathrm{Sm}\xrightarrow[22\ m]{\beta^-}{}^{155}\mathrm{Eu}\ .$$

The most serious difficulty in producing ^{155}Eu is minimizing the production of 16-y ^{154}Eu impurity simultaneously produced by the reactions

$$^{152}\text{Sm}(n,\gamma)^{153}\text{Sm} \xrightarrow[47\ h]{\beta^-} {}^{153}\text{Eu}(n,\gamma)^{154}\text{Eu}\ .$$

Since the saturation yield of ^{155}Eu is only 0.5 Ci per g of ^{154}Sm, chemical separation of product and recycling of target are necessary. The ratio of ^{154}Eu:^{155}Eu can be minimized by use of targets with a low ^{152}Sm:^{154}Sm ratio, short irradiation time, and frequent target reprocessing to remove ^{153}Eu.

Irradiation of enriched ^{154}Sm containing 98.5% ^{154}Sm and 0.5 to 1.25% ^{152}Sm at 2 x 10^{14} neutrons/cm^2.sec for 2 days should yield 0.17 Ci of ^{155}Eu per g of samarium. The computed ^{154}Eu:^{155}Eu ratio is (3-8) x 10^{-4}. Although shorter irradiation periods would yield a lower ^{154}Eu:^{155}Eu ratio, the resultant yield of ^{155}Eu would be impracticably low.

Cobalt-57 can be produced in the ORNL 86-Inch Cyclotron at ~25 mCi per beam hour.

Experimental Extracorporeal Blood Irradiator

An experimental blood irradiator (Fig. 4) was designed and fabricated for the Medical Research Division of Brookhaven National Laboratory. The blood of humans or animals can be shunted from an artery by 0.104-in.-dia silastic tubing through the irradiator for exposure and back into a vein. The volume of the tube in the irradiator is small (1.8 ml). Because the path through the irradiator is straight and the tubing is of small diameter, forces caused by expansion or constriction are minimized, and the radiation is absorbed more homogeneously. The blood and the silastic tubing, which it contacts, are compatible.

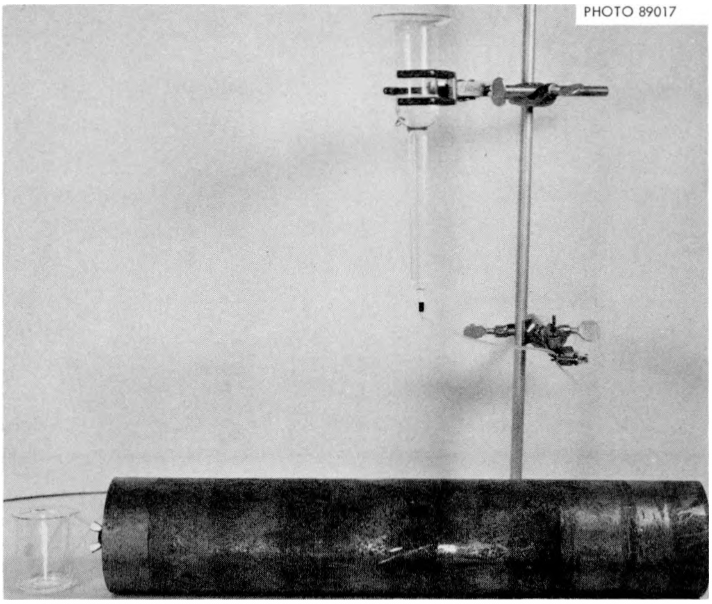


Fig. 4. Strontium-90 Extracorporeal Blood Irradiator.

The source for the irradiator is fabricated by ion exchange of ^{90}Sr-^{90}Y into molecular sieve material which is then formed into ceramic cylinders, 0.032 in. high by 0.032 in. dia, by firing at ~1100°C. These radioactive ceramic cylinders are placed into a stainless steel tube of 16-in. length, 0.067-in. OD, and 0.006-in. wall thickness. Small stainless steel plugs are pressed into the ends of the tubing and sealed by welding. The 2.68-Ci source is fastened slightly off center inside a 1-in.-dia graphite cylinder. The graphite cylinder and source are placed inside a 1-1/2-in.-thick lead cylindrical shield, 20 in. long by 4 in. dia. The silastic tubing can be threaded through the lead shield and through the graphite holder. The tubing inside the graphite is in very close contact with the radioactive source. The graphite cylinder shields the beta radiation, and the outer lead covering shields the bremsstrahlung radiation produced by the graphite. The radiation along the length of the cylinder is <20 mr/hr. At the ends the scattered beta radiation is 100-110 mr/hr. When the tubing and solution are placed in the irradiator, the radiation is reduced to <20 mr/hr at the ends.

The radiation-absorbed dosage was determined by the Fricke method,[7] by which the oxidation of Fe^{2+} to Fe^{3+} can be measured. The volume of 1.8 ml will receive an absorbed dose of 3.69×10^5 rads if it remains in the irradiator for 1 hr. At a flow rate of 100 ml/min, the blood stream will absorb energy which, if homogeneous, is calculated to be ~110 rads.

This irradiator weighs 101.5 lb and is not considered to be a portable unit. The patient must be immobilized during treatment. Units which weigh less and contain less radioactivity can be fabricated if medical experiments indicate that they are needed.

Iron-55 Source for Use in X-Ray Diffraction

An ^{55}Fe source was prepared for use in an X-ray diffractometer. Iron-55 decays by electron capture to stable ^{55}Mn; in ~30% of these transitions, a 5.9-kev manganese K X ray is emitted.

Iron-55 was formed by proton bombardment of ^{55}Mn in the ORNL 86-Inch Cyclotron. Manganese-54, which formed simultaneously, served as a tracer for manganese during chemical separation of iron from the target. The iron was separated from the manganese by solvent extraction. No ^{54}Mn was detected in the iron fraction. A total of 416 mCi of ^{55}Fe was recovered. The iron fraction was dissolved in 2 ml of an electroplating solution of saturated ammonium oxalate, 0.15 M in H_2SO_4. The Plexiglas electroplating cell exposed a rectangular cathode area of 2 by 10 mm; 50 ma was passed through the system for 90 min. Ninety-five percent of the ^{55}Fe was deposited onto platinum foil. The source was covered with a thin film of aluminum to prevent smearing.

Sources for Use in Radiographic Cameras

Two types of radiographic cameras were designed and fabricated: one for use with ^{147}Pm and ^{169}Yb and the other for use with ^{241}Am and ^{57}Co. The characteristics of the sources and the output of the cameras are given in

Table 5. The advantage of the radiographic camera is portability. This radioactive device can be transported and used with very little exposure to the operator. When used with fast developing Polaroid film, a positive print can be obtained within a few minutes after exposure.

Table 5. Source Characteristics and Output of Radiographic Cameras

Source	Activity, Ci	Source dimensions, in.		Radiations, kev	Exposure[a] at 16 in., mr/hr
		Height	Diameter		
$^{147}Pm_2O_3$	100-300	3/8	1/2	38 X ray	90-280
^{169}Yb	1	3/8	1/2	63 gamma-50 X ray	600
$^{241}Am_2O_3$	2	1/2	1/2	60 gamma	90
^{57}Co	0.5	1/2	1/2	124 gamma	250

[a] With shutter open.

The boxlike camera (1-1/2 x 1-1/2 x 2-1/4 in.) shown in Fig. 5 is designed for use with ^{147}Pm (it can also be used with ^{169}Yb) and shielded with a minimum thickness of 3/4 in. of tungsten-copper alloy, which reduces the outside radiation to $\leq$20 mr/hr. The window of the encapsulated source is made of 0.015-in. aluminum or 0.016-in. stainless steel. For use with ^{169}Yb, an additional 1/2 in. of shielding is used to reduce the outside radiation to <50 mr/hr. The camera is designed so that the source remains fixed, and the shutter opening is revolved into position by moving the tungsten-copper alloy shield. A standard camera shutter release was modified to lock the shutter in an open position and to release the shutter so that timed exposures can be made.

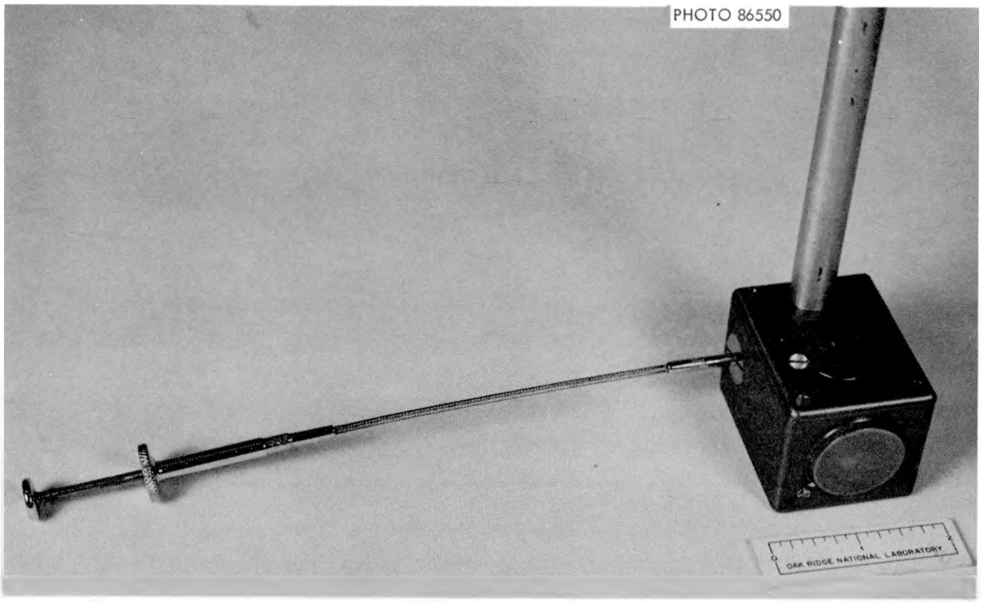


Fig. 5. Promethium-147 Radiographic Camera.

The other camera (Fig. 6) was specifically designed for use with ^{241}Am but can also be used with ^{57}Co. The window of the stainless steel capsule is made of 0.010-in.-thick stainless steel. The shutter fabricated from 3/4-in. thick tungsten-copper alloy operates by a simple mechanism and moves perpendicular to the direction of the radiation. The shutter can be locked in an open position and released for timed exposures.

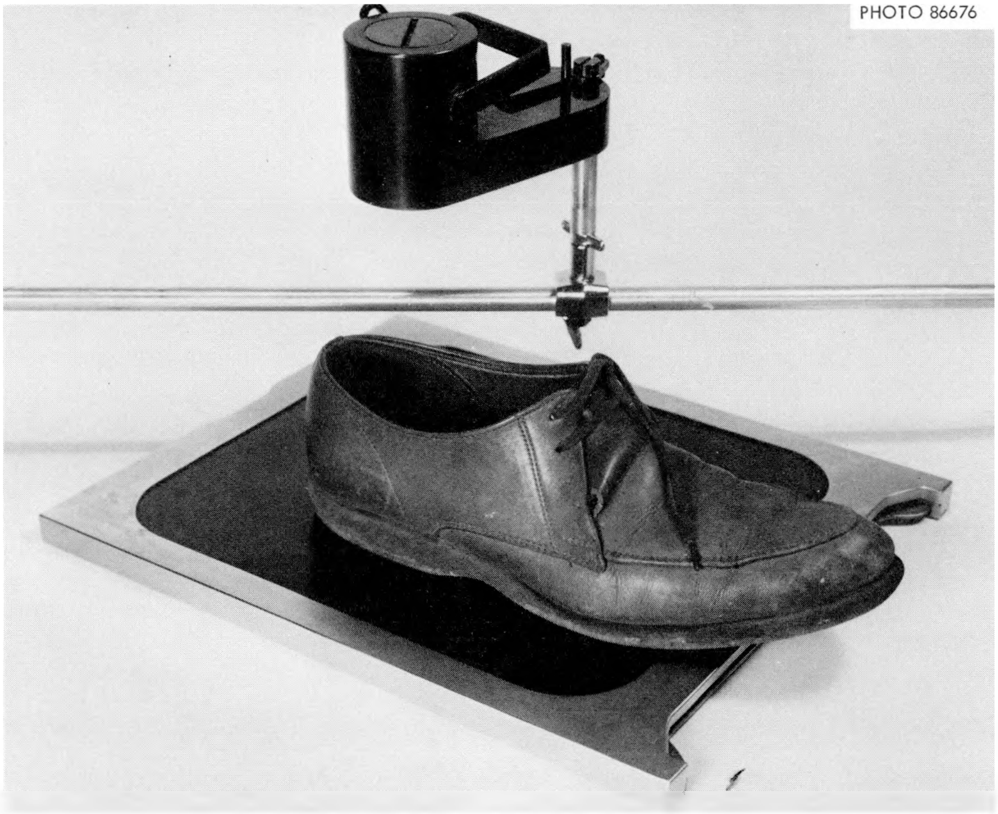


Fig. 6. Americium-241 Radiographic Camera for Inspection of Contraband.

Preparation of $^{90}SrSiO_3$ Microspheres for Use in Beta Sources

A process was developed for producing high integrity beta sources consisting of stainless steel capsules containing strontium silicate spheres embedded in an aluminum matrix. The process consists of three steps: (1) production of the strontium silicate feed (Fig. 7) for the sphere forming furnace, (2) formation of the spheres at 2000°C, and (3) encapsulation of the spheres in molten aluminum.

Strontium silicate was precipitated from an aqueous nitrate solution with a commercially available organic quaternary ammonium silicate. The strontium silicate was washed, dried, calcined, and fused. The fused mass was crushed to a particle size ranging from 50 to 100 mesh. Strontium silicate spheres (Fig. 8) were formed by dropping the feed through a 2-in.-dia graphite tube heated to ~2000°C.

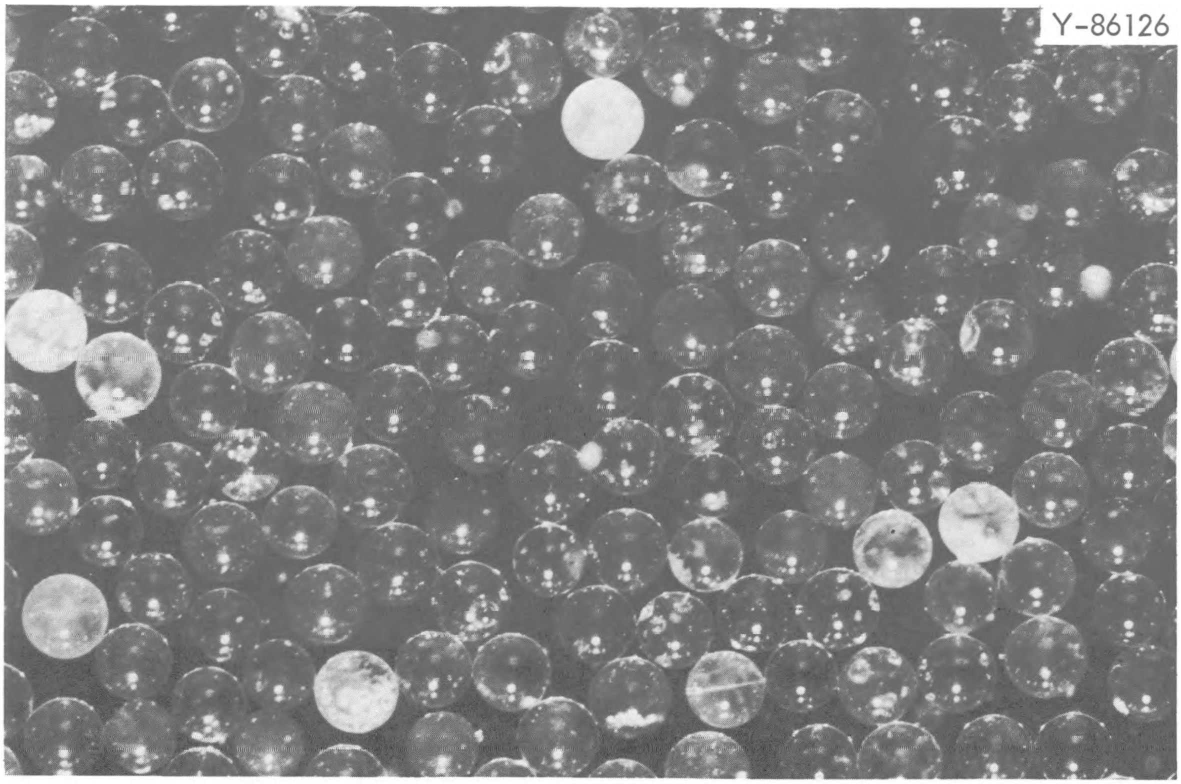


Fig. 7. Strontium Silicate Minibead Furnace Feed, 90X.

Fig. 8. Strontium Silicate Minibeads, 40X.

Source capsules were filled by passing strontium silicate spheres through a small hole drilled in one end plug. Molten aluminum was forced into the capsule by atmospheric pressure, and a matrix completely enclosing each sphere was formed. The final seal was made by electric arc fusing a stainless steel plug (usually 0.040 in. dia) in the fill hole.

The feasibility of the process for large-scale operation was demonstrated by production of 17,000 Ci of $^{90}SrSiO_3$ spheres in the Fission Products Development Laboratory. This material was loaded into rectangular tubing for use in chemical process irradiators. The properties of the $^{90}SrSiO_3$ sources are given in Table 6.

Table 6. Properties of Strontium-90 Silicate Sources

Spheres

Diameter, μ	125 to 375
Density, g/cm^3	3.5
Weight, g/cm^3	2.18
Weight of spheres with Al, g/cm^3	3.18
Specific activity, Ci/g	33.7
Specific activity, Ci/cm^3	74.14

Source Size and Loading

Cross section of rectangular tube, in.	Loading, Ci/in. of length	Output, Ci/in. of length
0.065 by 0.375	29.6	~7.7
0.085 by 0.375	38.6	~10.0

A chemical process irradiator and its tube source plaque are shown in Fig. 9. Rectangular and finned-type source forms (Fig. 10) have also been fabricated for investigation.

The physical tests made on prototype sources containing nonradioactive strontium silicate showed them to possess excellent physical properties. The sources remained leak free after exposure to high temperatures (1700°F for 1 hr) and when subjected to external pressure (20,000 psig), to high crush forces (2000 lb for 1 hr), and to high shear forces (1000 lb for 1 hr).

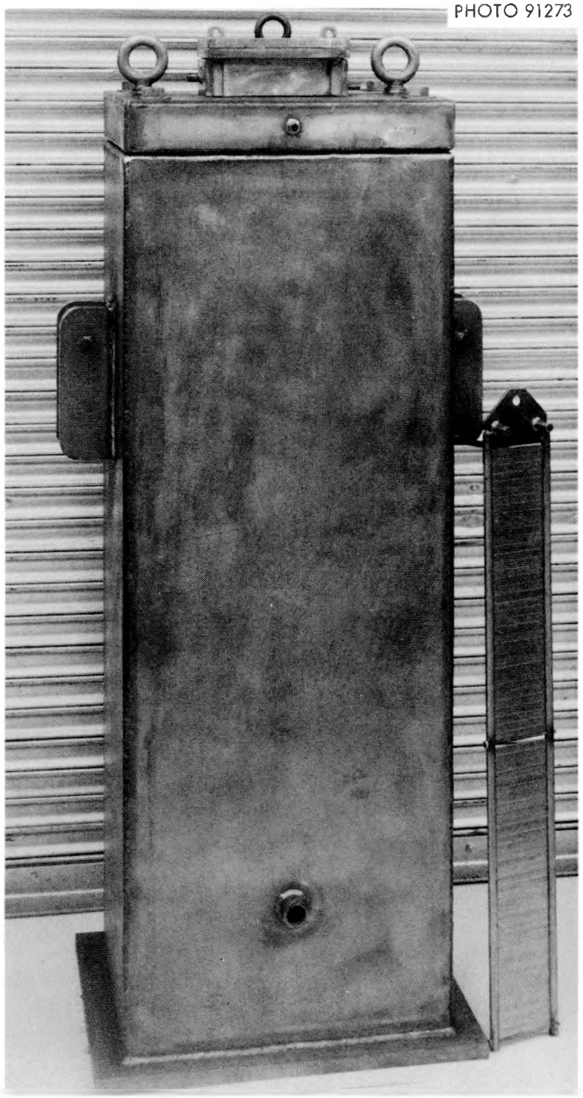


Fig. 9. Chemical Process Irradiator and Tube Source Plaque.

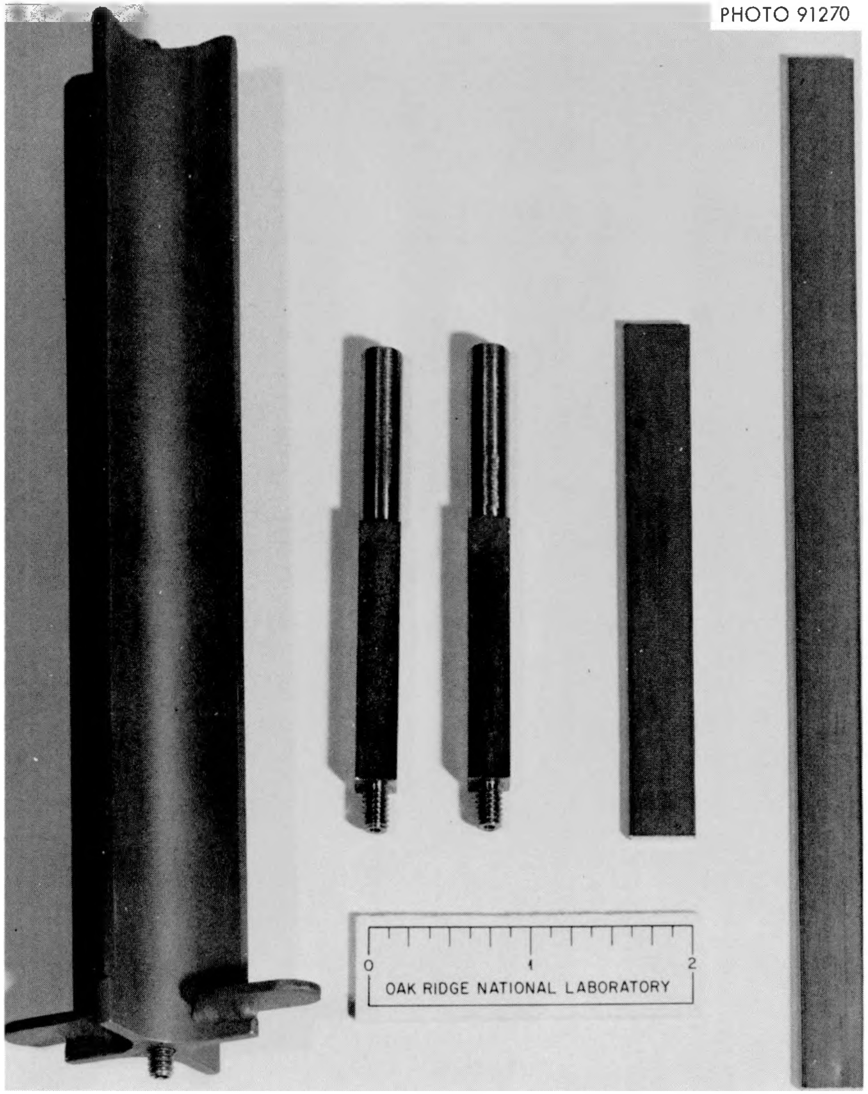


Fig. 10. Example of Source Geometry Possible Using $^{90}SrSiO_3$ Minibeads in an Aluminum Matrix.

RADIOISOTOPE SOURCE SAFETY TESTING

The Radioisotope Source Safety Testing Program is concerned with determining the reliability of radioactive sources to contain radioactive material under conditions of normal use as well as in the event of an accident. In addition to sources, shipping packages used to transport sources and radioactive materials are tested under simulated accident conditions. Investigations provide data which enable source manufacturers and regulatory agencies to evaluate existing and future source and shipping package designs from the standpoint of safety to the users and the general public. During investigation, simple tests are devised which can be used by manufacturers for quality control purposes.

Source Applications Guide

An applications rating guide was developed to determine the required source classification[8] for the intended application of a source and the expected environmental conditions during its operating life. The guide was compiled after studying actual field experience of source users with sources in the individual classification groups and the results of laboratory tests simulating various use conditions.

The Source Applications Guide establishes the required source capsule classification by evaluating the key containment factors involved in source usage and relating them to the containment reliability described by the Source Capsule Classification System. The purpose of the guide is to assist source users, manufacturers, and licensing authorities in determining the source capsule classification required for a specific source use and use environment.

The Guide consists of two parts. In Part I a minimum classification is determined, based entirely on operating conditions of the source in normal use. In Part II a point rating system is used to establish a second classification which is based on the consequences of release; fire, explosion, and accident potential of the source use environment; and protection offered by devices. The required source capsule classification is obtained by comparing the classifications obtained in Part I and II and selecting the highest temperature and structural classification.

Special Form Source Testing

Tests are underway to determine if certain selected ORNL source capsules meet the USAEC test requirements for special form material. The test capsules are normal source capsules except that nonradioactive CsCl was used instead of the radioactive material and the atmosphere inside the inner capsule is a mixture of 65% argon—35% helium to facilitate leak testing. Impact percussion, heating, and immersion tests are required for certification as special form materials.

The impact test consists of dropping the capsule from a height of 30 ft onto a flat horizontal target in a position to produce maximum damage.

The percussion test consists of placing a capsule on a 1-in.-thick sheet of lead and impacting with a 3-lb steel weight dropped from a height of 40 in. The heating test consists of heating the capsule in air to 1475°F and holding at that temperature for 10 min. The immersion consists of immersing the capsule for 24 hr in water at room temperature. It is also necessary to demonstrate that the capsule is constructed of material which does not dissolve or convert into dispersible reaction products by >0.005 wt % of material during one week's immersion in water at 68°F or in air at 86°F.

After each test, the test capsule is leak tested by two methods: the helium mass spectrometer method with a sensitivity of 1×10^{-8} atm.cm/sec and the vacuum leak test.

Source Capsule Leak Definition

Four doubly encapsulated CsCl sources, each containing a 0.34-g nonradioactive CsCl pellet (equivalent to 8.5 Ci of ^{137}Cs), were leach tested by the ORNL Source Capsule Classification Test Procedures. A single hole was drilled in each capsule; the holes ranged from 0.5 to 2.5 mils in diameter.

The sources were immersed in water, and the pressure was alternately decreased and increased. The leach water was then analyzed for cesium by flame photometry. When the source capsules were opened after the test, the CsCl pellets had almost completely dissolved, and the void spaces in the capsules were nearly filled with water. Flow rates were measured before the test to ascertain that the capsule leak holes were not plugged and after the test to determine if plugging occurred during the test. The leak holes of the outer capsules were free of plugging after the test. The amount of plugging of the inner capsule leak holes and the amount of cesium leached from each source are given in Table 7. The sizes of the leak holes are stated as air leak rates measured at a pressure drop of 15 psi across the leaks. Positive indications of leaks were obtained for each of the sources.

Table 7. Results of Vacuum Leach Test on Doubly Encapsulated Nonradioactive CsCl Sources

Leak hole size, ml of air/sec		Cesium leached	Amount of inner
Inner capsule	Outer capsule	from source, μg[a]	capsule plugging, %
0.098	0.156	5	98
0.215	0.160	27,800	42
0.377	0.185	10	8
0.606	0.202	37,600	3

[a] One microgram of CsCl is equivalent to ~25 μCi of ^{137}Cs.

Examination of Old Sources

Two ^{137}Cs sources that had been in storage were examined. One was a duplicate of the sources used in a Peruvian insect irradiator, and the other was a duplicate of sources used in a Dow Chemical Company gamma-activated, organic reaction process. Both sources were measured and found to be bulged. Gas samples of the sources indicated that the duplicate of the Peruvian sources leaked and the duplicate of the Dow sources was still intact. Metallographic examination of the outer and inner capsules of the Peruvian source showed a hole in the outer closure weld. The leak in the inner capsule, though not located, was probably obliterated during gas sampling.

Compatibility of Molten CsCl with Various Capsule Materials

Test capsules made of 304L and 316L stainless steel, Inconel, and Hastelloy C were exposed to cesium chloride for 2 hr at 1300°C to determine the compatibility of the various capsule materials with cesium chloride under conditions which may exist in the event of a fire. A nominal analysis of the test capsule materials is given in Table 8.

Table 8. Nominal Composition of Alloys
Used to Contain CsCl for 2 hr at 1300°C

Alloy	Nominal composition, wt %								
	Fe	Ni	Cr	Mo	W	Mn	Si	Co	C
304L SST	70	8-10	18-20	–	–	–	–	–	0.03
316L SST	65	10-14	16-18	2-3	–	–	–	–	0.03
Inconel	7	75	15	–	–	1.0	0.5	–	0.15
Hastelloy C	4-7	55	14.5-16.5	15-17	3-4.5	1.0	1.0	2.5	0.08

The stainless steel and Inconel capsules were made from tubing; the Hastelloy capsule was machined from a rod. A pressed pellet of cesium chloride was loaded into each of the capsules in an argon atmosphere, and the capsules were sealed by electron-beam welding. Each capsule was placed into individual Inconel protective containers which were also electron-beam welded.

Visual examination of the capsules after testing disclosed that the 304L and 316L stainless steel and the Inconel capsules did not leak, but that the Hastelloy C capsule did leak. Metallographic examination of cesium chloride samples from each of the capsules showed no evidence of interaction between cesium chloride and 304L or 316L stainless steel or Inconel. The cesium chloride did interact with the Hastelloy C, however, and intergranular penetration of the bottom end plug and the welded region between the capsule wall and the end plug was observed. Analyses of the cesium chloride before and after the test are given in Table 9.

Table 9. Chemical Analyses of CsCl Before and After Contact With Several Container Materials for 2 hr at 1300°C

Analysis	Concentration, ppm								
	Fe	Ni	Cr	Co	Mn	Mo	Si	P	S
Before test	–	–	–	–	–	–	–	1	112
After test									
In 304L SST	370	70	110	–	–	–	–	–	–
In 316L SST	170	160	60	–	–	80	<40	–	–
In Inconel	320	260	110	–	80	–	<40	–	–
In Hastelloy C	120	340	110	<40	50	<40	130	–	–

Shipping Container Testing and Evaluation

Sugarman Cask Fire and Impact Shield

A heat transfer study was performed on a Sugarman shipping cask in combination with the fire and impact shield in which it will be shipped. The shield is constructed of two thicknesses of 2- by 6-in. maple boards (total thickness, 4 in.). A 400-w heater located inside the cask was connected to a Variac to simulate the heat output of various radioisotope shipments. Temperature readings were obtained for nine heat input conditions between 102 and 371 w by using thermocouples attached to the inner and outer surfaces of the cask and the fire-impact shield. The values for the inner and outer cask surfaces ranged from 60 and 55°C, respectively, for 102-w to 165 and 152°C for 371-w input. The corresponding temperatures for the inner and outer surfaces of the wooden shield were 45 and 32°C to 121 and 45°C, respectively. Ambient air temperatures during the tests varied from 25 to 31°C.

The container was impact tested after loading with a cask and lead brick to a total weight of ~3300 lb, and two drop tests were performed. One corner of the shield impacted the test pad from the 30-ft drop. The entire bottom of the box was splintered. Severe cracking of the wood on all four sides was evident. The second drop was from 40 in. onto a steel bar ~15 cm in dia. The bottom center of the shield impacted the bar. Extensive damage was sustained. Extensive bending of the entire steel frame around the wooden box was evident. The wooden bottom was badly punctured and severely splintered.

The container was then fire tested according to the procedures specified by the IAEA.[9] The burn pits consisted of three concentric circular tanks with diameters of 6, 12, and 16 ft and with ~130, ~145, and ~165 gal, respectively, of fuel oil floating on water. A water-cooled stand was placed over the center pit. The fire and impact shield was placed on the stand and 12 thermocouples were used to obtain temperatures at various points during the test (Fig. 11).

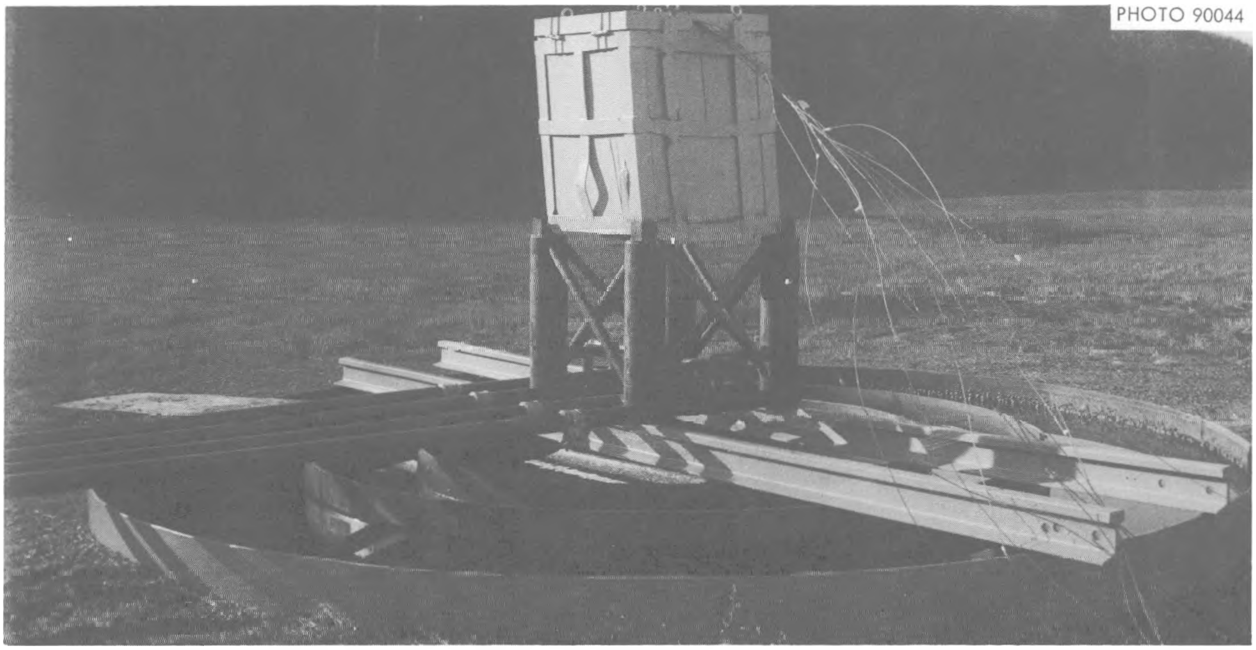


Fig. 11. Sugarman Fire and Impact Shield Prior to Start of the IAEA 30-Min Fire Test.

Although the box maintained its integrity during the 30-min oil fire (Fig. 12), it continued to burn after the oil was extinguished and was finally consumed (Figs. 13 and 14).

Fig. 12. IAEA 30-Min Fire Test In Progress.

PHOTO 90047

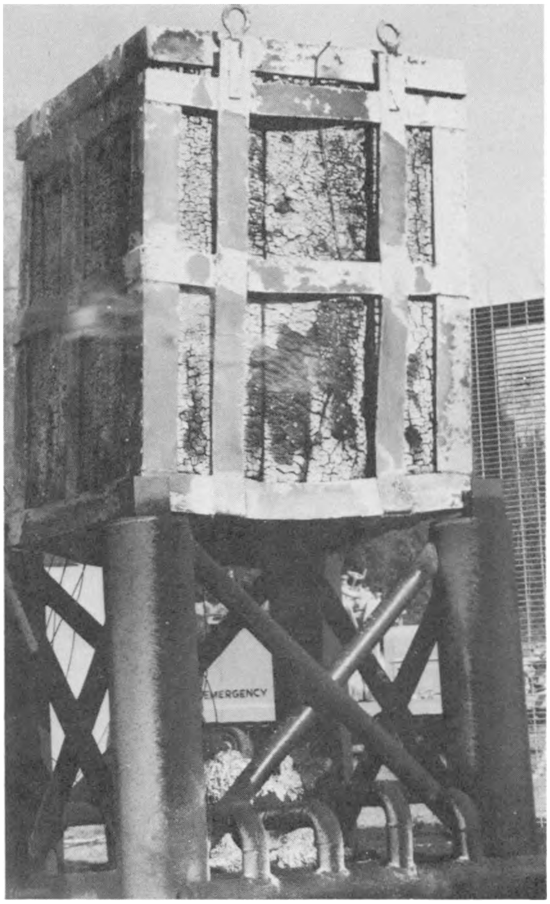


Fig. 13. The Sugarman Fire and Impact Shield After the 30-Min Fire Was Extinguished. Some of the melted lead that ran out and the beginning of the self-ignition of the charred wood shield can be seen.

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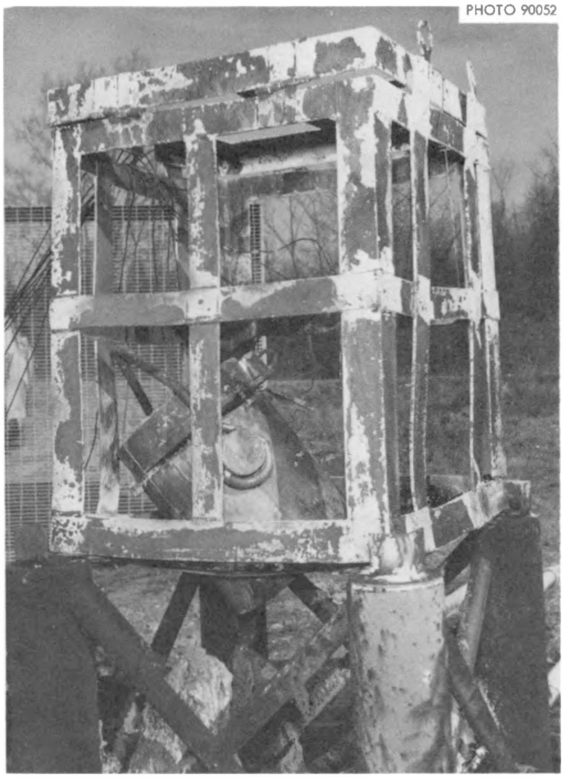

Fig. 14. The Sugarman Fire and Impact Shield 80 Min After the Oil Fire Was Extinguished. The substitute cask ruptured, the additional lead bricks completely melted, and the entire wood shield was consumed.

Since the present testing procedures specified in the regulations do not permit the fire shield itself to be extinguished after the 30-min oil fire, the present design was considered inadequate, and the wooden fire and impact shield was redesigned. The frame was modified to afford more protection to the wooden surfaces. The wooden box will be constructed of an outer layer of maple ~2 in. thick. Two thicknesses of 1-in. plywood will form the interior layer of the box. The outer surface will be painted with an intumescent paint. Intumescent paint will be applied between the surfaces of the plywood. A fire retardant paint may be used as an undercoating for the intumescent paint. Two shields of the new design will be built and tested.

Shipping Cask Evaluations

The structural and heat transfer calculations for the large uranium-shielded shipping cask to be used for shipments up to 400,000 Ci have been completed and were submitted to the AEC for approval.

Structural and heat transfer calculations for the double-envelope lead-shielded cask (75,000 Ci) were approved by AEC-ORO, and a special permit for the use of this cask in national and international commerce was issued by the Department of Transportation.

A fire and impact shield for radioactive gas cylinders similar to the ORGDP UF_6 phenolic resin foam shipping containers was designed and fabricated by ORGDP and approved by AEC-ORO.

Tungsten-Shielded Cask

A small stainless steel-clad, tungsten alloy-shielded cask was designed, fabricated, and tested as a prototype shipping container. This cask has a 2-in.-thick cylindrical radiation shield (equivalent to 4 in. of lead) with a hemispherical base instead of the conventional flat bottom. It weighs 380 lb and has been tested for a thermal capacity of up to 150 w. It is presently being reviewed by AEC-ORO for approval as a shipping container.

Irradiator Evaluations

The AEC Division of Isotopes Development requested ORNL to evaluate four irradiators to determine if the irradiators meet current AEC, ICC, and IAEA regulations concerning radioactive materials shipping containers. The four were the Shipboard Irradiator, Mobile Gamma Irradiator, Portable Cesium Irradiator, and Bulk Grain Irradiator. The evaluations of the Shipboard Irradiator and the Mobile Gamma Irradiator were completed. The evaluation of the Portable Cesium Irradiator was ~75% complete at the end of the year. A study (~75% complete) is being made to determine if it is economically feasible to modify the Bulk Grain Irradiator to meet the new requirements.

ISOTOPES INFORMATION CENTER

The objectives of the Isotopes Information Center (IIC) include:

1. Writing state-of-the-art reviews and other production and applications brochures, with particular emphasis on the areas of interest to the Division of Isotopes Development (DID).

2. Publishing the quarterly Technical Progress Review, Isotopes and Radiation Technology.

3. Establishing a continuing and up-to-date collection of indexed material covering the production and applications of isotopes and amenable to retrieving the information necessary to publish the above-mentioned state-of-the-art reviews and Isotopes and Radiation Technology.

4. Providing technical and editorial assistance to DID in the preparation of talks, annual reports, etc.

5. Answering inquiries for information concerning the production and applications of isotopes.

6. Participating in meetings, seminars, and panel discussions (i.e., miscellaneous information-type activities appropriate to the functions of AEC-DID, the ORNL Isotopes Development Center, and the ORNL Isotopes Division).

State-of-the-Art Reviews

Work was started on several state-of-the-art reviews including "Radioisotopes in the Textile Industry," "Technetium-99," "Kryptonates," "Iodine-132," and "Isotopes in Diffusion Studies." One review, "Isotopic Methods for Oceanographic Measurements," was completed and is being published.

Isotopes and Radiation Technology

During 1967, Vol. 4 of Isotopes and Radiation Technology was completed, one issue of Vol. 5 was published, the second issue was galley-proofed, the draft of the third issue was printed as ORNL-TM-2001, and the draft of the fourth issue was ~50% completed. Difficulty is still being experienced in getting persons other than IIC staff to author articles. However, a bright note is that more articles are being submitted (rather than requested) than previously, and authors are writing for the second time. Reprints are being used at an increasing rate; ~300 were distributed during the year. During the year >30 articles were written by IIC staff members for Isotopes and Radiation Technology.

Information Center Library

Toward the end of the year, the IIC completed its first 10,000 accessions (Fig. 15). The scope of the system is being reevaluated to determine what

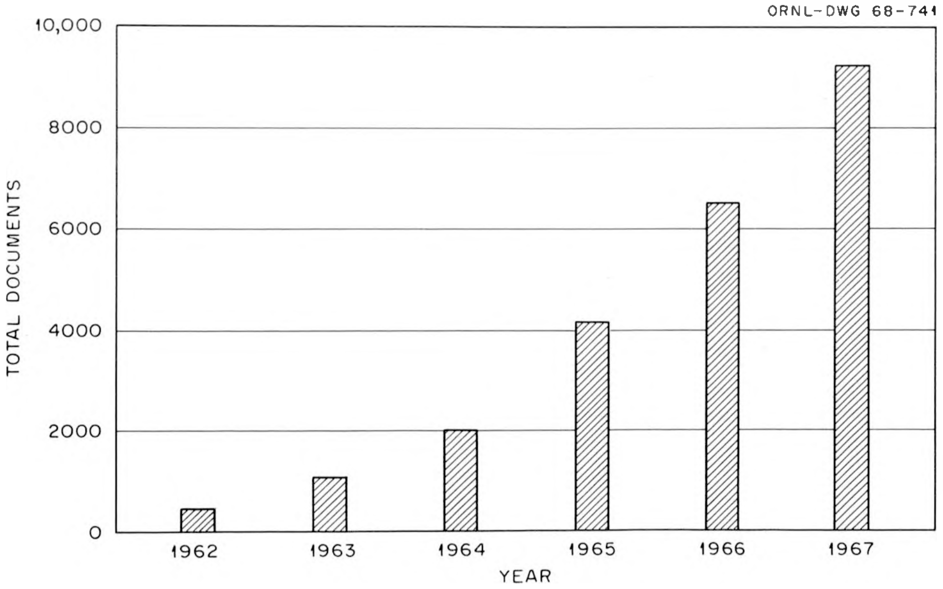

Fig. 15. Growth of IIC Accessions, 1962-67.

new terms need to be added and what terms can be deleted. In general, the emphasis on medicine will be decreased and less attention will be given to routine applications than in the past. The IBM MTST-DigiData-computer program is nearly completed, and preliminary results indicate that there will be no major problems. The chemical-patent tapes purchased from Information for Industry were received, and a program is nearly completed for obtaining a printout of patents dealing with radiation processing.

DID Technical and Editorial Assistance

Assistance was again provided to the DID for preparing the annual report to Congress, a paper was written for presentation at the 8th Japan Conference on Radioisotopes, and help was provided in editing the summaries of all DID contracts.

Handling Requests for Information

Approximately 650 letters were written in response to technical inquiries, and numerous other responses were made by telephone and in person (Fig. 16). Approximately 9000 reprints of various articles were sent to authors and other requesters or distributed at meetings. Packets of food-irradiation material were made available at the Oak Ridge Food Irradiation Symposium. Twenty-two papers and ~150 letters were translated. IIC and Isotopes and Radiation Technology flyers were mailed to >1700 members of the Atomic Industrial Forum to acquaint them with the services and activities of the IIC. Approximately 130 letters and brochures were sent to state representatives for the State Technical Services Program, offering IIC assistance in furnishing literature, speakers, etc. There were >100 visitors to the IIC, including representatives from ten foreign countries (France, Sweden, Australia, Finland, Denmark, Brazil, South Africa, Austria, Germany, and England).

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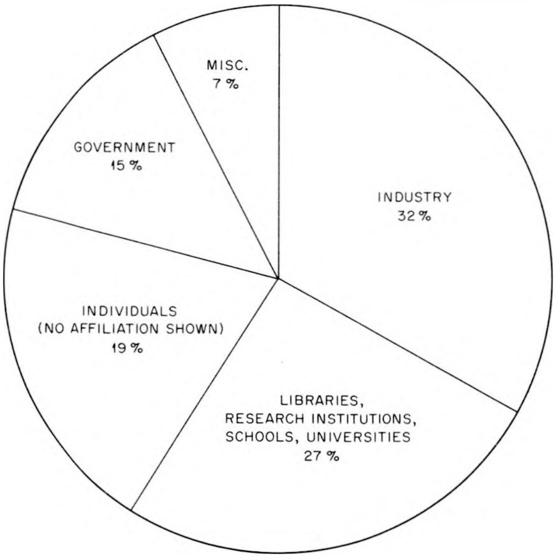

Fig. 16. Origin of ~1000 Inquiries Received by IIC During 1967.

Miscellaneous Activities

Several articles were written for publication, including a chapter on "Stable Isotope Preparation and Applications" for Survey of Progress in Chemistry; a chapter on "Radioisotopes" for The Book of Popular Science; two chapters on "Nuclear Data" and "Radionuclides for Industrial Applications," for Handbook of Radioisotopes; and "Availability of Uses for Radioisotopes in Chemical Processing" for Chemical Engineering. Other publications include two "Isotope Developments" (Nos. 4 and 5, "Ocean-Bottom Sediment Density Gage" and "Suspended-Sediment Concentration Gage") and the reports listed under "Publications" (page 39). Page proof was completed on "Special Sources of Information on Isotopes" (ORNL-IIC-12).

Staff members participated in the Columbus Aerospace Meeting (Program Committee) and the Gatlinburg Analytical Chemistry Meeting (chairman of discussion panel); they presented talks at the Columbus Aerospace Meeting, to the Missouri AEC, at the Latin American Isotopes Conference (Los Angeles), at DePauw University and the University of Arkansas, at the ORINS Isotope Technology Course, at the ORAU-sponsored Engineering Faculty Orientation Meeting, and at two meetings sponsored by the New York State Technical Services Board. Members also helped in a one-month training of a visiting information center specialist.

ISOTOPES PRODUCTION AND SALES

Fission Products Development Laboratory Operations

Shipments Received and Production

During 1967, 1,040,000 Ci of ^{137}Cs feed was received; ~800,000 Ci was processed through the mainstream accumulation step; 400,000 Ci of ^{137}CsCl product was prepared; and ~315,000 Ci of ^{137}CsCl was pressed into pellets. No ^{90}Sr feed was received or processed to $^{90}SrTiO_3$ for inventory. Approximately 230,000 Ci of ^{90}Sr was converted to fuel pellets for heat sources.

Cesium-137

Cesium-137 was shipped from Richland, Washington, on a routine basis in the Decalso-loaded Shielded Transfer Tanks (STT), Model II. A new STT (Model III) containing 110 gal of AW500 ion exchanger, compared with 400-gal Decalso beds in the Model II casks, was placed in service. However, due to the higher sorptive capacity of AW500 for ^{137}Cs, both types of casks can be loaded with ~50,000 Ci.

Comparative stripping tests were made with the Model II and Model III casks. It was found that the higher sorptive capacity of AW500 is balanced by a proportionally lower elution efficiency so that the ^{137}Cs concentrations in the effluent solutions from both types of casks are essentially the same. If AW500 were used in the larger Model II cask, the maximum approved limit of 90,000 Ci could be shipped at reduced shipping costs. Since the present rate of processing can be maintained adequately with existing casks, no plans have been made to replace the Decalso in the Model II casks.

No major changes in the cesium-ammonium alum cocrystallization process were made. The ^{137}Cs alum concentrate is being stored in solution form in amounts of up to 1 MCi rather than being processed directly to final product to permit purification, processing, and product preparation on a campaign basis. The entire ^{137}Cs crystallizer system operated throughout the year without in-cell maintenance.

Cesium-137 was purified by the oxalate crystallization process on a campaign basis in batch sizes ranging from 100,000 to 250,000 Ci. Difficulties were encountered in handling the concentrated ^{137}Cs alum feeds, and yields averaged <50%. Full-scale tests are being made to determine whether the first of two oxalate crystallization steps can be eliminated without affecting product quality. This will require operation at lower ^{137}Cs concentrations in the purification cycle to prevent cocrystallization of aluminum oxalate, and more batches will be required for a given throughput. The amount of ^{137}Cs in recycle streams, however, should be much lower, and the processing time thus saved should compensate for the additional incremental batch times.

Strontium-90

Due to the large amount of $^{90}SrTiO_3$ on hand (~3 MCi), conversion of ^{90}Sr to $^{90}SrTiO_3$ for inventory was not required. A program for upgrading $^{90}SrTiO_3$ was initiated. Since much of the present inventory is >3 years old, decay of ^{90}Sr has significantly reduced the power concentration. A large fraction of this stored $^{90}SrTiO_3$ contains excess titanium and can be blended with freshly prepared ^{90}SrO and recalcined to improve the power concentration.

The preparation of ^{90}Sr pellets by hot pressing for use as heat sources was adapted as the standard method. Pellets of various sizes were made, ranging in diameter from 0.25 to 2.86 in. Since the hot-press technique is still relatively new (for in-cell operations), several procedural modifications were made as experience dictated. Procedures have evolved to a point from which reasonably accurate predictions of time, costs, and product quality can be made for specific orders. A summary of the ^{90}Sr pellets prepared to fill customer orders is given in Table 10.

In addition to the 32 pellets described in Table 10, seven others were made. One 85-w platinum-sheathed pellet and one 15-w unsheathed pellet were made for use in a 100-w UKAEA source. The pellets were loaded into a source liner, but the liner was later defueled for metallurgical inspection. The two fuel pellets were replaced by a single 100-w pellet. Two ^{90}SrO pellets (62 w, 1.63 in.) were pressed. One broke during ejection; the other broke while being loaded into the source liner. On the first pressing of a 200-w pellet for a SNAP-21 source, a break in the press collar necessitated an early shutdown of the press, and a poor pellet resulted. The pellet was re-pressed without difficulty. During the other two pressing operations (an "LCG" type and a "RIPPLE" pellet), the hot press failed due to breakage of the graphite die bodies. In both cases it was necessary to install a new hot press. Including test operations, the average operational life for hot presses has been 15 cycles. This can be expected to improve somewhat as more experience is gained with this system.

Table 10. Strontium-90 Pellet Fabrication During 1967

Customer	Pellet dia, in.	No. of pellets	Total ^{90}Sr Watts	Total ^{90}Sr Curies
Martin Company (LCG-25)	2.86	4	742	109,100[a]
3M Company (SNAP-21)	2.70	2	374	55,000[a]
UKAEA (RIPPLE)	2.00	5	304	44,700[a]
UKAEA (RIPPLE)	1.34	2	51	7,500[a]
Mitsubishi Int.	1.63	1	62	9,100[b]
NUMEC	0.76	6	50	7,400[c]
NRDL	0.25	12	1	200[d]
Totals		32	1584	233,000

[a] $^{90}SrTiO_3$, platinum sheathed.
[b] ^{90}SrO, platinum sheathed.
[c] $^{90}SrTiO_3$, unsheathed.
[d] Nine $^{90}SrTiO_3$ and three ^{90}SrO pellets, all unsheathed.

Special Projects

Source Materials

Two $^{137}CsCl$ sources containing rectangular pellets encapsulated in stainless steel were defueled so that the capsules could be inspected. The fuel in one of these sources contained ~50% inert cesium chloride; it was discarded. The other, fully active $^{137}CsCl$ was returned to inventory after sampling.

Three $^{144}CeO_2$ fueled sources used in an experiment for General Motors-Allison Division at ORNL were returned to the FPDL at the end of the experiment. The sources were defueled and the fuel was put into storage. A small fraction of the fuel was used to prepare other sources.

The $^{90}SrTiO_3$ fuel recovered from four 5-year-old SNAP-7A sources was assayed calorimetrically. The power concentration of the fuel agreed exactly with the original loading data corrected for decay. Fractions representing two different batches of this fuel were reserved for experimental work; the remainder was returned to storage.

The fuel pellet from a 200-w experimental ^{90}SrO source prepared in January 1966 was reprocessed to recover the ^{90}Sr. Samples of the platinum protective sheath from this pellet were prepared for metallurgical examination. Several batches of aged ^{90}SrO were reprocessed.

The four SNAP-7A capsules were opened and defueled. There was no evidence of mechanical damage or chemical attack to the capsules. Of the 24 pellets contained in the four capsules, only one was badly broken. Metallurgical samples were prepared from each capsule and transferred to HRLEL for inspection. Samples of the ^{90}SrTiO$_3$ were obtained, and the remainder of the fuel was stored at the FPDL.

The heat output of ^{90}Sr was determined to be 6.86 ± 0.40 w/kCi. Three calorimeter standards were fabricated, calibrated, and delivered to the FPDL.

A separation of ^{238}Pu was made from ^{242}Cm and ^{241}Am with good results. Approximately 240 mg of ^{238}Pu was recovered by ion exchange using Dowex 1 resin (nitrate form). Mass analysis of the product showed it to be 99.59% pure.

Cesium-137 Chloride Pelletization

Two campaigns of ^{137}CsCl pellet fabrication were carried out, both involving 1-in.-wide by 1/4-in. thick pellets. In the first campaign, 196 pellets ~1 in. long were made; the total ^{137}Cs content of these pellets was 100,000 Ci. Since the sources made from these pellets would be loaded to full capacity with cesium chloride, close control of pellet lengths and individual pellet curie contents was not required. The second campaign included pressing of 213,000 Ci of ^{137}CsCl into 708 pellets. In this case the sources would be loaded in definite patterns using spacers and various lengths of pellets. Strict tolerances were thus placed on the pellets, and it was necessary to weigh each individual powder increment before pressing as well as to weigh and measure each finished pellet. Pellets were made of 1-, 3/4-, and 1/2-in. lengths.

Maintenance

A major revision and maintenance program in the manipulator cells was in progress at the beginning of 1967. This program included repair and minor modification of the ^{90}Sr processing equipment in two cells, installation of new processing equipment in the ^{137}CsCl preparation cell, and conversion of an existing process cell to two manipulator cells. The two new manipulator cells are not directly connected with the other FPDL cells. They will be available for use in special projects where ready access and relative ease of equipment installation is an advantage.

The main cell ventilation system was improved by the installation of a new bank of absolute filters in series with the existing system. A new above-ground filter house was constructed. It contains three separate banks of absolute filters which can be individually valved out of the system for filter replacement or control of cell ventilation. The first set of filters to be installed showed filtration efficiencies better than ORNL standards. In addition to the improvement of the main filter system, each of the manipulator cells on which maintenance was done was equipped with improved in-cell filters.

Improvements to equipment included installation of manipulators in the decontamination cell, installation of a separate sampling station for the tank-farm vessels, and addition of a self-contained shield for the by-pass off-gas filter. An existing STT, Model II, was loaded with Decalso ion exchanger and put into service to replace a Model II STT which had been taken out of service due to a suspected leak and a low efficiency of loading.

Radioisotope Sales Report — Calendar Years 1966 and 1967

In 1967, 3955 shipments totaling 3,669,066 Ci were made, an increase of ~40% in total curies shipped. However, nearly 50% of the curies shipped were ^{60}Co and ^{137}Cs. Strontium-90 shipments accounted for another 40% of the total quantity shipped during the year. The balance of radioisotopes shipped decreased markedly this year, since private industry now supplies the bulk of these radioisotopes. The gross income was $2.17 million, a decrease of ~7% compared with 1966.

Sales of stable isotopes and other special research materials amounted to $2.09 million during the year, an increase of ~3% over the 1966 sales.

SALES REPORT — CALENDAR YEARS 1966 AND 1967

Account No.	Isotope	Calendar Year 1966					Calendar Year 1967				
		Sales Proceeds	Fund Cost of Sales	No. of Shipments	Millicuries Shipped	Fund Cost of Production	Sales Proceeds	Fund Cost of Sales	No. of Shipments	Millicuries Shipped	Fund Cost of Production
Inventory Items											
3651	Carbon-14	$ 205,611	$ 124,285	83	47,259	$ 218,248	$ 228,786	$ 154,800	75	62,335	$ 137,702
3652	Cesium-137	23,429	176,060	155	83,148,359	699,762	130,190	1,410,631	124	937,531,249	1,131,123
3653	Cobalt-60	594,341	297,363	22	2,009,948,448	307,270	316,154	306,462	10	1,797,488,000	250,738
3654-104	Promethium-147	56,318	257,712	549	231,527,451	227,918	30,162	94,313	499	113,424,152	116,791
3654-106	Technetium-99	45,208	43,522	34	1,130,727 mg	843	1,599	10,890	20	33,278 mg	390
3654-107	Krypton-85	112,132	93,426	161	5,334,289	19,267	144,447	103,356	181	6,598,054	27,876
3654-288	Chlorine-36	19,762	16,192	64	60	17,607	27,358	21,503	48	78	34,583
3654-327	Nickel-63	23,833	7,085	36	4,135	3,386	39,530	8,944	27	8,325	14,334
3654-365	Thallium-204	3,089	3,398	17	3,948	6,582	3,363	3,428	12	6,122	3,888
3662	Strontium-90	30,939	211,320	111	108,023,189	1,185,302	107,261	1,314,012	57	535,130,441	4,096,873[a]
3663	Tritium	188,554	98,327	201	166,753,560	197,831	256,026	174,409	224	275,107,924	90,456
Total Inventory Items		$1,303,216	$1,328,690	1,433	1,130,727 mg 2,604,790,698	$2,884,016	$1,284,876	$3,602,748	1,277	33,278 mg 3,665,356,680	$5,904,754
Major Products											
3654-117	Xenon-133	$ 41,568	$ 36,582	289	2,332,760		$ 62,151	$ 48,035	446	2,718,226	
3654-140	Iodine-131	15,624	48,794	530	41,499		12,456	10,738	310	16,985	
3661-266	Barium-133	15,058	9,614	43	100		9,205	6,555	24	61	
3661-284	Calcium-47	67,222	38,041	211	1,765		47,982	25,547	190	189	
Total Major Products		$ 139,472	$ 133,031	1,073	2,376,124		$ 131,794	$ 90,875	970	2,735,461	
Minor Fission Products											
3654-100	Barium-140	$ 4,245		51	559		$ 5,924		66	766	
3654-101	Strontium-89	15,566		115	1,148		14,795		76	1,086	
3654-103	Cerium-144	2,655		89	165,695		2,159		82	862,487	
3654-109	Gross Fission Products	444		9	78		300		10	45	
3654-110	Zirconium-95—Niobium-95	6,949		69	1,208		4,826		54	837	
3654-111	Niobium-95	1,276		29	54		1,975		28	79	
3654-112	Praseodymium-143	275		6	6		175		3	4	
3654-113	Neodymium-147	1,625		18	32		1,200		11	24	
3654-114	Ruthenium-103	1,400		22	56		1,275		16	51	
3654-115	Ruthenium-106	8,358		46	326		3,288		35	129	
3654-116	Yttrium-91	8,475		54	5,444		10,342		41	6,661	
3654-120	Molybdenum-99, Fission Product	1,151		14	7,669		9,059		118	57,855	
3654-121	Tellurium-132—Iodine						775		16	150	
Total Minor Fission Products		$ 52,419	$ 60,905	522	182,275		$ 56,093	$ 43,161	556	930,174	
Developmental Fission Products											
3654-105	Europium-155	$ 440		2	22		$ 105		2	5	
3654-108	Iodine-129	6,085		29	6,173 mg		7,110		24	7,873 mg	
Total Developmental Fission Products		$ 6,525	$ 3,538	31	6,173 mg 22		$ 7,215	$ 5,821	26	7,873 mg 5	

Account No.	Isotope	Calendar Year 1966 Sales Proceeds	Calendar Year 1966 Fund Cost of Sales	Calendar Year 1966 No. of Shipments	Calendar Year 1966 Millicuries Shipped	Calendar Year 1967 Sales Proceeds	Calendar Year 1967 Fund Cost of Sales	Calendar Year 1967 No. of Shipments	Calendar Year 1967 Millicuries Shipped
Special AEC Approved Production Services[b]									
3661-102	Cerium-141	$ 32		1	10	$			
3661-160	Iridium-192	6,325		25	586,760	628		17	28,638
3661-194	Cobalt-60 Gamma Service Irradiations	5,149		3	3 S.I.	6,961		5	5 S.I.
3661-210	Phosphorus-32, HSA, Processed	15,406		360	10,952				
3661-252	Antimony-124	282		10	54				
3661-254	Antimony-125	410		2	4				
3661-260	Arsenic-76	1,935		38	190				
3661-272	Bromine-82	1,015		26	258	360		1	3
3661-273	Cadmium-109	612		4	5				
3661-274	Cadmium-115	91		3	12				
3661-276	Cadmium-115m	396		7	12				
3661-278	Calcium-45	580		8	62				
3661-285	Cesium-134	35		4	12				
3661-296	Copper-64	14,452		55	7,106	12,570		38	5,700
3661-302	Gold-198	1,026		25	5,925				
3661-314	Iron-55	300		3	6				
3661-315	Iron-59	3,711		41	123				
3661-317	Lanthanum-140	170		8	44				
3661-321	Mercury-197	930		3	620				
3661-323	Mercury-203	372		15	270				
3661-325	Molybdenum-99	4,566		32	22,200				
3661-333	Potassium-42	4,568		144	1,451				
3661-347	Selenium-75	25		1	10				
3661-348	Silver-110	301		12	104				
3661-356	Sodium-24	4,869		83	372				
3661-358	Strontium-85	1,050		10	21				
3661-360	Sulfur-35	4,223		87	3,877	1,750		3	110
3661-366	Tin-113	392		5	11				
3661-374	Zinc-65	15		2	6				
3661-379	Chromium-51	10		4	5				
3661-381	Selenium-75	799		7	32				
Total AEC Approved Production Services		$ 74,047	$ 56,940	1,028	3 S.I. 640,514	$ 22,269	$ 12,587	64	5 S.I. 34,451
Miscellaneous Developmental Items									
3661-250	Antimony-122, HSA	$ 34		3	3	$ 279		5	31
3661-256	Argon-37, CF	1,655		14	73	2,957		13	77
3661-264	Barium-131, HSA	898		9	9	1,170		9	8
3661-270	Bismuth-210, HSA	595		7	39				
3661-300	Gallium-72, HSA	2,245		39	370	1,901		37	197
3661-305	Iodine-130, HSA					100		4	16
3661-331	Palladium-109, HSA	459		30	300	40		2	12
3661-335	Praseodymium-142, HSA	25		1	–	225		3	30
3661-337	Rhenium-186, HSA	270		10	60	275		9	42
3661-343	Samarium-153, HSA	109		5	29	365		9	62
3661-350	Silver-111, Processed	459		11	62	416		7	31
3661-367	Thulium-170	600		6	171	450		3	1,300
3661-370	Tungsten-187, HSA	1,065		26	118	439		7	31
Total Miscellaneous Developmental Items		$ 8,414	$ 36,679	161	1,234	$ 8,617	$ 17,374	108	1,837

Account No.	Isotopes	Calendar Year 1966				Calendar Year 1967			
		Sales Proceeds	Fund Cost of Sales	No. of Shipments	Millicuries Shipped	Sales Proceeds	Fund Cost of Sales	No. of Shipments	Millicuries Shipped
	Minor Processed Material Items								
3661-295	Cobalt-60, HSA, Processed	$ 2,293		37	699	$ 855		16	200
3661-298	Europium-152—Europium-154, HSA	970		18	60	1,405		18	68
3661-306	Hafnium-181, HSA	837		25	293	924		31	224
3661-308	Indium-114, HSA	1,525		25	179	1,350		29	136
3661-311	Iron-55—Iron-59, HSA	210		2	4				
3661-329	Osmium-191, HSA	265		3	14	40		1	2
3661-339	Ruthenium-86, HSA	4,804		219	6,178	4,510		199	5,582
3661-345	Scandium-46, HSA	1,306		33	1,010	1,664		28	768
3661-363	Tantalum-182, HSA	800		19	133	515		8	51
3661-368	Tungsten-185, HSA	742		13	132	402		7	47
3661-372	Yttrium-90, HSA	1,800		16	513	105		4	14
3661-386	Phosphorus-33					500		2	1
3661-400	Iodine-123					5,150		49	206
Total Minor Processed Material		$ 15,552	$ 46,913	410	9,215	$ 17,420	$ 27,226	392	7,299
Total Radioisotopes		$1,599,645	$1,666,696	4,658	3 S.I. 1,136,900 mg 2,608,000,082	$1,528,284	$3,799,792	3,393	5 S.I. 41,151 mg 3,669,065,907
	Service Irradiations								
3658-190	ORNL 86-Inch Cyclotron	$ 250,365	$ 212,709	155	154 S.I.	$ 164,904	$ 142,563	119	119 S.I.
3658-193	Reactor	82,720	54,462	445	570 S.I.	65,347	47,976	375	389 S.I.
3658-196	Naval Research Laboratory[c]	91,465	87,492	21	7 S.I.	82,072	80,109	24	11 S.I.
Total Service Irradiations[d]		$ 424,550	$ 354,663	621	731 S.I.	$ 312,323	$ 270,648	518	519 S.I.
	Other Services								
3659-203	Special Services	$ 130,509	$ 105,238	16		$ 18,944	$ 15,006	30	
3659-204	Target Preparations — Radioisotopes	1,260	9,864	5		8,585	5,117	3	
3659-205	Cobalt-60 Source Selection	8,669	6,577			7,478	5,350	3	
3659-206	Fission-Product Source Fabrication	14,945	54,435			173,316	101,619	2	
3659-209	Incoming Transportation Charges — Customer Service	71	1,238			1,302	1,485		
3663-521	Tritium Target Fabrication	6,635	12,567	19		9,275	9,054	5	
Total Other Services		$ 162,089	$ 189,919	40		$ 218,900	$ 137,631	43	
Packing and Shipping		$ 145,873	$ 136,115			$ 111,171	$ 80,879		
Isotopes Sales Department			$ 112,218				$ 68,690		
TOTAL		$2,332,157	$2,459,611	5,319	734 S.I. 1,136,900 mg 2,608,000,082	$2,170,678	$4,357,640	3,954	524 S.I. 41,151 mg 3,669,065,907

HSA — High Specific Activity S.I. — Service Irradiations

[a] Includes $3,930,271 cost for 1,105,000 curies of ^{90}Sr transferred to ORNL through OROO from Quehanna.

[b] Represents materials and services for which ORNL has been withdrawn from routine production and distribution. These materials and services are furnished only when materials of customer's specifications are not available commercially.

[c] Fund costs only are billed.

[d] Does not include "Cobalt-60 Gamma Service Irradiations"; these are listed under "Special AEC Approved Production Services."

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