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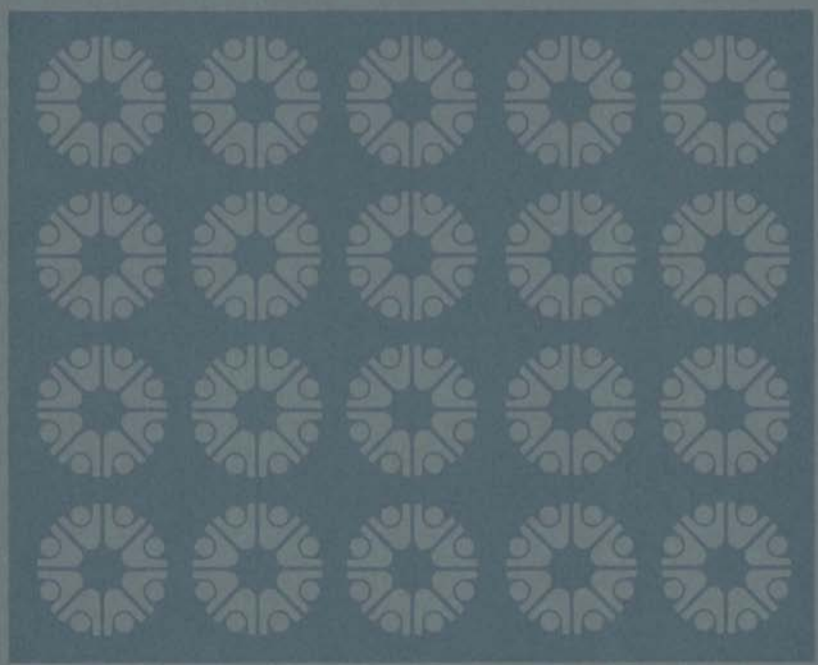
Richland, Washington 99352

AEC Research and Development Report

PACIFIC NORTHWEST LABORATORY DIVISION OF ISOTOPES DEVELOPMENT PROGRAMS QUARTERLY REPORT: FEBRUARY - APRIL 1971

June 1971



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PACIFIC NORTHWEST LABORATORY
DIVISION OF ISOTOPES DEVELOPMENT PROGRAMS
QUARTERLY REPORT: FEBRUARY-APRIL 1971

Compiled by J. E. Hansen

June 1971

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PACIFIC NORTHWEST LABORATORIES
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PACIFIC NORTHWEST LABORATORY
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INTRODUCTION

This report, BNWL-1308-6, is the 18th report of a series initiated in 1966 to provide detailed and timely information on the results of the isotopic fuels program and related research being conducted at the Pacific Northwest Laboratory by Battelle-Northwest for the U.S. Atomic Energy Commission. The previous reports of the series are BNWL-385, 466, 570, 680, 681, 781, 877, 924, 1010, 1085, 1177, 1256, and 1308-1 through 1308-5.

This report covers the three-month period, February through April 1971.

SUMMARY

ENCAPSULATED WASTE MANAGEMENT CESIUM AND STRONTIUM AS PROCESS RADIATION SOURCES

The recently announced closure of the Hanford KE Reactor should have no appreciable effect on the Hanford Waste Management Program (HWMP). ARHCO has begun construction on the encapsulation portion of the Waste Management Plant. Production of fission product SrF_2 may result in complex mixtures containing minor fractions of NaF, CaF_2, BaF_2, and perhaps KF and NaCl. Evaluation of these mixtures is proceeding to determine their possible effect on the melting point and compatibility of the Hanford Waste Management products.

UTILIZATION OF FISSION PRODUCT RHODIUM, PALLADIUM, RUTHENIUM AND TECHNETIUM

Determination of the half-life of ^{102m}Rh has been completed. The dose rates from fission product palladium are being determined by the use of a palladium foil representing an infinitely thick slab. Investigation of future supply and demand for the platinum metals is continuing. The effects of radiological hazards on the marketability of reactor produced platinum metals is being investigated. Calculations with respect to the potential hazards from ^{107}Pd indicates that the radiological hazard may not be biologically significant, neither in terms of population dose nor individual dose.

EVALUATION OF LARGE SCALE USES FOR PURIFIED FISSION PRODUCT XENON

The stock of purified fission product xenon at ORNL is being further processed to remove major impurities, including

moisture, oxygen, and carbon dioxide. Additional expressions of interest on the availability of the xenon have been obtained.

ISOTOPE PRODUCTION EVALUATIONS

A series of parametric market evolution cases is being prepared to simulate various possible ways that ^{137}Cs and ^{90}Sr markets might develop. A series of computer programs has been developed to carry out these calculations. Final results are being developed in terms of price requirements for profitable recovery of cesium and strontium from spent power reactor fuels. A revised version of the NUCLIDE TABLES is being prepared.

PROMETHIUM-146 BURNOUT

Irradiation conditions and target matrix requirements to produce promethium containing less than 0.1 ppm have been investigated. In addition to detailed examination of literature related to γ,n and n,2n reactions in neptunium and promethium, a major basis for determining the irradiation conditions and target matrix requirements has been obtained from Savannah River and Hanford irradiations of ^{237}Np to produce ^{238}Pu containing low levels of ^{236}Pu. Computations have been started to determine the maximum high energy gamma and/or neutron flux that can be allowed to achieve a desired ^{146}Pm burnout of about 80%. Conditions necessary to achieve 80% ^{146}Pm burnout, or a ^{146}Pm level of 0.05 ppm are reported.

NEUTRON RADIOGRAPHY USING ^{252}CF

Development of a proportional counter for imaging of thermal neutrons has progressed to the evaluation stage. Other techniques for neutron imaging with counters have been investigated, including other types of position-sensitive proportional counters, solid state detectors, and channeltron devices.

Advanced concepts for a portable shield have been developed and a simple facility has been constructed for testing shielding materials in beam extraction geometries.

ADVANCED SOURCE DEVELOPMENT

Four of the ten alpha-photon radiation sources prepared in the previous quarter were tested at ORNL. The test results are yet to be formally reported but preliminary results are encouraging. In most tests the sources performed better than previous PNL alpha-photon sources. Some areas for improvement are still indicated. Test results will be used to improve the next generation of alpha-photon sources. A new tuning network was procured for the glovebox sputtering system. It will be installed in May, and should result in a significant increase in experimental efficiency.

CIRCULATORY SUPPORT SYSTEMS

Measurements of the neutron dose rates in a large tissue fluid phantom were completed using ^{252}Cf as a substitute for $^{238}PuO_2^{16}$ in a Rankine power source prototype. Similar measurements are near completion for a Remab phantom.

Neutron and photon dose rates were calculated for a 30-W $^{238}PuO_2^{16}$ source placed in the Rankine device. When adjustment is made for similar tissue thickness and emission rates, the calculations agree well with those previously made for LASL-1, a 30-W, ^{238}Pu metal source.

Engineering studies to develop radioisotopic blood heaters in support of PNL's "Biological Effects of Intracorporeal Radioisotope Heat Source" study were initiated.

NUCLEAR TECHNIQUES FOR SEABED MINERAL EXPLORATION

Variations in detection efficiency for elements in substrates of various densities, cross sections, and over burdens are being investigated. A subcritical multiplier for ^{252}Cf neutrons was evaluated for seabed mineral analysis. An evaluation of the primary gamma rays from molybdenum ores in seabed was completed. The prototype designs of the nuclear probe underwater assembly and the electronics packages for data accumulation and analysis have been finalized.

ENCAPSULATED WASTE MANAGEMENT CESIUM AND STRONTIUM AS PROCESS RADIATION SOURCES

H. H. Van Tuyl and W. E. Keder

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

The goal in FY-1971 is to develop equipment for fabricating smaller diameter sources in the HWMP facility. The incremental cost of producing smaller sources will be evaluated. Prototype sources will be fabricated with nonradioactive feeds, including loading capsules, welding, and NDT. The sources will be tested to determine mechanical integrity. Potential customers will be contacted to stimulate interest in the HWMP sources.

The recently announced closure of the Hanford KE Reactor should have no appreciable effect on the Hanford Waste Management Program (HWMP). Accumulation of cesium will continue as a partially purified crude. Ion exchange purification equipment for cesium is being installed in B-Plant, and final purification of cesium is expected to start in FY-1972. All of the required strontium purification is being done currently, but flowsheets are still being changed occasionally to obtain higher purities, better recovery, or lower operating cost. Good product purity can be achieved even with existing technology. One of the strontium product tanks contains about 12 MCi of strontium of 95% chemical purity. The ^{90}Sr content is 53.7 wt% based on total cation content. This material is better than any solvent extraction material previously shipped to ORNL.

Construction has been started by ARHCO on the encapsulation portion of the Waste Management plant. The facility is expected to be ready for cold testing in CY-1973, and for full hot operation in January 1974. At the present processing rate, the waste storage tanks will have been completely processed in about 1976. A few additional years will probably be required for conversion to solids and encapsulation.

Production of fission product SrF_2 may result in complex mixtures containing minor fractions of NaF, CaF_2, BaF_2, and perhaps KF and NaCl. Decay of the strontium isotopes will introduce significant quantities of zirconium into the mixture. The behavior of the solid material will depend upon solid solution formation which tends to lower freezing point a reasonable amount; eutectic formation, which usually results in a much greater freezing point depression; and compound formation. In addition, actual chemical reaction may be important in the mixture.

BaF_2 forms solid solutions with both CaF_2 and SrF_2, which have broad, shallow-freezing point minima at equimolar concentrations (Reference 1, Vol. I, p. 128, 134). One would estimate that CaF_2 and SrF_2 would form solid solutions together and that perhaps the freezing point would be slightly less than for pure SrF_2. On the other hand, SrF_2-$SrCl_2$ form two eutectics: 14 and 58% SrF_2 which freeze at 763 and 944 °C, respectively, with a maximum of 960 °C between these compositions at 50% SrF_2. NaF-SrF_2 form a eutectic mixture also, at 50 mole % SrF_2, which melts at 856 °C. That is to say, introduction of a small amount of NaF into a SrF_2 sample will lower the freezing point about 550 from 1400 °C. As a SrF_2 rich SrF_2-NaF melt cools, pure SrF_2 should crystallize out until at 856 °C the equimolar eutectic solidifies as a separate phase. This should be the case no matter what the proportion of the original mixture. For example at 1000 °C the melt should have a ratio $2SrF_2$ to 1NaF.

In its simplest form the encapsulated SrF_2 would consist of strontium fluoride, zirconium daughter, and a certain amount of impurity sodium fluoride. The introduction of daughter zirconium into the SrF_2 system produces significant complexity. The resulting zirconium compound of course has the stoichiometry ZrF_2. One would expect the presence of ZrF_2 in SrF_2 to produce phase behavior similar to mixtures of other divalent fluorides. If a series of solid solutions is formed, this would probably mean a maximum freezing point depression of only a few hundred degrees. In the absence of SrF_2-ZrF_2 solid solutions, a eutectic with a much lower freezing point might be obtained.

ZrF_2 has been reported and its properties are said to be those of group IV-A difluorides.[2] It appears to disproportionate however. The indication here is that one could expect a change of behavior when a sample of old encapsulated SrF_2 was brought to 800 °C for the first time. Probably this would mean a decrease in melting point as the ZrF_4 is formed and makes eutectic mixtures with other fluorides. Note too that ZrF_2 will oxidize easily if there is any oxidizing agent present.

The chemistry of the fluorides of zirconium is of interest in the effort to estimate the Zr species which will control the phase behavior of aged SrF_2. ZrX_2 and ZrX_3 compounds have been reported for all the halides, but the fluorides are less well described than the others. Zirconium metal will reduce $ZrCl_4$ to $ZrCl_3$ at 450 °C and to $ZrCl_2$ at 675 °C. ZrF_3 is reported to exist, and ZrF_4 has been reduced at 350 °C by atomic hydrogen to ZrF_2. The latter compound disproportionates at 800 °C. This would indicate that daughter zirconium would exist as ZrF_2 in the beginning and as a mixture of metal and ZrF_4 if the material in the capsules got hot enough.

The phase behavior might change significantly above a temperature at which disproportionation of ZrF_2 takes place. ZrF_4 itself melts at about 920 °C, which is significantly lower

than what one would estimate for ZrF_2. It forms a series of compounds with alkali[3] and (presumably) alkaline earth fluorides, which form eutectics with each other, some melting as low as 500 °C. The phase diagrams of ZrF_4 with the alkali halides would indicate, however, that at lower ZrF_4 concentrations the lowest melting point mixture would probably be a eutectic with the compound M_3ZrF_7 or a compound containing whatever cations are available (perhaps Sr^{+2}). Such a eutectic could melt below 700 °C, and if there were several different cations, maybe well beyond that.

A careful analyses for alkali halides should be made and the effect of ZrF_2 should be worked out. The reactivity of this material with the containers is also worth checking, and the behavior (solubility) of zirconium metal itself would be worth knowing. The presence of alkali fluorides is the most serious problem in the effort to hold the melting point up, however.

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UTILIZATION OF FISSION PRODUCT RHODIUM, PALLADIUM, RUTHENIUM, AND TECHNETIUM

The traditional view of fission products as waste materials has been supplanted by their role as a national resource. Pursuant to this revised status, a resource strategy is now being formulated to direct optimum technological and economic application of the fission products Rh, Pd, Ru, and Tc created in operation of nuclear power reactors. The program seeks to evaluate the potential of these artifacts as supplements to the natural reserve of precious metals, and emphasizes the practical aspects of their safe use. The program comprises the following tasks:

1. *Evaluation of rhodium and palladium produced in power reactors.*
2. *Technology for recovery of Rh, Pd, Tc, and Ru.*
3. *Market analysis of Rh, Pd, Tc, and Ru.*
4. *Economic analysis of Rh, Pd, Tc, and Ru production from nuclear wastes.*
5. *Fabrication process development for nuclear source (Rh, Pd, Tc, and Ru) products.*

^{102m}Rh HALF-LIFE

F. P. Roberts

The determination of the half-life of ^{102m}Rh has been completed, and a final report is being prepared.

RADIATION CHARACTERISTICS

F. P. Roberts

Dose Rates from Fission Product Palladium

The ^{107}Pd in fission product palladium decays with the emission of a 0.035 MeV beta and no gamma. Because of the very low energy of the beta only those atoms decaying at or near the surface contribute to the dose rate. Because only a limited amount of fission product palladium is available the actual measurement of the dose rate is being carried out using a foil

prepared by electrodepositing 0.5 grams of highly purified fission product palladium on a 3 inch by 3 inch sheet of nickel. This amount of palladium results in a thickness of 8.4 mg/cm^2 which greatly exceeds the range of the ^{107}Pd beta, and the foil can therefore be treated as if it were an infinitely thick slab of palladium for the dose rate measurement. The dose rate will be determined in an extrapolation chamber both with and without a 7 mg/cm^2 window. The window is the same thickness as the average thickness of the human skin. Both the total dose and the dose through the skin can be ascertained in this way.

MARKET ANALYSIS FOR Rh, Pd, Tc and Ru

J. C. Sheppard

Supply of Platinum Metals

Examination of the current literature indicates that the major sources of the platinum metals are well defined. Study of the "Economic Geology of the Platinum Metals" by Mertic indicates that the prospects of a major, high grade platinum metal find, comparable to the Mirensky Reef in South Africa, are unlikely. In this connection, it is interesting to note that a recent discovery in the Rudall River area of West Australia by the North West Oil and Mineral Company amounts to less than ten percent of the world's known platinum metals reserves. Future discoveries must approach the presently known reserves to be truly significant.

Analysis of the demand for platinum metals using the Lotka-Volterra logistic growth curve indicates that there may be a world shortage of platinum metals in the period beyond 1980. Further analysis suggests a shortage of palladium, but the situation with respect to rhodium and ruthenium requires more study. Examination of "Mineral FActs and Problems, 1969" indicates general agreement with the above analysis.

The amount of palladium, rhodium, and ruthenium that will be available through production in power reactors will depend greatly on the timing and numbers of Liquid Metal Fast Breeder Reactor, because light water moderated reactors produce an order of magnitude less platinum metals than a reactor using plutonium as the fuel.

The only new large use of platinum metals might be the use of platinum in catalytic air pollution control devices. Both the major producers of platinum metals (Johnson-Mathey and Engelhard Industries) expect that platinum (and possibly the other platinum metals) will figure prominently in the solution of the air pollution problem. In the case of palladium, changing electrical switch technology due to the increasing use of electronic switches may significantly alter the future demand for this metal. The demand for rhodium is expected to increase due to its use as an alloying agent in various catalysts in the production of nitric acid and in petroleum reforming. The situation with respect to ruthenium requires more study.

Dispersion and Radiological Hazards

The marketability of reactor-produced platinum metals will be effected by the hazards associated with the small amounts of radioactivities which are present. Part of this study is the evaluation of these hazards both from the standpoint of the user and the potential for dispersion of unsafe amounts of the radioactivities into the environment.

Calculations with respect to the potential hazard from ^{107}Pd (7×10^6 year half-life and 0.035 MeV beta) indicates that the radiological hazard may not be biologically significant, both in terms of population dose and individual dose. This is because of the chemical inertness of palladium metal and the very low beta energy which leads to considerable self-shielding

and absorption in biologically insignificant regions such as the skin. Tests by Engelhard Minerals and Chemical Corporation (briefly reported in the "Minerals Year Book of 1969") are in accord with this conclusion. It is in disagreement, however, with that of Newman and Smith of Johnson Mathey who claim possible skin damage from absorption of the 0.035 MeV beta of ^{107}Pd. More study is required here also. The situation with respect to reactor produced rhodium and ruthenium are being studied at this time and no conclusions have been made.

EVALUATION OF LARGE SCALE USES FOR PURIFIED FISSION PRODUCT XENON

C. A. Rohrmann

Of all the fission products, xenon has the highest fission yield. Although many of the xenon isotopes are radioactive, they all have such short half-lives that their activity would be insignificant after allowing time for normal fuel reprocessing.

In the past, the extreme rarity of xenon in nature has clearly inhibited its consideration for specific large scale uses even where its properties give it definite and unique advantages. But now, xenon could be made available in surprisingly large quantities. Perhaps the most important and beneficial use of xenon would be for human surgical anesthesia. Another of the potential uses is as a filler gas in incandescent light bulbs with significant increase in both brightness and efficiency.

The scope of the PNL program is to: (1) establish the quality of xenon which can be accepted or generally licensed for consumer uses, (2) determine what industries would be interested in evaluating fission product xenon for large scale applications, (3) distribute specimens to participating evaluators, (4) compile evaluation results, and (5) prepare a final report on the results of this total effort.

Further clean up of the stock of purified fission product xenon at ORNL has been authorized. Although the ^{85}Kr content is reported to be less than one microcurie per liter of xenon (STP), the other major impurities which are to be reduced include oxygen, carbon dioxide, and moisture. The further purification also includes analysis of the gas.

The report on fission product xenon, its properties, applications, and potential availability appeared as the lead article in the Vol. 8, No. 3, 1971, issue of Isotopes and Radiation Technology.

Contacts on this project continue to generate responses of interest. In this period, representatives of a major producer of atmospheric xenon visited and expressed great interest in the project and suggested consideration of their facilities for additional purification, if required, for specific uses. Interest by NASA personnel has also developed in view of their rather impressive requirements in the space power Brayton cycle system. Interest for medical and electronic applications has also continued via inquiries during this period.

ISOTOPE PRODUCTION EVALUATIONS

R. W. McKee, B. M. Cole, R. G. Rau and W. W. Waddel

This program is designed to evaluate major aspects of isotope production. Until recently, the principal source of isotopes has been the AEC production reactors. Now, however, power reactors are rapidly becoming important sources of isotopes. Power reactors also offer attractive prospects for target irradiations. The future availability and cost of these unique isotopes depend on full and efficient utilization of the production potential of power reactors, and the inherent value of these materials can have a significant impact on nuclear power costs.

The scope of this program includes analysis of fast and thermal reactor fuel cycles within the framework of an expanding nuclear power industry to identify the sources, quantities, timing and cost of production of isotopes of potential importance.

A new task to develop an Isotope Data System was initiated in FY-1971. The objective is a computerized, quick-response system for answering queries on isotope properties, availability, applications, shielding requirements, decay compositions, toxicity, and other factors.

FISSION PRODUCT PROFITABILITY STUDIES

A series of parametric market evolution cases are being prepared to simulate various possible ways that ^{137}Cs and ^{90}Sr markets might develop. The markets might be supplied either through sales of fission products recovered in the Hanford waste management program or through sales of fission products recovered from processing of spent power reactor fuels in a private facility.

A series of computer programs have been developed to carry out these calculations. One code called RASOR, computes a replenishment schedule for either a one- or two-component radiation source material. Certain constraints can be specified.

The other code, called RAMARK, combines the results from RASOR with one of several optional market projection equations. For a base market defined in terms of any given radiation source, the code apportions the market among up to three source materials and identifies the separate market requirements for each one. These programs are combined with the CAESR program (described in a previous quarterly) so that the economics of various market situations can be examined.

The final results are being developed in terms of price requirements for profitable recovery of ^{137}Cs and ^{90}Sr from spent power reactor fuels. The objective is to identify the time and circumstance when ^{137}Cs and ^{90}Sr can be recovered in private facilities and marketed at a price that would be competitive with other alternatives.

NUCLIDE TABLES

The Nuclide Data File has been completely revised and updated and work is progressing towards publication of a revised version of the "Nuclide Tables". This work has involved preparation of approximately 15,000 punched cards to accommodate the revisions. Among the more important additions to the file are: experimental gamma photon data in addition to the decay scheme transitions, identification of daughter nuclides for all radioactive decays, and references for the sources of data.

PROMETHIUM-146 BURNOUT

J. C. Sheppard

The objective of the ^{146}Pm burnout program is to estimate by computation the extent and technical feasibility of significantly reducing the ^{146}Pm content in promethium heat source material by neutron irradiation. The goal will be to establish irradiation conditions and target matrix necessary to produce promethium containing less than 0.1 ppm.

Much of this quarter's effort was devoted to the examination of literature related to γ,**n** and n,2n reactions in neptunium and promethium. Data for n,2n reaction in the rare earth region indicate that $^{147}Pm(n,2n)^{146}Pm$ reaction has a cross section of about 5 mb for neutrons greater than 7.7 MeV. This cross section is too crude to make an estimate of the fission spectrum weighted cross section. However, it is comparable to the 15 mb value observed for the $^{236}Np(n,2n)^{236}Np$ reactions.

Information on the $^{147}Pm(\gamma,n)^{146}Pm$ is slightly better. In the rare earth region the giant resonance for γ,n reaction occurs near 14 MeV, has a width at half maximum of about 5 MeV, and a maximum cross section of 400 mb. From these data the $^{147}Pm(\gamma,n)^{146}Pm$ cross section is estimated to be about 5 mb for gammas greater than 7.7 MeV. The uncertainty in this estimate is how rapidly the cross section changes with increasing gamma energy.

These estimates of the cross sections provide part of the data required to estimate the extent of ^{146}Pm burnout in the presence of ^{147}Pm. Computer computations using these cross sections to estimate optimum irradiation conditions are in the computational section of this report.

Considerable data from Savannah River and Hanford irradiations of ^{237}Np to produce ^{238}Pu containing low levels of ^{236}Pu are available. Both Hanford and Savannah River have produced ^{238}Pu containing 0.15 ppm ^{236}Pu. This reduction of the ^{236}Pu level by a factor of seven or eight was achieved largely by the proper choice of target matrix (graphite or zirconium) to minimize the high energy gamma flux within the target and by location of the target in a reactor position that reduced contributions from fast neutrons and high energy gammas to a minimum.

The approach above is directly applicable to the ^{146}Pm burnout experiments because the $^{147}Pm(\gamma,n)^{146}Pm$ and $^{147}Pm(n,2n)^{146}Pm$ contribute significantly to the production of ^{146}Pm. Further examination of Hanford and Savannah ^{238}Pu data revealed the following:

1) Structural parts (walls) of the Savannah River reactors make large contributions to the production of ^{236}Pu via the $^{237}Np(\gamma,n)^{236}Np$ reaction. These high energy gammas (>8 MeV) are produced by n,γ reactions on iron, chromium and nickel. This gamma flux decreases linearly with distance from the reactor wall. This suggests the obvious approach of performing irradiations as far from the walls as possible. Savannah River has done this!
2) As mentioned above, it is very important to use a target matrix that doesn't produce high energy gammas. Savannah River has shown that, in addition to zirconium and carbon, magnesium should be a good target matrix. Savannah River has also shown that when aluminum must be used the amount should be kept to a minimum. Savannah River showed that contribution of ^{236}Pu from the aluminum matrix was almost proportional to the amount of aluminum present - a 38% reduction in the amount of aluminum present resulted in a 33% reduction in the ^{236}Pu level. These data explain

in part the failure to significantly reduce the ^{146}Pm level in the Hanford and Savannah and Hanford ^{146}Pm burnout experiments - the capture gammas (7.72 MeV) from aluminum contributed significantly to the $^{147}Pm(\gamma,n)^{147}Pm$ reaction and could have been reduced by a reduction in the amount of aluminum present.

3) Savannah River data indicate that the γ,n reaction contributes 90% of the ^{236}Pu under optimum irradiation conditions. This is probably the case for the ^{146}Pm also.
4) Iron, nickel, and chromium impurities in aluminum cladding can increase the ^{146}Pm and ^{236}Pu levels by about ten percent.
5) Savannah River has considered bismuth as target shielding material. Use of 1 inch of bismuth (and presumably lead) would reduce the production of ^{236}Pu (and ^{146}Pm) from external (wall, etc.) sources by one fourth.

There is a direct correspondence between the above observations and the ^{146}Pm burnout experiments because the contaminants come from γ,n and n,2n reactions which differ only in the reaction thresholds and cross sections. Rough calculations indicate that irradiation conditions that produce one ppm ^{236}Pu in ^{238}Pu should produce promethium containing <0.5 ppm ^{146}Pm on a weight basis (<0.25 ppm on a disintegration basis). Since Savannah River has developed extensive data on optimum ^{238}Pu irradiations, the promethium data were normalized to the 1 ppm ^{238}Pu data to demonstrate the importance of the various contributions (distance, target matrix, n,2n; γ,n, etc). The results of this normalization are shown in Table 1 and Figure 1.

Computations have been started to determine the maximum high energy gamma and/or neutron flux that can be allowed to achieve a desired ^{146}Pm burnout of about 80%.

TABLE 1A. ppm ^{146}Pm in Promethium-Aluminum Matrix

(normalized to Savannah River Data)

Distance from Reactor Center, cm		163	210	222	235
$^{147}Pm(n,2n)^{146}Pm$		0.03	0.01	0.006	0.003
$^{147}Pm(\gamma,n)^{146}Pm$	external	0.02	0.03	0.05	0.18
$^{147}Pm(\gamma,n)^{146}Pm$	internal	0.07	0.07	0.07	0.07
	TOTAL	0.12	0.11	0.13	0.25

TABLE 1B. ppm ^{146}Pm in Promethium-Graphite (or Zirconium, or Magnesium) Matrix

Distance from Reactor Center, cm		163	210	222	235
$^{147}Pm(n,2n)^{146}Pm$		0.03	0.01	0.006	0.003
$^{147}Pm(\gamma,n)^{146}Pm$	external	0.02	0.03	0.05	0.18
$^{147}Pm(\gamma,n)^{146}Pm$	internal	0.01	0.01	0.01	0.01
	TOTAL	0.06	0.05	0.07	0.19

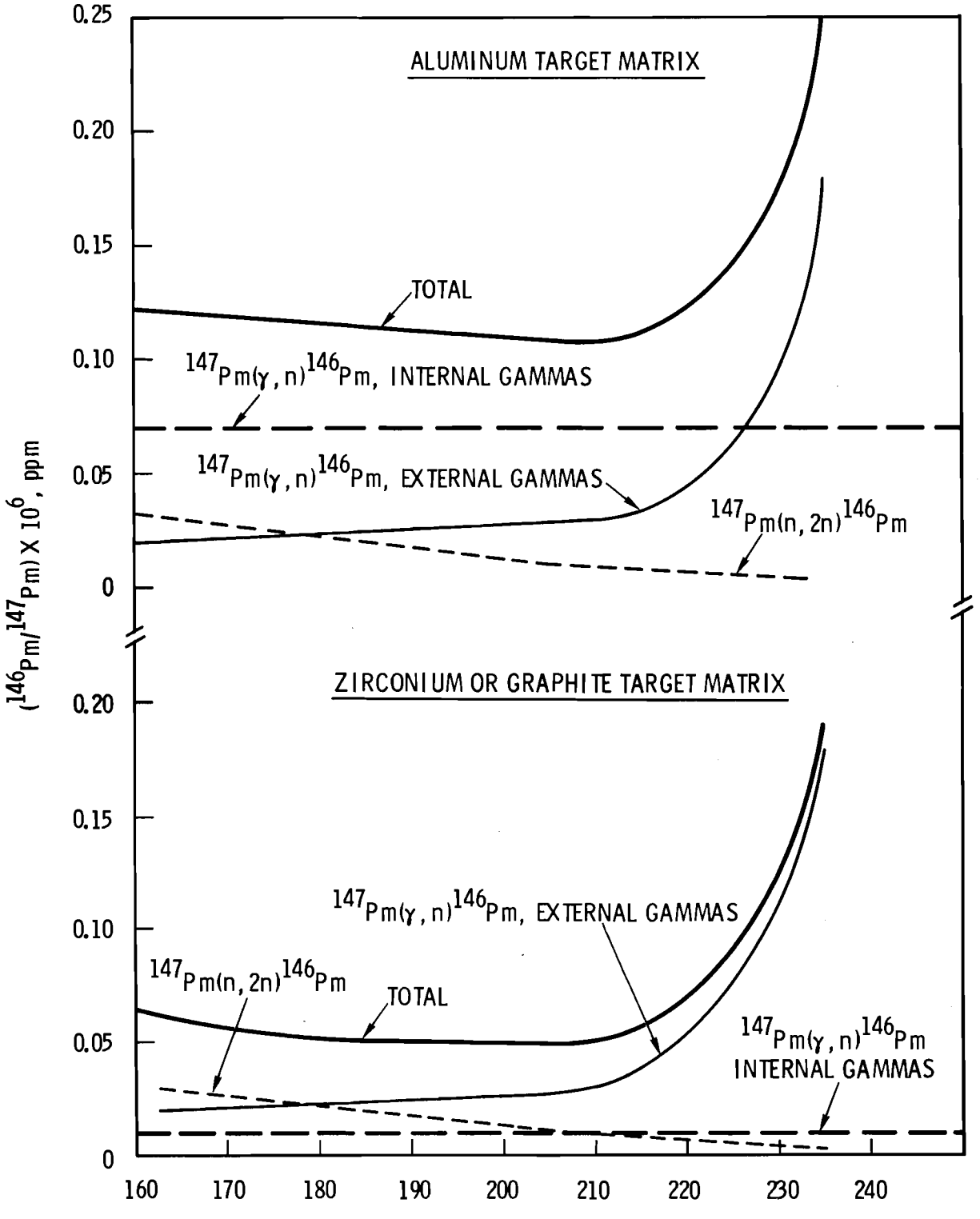

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FIGURE 1. Target Distance from Reactor Axis Versus ^{146}Pm Content

Calculations were made as a function of an assumed back-reaction rate, the ^{146}Pm neutron cross section, and time. It was also assumed for simplicity that only the γ,n reaction contributed to the back reaction. Since Savannah River has shown that the γ,n reaction dominates for conditions necessary to achieve low levels of ^{236}Pu contamination in ^{238}Pu (and presumably also for promethium containing low levels of ^{146}Pm), this assumption is considered valid. These calculations were made for an initial ^{146}Pm level of 0.25 ppm. Table 2 reports the conditions necessary to achieve 80% ^{146}Pm burnout or a ^{146}Pm level of 0.05 ppm.

TABLE 2. Limiting High-Energy Gamma Flux ($E\gamma$ > 7.7 MeV) for the Production of 0.05 ppm ^{146}Pm

Neutron flux, ϕ, n/sec/cm^2	Cross Section,[a] barns	Gamma flux, $\phi\gamma$,[b] γ/sec/cm^2	Percent ^{147}Pm Burnout $\phi\gamma$[c]	Percent ^{147}Pm Burnout $\phi\gamma=0.01\phi$[d]
4×10^{14}	2000	1.1×10^{13}	42	28
	3500	2.2×10^{13}	28	15
	5100	3.2×10^{13}	20	10
	6700	4.5×10^{13}	18	<10
	8400	6.4×10^{13}	18	<10
2×10^{14}	2000	5.6×10^{12}	45	33
	3500	1.1×10^{13}	30	17
	5100	1.6×10^{13}	21	11
	6700	2.2×10^{13}	20	<10
	8400	3.2×10^{13}	20	<10
10^{14}	2000	2.8×10^{12}	51	35
	3500	5.6×10^{12}	33	20
	5100	8.0×10^{12}	24	13
	6700	1.1×10^{13}	21	<10
	8400	1.6×10^{13}	21	<10
5×10^{13}	2000	1.4×10^{12}	56	39
	3500	2.8×10^{12}	39	23
	5100	4.0×10^{12}	28	15
	6700	5.6×10^{12}	23	<10
	8400	8.0×10^{12}	22	<10

a. Measured values of the ^{146}Pm n,γ cross section have a range of 3500 to 8400 barns.

b. The limiting gamma flux for conditions necessary to achieve greater than eighty percent ^{146}Pm burnout with a given ^{146}Pm n,γ cross section and a 5 mb γ,n cross section for ^{147}Pm.

c. The maximum ^{147}Pm burnout expected for conditions stated in (b).

d. The maximum ^{147}Pm burnout expected for conditions where $\phi\gamma(E\gamma > 7.7\ MeV)$ is less than 0.01ϕ.

Careful selection of target matrix and reactor position are expected to result in conditions where $\phi\gamma(E\gamma > 7.7$ MeV) will be less than 0.01ϕ. The Savannah River and Hanford ^{238}Pu experiments have demonstrated that this is possible. Examination of the table above shows that the limiting gamma flux is greater than experimentally attainable conditions, even for the 2000 barn condition, indicating that it is quite feasible to produce promethium containing less than 0.1 ppm ^{146}Pm. Under optimum conditions these data also indicate that 10 to 20% of the initial promethium will be destroyed by neutron capture and decay.

NEUTRON RADIOGRAPHY USING ^{252}Cf

K. L. Swinth

The objective of this program is to develop and investigate low-flux neutron radiography techniques and applications using ^{252}Cf. Previous efforts have led to the development of a portable shield and exposure facility for ^{252}Cf thermal neutron radiography. Evaluation of this facility is underway and will lead to recommendations for future designs. Applications of ^{252}Cf neutron radiography will be identified and efforts will be extended to fast neutron radiography.

The major effort in the program is development of techniques for low-flux imaging. A position-sensitive proportional counter is under development that provides for instantaneous readout of data representing the imaged object. Various converter foils, such as Eu and ^{6}LiF thermoluminescent foils, are under investigation, as a means of radiographically recording the images.

IMAGING TECHNIQUES

Development of a proportional counter for imaging thermal neutrons has progressed to the evaluation stage. Availability and cost of converter and collimation materials necessitated some compromises in the design; however, the counter will serve its purpose of evaluating the technique of counter imaging. Figure 2 illustrates the operation of the counter. A converter foil converts the thermal neutrons to alpha particles which are collimated and then detected in a position-sensitive proportional counter. Figure 3 is a photograph showing the interior of the counter which is encased in a sealed chamber during operation. Gas pressure, degree of collimation, converter foils, and anode-cathode spacing can be varied in this counter to determine the optimum counter configuration. After

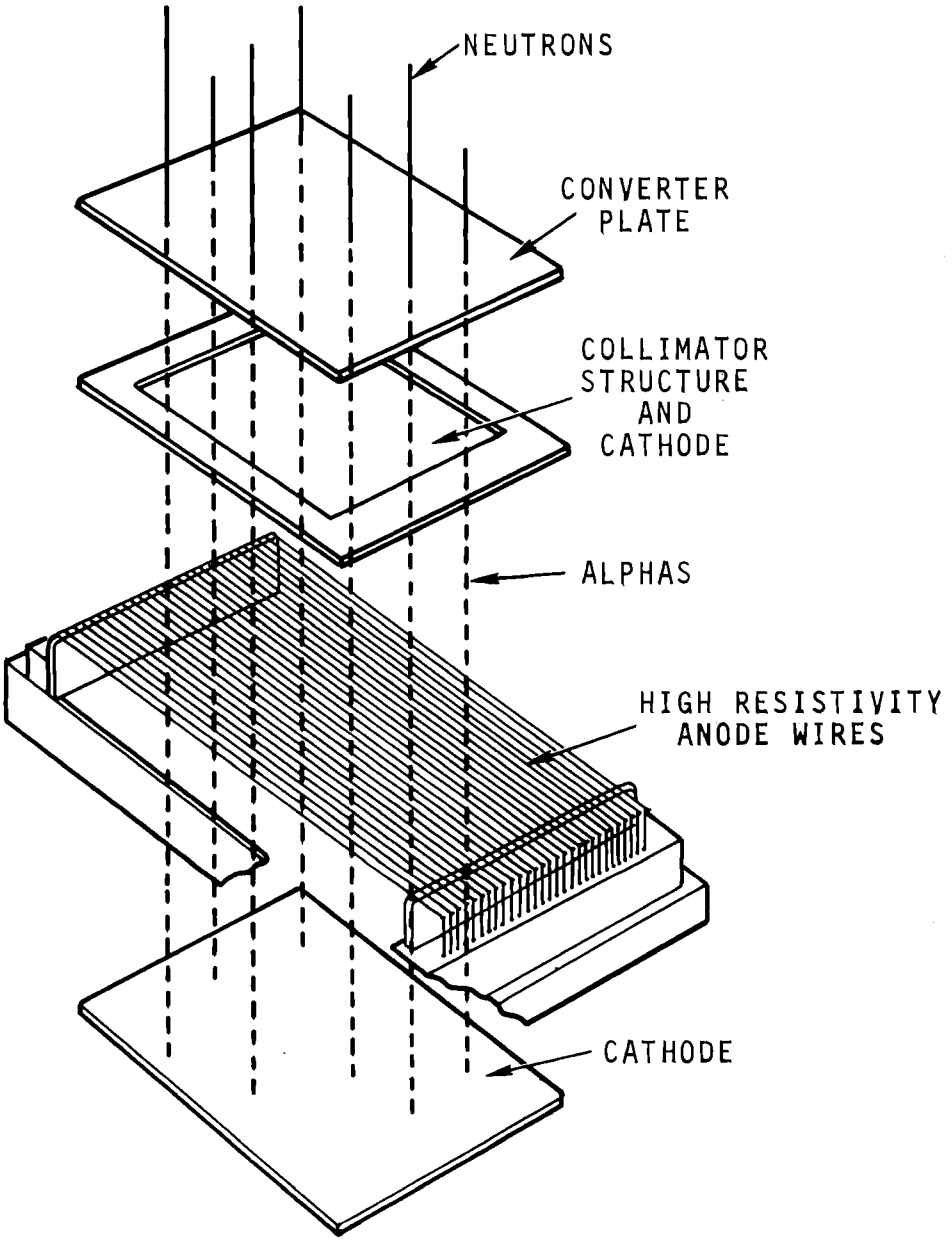


FIGURE 2. Sketch Illustrating the Operation of the Two-Dimensional Neutron Imaging Counter

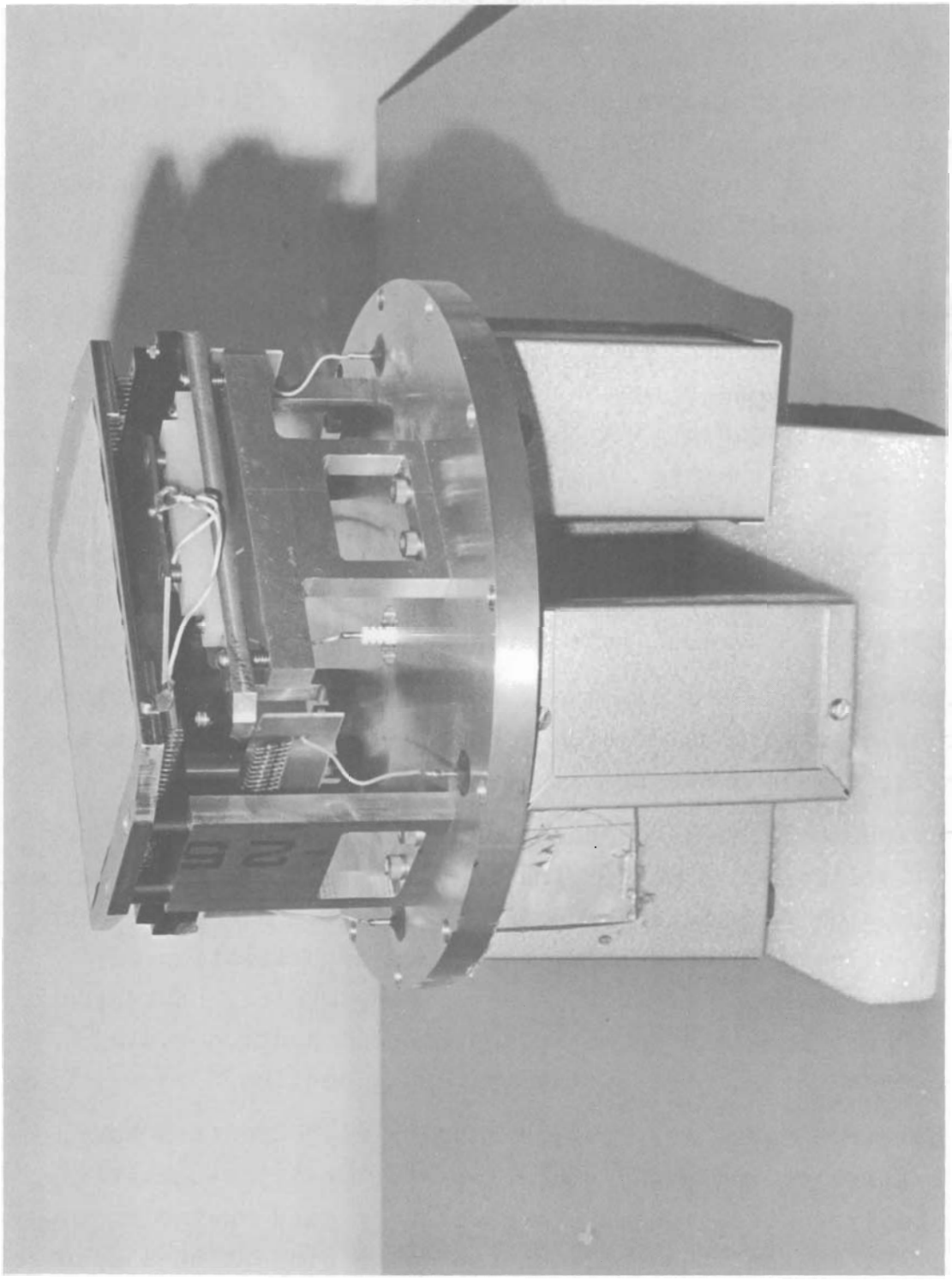


FIGURE 3. Photograph Showing the Interior of the Imaging Counter

optimization of counter parameters, a much simpler counter could be built using circuit board fabrication techniques.

While the large imaging counter was being completed a small one-dimensional counter was fabricated for testing of electronics. Figure 4 shows the spatial distribution of alpha interactions from a collimated ^{241}Am source with this counter. The measured resolution was 4 mm full width at half-maximum (FWHM) which agreed with the results predicted from geometrical consideration of the collimator and counter geometry. Later tests with a smaller hole collimator yielded a resolution of 2.6 mm FWHM. This is a further indication that the present resolution is limited by the test source and not by the electronics. Tests with this counter have reaffirmed the prediction that the use of a gadolinium converter will not work. The gas gain needed to detect the 70 keV electrons from gadolinium makes the counter highly sensitive to gamma radiation and results in background interference.

Figure 5 shows a preliminary test with the two-dimensional counter using a collimated alpha source. Resolution measurements using this counter are presently in progress.

Previous efforts have shown that europium foils offer an excellent medium for transfer imaging with low intensity sources; however, the difficulty of handling such foils, makes the technique expensive. To circumvent the rapid deterioration of these foils, a simple dry box is being constructed. Europium will also be a good candidate for epithermal neutron radiography because of its high epithermal cross section.

Other techniques for neutron imaging with counters have been investigated including other types of position-sensitive proportional counters, solid-state detectors and channeltron devices. At present the position-sensitive proportional counter is the most promising technique due to the limited areas available with other counters.

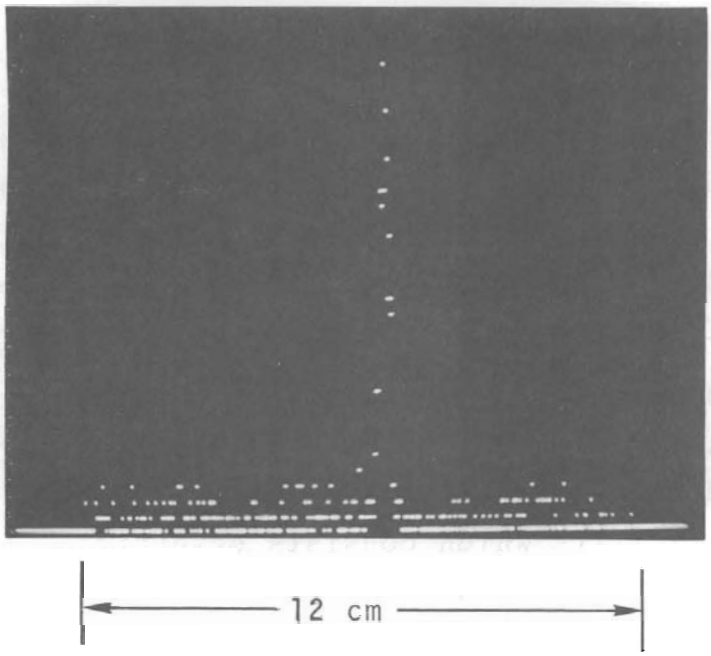
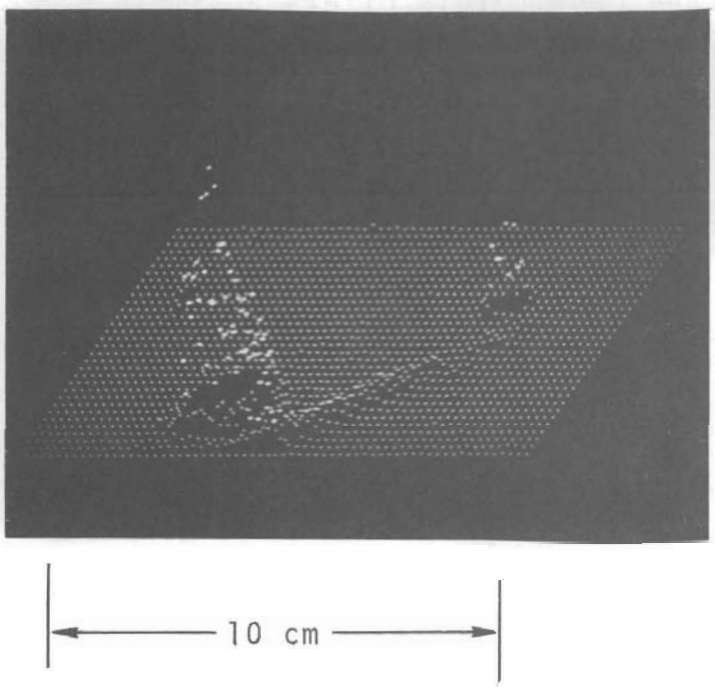
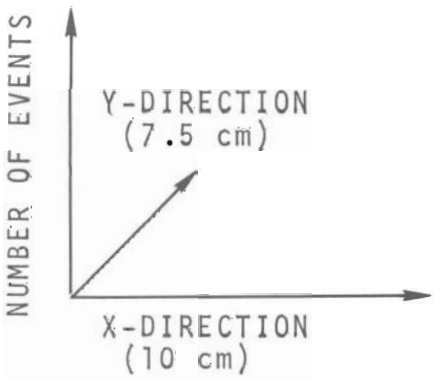


FIGURE 4. Spatial Resolution of One Dimensional Test Counter for Collimated Alpha Source. The width of the peak is 2.6 mm full width at half maximum.





COLLIMATED ALPHA SOURCE

UNCOLLIMATED SOURCE

FIGURE 5. Tests on the Two-Dimensional Counter with an Alpha Source

SOURCE HOLDER DEVELOPMENT

Evaluation of the present source holder has included comparison of moderators and gamma filters. Replacement of the Zr H_2 moderator with Ti H_2 results in a lower neutron flux and a higher gamma to neutron ratio. Paraffin with lead filters compares favorably to the Zr H_2 with lead gamma filters.

Advanced concepts for a portable shield have been developed and a simple facility has been constructed for testing shielding materials and beam extraction geometries. Figure 6 shows a sketch of this facility which consists essentially of a water tank into which the source can be lowered or raised remotely. The facility was designed to handle the 2 mgm ^{252}Cf source expected in the future.

The concept presently under consideration for a portable shield consists of a fiberglass shell that can be filled with water for shielding and neutron moderation. This will allow easy positioning of the empty shell after which a source can be loaded remotely from a storage cask. This offers a simple and light shield that can store small sources (<250 μgm) and utilize much larger sources stored in a separate cask. It is expected that this new shield can be kept below 100 lb for handling as opposed to the 350 lb of the present shield.

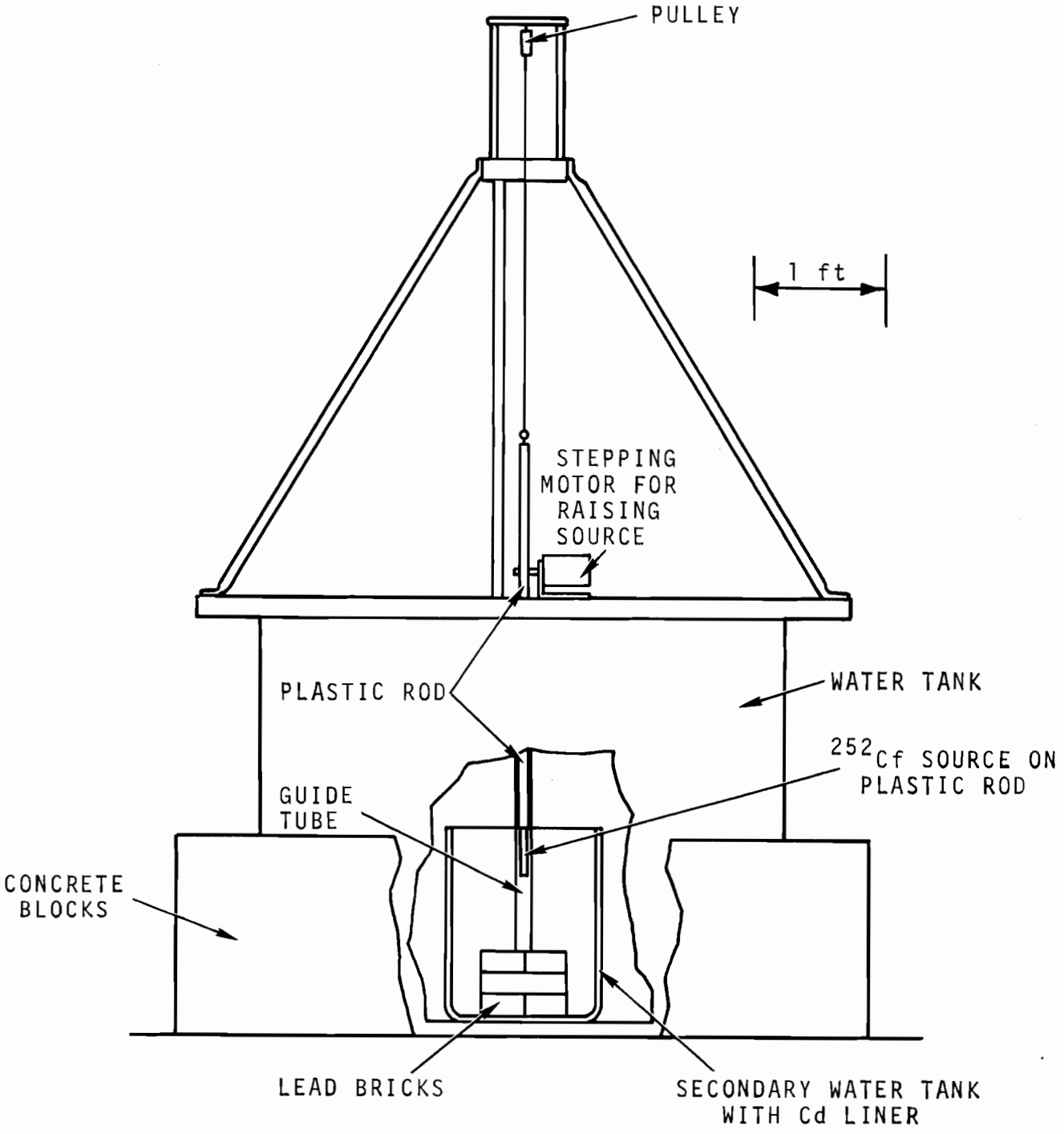


FIGURE 6. Sketch of Shield for Testing Neutron Shielding Configurations and Beam Extraction Geometries

ADVANCED SOURCE DEVELOPMENT

J. H. Jarrett

The objective of this program is to evaluate present and future heat and radiation source application requirements and to relate them to present and future encapsulation technology.

The goal of the current work is concerned primarily with the development of thin-film encapsulation and evaluation techniques for alpha photon sources. Emphasis will be directed toward the technique of sputtering as a means to obtain thin films of radioisotope and containment materials.

The preparation of ten alpha photon radiation sources in a single run using rf sputtering techniques was completed in late 1970. Source preparation detail may be found in the previous report in this series, BNWL-1308-5, distributed in March, 1971. In early 1971 four of these sources were sent to ORNL for testing and evaluation in the AEC-DID sponsored Source Safety Testing Program. The testing has been completed and as with previously tested PNL sources, it is anticipated that the results will be discussed in detail in ORNL reports to DID. Indications are that in most tests the sources performed as well or better than previously tested PNL prepared alpha photon sources. Some areas for source improvement are still indicated. Significant contamination was released in the simulated fire test where, in one hour, the source is heated to over 900 °C in an air atmosphere. Our preliminary evaluation of these results is that the use of copper (which oxidizes fairly readily, and can form a poorly adhering oxide layer) in the $^{241}AmO_2$-Cu active layer should be discontinued, and that it should be replaced by a material more resistant to oxidation such as Inconel 600. A complete analysis of the test results will be made when the ORNL discussion becomes available.

One of the ten alpha photon sources prepared in late 1970 was placed in boiling water at PNL for about eight weeks. At the end of this period the smearable contamination on the source surface was measured at 500 dis/min. The total amount of contamination released to the water was only about 22,000 dis/min or 1×10^{-5} mCi. The source surface discolored (darkened) during the test but contamination release was minimal.

A new tuning network was procured for the glovebox sputtering system. Installation of the network in the glovebox is anticipated in May. The new network is much improved over the one in present use. The result should be greater experimental efficiency and improved control of operating variables.

CIRCULATORY SUPPORT SYSTEMS

F. T. Cross, J. C. Sheppard and B. D. Bingham

The main objective of this program is to determine the dose from radioisotopic heat sources suitable for circulatory support systems. This is accomplished by measuring the dose rates within and surrounding various tissue equivalent phantoms.

Part of the program concerns the prediction of the measured dose rates by the development of computational codes. The computer codes are also used in parametric studies and for the prediction of the in-phantom dose rates from sources not available for measurement.

Another part of the program concerns the development of radioisotopic blood heaters for use in an animal study aimed at assessing the combined effects of heat and radiation in Hanford miniature swine implanted with 50-W plutonium heat sources.

DOSIMETRY STUDIES

Measurements of the neutron dose rates from the Rankine dosimetry model (a prototype circulatory support heat source subsystem, see Figure 7) placed in a large phantom filled with tissue-fluid were completed. Similar measurements with a Remab phantom are near completion. Californium-252 is being used as a substitute for the $^{238}PuO_2^{16}$ source that will eventually be placed in the Rankine device. The measurements will be included in subsequent reports when the data are analyzed and a ^{252}Cf to ^{238}Pu normalization factor is established.

Photon dose rates were calculated for a 30-W $^{238}PuO_2^{16}$ source placed in the Rankine dosimetry model which, in turn, is placed in a large tissue fluid phantom. The dosimetry device material thicknesses and compositions were taken from Hittman Associates drawings 3017D196 and 3017D200. The encapsulated source dimensions, density and compositions were obtained from LASL's January 15, 1971 Progress Report, CMB-1797 and by communication with LASL.

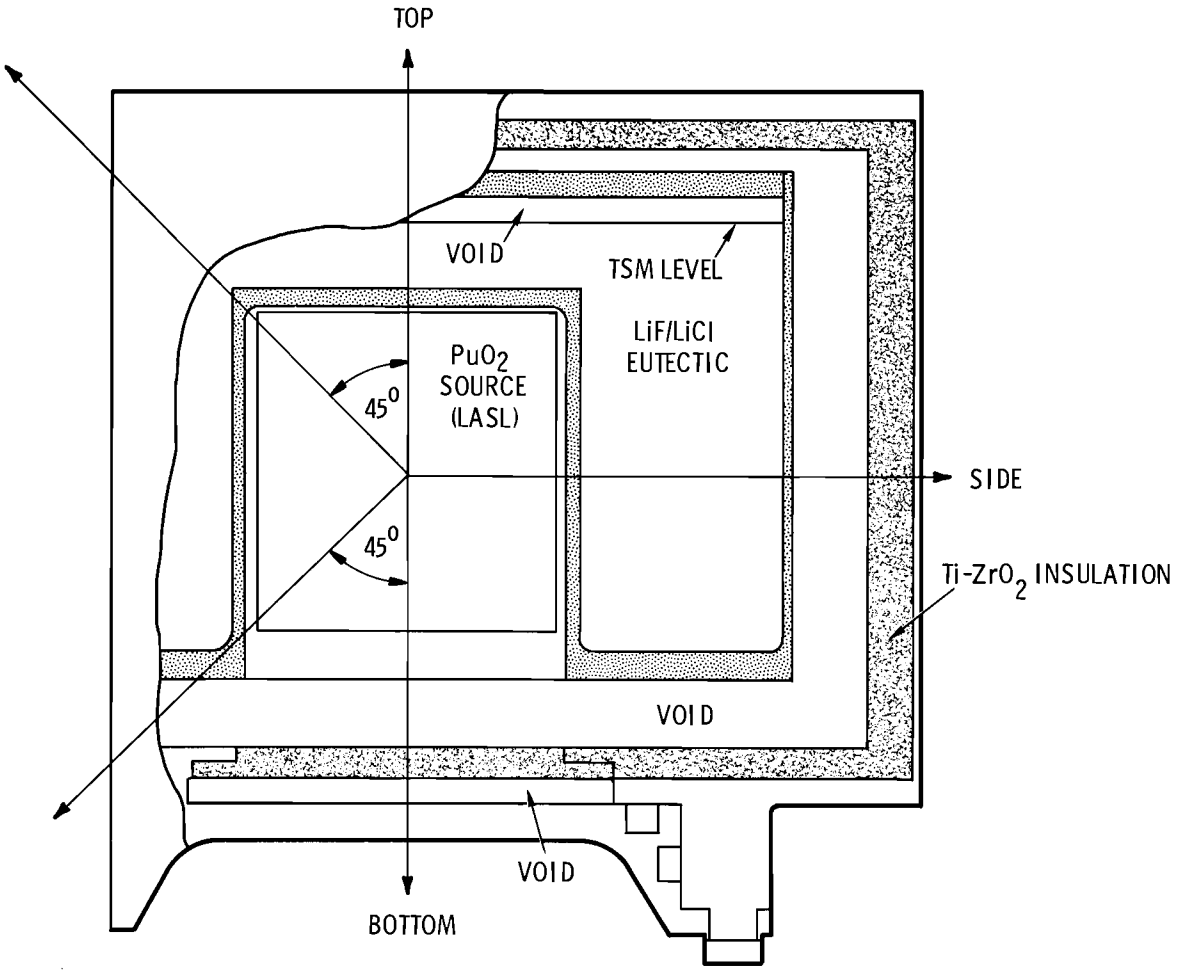


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FIGURE 7. Rankine Dosimetry Model

The right cylindrical dosimetry device was described by 22 planes, normal to the "Z" or axial axis, intersecting 13 concentric cylindrical surfaces. The "Z" axis was the center axis of the cylinder. Ten elements (Pu, Ta, Zr, Fe, Ti, Cl, F, O, Li and H) describe the 16 compositions used in the calculations. Photon dose rates were calculated for both the solid and liquid states of the contained LiF/LiCl eutectic. The buildup factors of lead were used and the origin was taken at the source center. Results of the calculations are presented in Table 3.

TABLE 3. ^{238}Pu Photon Dose Rates in Tissue from Rankine Dosimetry Model

Distance, cm	LiF/LiCl Eutectic Cold (Dose rate, mrad/hr)	LiF/LiCl Eutectic Hot, molten (Dose rate, mrad/hr)
"Z" axis		
3.8634 (top)	48.16	45.52
5.0000	26.15	24.73
10.0000	4.55	4.30
15.0000	1.44	1.35
20.0000	0.58	0.54
-3.5181 (bottom)	60.27	60.27
-5.0000	26.32	26.32
-10.0000	4.58	4.58
-15.0000	1.45	1.45
-20.0000	0.58	0.58
X, axis		
5.0445 (side)	27.56	29.25
10.0000	5.01	5.33
15.0000	1.60	1.70
20.0000	0.65	0.69
X, Z axis (10 cm 45°)		
7.0711, 7.0711 (towards top)	3.81	3.56
7.0711, 7.0711 (towards bottom)	3.73	3.79

As expected, the dose rates out the bottom do not change when the eutectic melts since the photons do not traverse the eutectic. The increase of the dose rates out the side reflects the 24 percent reduction in the density of the LiF/LiCl eutectic when it melts. The reduction of the dose rates out the top can be attributed to an increase in the thickness of the LiF/LiCl eutectic and a reduction of the void thickness above the eutectic when it melts. This is contrasted to a constant

LiF/LiCl thickness but reduced density for the side. Although the photon dose rates from the previously calculated LASL-1 source are not strictly comparable, they are in good agreement with these calculations when adjustment is made for similar tissue thicknesses.

Similar calculations were made of the neutron dose rates and the photon dose rates from ^{236}Pu assuming 0.25 ppm for this impurity. The coordinates, compositions and lead buildup were identical to the ^{238}Pu calculations. The 0.25 ppm ^{236}Pu and a neutron emission rate of 2×10^5 n/sec were used to conform with the LASL determined values for the 30-W $^{238}PuO_2^{16}$ source LASL 30 C6. These calculations show similar features to the ^{238}Pu calculations - slight changes due to the thickness and density of material traversed by the photons and neutrons. Also the neutron dose rates agree well with calculations using the simplified equation reported in BNWL-1489. This equation was generated for predicting the measurements of a 30-W metal source and assumes a constant tissue attenuation of about 11.4% per cm in addition to an inverse square fall off of the dose. Results of the calculations are presented in Table 4.

RADIOLOGICAL ENGINEERING STUDIES

Engineering studies to develop radioisotopic blood heaters in support of PNL's "Biological Effect of Intracorporeal Radioisotope Heat Source" study were initiated this past quarter. Heater geometry, materials, and temperature changes in the bulk stream and boundary layer of the blood were investigated. The conceptual design of a telemetry system for monitoring blood temperature changes in the host animals was completed. In addition, two surgical shams were fabricated for implantation in miniature swine. A third sham with end pieces incorporating hydraulic screw couplings to simplify surgery is being fabricated

TABLE 4. Neutron and ^{236}Pu Photon Dose Rates in Tissue from Rankine Dosimetry Model (mrad/hr)

Distances, cm	Neutron Dose Rate		0.25 ppm ^{236}Pu Photon Dose Rate, 10 yr Avg	
Top	Cold	Hot	Cold	Hot
3.8634	11.40	10.97	21.29	20.58
5.0000	6.10	5.85	11.35	11.58
10.0000	0.92	0.88	2.41	2.33
15.0000	0.23	0.22	0.88	0.85
20.0000	0.064	0.061	0.41	0.40
Bottom				
-3.5181	14.12	14.12	26.29	26.29
-5.0000	6.05	6.05	12.00	12.00
-10.0000	0.91	0.91	2.42	2.42
-15.0000	0.22	0.22	0.89	0.89
-20.0000	0.063	0.063	0.41	0.41
Side				
5.0445	5.93	6.18	12.08	12.74
10.0000	0.92	0.97	2.56	2.65
15.0000	0.23	0.24	0.94	0.97
20.0000	0.064	0.069	0.44	0.45
45°				
10.0000 (Top)	0.94	0.89	2.16	2.08
10.0000 (Bottom)	0.93	0.94	2.13	2.15

for implantation in the near future. The final thoracic heater design and shop drawings were completed and fabrication of a bench model device for test will be initiated upon receipt of materials.

An analysis of the plutonium heat source centerline temperature in a fatality (death) situation was completed. The model used for analysis was a composite tissue cylinder of size

equivalent to a Hanford miniature swine containing a 50-W $^{238}PuO_2^{16}$ capsule. An account was made of the heat losses both radially and axially. The temperatures determined at various locations are shown in the accompanying diagram (Figure 8). Briefly, the predictions are for a centerline temperature of 972 °F assuming an ambient temperature of 75 °F. It is recommended, therefore, that a Pt-20 Rh corrosion-resistant liner be employed as the source compatability container.

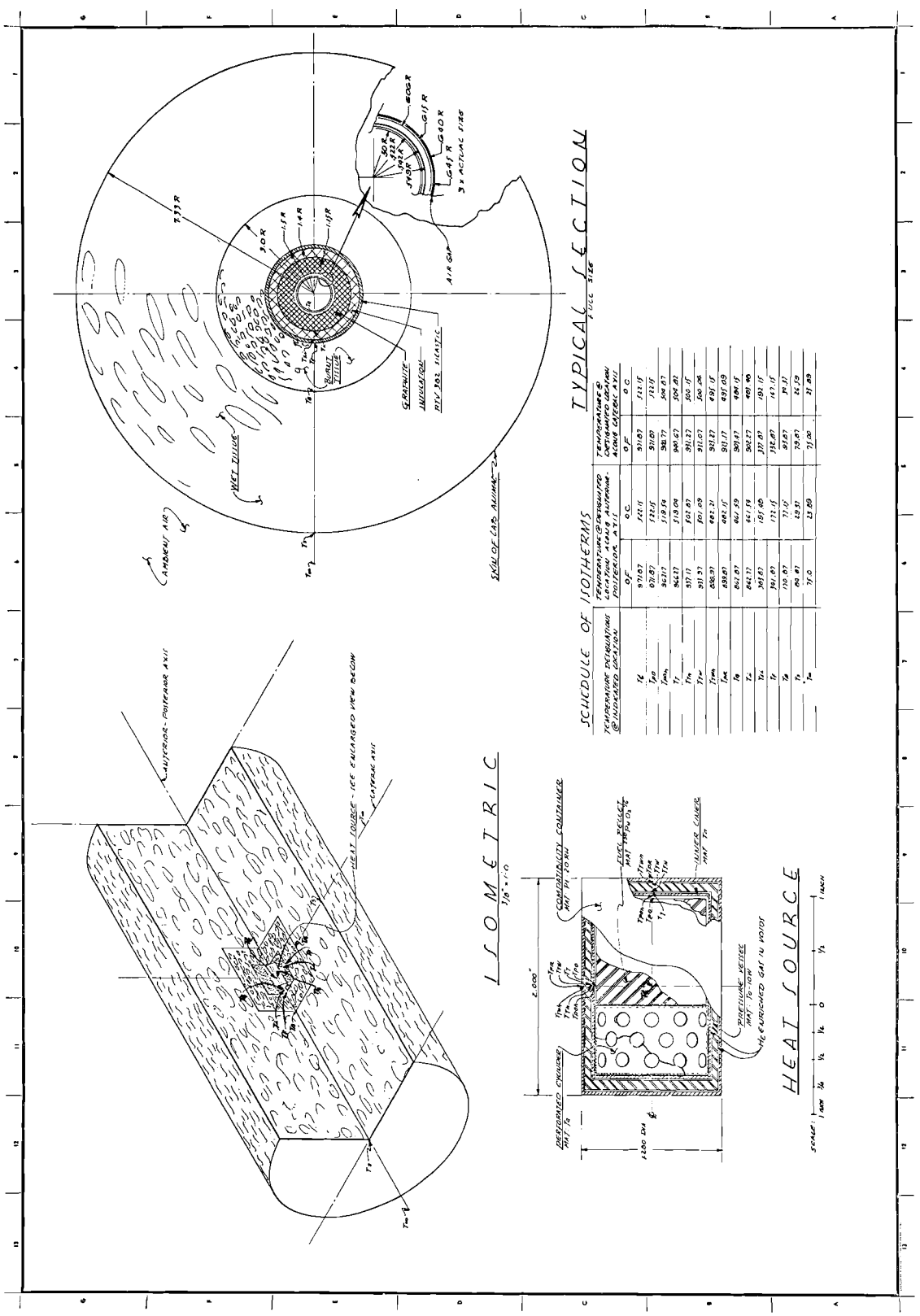

FIGURE 8. Calculated Heat Source and Surrounding Tissue Temperatures in Fatality Situation

NUCLEAR TECHNIQUES FOR SEABED MINERAL EXPLORATION

R. W. Perkins and N. A. Wogman

In recent PNL studies of the composition of deep ocean sediments, it has been demonstrated that various mineral deposits and commercial-grade ores can readily be identified by neutron activation and instrumental analysis. From preliminary analysis, it is evident that the elemental composition of most mineral deposits can be easily identified at concentrations well below those of economic importance. In-place experience with ^{252}Cf as a neutron source has confirmed these conclusions. Two methods for in-place seabed mineral exploration are being investigated. The first involves placing a neutron source and detector system on the ocean floor. The second method involves bringing the samples on-board ship and employs a shipboard ^{252}Cf irradiator and a Ge(Li) spectrometer. The former technique has the advantage of very rapid analyses, and the latter technique has the advantage that mineral depth-profiles can be defined from core sampling.

During the past quarter studies have included the variations in detection efficiency for elements in substrates of various densities, cross sections, and overburdens. The detection of gamma rays in the range of 0.1 to 2.7 MeV have been considered in the matrices. All measurements utilize the mockup facility containing a 1.8 mg ^{252}Cf source and simulate the actual seabed irradiation and counting conditions. The variations were measured in 200 lb specimens using mixtures of iron powder and dunite to produce matrix densities varying from 1.7 to 4.0 grams per cm^3. The elements of interest which on neutron capture emit gamma rays in energy ranges from 0.1 to 2.7 MeV were homogeneously mixed into these substrates, irradiated in a simulated sea floor condition and analyzed for their specific element. The measurement efficiency which is a function of sample density, cross section, hydrogen content, and the energy of the emitted gamma ray, is reduced only 25%

for 1.8 MeV gamma rays as the matrix density rises from 1.7 to 4.0. Elements emitting gamma rays near 100 keV have their measurement efficiencies decreased about 1.5 fold as the sediment density increases from 1.7 to 4.0. If these elements are beneath 1 inch of sediment with a density of 1.7 and containing 1% cadmium, their detection sensitivity is reduced 4.6 fold. The 1 inch of sediment alone decreases the detection limit four-fold, thus the high capture cross section produced by the 1% cadmium in this surface layer has relatively little effect on the overall detection limit. Studies of the detection efficiency versus variations in the distance and hydrogen content are continuing and will allow the maximum effect of non-uniform seafloor conditions to be determined.

In work related to this program, a subcritical multiplier for ^{252}Cf neutrons was evaluated for seabed mineral analysis. A mockup composed of a homogenous mixture of PuO_2 in polystyrene reflected with Plexiglas was employed. The hydrogen to plutonium ratio was 15. The effective multiplication constant was about 0.975 and was obtained from the cuboidal core assembly which was 9 × 9 × 7 inches. Simulated ocean floor mineral samples were irradiated with 30 to 40 fold flux increase over that available without multiplication. The investigation demonstrated that a subcritical assembly could be used to enhance the thermal flux from a ^{252}Cf neutron source in the designed seabed probe.

An evaluation of the primary gamma rays from molybdenum ores and those most sensitive for its measurement in the seabed was completed. Some 150 gamma rays are emitted following the irradiation of natural molybdenum ore.

The prototype designs of the nuclear probe underwater assembly and the electronics packages for data accumulation and analysis have been finalized. Bids for the electronic packages have been received and the purchase order will be issued during May.

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Technical Information Files (5)
Technical Publications (2)