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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1974
TO THE USAEC DIVISION OF BIOMEDICAL
AND ENVIRONMENTAL RESEARCH
PART 4 PHYSICAL AND ANALYTICAL
SCIENCES

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

FEBRUARY 1975

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ANNUAL REPORT FOR 1974
TO THE
USAEC DIVISION OF BIOMEDICAL AND ENVIRONMENTAL RESEARCH
PART 4 PHYSICAL AND ANALYTICAL SCIENCES

By

J. M. Nielsen
with Major Contributions from
Staff Members of
Radiological Sciences Department
Physics and Instrumentation Department

February 1975

On January 19, 1975, research and development programs of the U.S. Atomic Energy Commission (AEC) became part of the newly formed Energy Research and Development Administration (ERDA). In this report, since it refers to work done in 1974, most references are to AEC programs.

BATTELLE
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RICHLAND, WASHINGTON 99352

PREFACE

The Annual Report for 1974 to the U.S. Atomic Energy Commission's Division of Biomedical and Environmental Research is organized by major program categories according to our schedule-189 submissions. Each part is directed toward a particular DBER Research and Development Program: Part 1 to Biomedical Programs, Parts 2 and 3 to Environmental Programs, and Part 4 to Physical and Technological Programs. Each part of the Annual Report comprises project reports authored by scientists from several research departments, reflecting the interdisciplinary nature of the research effort. The Annual Report consists of four parts:

Part 1	Biomedical Sciences	Program Manager: R. C. Thompson Editor: D. L. Felton
Part 2	Ecological Sciences	Program Manager: B. E. Vaughan Editor: J. L. Engstrom
Part 3	Atmospheric Sciences	Program Manager: C. E. Elderkin Editor: E. L. Nichols
Part 4	Physical and Analytical Sciences	Program Manager: J. M. Nielsen Editor: G. M. Garnant

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1956	HW-47500
1957	HW-53500
1958	HW-59500
1959	HW-63824, HW-65500
1960	HW-69500, HW-70050
1961	HW-72500, HW-73337
1962	HW-76000, HW-77609
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1973	BNWL-1850, Pt. 1-4
1974	BNWL-1950, Pt. 1-4

W. J. Bair
Program Director, Life Sciences

FOREWORD

Part 4 of the Pacific Northwest Laboratories Annual Report for 1974 to the USAEC Division of Biomedical and Environmental Research continues the same format as for 1973 and includes those programs funded under the title Physical and Analytical Studies. Included are the programs in Instrumentation Technology which cover radiological chemical methods and instrument development and studies, and the development of nuclear techniques for application to medical studies; programs in Radiological and Chemical Physics which include radiation physics, radiation biophysics and radiation dosimetric research; studies related to the development of nuclear powered prostheses and the medical uses of isotopes; and research on the artificial heart.

Julian M. Nielsen, Manager
Radiological Sciences Department

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INSTRUMENTATION TECHNOLOGY

- **RADIATION INSTRUMENTATION—RADIOLOGICAL CHEMISTRY**
- **FAST REAL-TIME MEASUREMENT OF EXTREMELY LOW-LEVEL AIRBORNE PLUTONIUM**
- **ANALYTICAL TECHNIQUES FOR MEASUREMENT OF ^{99}Tc IN ENVIRONMENTAL SAMPLES**
- **NUCLEAR TECHNIQUES IN MEDICINE**

- **RADIATION INSTRUMENTATION—RADIOLOGICAL CHEMISTRY**

The development and utilization of radioanalytical technology and instrumentation allows studies of the environmental behavior of trace amounts of radionuclides and their stable element counterparts. During the past year, emphasis has been placed on improving the sensitivity of radioanalytical analysis systems and techniques as well as feasibility studies which focus the future direction of the program.

Low-Level Radiological Chemistry Laboratory Techniques

In Situ Monitoring

Activation Analysis Technology

Chemical and Molecular Composition Technology

Environmental Pollution Analysis

Low-Level Radiological Chemistry Laboratory Techniques

Newly developed electronic and computer technology together with improved low-background construction materials have aided in developing more sensitive radiation instrumentation. Backgrounds have been reduced by tenfold at the Th and U photopeak areas, as well as twenty- to one-hundredfold in the ^{40}K photopeak region. The employment of pulse pileup rejection systems on Ge(Li) and Si(Li) spectrometers has allowed count rates with system dead time of 40% to be used with less than a 2% live-time loss. High sensitivity x-ray analysis, coupled with computer techniques, allowed measurements of specific tracers for resuspension studies. A feasibility study of laser excitation fluorescence and ionization analysis was completed, along with a study of the techniques required for the measurement of long half-life species from power reactors, such as ^{129}I , $^{59-61}\text{Ni}$, $^{93}\text{Zr/Nb}$, ^{99}Tc , ^{75}Se , ^{135}Cs , and ^{151}Sm .

Potential of Laser Excitation for Radionuclide Analysis

M. R. Petersen, L. A. Rancitelli and N. A. Wogman

With the growing application of nuclear power generation, the need is increasing for improved sensitivity in the measurement of many radionuclides. Essentially all the AEC's studies of the environmental behavior of radionuclides could be simplified if measurement sensitivities could be increased by a few orders of magnitude. Through recent developments in tunable lasers, it is now possible to selectively excite isotopic forms of elements and molecules of interest to fluorescent energy states or through multistep processes to ionization. The feasibility of using laser-induced activated species for analysis of gaseous forms of radionuclides appears most promising.

An example of the potential improvement in sensitivity can be shown in the case of ^{14}C . With its 5730 year half-life, the probability is that one out of two atoms will decay during the half-life period of 1.8×10^{11} sec; thus, one requires approximately 10^3 (about 0.4 d/m) atoms to make an accurate measurement of the ^{14}C content in a sample. By the proposed laser exci-

tation technique, it may be possible to measure as little as 1 to 100 atoms of ^{14}C . This would improve the current sensitivities by 7 to 9 orders of magnitude and could extend ^{14}C dating from the nominal 50,000 years presently available to as much as 200,000 years.

The basic principles involved in laser excitation analysis are relatively simple. An atom or molecule in one energy state can be selectively promoted to a higher energy state by resonant absorption of a laser-produced photon. The real advantage in tunable laser excitation is that a very large fluence of nearly monoenergetic photons are produced to excite a particular isotope of an element or molecule. By coupling this excitation technique with high-sensitivity photon and gas counting methods, it should be possible to improve measurement sensitivities for radionuclides, especially long-lived ones, by many orders of magnitude. Some of the radionuclides of interest include ^{85}Kr , $^{133-135}\text{Xe}$, $^{37-39}\text{Ar}$, $^{129-131}\text{I}$, ^{14}C , ^3H and the transuranium elements.

The proposed technique consists of exciting the gaseous species of interest in a multistep fashion in which at least one selective step

is by a tunable laser. The other steps may consist of another tuned laser or a nonselective excitation source such as dc gas discharge, microwave, or high energy particle irradiation. There is a variety of possible energy transfer schemes between excited states which allow fluorescence or ionization to occur, as are shown in Figure 1.1. Fluorescence could be detected using sensitive photomultipliers, and ion pairs formed during photoionization could be detected by proportional counting methods.

In the case of solid substances such as ^{99}Tc , ^{129}I and the transu-

ranium elements vaporization is necessary prior to excitation. For analysis of these solid substances it appears that it will be necessary to carry them through preliminary chemical separation before vaporizing them in a reduced form. The analysis of solids may therefore be somewhat more difficult than that of gases.

The development of a laser excitation technique followed by fluorescent or ionization analysis procedures should greatly improve the measurement sensitivities of numerous trace substances and is expected to provide a major breakthrough in

Neg 747716-1

FIGURE 1.1. Energy Transfer Schemes in the Utilization of Laser Excitation Analysis.

sensitivity for the measurement of many long-lived radionuclides.

Improved Multidimensional Gamma Ray Spectrometer Sensitivity
N. A. Wogman

Improving the sensitivity of highly efficient gamma-ray spectrometers has, for the past 10 years, been mainly limited by background photons from the ^{238}U and ^{232}Th decay chains, and from natural ^{40}K . Reduction of the contribution from these sources is essential to the development of ultra-low background systems. Methods of optimum detector operation and our studies of potential cladding material for the detectors should reduce the contributions to the background of ^{40}K , ^{238}U and ^{232}Th by more than tenfold.

Our studies with light pipes indicate that extension of their lengths from the present 10 cm to a length of 20 cm can reduce the phototube ^{40}K contribution to the system by approximately tenfold.

Since the primary source of ^{238}U and ^{232}Th is the reflective coating and crystal housing material, specimens of construction materials were analyzed to find those of low natural radioactivity. Titanium metal was found to be very pure containing less than 5% of the U and Th in present detector cladding, and methods were investigated to fabricate Ti cans for the housing of a 30-cm-diameter by 20-cm-thick NaI(Tl) system. The Ti material should reduce this source of heavy element background by at least twentyfold. Low background materials that might replace the MgO which presently serves as a reflective coating between the housing and crystal were investigated. Some of the materials considered are compared with MgO in Table 1.1. These items are proprietary and are therefore simply identified by number. The data clearly illustrate the reflective coating backgrounds which can be achieved for satisfactory reflective coating materials. The magnesium oxides normally found in reflective coatings usually contain 100 to 300 ppb Th and 70 to 200 ppb U. As is seen in Table 1.1, some of the coatings have less than 1 ppb Th and less than 3 ppb U. Therefore these reflective coatings contain

TABLE 1.1. Th and U Concentrations in Selected Reflective Material

Sample	Th, ppb	U, ppb
10 MgO ¹	100-300	70-200
16	0.65	<3
17	0.6	<3.5
18	25	71
19	104	95
19A	70	91
15	4	12

¹Commercial Reflective Coatings.

less than 1/50 of the radioactivity in present coatings. The total reduction in Th and U background is as yet unknown, since the systems must be installed in lead caves with shielding similar to those of our present systems. The selection of these construction materials should allow a substantial improvement in background in future low-level gamma-ray spectrometry systems and other low-level counting systems.

With these reduced backgrounds the relative contribution of cosmic-ray interactions increases. As these systems are tested the optimum methods for further reduction of the cosmic ray contributions will be evaluated.

During the investigation of methods to reduce backgrounds a study was initiated of the resolution loss which occurred in a NaI(Tl) crystal system as its phototube quantity was varied and as its light pipe configuration was changed.

Thirty-cm-diameter by 20-cm-thick NaI(Tl) crystals generally utilize seven 7.6 cm phototubes. These are typically found in an array around the edges of the crystal, with one tube at the center. As is shown in Table 1.2, this type of crystal has a ^{137}Cs resolution varying from 8.9-13.1% as the number of phototubes vary from one center tube to six around the edges. The ^{40}K contribution from the phototubes could be reduced by greater than 50% if the scientist could accept a 1% change

TABLE 1.2. ^{137}Cs Resolution of a 30 x 20 cm Thick NaI(Tl) Crystal Under Various Phototube Configurations.

Phototube ² Configuration	Resolution, % FWHM ¹		
	No Light Pipe	30-cm-diameter x 10-cm-thick NaI Light Pipe	7-cm-diameter x 30-cm-long Quartz Light Pipe
One Center	13.1		18.0
Two 180° opposed	11.1		14.0
Three 120° opposed	10.0	12.0	
Four 90° opposed	9.9	10.7	11.5
Four; 3-120° opposed, 1 center	9.2		
Five; 4-90° opposed, 1 center		10.5	11.1
Six 60° opposed	8.9	10.4	
Seven 60° opposed, 1 center	9.2	10.4	

¹Cesium-137 source 40 cm away from the crystal center line.

²Selected RCA 8054 Tubes.

³Each quartz light pipe was associated with one phototube and was covered with a Harshaw Chemical Co. proprietary reflective material.

in system resolution. Resolution also decreases if a light pipe is introduced between the phototubes and the primary crystal. With a 10-cm-thick pure NaI light pipe, the resolution decreases about 10%. For a system with 30-cm-long, 7-cm-diameter quartz light pipes on top of the primary 30-cm-diameter, 20-cm-thick NaI(Tl) crystal the resolution was greater than 11%. This cannot be tolerated unless the investigator is studying only a few radionuclides.

A gamma-ray spectroscopist interested in measuring the primordial radionuclides ^{40}K , ^{232}Th , and ^{238}U might require such a low background for these isotopes that he would allow the utilization of systems with resolutions exceeding 10%.

This study indicates that for large detector systems, 4 phototubes provide resolutions of about 9-11%. A system consisting of a primary detector, 10 cm NaI light pipe and 30 cm quartz light pipe with 4 tubes will provide a resolution of about 13%, yet move the source of the ^{40}K , the phototubes, 40 cm away from the detector in addition to the twofold

^{40}K reduction through fewer tubes. It is estimated that the overall reduction would be of the order of twentyfold. A further reduction of ^{40}K is anticipated, since this design allows Pb to be placed above the NaI(Tl) detector, but below the glass phototubes, providing 30 cm of high-density shielding.

Improvements in Ge(Li) Spectrometry N. A. Wogman and H. L. Nielson

A problem is often encountered in the use of Ge(Li) detector systems for the analysis of standards and unknowns with vastly different disintegration rates. To determine the degree to which analysis errors may be generated by high counting rates and investigate the improved prototype instrumentation which might circumvent this dead-time problem, a study was undertaken on the effect of changing amplifier pulse shapes, and the use of base line restoration and pulse pileup rejection on active Ge(Li) systems. These relationships were evaluated by observing the effect on the 1332 keV photopeak of a standard ^{60}Co source at various dead times.

A ^{137}Cs source was positioned at various distances from the diode while the ^{60}Co remained at a specific point, thus increasing the number of pulses reaching the analyzer and increasing the analyzer dead time. Replicate measurements were taken with each system at amplifier shaping time constants of 1, 2 and 4 μsec as well as studying the effect of base line restoration. A set of measurements was also made with a counting system at a 4- μsec shaping time constant, with and without a pulse pileup rejection.

A series of different analyzer components was employed in this evaluation. The first system consisted of a 130 cm^3 Ge(Li) coaxial diode with a model RG11C Princeton Gamma-Tech preamp. This diode has an efficiency of 25.8% relative to a 3 in. NaI(Tl) crystal. A Northern Scientific Model NS715 Analyzer with

a built-in ADC was employed with a Model 1413 Canberra amplifier and a 1464 Canberra pulse pileup rejector. A second counting system utilized a Nuclear Data analyzer, a Northern Scientific NS629 analog digital converter, and a 1413 Canberra amplifier. The Ge(Li) diode was an 80 cm^3 , 12% efficient Princeton Gamma-Tech diode with an RG11C preamp.

As can be seen from Figure 1.2, the 130 cm^3 diode system resolution changes by over twofold as the dead time is increased from 8 to 42%, with a shaping time of 4 μsec and no pulse pileup rejection. When the pulse pileup rejector was added to the system, the resolution was degraded by only some 25% for the same dead time variation.

As is shown in Table 1.3, resolution degradation also occurs with changes in amplifier shaping times.

FIGURE 1.2. Comparison of Ge(Li) Resolutions With and Without Pulse Pile Rejection for Various Amplifier Shaping Times as a Function of System Dead Time.

TABLE 1.3. Resolution as a Function of System Dead Time [130 cm³ Ge(Li)]

Shaping Time	Percent D.T.	Resolution keV
1 μ sec	10	3.38
	12	3.44
	20	3.59
	32	4.10
	62	~ 7
2 μ sec	10	3.15
	12	3.40
	20	3.63
4 μ sec	10	3.20
	12	3.28
	20	3.65
	32	76

At 1 μ sec with a dead time increasing from 10-62%, the resolution changes by greater than twofold. This also occurred with a 4 μ sec pulse shape time constant for this system. A satisfactory time for the system, seemed to be about 2 μ sec. Figure 1.3 shows the effect on ⁶⁰Co counting for the 80 cm³ system with increase in count rate of ¹³⁷Cs. With a 4 μ sec time constant, using 90% of the peak, it was found that a ⁶⁰Co rate of 48 cps changed by almost threefold with the addition of

650 cps ¹³⁷Cs. Using a 1 μ sec amplifier shaping time, a twofold decrease in the ⁶⁰Co count rate was noticed at 3700 cps ¹³⁷Cs. Data obtained from the 2 μ sec time constant from the amplifier pulse shape fell between 1 and 4 μ sec data. Approximately a 10% change in count rate was found to be due to the effect of base line restoration. Pulse pileup rejection with microsecond pulse shape timing almost eliminates the problems portrayed in Figure 1.3. A 40% count rate dead time can be tolerated with less than a 2% change in the ⁶⁰Co count rate if a pulse pileup rejector is employed (Canberra 1468).

Problems similar to that discussed were observed on other systems. The actual variations could not be predicted from one system to another because of differences in electronics. The data do indicate, however, that pulse pileup rejection and base line restoration are important for Ge(Li) or Si(Li) systems where high count rates are ($\leq 50\%$ dead time) required. The systems employed at Battelle-Northwest for neutron activation analysis programs are presently being converted to use this type of instrumentation where the high dead times can be expected in the analytical phase of DBER programs.

FIGURE 1.3. A Comparison of the Count Rate Changes for a ⁶⁰Co Source at Various Dead Times with Changing Electronics Parameters for a (Ge(Li) Spectrometer.

Molybdenum as a Tracer for
Resuspension Studies
J. D. Ludwick

Resuspension of deposited radionuclides is a problem receiving considerable attention. Of particular interest is the resuspension of long-lived radionuclides such as the plutonium isotopes and ^{241}Am . To permit studies of resuspension materials, we have developed an analytical technique in which the relatively high density material, calcium molybdate (4.35 g/cm^3) serves as a tracer. In the actual resuspension program, this material is generated as an aerosol from a newly formed precipitate and deposited over the test area. Subsequent collections downwind from this area are used to determine resuspension as a function of time and meteorological conditions. From a tracer detection point of view, molybdenum was chosen because it is reasonably rare in nature, the calcium molybdate can be separated from soils in a manner which does not remove a significant amount of the natural molybdenum, and we can measure it with a very high degree of sensitivity ($5 \times 10^{-8} \text{ g}$) through an x-ray fluorescence technique.

Basically, the procedure involves the following steps. Soil samples or filters on which particle size separates are collected and leached with 3N HNO_3 . Following evaporation to dryness and dissolution in $0.5\text{N H}_2\text{SO}_4$, molybdenum is extracted into an 0.5% α -benzoin-oxime in chloroform. This organic extract is evaporated and the residue is subsequently mounted on a mylar film within a 1.2 cm circle which is defined by a ring of silicon grease. The mylar film premounted on a 35-mm slide holder, is analyzed by x-ray fluorescence in which the x rays are generated from a secondary silver target. This relatively simple procedure takes advantage of the inherent high sensitivity of x-ray fluorescence analysis to permit rapid and sensitive measurements for a realistic resuspension study.

Measurement Techniques for Long-Lived
Radionuclides from Power Reactors
N. A. Wogman

A high exposure of nuclear power reactor fuels will produce large

amounts of long-lived fission and activation products such as ^{59}Ni , ^{63}Ni , ^{79}Se , ^{93}Zr , ^{99}Tc , ^{126}Sn , ^{129}I , ^{135}Cs , and ^{151}Sm . In this study we have considered the possible methods of analysis of these radionuclides and compared the possible sensitivities which might be achieved. Since these radionuclides will be of concern for hundreds to millions of years, methods for determining their environmental behavior are important. Table 1.4 provides a summary of the nuclear decay properties of the radionuclides together with potential measurement sensitivities by existing and future possible analytical techniques.

For measuring all of these long-lived species we considered the techniques of mass spectrometry, neutron activation analyses, direct counting techniques and future laser excitation analysis. For example, a three-stage mass spectrometer will differentiate ^{133}Cs , ^{134}Cs , ^{135}Cs and ^{137}Cs . In normal mass spectrometers the distribution around the stable ^{133}Cs peak would normally interfere with the analysis of the ^{135}Cs and isotope separation would be needed before quantitative analysis was attempted. With the three-stage system 10^6 atoms of ^{135}Cs per mole of ^{133}Cs may possibly be detectable. Using this technique a precise value for the isotopic relationship between ^{135}Cs and ^{133}Cs can be obtained thus allowing ^{133}Cs to be used as a standard for ^{135}Cs . Similar techniques may be used for the other long-lived species. Laser excitation analysis may provide a one-hundredfold better sensitivity. Another possibility with poorer sensitivity would be the chemical separation of Cs, the isotope separation of ^{135}Cs , and its analysis by beta counting.

For ^{99}Tc , picograms of material may be analyzed by a three-stage mass spectrometer; however, the best to date is nanograms. Part of the problem is that filament ionization is less than 0.1% efficient. For quantitative analysis this procedure must use an isotopic dilution technique with ^{97}Tc . The radiochemical analysis of ^{99}Tc may be accomplished by analyzing its Ru daughter K x rays or more sensitive analysis may be accomplished with low background beta counting of separated Tc. Although neutron activation analysis techniques have been

TABLE 1.4. Decay Characteristics and Potential Detection Sensitivities for Some Long-Lived Radionuclides.

Radionuclides	$t_{1/2}$ (year)	Type of Decay	Predominant Energy	Detection Sensitivities dpm					
				Beta ⁺	x ray ⁺	γ ray	N.A.A.	Mass Spec. ⁺⁺	Laser ⁺⁺⁺
⁵⁹ Ni	7.5×10^4	E.C.; Co x ray	6.925 keV ^c 7.649 keV γ continuum to 1.065 MeV	-	2	1000	-	10^5	10^{-6} to 10^{-7}
⁶³ Ni	92	β^-	66 keV 100%	1	-	-	-	10^8	10^{-3} to 10^{-4}
⁷⁹ Se	6.5×10^4	β^-	160 keV 100%	1	-	-	-	-	10^{-6} to 10^{-7}
⁹³ Zr	1.53×10^6	β^-	0.6 MeV (>95%)	0.5	-	-	-	10	10^{-8} to 10^{-9}
		^{93m} Nb γ	0.9 MeV (<5%) 30.4 keV ^d	-	200 ^a	-	-	-	-
⁹⁹ Tc	2.1×10^5	β^- Ru x rays	0.292 100% 19.236 & 21.646 keV ^e	0.5	-	-	-	.05	10^{-7} to 10^{-8}
				-	2500	-	-	-	-
¹²⁶ Sn	10^6	β^-	0.250 MeV 100%	0.5	-	-	Possible ^b	10^4	10^{-8} to 10^{-9}
		¹²⁶ Sb γ	17.7 to 87.56 keV ^f	-	2	-	-	-	-
		¹²⁶ Tc γ	0.29-0.99 MeV ^g	-	-	-	-	-	-
		γ, γ	0.75 MeV by 0.667 MeV 91% ^h	-	-	1	-	-	-
¹²⁹ I	1.57×10^7	β^- γ^- e^-	0.150 MeV 100% 0.038 Xe x rays ⁱ 0.005, 0.034 keV ^j	0.8	-	-	10^{-5}	10^{-8} to 10^{-4}	10^{-9} to 10^{-10}
				-	20	-	-	-	-
¹³⁵ Cs	2.3×10^6	β^-	0.210 100% ^j	0.8	-	-	10^{-4}	5×10^{-7}	10^{-8} to 10^{-9}
¹⁵¹ Sm	87	β^-	76 keV-98.34% 54 keV 1.66% 21.54 keV 1.66% ^k	1	-	-	-	-	10^{-3} to 10^{-4}
		γ		-	200	-	-	-	-

⁺Chemical separation of a sample followed by analysis.

⁺⁺Three stage mass spec. - assumed some interference has been taken into account.

⁺⁺⁺Possible future development may reduce this estimate; assume 10^4 to 10^5 atoms can be detected.

^aAllow growth of ^{93m}Nb daughter.

^bCross sections have not been determined for neutron capture.

^c100% E.C., Internal Bremsstrahlung End Point at 1.065 MeV.

^dFrom daughter ^{93m}Nb, $t_{1/2} = 13.6$ year.

^e 3.89×10^{-4} K shell ionization per beta decay (Ru x rays)

^f¹²⁶Sb, meta stable states of $t_{1/2} = 19$ and 12.4 min

^g¹²⁶Te daughter of ¹²⁶Sb

^hRequires long coincidence gate 4 and 0.6 ps $t_{1/2}$ states.

ⁱ $e_K/\gamma = 22$, $k/L = 10$

^jNo γ

^k100-fold more photons than Eu x rays between 40 and 42 keV, e^- 1.7%, $e_L/\gamma = 20$.

developed, the short half-life of the neutron activation product, ¹⁰⁰Tc ($t_{1/2} = 16$ sec) makes this method less desirable.

Various methods have been considered for ¹²⁹I, one of which utilizes neutron activation of ¹²⁹I to ¹³⁰I. Iodine-130 decays with several coincident gamma-rays, thus allowing multiple gamma-ray coincidence counting techniques for background reduction and discrimination against interfering activities. Activation analysis with 10^{13} n/cm²/sec results in 1 d/m of ¹³⁰I per 10^{-5} d/m of ¹²⁹I. This technique can be applied to water samples, tissues and particulate or gaseous iodine. Techniques which analyze ¹²⁹I ($t_{1/2} = 1.57 \times 10^7$ years) directly in environmental milk and water samples require elemental iodine separation by solvent extraction, followed by liquid scintillation counting of

the low energy beta emissions (100% $\beta^- = 0.150$ MeV). This provides an analytical sensitivity of 0.2 d/m ¹²⁹I per liter for a four-liter sample or 2×10^{12} atoms.

Thin beta sources can be made on electrospayed ion exchange resin for 4 π analysis, which is most useful for low-energy pure beta emitters such as ⁶³Ni ($t_{1/2} = 92$ years, $E_\beta = 66$ keV). These finely ground ion exchange pads allow 4 π counting efficiencies to reach 0.98 for energies such as 300 keV with reproducibility to within $\pm 0.2\%$. Because ⁶³Ni decays directly to the ground state of ⁶³Cu by beta transition with a maximum energy of only 66 keV, the self-absorption correction introduces some uncertainty in conventional 4 π beta counting. An efficiency-tracer technique allows this correction to be well established. In this technique, samples of ⁶³Ni and

^{65}Ni ($t_{1/2} = 2.52$ hr) are mixed to produce a Ni spraying solution. Tracers such as ^{134}Cs or ^{60}Co ($E^- = 0.314$ MeV) are also prepared. The ^{65}Ni allows the plating efficiency of the ^{63}Ni to be determined. After the ^{65}Ni decays, the ^{134}Cs which has been placed on the source is determined by relative gamma-ray counting against standardized ^{134}Cs . By using various gold-coated VYNS films over these standard sources, the 87 keV beta branch of ^{134}Cs can be investigated and an absorption relationship developed for the specific weight of ^{63}Ni that has been analyzed. This allows the analysis of about 1 d/m of ^{63}Ni or 7×10^7 atoms.

Selenium-79, with a half-life of 6.5×10^4 years, decays 100% by negative beta emission, with an energy of 160 keV to the ground state of ^{79}Br (a stable radioisotope). It can be radiochemically detected at the 1 d/m level only by chemical separation and low-level beta counting.

Nickel-59, a 7.5×10^4 year half-life isotope, decays to ^{59}Co , by electron capture. The gamma continuum end point energy is 1.065 MeV, and there are cobalt K x rays. For low-level analysis, chemical separation and x-ray detection would allow 2 d/m to be determined. This should be compared to mass spectrometry, but little data exists.

Zirconium-93 (1.53×10^6 years half-life) decays through two beta emissions, one $\leq 5\%$ at 0.09 MeV to stable ^{93}Nb ; the other $\geq 95\%$ at 0.6 MeV to an excited ^{93}Nb state. The energy of the gamma of the excited ^{93}Nb is 30.4 keV with a half-life of 13.6 years. No unconverted gamma-rays have been observed. Chemical separation followed by beta analysis will allow 0.5 d/m to be measured.

Samarium-151 decays with an 87-year half-life through beta emission, to ^{151}Eu . Chemical separation and low-level beta analysis would allow 1 d/m to be determined.

Tin-126 with its 10^6 -year half-life decays 100% by beta emission to the excited state of ^{126}Sb , which is itself radioactive with meta stable state half-lives of 19 and 12.3 min. The photons of ^{126}Sb range in energy from 17.7 to 87.56 keV. Antimony-126 decays to ^{126}Te energy states that

produce a cascade of gamma-rays ranging in energy from 0.29 to 0.99 MeV.

After chemical separation of the ^{126}Sn has occurred, the daughter ^{126}Sb is allowed to come to equilibrium. The analysis of the ^{126}Sb coincident gamma-rays allows measurements to be made with sensitivities approaching 0.5 d/m for beta counting, 2 d/m for x-ray analysis, and 1 d/m for gamma-ray analysis. It is possible to analyze ^{126}Sn by neutron activation analysis mass spectrometry, and through the use of a laser technique.

In the future, the degree of analytical or chemical difficulty of the above mentioned techniques must be compared to the ease of analysis that may become available through the use of laser excitation followed by fluorescence analysis or ionization detectors of the excited species. This developing laser technique may also allow the limits of detectability to be so low that one would use it as a matter of choice, though it may require more sample preparation.

Computerization of Laboratory Counting Instruments J. R. Kosorok

A continuing program is maintained to develop and improve computer control of counting systems and calculation of radionuclide or trace element concentrations. A rapid and effective procedure for quantitative analysis of Ge(Li) gamma-ray spectra involves among other things a comparison of the sample weight and nuclide spectrum peak area of the sample with that of a known standard. The currently operating computer programs that efficiently perform this type of analysis are being improved to also provide information on spectral data reliability. The new program will determine all the statistical uncertainties associated with the measurement of each radionuclide.

In an effort to maximize counting efficiency and time, an automatic sample changer has been designed and is being built which will consecutively move samples into position under control of a digital device and will control spectrum collection and computer storage. Our development of this system became necessary after an investigation of commercially available units

indicated they could not be adapted to the existing system.

In x-ray energy spectrometry there is a continuing need for computer programs of various degrees of sophistication. We have developed a simplified program that provides quantitative results for elements with

non-overlapping x-ray lines. This simplified direct approach meets most of the normal requirements for research sample analyses without the uncertainties added by more complex programs. A compromise between this program and more complex Livermore programs will most likely be our ultimate choice.

In Situ Monitoring

Systems for in situ analysis of radionuclides around storage areas and nuclear sites are being developed. The development and initial use of a Ge(Li) system for down well radioactivity measurements was completed. Design criteria were developed and construction begun on a ^{90}Sr counter for use on natural soils.

Down Well In Situ High Resolution Gamma-Ray Spectrometry*

H. L. Nielson, H. G. Rieck and
R. L. Brodzinski

Field and laboratory studies using Ge(Li) and intrinsic germanium gamma-ray spectrometry techniques have demonstrated the feasibility of making rapid in situ concentration measurements of underground high-level radioactive wastes. This particular technique was developed specifically for application to the study of liquid wastes leaked into the soil from underground storage tanks located on the Hanford reservation, although its applicability to the remote determination of high-level radioactivities for other purposes is immediately obvious.

The technology developed uses a 20 cm^3 Ge(Li) diode (Figure 1.5) or a 0.5 cm^3 intrinsic germanium diode, with and without lead shielding, to measure radionuclides in the soil over a concentration range of 10^8 . Each diode is adapted to a cylindrical housing capable of passing down a 6-in. well casing. The probe is positioned at various depths in the wells which are strategically located around the underground storage tanks, and the data from counting intervals of about 10 min are recorded in a 2000 channel pulse height analyzer. A parallel line printer is used for a paper-tape printout of the gamma-ray spectrum after each count. In addition to the identity and the concentration of each radionuclide present, other pertinent information obtainable from the collected data includes:

- the relative motion, including the rate and direction, for each radionuclide present in the soil,

- the long-term soil retention characteristics of each radionuclide,
- the direction of the radioactivity relative to the diode position,
- the detection of "channeling" or "wicking" alongside well casings, and
- the location of the leak on the surface of the tank.

The technological capabilities of rapid in situ measurements of radionuclides was recently demonstrated in a field study investigating a leak in an underground storage tank located in the T Farm of the 200-West Area on the Hanford reservation. Specific details obtained from analyses of the data collected in this study included:

- the identity and concentration of each radionuclide present in the soil at various distances from the leak,
- the relative mobility of each radionuclide through the soil,
- the extent of both the horizontal and vertical migration of each radionuclide, and
- the location of the leak on the surface of the tank.

To take advantage of the total capability of the in situ technology, replicate measurements over an extended time period would be necessary.

Perhaps the most significant conclusion from the studies conducted is that (with an optimum working technology for in situ measurements of high-level radioactive wastes) field data can be rapidly collected, analyzed, and used to determine if a tank is leaking, and the extent of the leak.

* Only partially costed to DBER

FIGURE 1.5. Battelle's Well Logging Probe.

Although the technique has been developed specifically for application to problems associated with underground storage of high-level liquid wastes, many other in situ investigations can be accomplished with little or no modification to the existing equipment. Some prime examples include identification of radionuclides and concentrations in and around cribs, ponds, trenches, and dry-waste burial grounds. Only a slight modification is necessary for application of the technique to measurements under water, such as the determination of the radionuclide concentrations in a well filled with ground water. In addition, modifications to the system are being considered which will render it applicable to in situ underground neutron activation analysis when used in conjunction with a ^{252}Cf source.

Development of an Environmental Strontium-90 Counter

N. A. Wogman and J. A. Campbell

Strontium-90 is one of the more prominent radionuclides produced in

the nuclear power industry. It is also of major interest since when it is ingested, it concentrates within the skeletal system. In this study we describe the development of an environmental ^{90}Sr counter which is designed to make in situ measurements of the ^{90}Sr level in soil. In the design of the ^{90}Sr environmental counter, several factors were considered. Since the decay of ^{90}Sr is by the emission of beta particles, it was necessary to devise as selective a method as possible of measuring these beta particles in the presence of other beta and gamma radiation which would be emitted by other artificial and natural radionuclides. In Table 1.5 the relative concentrations of some of the long-lived radionuclides produced by the nuclear industry are shown for various decay periods following discharge from fuel processing plants. While one would encounter serious problems in selective measurement of ^{90}Sr after periods of just a few years, it is evident that after a decay period approaching 10 years, the expected abundance of most of the long-lived radionuclides is small compared to that of ^{90}Sr .

After 10 years of decay, the ^{137}Cs : ^{90}Sr ratio is 1.4, with ^{134}Cs and ^{147}Pm being about 1/10th of the ^{90}Sr , ^{154}Eu 1/20th that of the ^{90}Sr , and ^{151}Sm and ^{155}Eu 1/50th that of the ^{90}Sr . Thus, the ^{137}Cs and ^{90}Sr in old wastes predominate, with other species being less than 1/10th of the total radioactivity.

In the design of the ^{90}Sr counter, a low-background polycarbonate plastic dish 2.5 cm thick by 15 cm diameter was used for the counter body. Two 1/8 in. holes for gas ports and 10 small holes for stringing the 1 mil anode wire were drilled into the side of the counter cavity. The 1 mil stainless steel anode wire is stretched across the face of the counter five times with a wire/wire separation of 2.5 cm. The inside surface of the counter is coated with vapor deposited 99.999% pure Ag to form a cathode wall. The counter window of 0.3 mil aluminized mylar fits over the front of the counter and is held in place with a thin, plastic ring.

A beta counter similar to the one presently under construction, but smaller and containing more mass, was used to obtain beta absorption curves

TABLE 1.5. Total Curies of Major Radioactive Species¹
Produced for the Entire U.S. Nuclear Economy by 1974
and the Amounts Remaining After 1, 10, and 100 Year
of Decay.

	0 Decay	1 Year Decay	10 Year Decay	100 Year Decay
⁸⁹ Sr	5.28×10^6	-	-	-
⁹⁰ Sr	$3.09 \times 10^{7^2}$	3.01×10^7	2.41×10^7	2.60×10^6
⁹⁰ Y	$3.11 \times 10^{7^2}$	-	-	-
⁹¹ Y	9.87×10^6	-	-	-
⁹⁵ Zr	$1.87 \times 10^{7^2}$	3.93×10^5	-	-
^{95m} Nb	3.97×10^5	-	-	-
⁹⁵ Nb	3.79×10^7	-	-	-
¹⁰³ Ru	3.67×10^6	-	-	-
¹⁰³ Rh	3.68×10^6	-	-	-
¹⁰⁶ Ru	$9.60 \times 10^{7^2}$	4.84×10^7	-	-
¹⁰⁶ Rh	9.60×10^7	-	-	-
^{110m} Ag	4.72×10^5	-	-	-
¹²⁵ Sb	$2.60 \times 10^{6^2}$	2.02×10^6	-	-
¹²⁵ Te	1.07×10^6	-	-	-
^{127m} Te	6.57×10^5	-	-	-
¹²⁷ Te	6.50×10^5	-	-	-
¹³⁴ Cs	6.52×10^7	4.66×10^7	2.26×10^6	-
¹³⁷ Cs	4.30×10^7	4.2×10^7	3.42×10^7	4.3×10^6
^{137m} Ba	4.02×10^7	-	-	-
¹⁴¹ Ce	1.92×10^6	-	-	-
¹⁴⁴ Ce	$1.59 \times 10^{8^2}$	6.53×10^7	-	-
¹⁴⁴ Pr	1.59×10^8	-	-	-
¹⁴⁷ Pm	3.17×10^7	2.43×10^7	2.25×10^6	-
¹⁵¹ Sm	5.11×10^5	5.07×10^5	4.74×10^5	2.43×10^5
¹⁵⁴ Eu	2.71×10^6	2.5×10^6	1.2×10^6	-
¹⁵⁵ Eu	1.88×10^6	1.63×10^6	4.4×10^5	-

¹The table includes only those species which are in greater than 1% of the quantity of ⁹⁰Sr in any given year.

²Daughter radioactivity in equilibrium with long lived parent.

and determine optimum absorber thickness for selective ^{90}Sr counting (Table 1.6). For 10-year-old debris, ^{90}Sr and ^{137}Cs are the major species. If each produced 1000 c/m beta from a thin, massless source, the beta rate recorded would be 2000 c/m with no absorber. With a 280 mg Al/cm² absorber, 6 c/m ^{137}Cs and 150 c/m ^{90}Sr is detected. For a sample of soil

that is 1.2 cm thick containing the same amount of ^{137}Cs and ^{90}Sr as the massless source, the beta rate recorded is only 105 c/m, which illustrates soil absorption and the decrease in counting efficiency. With 280 Al/cm² absorbers, most of the ^{137}Cs electron emission is absorbed and the resultant count rate is primarily due to ^{90}Sr (5/6 of the total).

TABLE 1.6. Changes in a Beta Analysis System¹ Sensitivity Under Absorber Conditions for a Given Quantity of Radioisotope.

CASE 1

Source	Absorber mg Al/cm ²	Radionuclides, c/m		Σ^1
		^{137}Cs	^{90}Sr	
Thin, massless	0 ²	1000	1000	2000
	280	6	150	156
	1680	4	-	4
1.2 cm thick ³	0	29	76	105
	280	3	15	18
	1680	3	-	3

CASE 2

Source	Absorber Mg Al/cm ²	Radionuclides, c/m					Σ^1
		^{137}Cs	^{90}Sr	^{110}Ag	^{106}Ru	^{144}Ce	
Thin, massless	0 ²	1000	1000	10	10	10	2030
	280	6	150	0.9	4.9	2.8	165
	1680	4	-	0.5	0.02	-	5
1.2 cm thick ³	0	29	76	0.59	1.88	0.73	108
	280	3	15	0.37	.71	0.23	19
	1680	3	-	0.37	-	-	3

¹As described in this paper except constructed from thick Bakelite material.

²The initial activity was standardized to 1,000 c/m on a massless source.

³The same quantity of radionuclide used for massless source was spread uniformly throughout a 1.2-cm-thick pad of sample.

The question of the depth of material being analyzed can be determined qualitatively in the field for old fission wastes by the count rate ratio obtained through the use of various Al absorbers. If the material ($^{90}\text{Sr} + ^{137}\text{Cs}$) is on the surface of the soil, the count rate will be 13-fold less than 280 mg Al/cm² and 500-fold less with 1680 mg Al/cm². If it is uniformly distributed 1.2 cm deep, the count rate will drop only sixfold with 280 mg Al/cm and about 15-fold with 1680 mg Al/cm². Intermediate values will be obtained for

soil thicknesses between these two or if the material is at a depth greater than 1.2 cm. As is also shown in the table, 10 d/m of ^{106}Ru , ^{110}Ag , and ^{144}Ce which could be added from fresh fission debris to the environment would not mask the detection of ^{90}Sr by this technique. With these radioisotopes at 1% of the disintegration rate of ^{90}Sr , the error involved in the ^{90}Sr determination would be 3%. A system such as that described will be applied to the monitoring of the Hanford environs during 1975.

Activation Analysis Technology

Instrumental and chemical techniques were developed for the determination of rare element tracers which are being used for the study of atmospheric processes in storm systems. A comparison of the existing neutron activation analysis techniques with those possible by laser excitation indicates that much can be done if laser excitation fluorescence analysis can be used for future programs. A method was developed to analyze Pu concentrations at relatively low levels in high concentrations of beta-gamma radioactivity, using the spontaneous fission and (α, n) neutrons. The neutrons are measured by metal foil neutron activation in radiation fields too intense for normal electronic equipment.

Neutron Flux Determinations from Measurement of Activation Products in Metal Discs*

R. L. Brodzinski, J. A. Campbell,
N. A. Wogman

In order to measure plutonium concentrations at moderate to high levels in locations not readily amenable to ordinary analytical techniques (such as high-level radioactive waste storage tanks), a method using neutron-induced nuclear reactions in metal discs has been developed whereby the quantified radioactive products are related to the neutron fluence.

Since some of these reactions have characteristic minimum neutron energy thresholds, the flux of neutrons above any chosen energy can be determined as long as the reaction excitation function (cross section as a function of energy) is known.

Reactions that are currently being used are:

$^{63}\text{Cu}(n, \gamma)^{64}\text{Cu}$ for thermal neutrons

$^{63}\text{Cu}(\text{Cd shielded})(n, \gamma)^{64}\text{Cu}$ for epithermal neutrons

$^{58}\text{Ni}(n, p)^{58}\text{Co}$ for high energy neutrons

$^{48}\text{Ti}(n, p)^{48}\text{Sc}$ for high energy neutrons

$^{64}\text{Zn}(n, p)^{64}\text{Cu}$ for high energy neutrons

$^{24}\text{Mg}(n, p)^{24}\text{Na}$ for high energy neutrons

The calculated sensitivities as a function of energy for each reaction are listed in Table 1.7. As a reference point, it should be recalled that the cosmic ray neutron flux is $\sim 1.7 \cdot 10^{-3}$ n/cm²/sec at sea level.

Through use of the several different nuclear reactions simultaneously, it is anticipated that the neutron flux and energy spectrum can be determined at many locations of interest including nuclear waste tanks, cribs, ponds, and storage areas. It is also anticipated that this flux-energy spectrum can be used to define the source of the neutrons; that is, estimate the fraction coming from (α, n) reactions on oxygen, from (α, n) reactions on fluorine, and from spontaneous fission. This information should be sufficient to predict not only the plutonium concentration at the location in question but also lend information regarding the chemical composition of the system (percentage of oxygen and fluorine) and the americium concentration.

* Only partially costed to DBER

TABLE 1.7. Sensitivity Limits for Measuring Neutron Fluxes ($10^{-3}n\text{ cm}^{-2}\text{ sec}^{-1}$) by Induced Nuclear Reactions in Metal Discs 6-in. in Diameter by 1/4-in. Thick for Various Neutron Energy Intervals.¹

Nuclear Reaction	Thermal	Epithermal	Neutron Energy Interval (MeV)											
			≥3	≥4	≥5	≥6	≥7	≥8	≥9	≥10	≥11	≥12	≥13	≥14
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	0.6													
$^{63}\text{Cu}(\text{Cd})(n,\gamma)^{64}\text{Cu}$		1.3												
$^{58}\text{Ni}(n,p)^{58}\text{Co}$			62	31	23	18	17	16						
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$							570	260	160	100	78	62	56	52
$^{64}\text{Zn}(n,p)^{64}\text{Cu}$			62	29	23	19	17	16	16	16				
$^{24}\text{Mg}(n,p)^{24}\text{Na}$							12	6	4	4	4	4	4	4

¹Assumptions include a 10-day exposure of the nickel disc (all other reactions are for saturation conditions) and a decay period of 9 hr before determination of the induced radionuclides.

Rare Elements as Tracers of the Dynamic Processes in Storm Systems T. M. Tanner

A technique for the addition of trace amounts of rare elements to storm systems and their subsequent analysis in precipitation has been developed. Rare element tracers in the form of submicron aerosols are generated from aircraft-mounted burners and provide a means for obtaining information on air entrainment in convective storm systems. In this technique tracers are released near the edges of convective towers and the subsequently collected ground level precipitation sample is analyzed by neutron activation techniques.

A major problem in the analysis of the tracer-containing precipitation samples is that they also contain airborne dust and industrial pollutants. These materials interfere with the analysis of the tracers and may contain a significant amount of the elements which were used as tracers. Therefore, as a first consideration, the tracer elements selected should be low in natural abundance. Neutron activation analysis has been shown to be by far the most sensitive analysis technique for most all of the possible tracers. Based on the above criteria, the tracers which have been chosen and

used are Ta, Eu, In, Ru, Au, Ir and Re. These elements have low concentrations in average crustal material, and, with the exception of Ru, are easily measured at the nanogram concentration levels by neutron activation analysis as summarized in Table 1.8.

The detection limits shown in Table 1.8 will naturally vary somewhat with the type and amount of background material collected with the

TABLE 1.8. Detection Limits for Atmospheric Tracers Compared with Earth's Crust Levels.

Element	Crustal Avg. Conc. (ppm)	Det. Limit ¹
Ta	2	1 nanogram
Eu	1.2	1 nanogram
In	0.1	1 nanogram
Ru	0.01	5 micrograms
Au	0.004	0.2 nanogram
Ir	0.001	0.5 nanogram
Re	0.001	2.0 nanograms

¹Based on 3σ counting statistics with Ge(Li) analysis of induced gamma-ray activity in the instrumental neutron activation analysis (INAA).

precipitation samples. In some instances it has been necessary to perform radiochemical separations to analyze for some of the tracers in rain samples. This was true in the National Hail Research Experiment (NHRE) study where many tons of soil were swept into the storm systems by updrafts and subsequently redeposited in the precipitation samples. With separations procedures, the 3 σ detection limit can be lowered by a factor of about 5. However, radiochemical separations are time consuming and increase the cost of each sample analysis relative to strictly instrumental methods. Where precipitation samples are expected to be "dirty," only those tracers which rarely require separation (Ta, Eu, Au and Ir) should be used. Other elements not requiring separation should be found to replace those which do. Listed in Table 1.9 are elements which have relatively low concentration in crustal material and which were also considered as possible tracers. Of those listed in Table 1.9 Lu, Tl, Tm, Bi, Pd and Rh cannot be measured by INAA scheme because of their lack of suitable nuclear parameters for neutron activation and γ counting. The elements Cd, Se and Te are released to the atmosphere in large amounts by anthropogenic sources and could not be used in highly populated areas such as St. Louis, because of their highly variable atmospheric concentrations. They could, however, be used in remote areas. Platinum can be detected at about the 20 ng level from its ^{197}Pt daughter after a chemical separation and would then be suitable for some applications. Osmium with its (n, γ) product, ^{191}Os , may be used for INAA at about the 10 ng level.

TABLE 1.9. Other Rare Elements Which Might Serve as Atmospheric Tracers.

Element	Crustal Avg. Conc. (ppm)	Element	Crustal Avg. Conc. (ppm)
Lu	0.5	Pd	0.01
Tl	0.5	Pt	0.01
Tm	0.5	Te	0.01
Bi	0.2	Os	0.005
Cd	0.07	Rh	0.005
Se	0.05		

Techniques for Measuring Trace Elements in Fuels and Fuel Additives T. M. Tanner and L. A. Rancitelli

Essentially all liquid fuels contain various chemical additives which are designed to improve their combustion properties. In order to analyze the combustible materials we have developed a suitable technique for their neutron irradiation and subsequent measurement.

Samples are transferred from glass bottles in which they are originally collected to precleaned 30 ml polyethylene vials for irradiation. Two 12-to 15-ml aliquots of each sample are prepared and used for a short and long irradiation. The vials are then heat sealed with a hot high-purity quartz rod. Since the concentration of the elements are reported in $\mu\text{g/ml}$ and the viscosities of the samples vary greatly (from highly volatile fuels to heavy oils) it is necessary to determine the density of each sample. This is done by weighing a carefully measured volume of each. The irradiated samples can then be weighed and the elemental concentration determined in $\mu\text{g/ml}$.

For the short irradiations, the samples, standards and flux monitor solutions are exposed for two minutes in a rotating high-density polyethylene tube at the Washington State University reactor at a neutron flux of about $5 \times 10^{12} \text{ n cm}^{-2} \text{ sec}^{-1}$. After irradiation, the samples and standards are quickly transferred to preweighed counting containers and analyzed on a Ge(Li) γ -ray spectrometer. In this way the short lived isotopes: ^{28}Al (2.3 min), ^{80}Br (18 min), ^{49}Ca (8.8 min), ^{66}Cu (5.1 min), ^{38}Cl (37.3 min), ^{27}Mg (9.5 min), ^{56}Mn (2.58 hr), ^{24}Na (15 hr), ^{123}mSn (39.5 min), ^{25}mSn (9.5 min), ^{51}Ti (5.8 min), ^{52}V (3.8 min), ^{69}mZn (13.8 hr) and ^{116}mIn (54 min) can be measured.

From these measurements and using the standard computer method, the concentrations of the elements Al, Br, Ca, Cu, Cl, Mg, Mn, Na, Sn, Ti, V, Zn and In can be determined (Table 1.10).

For the long irradiation the samples and standards are exposed in a second polyethylene container for 2 hr at $5 \times 10^{12} \text{ n cm}^{-2} \text{ sec}^{-1}$, allowed to "cool" for a few days and then transferred to weighed containers and the

gamma-ray spectrum measured on a Ge(Li) detector for 10 min. From this spectrum the elements As, Au, Br, K, La, Na and Sm are determined (Table 1.10). After a decay period of about 3 weeks,

the samples and standards are again counted for 100 min each and the elements Ag, Cd, Co, Cr, Cs, Eu, Fe, Hf, Hg, Ni, Rb, Sb, Sc, Se, Ta, Th and Zn are determined.

TABLE 1.10. Concentration Ranges in EPA Motor Oils and Fuel Additives.

	<u>Motor Oils</u> (µg/ml)	<u>Fuel Additives</u> (µg/ml)
Ag	<0.08	<0.2
Al	1.4 - 3.7	<0.01 - 5.5
As	<0.01 - 0.3	<0.001 - 0.2
Au	<0.002	<0.0001 - 0.004
Br	0.06 - 1.3	<0.002 - 7.8
Cd	<0.1 - 0.4	<0.1
Cl	4 - 160	1 - 5890
Co	<0.0004-0.006	<0.0001-0.36
Cr	<0.1 - 0.28	<0.01 - 0.1
Cs	<0.02	<0.04
Cu	<1	<1 - 1.3
Eu	<0.0004	<0.0004
Fe	<2 - 8	<0.2 - 560
Hf	<0.02	<0.02
Hg	<0.04	<0.09
In	<0.002 - 0.12	<0.002 - 0.04
K	<0.6 - 14	<1 - 24
La	<0.002-0.014	<0.001 - 0.008
Mg	<20 - 1170	<20 - 4740
Mn	<0.04 - 0.3	<0.01 - 2.7
Na	3 - 15	0.02 - 17
Ni	<4	<4
Rb	<0.3 - 1.1	<0.01 - 2.8
Sb	<0.005-0.25	<0.001-0.06
Sc	<0.002	<0.002
Se	<0.06	<0.02
Sm	<0.001 - 0.034	<0.0002-0.002
Sn	<6	<0.4 - 140
Sr	<10	<10
Ta	<0.006	<0.02
Tb	<0.007	<0.02
Th	<0.009	<0.02
V	<0.01-0.04	<0.01-0.2
Zn	1040-1990	0.2-6390

Applicability of a Homogenized IAEA
Oyster Tissue as a Standard Reference
Material

L. A. Rancitelli

The IAEA is preparing standard reference materials which can be circulated through laboratories in member states to permit them to determine the reliability of their trace

element measurements in marine biota. To meet this objective a large sample of oyster tissue was freeze-dried and homogenized by the IAEA laboratory in Monaco. Our present work has been concerned with determining the homogeneity of this material for specimens of 0.1 and 0.25 g. Presented in Table 1.11 are sample concentrations of several trace elements which were

TABLE 1.11. Elemental Content in IAEA Oyster Tissue ($\mu\text{g/g}$).

Sample No.	Sample Weight (g)											
		La	Ni	Rb	Sb	Se	Sm	Sn	Ta	Tb	Th	Zn
1	0.1004	<0.4	<10	<3	<0.02	2.29 ± 0.25	<0.04	<20	<0.008	<0.02	<0.09	4780 ± 6
2	0.1002	<0.4	<10	<3	<0.02	2.34 ± 0.25	<0.04	<20	<0.008	<0.02	<0.09	4700 ± 6
3	0.1004	<0.4	<10	<3	<0.02	2.34 ± 0.25	<0.04	<20	<0.008	<0.02	<0.09	4630 ± 6
4	0.1001	<0.4	<10	<3	<0.02	2.17 ± 0.26	<0.04	<20	<0.008	<0.02	<0.09	4760 ± 6
5	0.1001	<0.4	<10	<3	<0.02	2.50 ± 0.26	<0.04	<20	<0.007	<0.02	<0.09	4730 ± 6
6	0.0998	<0.4	<10	<3	<0.02	2.33 ± 0.26	<0.04	<20	<0.008	<0.02	<0.10	4840 ± 6
Ave. ±		<0.4	<10	<3	<0.02	2.33 ± 0.11	<0.04	<20	<0.007	<0.02	<0.09	4740 ± 72
A	0.2496	<0.3	<8	<2	<0.01	2.27 ± 0.17	<0.03	<15	<0.005	<0.02	<0.06	4480 ± 4
B	0.2502	<0.3	<8	<2	<0.01	2.84 ± 0.18	<0.03	<15	<0.005	<0.02	<0.07	4580 ± 4
C	0.2499	<0.2	<8	<2	<0.01	2.65 ± 0.17	<0.03	<15	<0.005	<0.02	<0.07	4610 ± 4
D	0.2505	<0.3	<8	<2	<0.01	2.30 ± 0.17	<0.05	<15	<0.005	<0.02	<0.07	4340 ± 4
E	0.2497	<0.2	<9	<2	<0.01	2.51 ± 0.17	<0.03	<15	<0.005	<0.02	<0.07	4500 ± 4
F	0.2504	<0.2	<9	<3	<0.01	2.45 ± 0.18	<0.03	<15	<0.005	<0.02	<0.07	4590 ± 4
Ave. ±		<0.2	<8	<2	<0.01	2.50 ± 0.22	<0.03	<15	<0.005	<0.02	<0.06	4520 ± 101
Sample No.	Sample Weight (g)	Ag	Co	Cr	Cs	Eu	Fe	Hf	Hg	K		
1	0.1004	3.75 ± 0.12	0.238 ± 0.006	2.34 ± 0.39	<0.05	<0.002	194 ± 7	<0.05	<0.1	<5000		
2	0.1002	3.56 ± 0.12	0.260 ± 0.006	1.33 ± 0.41	<0.05	<0.002	196 ± 7	<0.05	<0.1	<5000		
3	0.1004	3.30 ± 0.12	0.243 ± 0.006	0.93 ± 0.41	<0.05	<0.002	192 ± 7	<0.05	<0.1	<5000		
4	0.1001	3.49 ± 0.12	0.230 ± 0.006	1.37 ± 0.41	<0.05	<0.002	192 ± 7	<0.05	<0.1	<5000		
5	0.1001	3.49 ± 0.12	0.274 ± 0.007	0.98 ± 0.43	<0.05	<0.002	169 ± 7	<0.05	<0.1	<5000		
6	0.0998	3.38 ± 0.12	0.250 ± 0.006	2.03 ± 0.45	<0.05	<0.002	185 ± 7	<0.05	<0.2	<6000		
Ave. ±		3.49 ± 0.16	0.249 ± 0.016	1.50 ± 0.57	<0.05	<0.002	192 ± 11	<0.05	<0.1	<5000		
A	0.2496	3.88 ± 0.18	0.246 ± 0.009	1.55 ± 0.52	<0.08	<0.002	184 ± 10	<0.06	<0.2	<4000		
B	0.2502	3.24 ± 0.18	0.226 ± 0.009	1.10 ± 0.52	<0.08	<0.003	207 ± 10	<0.06	<0.2	<4000		
C	0.2499	3.23 ± 0.18	0.204 ± 0.009	2.09 ± 0.53	<0.08	<0.002	194 ± 10	<0.06	<0.2	<4000		
D	0.2505	3.36 ± 0.18	0.261 ± 0.010	1.58 ± 0.56	<0.08	<0.002	197 ± 10	<0.07	<0.2	<4000		
E	0.2497	3.34 ± 0.18	0.238 ± 0.009	2.26 ± 0.57	<0.08	<0.003	178 ± 10	<0.07	<0.2	<4000		
F	0.2504	3.42 ± 0.18	0.298 ± 0.010	2.75 ± 0.60	<0.08	<0.002	211 ± 11	<0.07	<0.2	<4000		
Ave. ±		3.41 ± 0.24	0.246 ± 0.032	1.89 ± 0.59	<0.08	<0.002	195 ± 11	<0.06	<0.2	<4000		
Sample No.	Sample Weight (g)	Au	As	Br	La	Na	Sm					
1	0.1004	0.37 ± 0.1	9.9 ± 1	225 ± 2	<0.43	28,800 ± 100	<0.026					
2	0.1002	0.10 ± 0.1	9.9 ± 1	216 ± 2	<0.43	27,800 ± 100	<0.027					
3	0.1004	0.20 ± 0.1	10.4 ± 1	215 ± 2	<0.42	27,800 ± 100	<0.025					
4	0.1001	0.38 ± 0.1	9.5 ± 1	213 ± 2	<0.42	27,300 ± 100	<0.026					
5	0.1001	0.42 ± 0.1	10.0 ± 1	227 ± 2	<0.44	29,000 ± 100	<0.026					
6	0.0998	0.10 ± 0.1	10.5 ± 1	222 ± 2	<0.43	28,400 ± 100	<0.026					
Ave. ±		0.26 ± 0.1	10.0 ± 0.4	220 ± 6	<0.43	28,200 ± 700	<0.026					
A	0.2496	0.18 ± 0.06	10.2 ± 1	214 ± 2	<0.25	28,100 ± 100	<0.026					
B	0.2502	0.20 ± 0.06	10.8 ± 1	215 ± 2	<0.26	28,800 ± 100	<0.027					
C	0.2499	0.22 ± 0.06	10.3 ± 1	206 ± 2	<0.25	27,200 ± 100	<0.025					
D	0.2505	0.22 ± 0.06	10.1 ± 1	212 ± 2	<0.25	28,000 ± 100	<0.026					
E	0.2497	0.16 ± 0.06	10.0 ± 1	208 ± 2	<0.25	27,200 ± 100	<0.026					
F	0.2504	0.20 ± 0.06	9.7 ± 1	212 ± 2	<0.25	27,800 ± 100	<0.026					
Ave. ±		0.20 ± 0.02	10.2 ± 0.4	211 ± 3	<0.25	27,800 ± 600	<0.026					

measured at our laboratory by neutron activation analysis on six replicate samples of about 0.1 and 0.25 g. From these data we conclude that the sample is homogeneous on both a 0.1 and 0.25 g aliquot basis except in the case of iron. Here the relatively large variation on one of the 0.1 g samples and also on a 0.25 g sample

suggests that iron is simply inhomogeneously distributed throughout the sample. The IAEA Committee on Standards will use this data to evaluate the applicability of the oyster tissue as a standard reference for distribution to member state laboratories.

Chemical and Molecular Composition Technology

Freon was analyzed from samples collected in St. Louis, Missouri, and Quillayute, Washington, for use as a conservative tracer. In addition, a procedure using the complexation of a metal ion was developed which allows its detection by gas chromatography. Selenium in sea water has been measured in this manner and technique development is under way for complexes of Be, Cr and Al.

Gas Chromatographic Analysis of Environmental Pollutants

M. R. Petersen

Analysis of trace pollutants is necessary to evaluate the environmental impact resulting from an increase in power generation, industrial processes, transportation, and agricultural growth. Numerous methods and kinds of instrumentation find use in analyzing and monitoring environmental pollutants. Gas chromatographic techniques are widely used, particularly in analysis of trace organic compounds. Even within the limits inherent in the technique, gas chromatography offers excellent analytical sensitivity for a wide variety of compounds and unsurpassed resolving capabilities of complex mixtures.

In analyses of gases, sample preparation can often be very minimal. For example, fluorocarbons or light hydrocarbons in ambient air are usually analyzed by direct introduction of a known air volume into the chromatograph. By properly choosing the absorbent, operating conditions, and the optimum detector, one can separate the desired components sufficiently to obtain quantitative values with suitable standards. These types of procedures have been developed in our laboratory for Freon analysis in

air samples collected in St. Louis and at Quillayute, Washington.

When the concentration of the gaseous species of interest is too low for direct analysis, some type of concentrating method is necessary. Cold traps have reportedly been tried but often complicate the analysis, due to the water vapor collected as ice in the trap. A more suitable method, which we have adapted, consists of the use of selective absorbents. There are various kinds of synthetic resins available which have been used to absorb non-polar organic compounds from aqueous or gaseous phases. When the sampling is completed the concentrated material is then removed from the absorbent and the analysis carried out using gas chromatography.

Trace metal analysis is a recent innovative use of gas chromatography. This technique consists of complexing the metal ion with a suitable chelating agent which is readily detected using a gas chromatograph fitted with an electron capture detector. Presently the technique is very sensitive for a few complexes of beryllium, chromium, aluminum, and selenium. The technique is less sensitive for other metals which form complexes including cobalt, zinc, some lanthanides, iron, palladium, and nickel. A selenium procedure based on this approach has been developed to analyze sea water samples for selenium.

Environmental Pollution Analysis

To understand the physical and chemical behavior of inorganic and organic pollutants in the environment, it is essential that techniques be developed to sample and measure pollutants at their source, during their transport, and throughout the environment. To meet some of these needs: 1) a method was developed for the in situ analysis of trace element pollutants in sediments by x-ray fluorescence which can measure up to 30 elements at the 20-50 ppm levels; 2) plasma excitation techniques were developed for chemical species determination of As and Sb; 3) sampling methods and gas chromatograph techniques were developed for inorganic and organic gases; and 4) feasibility studies have been made of continuous injection mass spectrometry and laser excitation analysis of isotopic trace elements and molecules at levels of 5 to 10 orders of magnitude improvement.

Feasibility of Laser Excitation
Analysis of Trace Elements, Organic
Compounds, and Molecular Structure
M. R. Petersen, R. W. Perkins,
L. A. Rancitelli and N. A. Wogman

Due to the growing use of fossil fuel power generation, the need is increasing for improved sensitivity in the measurement of trace elements and trace organic compounds. Through the recent development of tunable dye lasers it is now possible to selectively excite elements or molecules which absorb photons in the visible and infrared light ranges. Presently in our laboratory different ways are being considered to extend selective excitation to species which absorb in the ultraviolet range and to apply novel techniques of detecting the excited species.

Light produced by a tunable dye laser has two superior properties compared to light from diffuse sources dispersed by a prism or grating. The photon flux is orders of magnitude higher and with suitable techniques the photons possess greater monochromaticity ($1:10^8$ compared to $1:10^6$ with the best grating spectrometers). These two properties can be used to selectively excite an atom or molecule from one energy state to a higher state by resonant absorption of the

laser-produced photons. By coupling this selective excitation with some other nonselective excitation source, many other species of interest which absorb in the ultraviolet (uv) and vacuum uv ranges of the spectrum can be selectively excited.

Selectively excited species can be detected in several ways. Fluorescence analysis is one widely used technique which is applicable for analysis of solids, liquids or gases. The sample is radiated with an intense laser beam which the species of interest absorbs and the resulting fluorescence photons are detected with high-efficiency photon counting systems. This of course has the potential of allowing a response from every atom or molecule which is present, at least in a gaseous sample. Since the excitation line is spectrally narrow and tunable, many different molecules can conceivably be analyzed without the necessity of complicated sample preparation.

Another very promising application of laser excitation is selectively photo-ionizing the species of interest and detecting the resulting ion pair by proportional counting techniques. The theoretical sensitivity attainable by this technique again exceeds by many orders of magnitude any present

analytical method for isotopes, trace elements or organic compounds. The selectivity of the laser-produced photons allows the selective excitation of isotopes of the same element, which makes possible isotopic analysis within elements and molecules. Selective photo-ionization appears possible only in the gas phase, so solids and liquids will have to be vaporized for analysis, which complicates the proposed procedure.

The use of laser excitation analysis is a potentially powerful technique for analysis of radio- and stable-isotopes, trace elements, organic compounds and their molecular structure. Use of the full sensitivity of this method could lower the detection limits for many substances by several orders of magnitude.

In Situ X-Ray Fluorescence Analysis on the Ocean Floor

N. A. Wogman, H. G. Rieck and J. R. Kosorok

Many toxic elements are released to the marine sediments and an in situ method to measure their distribution and thus allow an evaluation of the hazards to man is desirable. An energy dispersive x-ray fluorescence analyzer has been developed which permits in situ analyses for up to 30 elements at concentrations greater than 50 ppm in sediments at water depths down to 100 m.

The detector for this system is an 80 mm² Si(Li) diode which is enclosed in a 25-cm-diameter x 60 cm pressure housing. Cooling during underwater operation is provided by solid cryogen located within the pressure housing, and liquid nitrogen serves to freeze the cryogen prior to use. The preamplifier is located inside a watertight pressure housing on top of the detector assembly. Electrical connection to the amplifier housing is made through a five-pin marine electrical bulkhead connector.

A collimator located below the bottom surface of the diode contains the ¹⁰⁹Cd excitation source, and serves to direct excitation photons to the underlying sediment. The fluorescence x rays from the sediment reach the diode through a 20 mil Be plus a 12-mm-diameter, 5-ml-thick Be window. A further purpose of the collimator is to provide shielding between the

detector and the ¹⁰⁹Cd source. The collimator and source holder shown in Figure 1.6 were fabricated from 99.999 pure silver and have overall dimensions of 53.5 mm diameter and 13 mm thick. The collimator opening may be reduced to 5.1 mm by installing an insert. The bottom of the collimator assembly is 4 mm above the pressure hull Be window. The data acquisition system is built around a Nuclear Data 4420. A Nuclear Data 812 central processor controls data acquisition, storage, display and computation operations. X-ray data is stored on magnetic tape cassettes for historical record. The diode signal is fed through an optically coupled preamplifier to an amplifier. The amplifier output signal is processed by an analog to digital converter and the resulting digital signal is stored by the computer (see Figure 1.7). Connection between the diode preamplifier and the amplifier is through a multiple conductor underwater cable. Penetration into the preamplifier pressure housing is through a five-pin underwater bulkhead connector. The underwater cable contains wires for the ±24 volts dc power, the preamplifier signal, the reject pulse signal and a ground. Plus and minus 24 volts is

FIGURE 1.6. Collimator and Radioisotopic Source Holder for an In Situ X Ray Fluorescence Marine Analyzer.

FIGURE 1.7. Electronic Flow Diagram of the In Situ X Ray Fluorescence Marine Analyzer.

used in the preamplifier housing to provide power for the preamplifier and a high voltage generator to supply bias voltage to the diode. This circumvents the problem of supplying high voltage through the multiconductor cable. The signal and reject pulse conductors are coaxial cables. The ± 24 volts is supplied to the underwater cable from a Nim-bin chassis which also powers the amplifier and an ADC.

The x-ray detector as described was attached to the Beaver Mark IV research submersible to test feasibility of using the x-ray fluorescence system for mapping pollution in marine sediments. The detector assembly was attached to the aft end of the submarine using an "A" frame assembly attached to the skids which support the submarine when it rests on the ocean floor. The "A" frame pivoted around this connection and an anchor cable attached to the center part of the "A" frame was used to raise and lower the detector.

The cable was attached to a hydraulically operated winch which was

controlled by the pilot. During a typical measurement the submarine was positioned on the ocean floor, the anchor winch cable was lowered causing the "A" frame and detector assembly to rest on the ocean sediments, and the spectrum was recorded for a given time period. The "A" frame detector assembly had a negative buoyancy of about 16 kg and therefore plastic floats were attached to reduce this to 4 kg. This force would cause the diode to be pressed firmly into the sediment to insure complete H_2O displacement. After measurements were made the anchor winch cable was retrieved causing the "A" frame to raise, thus lifting the x-ray detector clear from the ocean bottom. The submarine then proceeded to a new station. The connection between the x-ray detector and the data acquisition system was made through the submarine hull by a 40-pin watertight penetrator. Security for the source was provided by a 1/4 in. steel cable permanently attached to the pressure housing-detector, extending back through the "A" frame to a rigid member of the submarine. A 28 to 110 volt ac inverter was used to supply

power for the data acquisition system using the submarine battery power. Spectra taken by the system on board Beaver allowed the determination of Cr, Fe, Ni, Cu, Zn, Pb, Ag, Br, Rb, Sn, and Zn at concentration levels down to 50 ppm.

Sampling and Analysis of Atmospheric Gases

M. R. Petersen and N. A. Wogman

A scheme of sample collection and analysis has been developed for the measurement of Freon 11, Freon 12 and CO in the atmosphere downwind from metropolitan areas. These inert tracers can serve an important role in the study of air mass movement and in predicting the transport and deposition behavior of air pollutants. These gases were chosen as tracers be-

cause all are emitted due to man's activity and are conservative gases; that is, they do not readily decompose, and mixing is the predominate dilution process. They have served as conservative tracers in the METROMEX study.

An air sampler was designed which consists of a lucite tube (5 cm I.D.) and plunger connected to a dc motor. The motor is geared so that the plunger can be withdrawn over 1 or 2 hr periods filling the 500 ml tube with a time-integrated air sample. A fitting was added to the bottom of the tube so the air sample could subsequently be removed through a stainless steel tube to an evacuated gas bottle. The system was mounted on a tripod for field use (Figure 1.8). The concentrations of tracers in

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FIGURE 1.8. A Time Integrating Air Sampler Operating at a METROMEX Field Station.

these integrated air samples appear to be consistent for samples obtained simultaneously.

We have demonstrated a satisfactory technique for analysis of the Freons using a gas chromatograph fitted with an electron capture detector. Optimal separation and analysis was achieved with a 6 ft column, packed with 10% UC-W98 on 80-100 Chromosorb W AWMCS, at room temperature using a W2 carrier gas flow rate of 15 ml/min. Standards of the Freons are prepared by multiple dilution of the pure gases in glass flasks to the range 0.1-1.0 ppb.

Only 3-5 ml of air is needed for Freon analysis; the remainder is used for carbon monoxide analysis with a Beckman CO Analyzer Model DIF 7000. It is a continuous, nondispersive infrared analyzer with special compensation for interference from water vapor. The instrument is connected to a closed-loop stainless steel system of tubing, valves and a manifold which can operate independently of the closed loop. After the gas bottle is connected to the evacuated manifold, the air sample is introduced; a pressure reading taken; and then the sample in the manifold is brought up to atmospheric pressure with CO-free air. The manifold containing the transferred sample is switched into the loop and the concentration was recorded on a strip chart. Standards are run in a similar manner using carbon monoxide-nitrogen mixtures from Matheson Gas Products. The concentration of carbon monoxide is computed by comparing the pen deflections of standard gas mixtures and air samples and applying the dilution factor obtained from the pressure reading.

The collection and analysis of these three gases, Freon 11, Freon 12 and carbon monoxide exemplify specialized instrumental techniques which have been developed and then applied directly to atmospheric transport of air pollutant studies.

Potential Methods for Real-Time
Computer-Aided Data Analysis
J. R. Kosorok

A computer can serve as a research tool and be applied as a storage device for data originating from many analytical instruments. It is thus

capable of aiding the scientist in his understanding of data. This past year two different computer-based procedures for providing computer-aided analysis of complex research data have been investigated.

In research projects involving the analysis of large numbers of samples, there is frequently the necessity for identifying trends in individual parameters or correlations among two or more parameters. This can be frequently performed by standard statistical analysis procedures, which are available in "canned" form on large computers in automatic data processing centers. Obtaining the results of these analyses involve preparation of the data on punched cards and then waiting a day or more for the results. Research projects involving short-lived phenomena or those requiring frequent reconfiguration of the experimental apparatus can often provide much more timely and better quality data if at least some of the more common statistical tests (involving regression analysis, for example) can be performed during the experimentation. Methods are being investigated to allow statistical analysis of data near the research area with minimal effort on the part of the researcher. Programs that will run on the typical small laboratory data collection computer and will interface to statistical analysis programs are being developed so that minimal computer operation or data handling are required by the researcher.

This method was applied in a recent study of trace elements in urban aerosols where correlation of concentration with location and time of sampling was required. The large number of measurements (approximately 3500) required processing with a "canned" statistical analysis program. In a second study the method has served to formulate a least squares fit of a series of analytical standards to develop a calibration curve for the x-ray fluorescence analysis of stable iodine in milk.

When data must be collected in a harsh environment, such as on the ocean floor, it is often difficult to obtain large amounts of data. Therefore, the maximum amounts of information must be extracted from whatever data can be obtained. Methods are being investigated for allowing a

user to interact with a laboratory computer system to enhance information by combining the reasoning ability of the user with the computational speed of the computer. Methods for providing stripping and transform analysis procedures on laboratory data are being investigated. X-ray fluorescence spectra collected during the New York Bight experiment required extensive analysis procedures to extract the maximum amount of information. Computer analysis programs of the type described here can reduce the time and manual effort required for such work by several fold.

Feasibility of Anodic Stripping Voltametry for Trace Element Analysis of Solutions

E. A. Crecelius

Anodic stripping voltametry (ASV) is an electrochemical analytical technique that will be used for the direct analysis of several elements (Pb, Cu, Cd, Zn currently under study) in natural waters. The ASV method is analytically attractive because the measurements are direct, rapid, sensitive to ppb levels, and can be used on environmental samples for pollution studies.

The ASV technique achieves great sensitivity by electrolytically reducing trace metal ions on a mercury electrode for a given length of time. The metals are then reoxidized (stripped) from the electrode by varying the potential in the positive direction. By monitoring the anodic current during stripping of the metal, the concentration of metals in solution can be determined.

The ASV system which should be installed early in 1975 will first be used to determine the amount of free and organically bound copper in the seawater used to cool a nuclear power plant. Elevated levels of copper occur in the cooling water as the result of corrosion of the copper alloy heat exchangers. The amount of free ionic copper and organically bound copper will be determined by analyzing water samples before and after photo-oxidation of the organic material.

Another application of this instrument will be to determine the concentrations of IO_3^- and I^- in solution by using the instrument in the differential pulse polarographic mode. This

technique applies a series of constant voltage pulses between a mercury dropping electrode and a counter electrode. The current flowing between the electrodes, at a specific potential is related to the concentration of a particular chemical species present in the solution. The concentration of IO_3^- will be determined and then I^- oxidized to IO_3^- and the sample reanalyzed.

Analysis of DDT and Its Degradation Products in the Atmosphere

M. R. Petersen

A procedure has been developed for the collection of particulate and gaseous DDT and its degradation products by an aircraft for subsequent laboratory analysis. These techniques have already been applied in an atmospheric transport study of the DDT used in a spraying program of the Tussock moth infestation in a portion of the Pacific Northwest forests. A sampling technique was designed for use on the Battelle aircraft to collect particulate and gaseous forms of DDT and its degradation products over and downwind of spray areas.

For sampling, a glass fiber filter (293 mm diameter; type A) was mounted on the holder of the aircraft sampler and a cylindrical polyurethane foam plug (140 mm diameter x 152 mm) was inserted behind the filter. The filters were first fired at 400°C overnight and the plugs were extracted (Soxhlet) with benzene for 24 hr in order to remove any interfering organic compounds. Samples were collected over the spray areas flying a specific pattern for up to 1 hr at flow rates between 1.7 and 2.8 scm/min.

To remove the adsorbed pesticides, the filters and foam plugs were extracted with hexane and the extracts were concentrated to a 0.5 ml. Samples collected on the plugs required a cleanup step consisting of column chromatography on neutral alumina, activity III. The analyses were performed on a gas chromatograph fitted with an electron capture detector.

In general, the pesticide levels decreased over the three day sampling period after spraying. The proportion of the gaseous phase, defined as that trapped on the backup urethane foam

plug, became greater over the particulate phase during the sampling period. The concentrations also decreased with increasing altitude. The proportion of the degradation product, DDE, increased with time. The highest levels of particulate DDT of about 500 ng/m^3 occurred near the ground several hr after spraying. Particulate concentrations diminished considerably in several days to about 5 ng/m^3 . Gaseous concentrations after two days were about 10 ng/m^3 . In similar samples collected two months later, the levels were below the detection limits of 50 pg/m^3 for both the gaseous and particulate phases of DDT and its degradation products.

This technique appears very effective for collecting and measuring the total DDT and its derivatives. The adaptation of sampling from an aircraft requires that the ambient DDT concentrations are at least 20-fold higher than the reported ambient levels which range $10\text{-}20 \text{ pg/m}^3$ in maritime air. To permit measurements at these levels would require a major redesign of the aircraft sampling system to increase flow rates without prohibitively lengthening the sampling period.

High Sensitivity X-Ray Fluorescence Analysis by Selective Separations E. A. Crecelius

Analytical techniques applicable to environmental pollution problems are being developed using x-ray fluorescence analysis combined with selective elemental chemical separations. X-ray fluorescence techniques can be used for rapid, multielement analysis of submicrogram quantities in a variety of sample matrices. Selective chemical separations are used to concentrate groups of elements or separate single chemical species. Separation techniques studied include exchange and chelating resins, liquid-liquid extractions and coprecipitations.

The first two analytical procedures which have been studied are for the analysis of iodide ion in milk. In the first method iodide ion is extracted from milk by anion exchange resin. It then is measured by x-ray fluorescence analysis of the iodine contained on the resin. The iodine $K \alpha$ x rays measured with a Si(Li) detector are produced by excitation from

either an Americium-241 radiation source or by a tungsten tube bombardment of neodymium secondary target. In the second method, the milk is freeze dried and pressed into a pellet which can be analyzed for total iodine directly by x-ray fluorescence. The limit of detection from these procedures is $10 \text{ } \mu\text{g}$ iodide and the precision (one standard deviation) for $50 \text{ } \mu\text{g}$ of iodide is $\pm 12\%$. By using 100 ml of milk the concentration of iodide can be determined over the normal range of iodide in milk, from $0.1\text{-}3 \text{ ppm}$ ($0.1\text{-}3 \text{ mg/l}$). The next step being developed in the iodide-milk procedures is an intercalibration of these procedures with a method that determines total iodine in milk.

Plasma Excitation Emission Spectrometry Applied to Chemical Speciation of Trace Metals in Solutions E. A. Crecelius

The chemical behavior and toxicity of an element depends to a large degree on its chemical species. Very few techniques are available for the determination of the chemical speciation of trace metals in solutions at low concentrations. The concentration and chemical speciation of several trace metals in solutions and biological matrices can be determined by plasma excitation emission spectrometry combined with the appropriate chemistry. This analytical technique is sensitive to nanogram amounts of arsenic, antimony, mercury, selenium, germanium and bismuth. Further developments will undoubtedly extend the number of elements which can be measured.

The instrumentation now in use is equipped with a direct current discharge excitation source (see Figure 1.9). The electrical arc creates a helium plasma capable of producing characteristic spectral emission lines from all elements in the helium carrier gas. The monochromator is tuned to the emission line of the element of interest. Chemical procedures have been developed to selectively volatilize from solution, and transport into the arc, gaseous compounds and volatile derivatives of compounds. Chemical species that can be determined include As^{+5} , As^{+3} , methylarsonic acid, dimethylarsinic acid, Sb^{+5} and Sb^{+3} . At a pH of three-four, sodium borohydride will reduce As^{+5} ,

Sb^{+3} and methylarsenics to volatile hydrides which are swept into a cold trap and subsequently swept into the detector. By controlled heating of the cold trap arsine, methylarsine and dimethylarsine peaks can be resolved. The sample can then be analyzed for As^{+5} or Sb^{+5} by adjusting the pH to one-two and adding more sodium borohydride. The detection limit for these chemical species is approximately one ng. Technique development is underway for other chemical species.

These chemical speciation techniques will be applied to environmental pollution problems. Of particular interest will be chemical transformations which occur after pollutants enter the aquatic environment. One group of chemical transformations that is under study is the biomethylation of arsenic, mercury and selenium.

Feasibility of Air Injection Mass Spectrometry for Organic and Inorganic Materials
M. R. Petersen

There is an increasing need for measurements of trace inorganic and organic materials in environmental

samples. New techniques are constantly sought which allow improved sensitivity and selectivity in these measurements. In a series of papers, W. D. Davis of General Electric Corporate Research and Development described results with air injection mass spectrometry which are promising not only in increasing sensitivity, but also in continuous monitoring of the composition of air particulates.

Davis' technique is described as follows. A small stream of air enters the ionization chamber of a mass spectrometer and impinges on a heated rhenium ribbon at 1000-2000°K. The ions form as a particle evaporates from the ribbon surface and are then accelerated, focused, separated, and detected as usual. Since rhenium in the presence of absorbed oxygen has a work function of 7.5 eV at 1100°K, ionization of the major components of air (N_2 , O_2 , H_2O , CO_2 , etc.) cannot occur and no interfering signals from these components are produced.

The reported minimum detectable amount of a particular element appears to depend on its ionization potential. As few as 10^3 atoms of the alkali metals were continuously detectable.

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FIGURE 1.9 Details of Volatilization Apparatus and D. C. Emission Discharge Cell

About 10^5 atoms were required for chromium and lead, which are in the 6.5-7.5 ev range; 7×10^6 atoms were necessary for copper (I.P. = 7.7 ev). Germanium, tungsten, cadmium, and zinc were not detected, which indicates the cutoff limit on the ionization potential was about 8 ev.

The versatility of air injection mass spectrometry results from the ionizing source. A rhenium ribbon allows selective ionization of elements having I.P. less than 8 ev. If other materials such as platinum were used, other additional elements may be similarly ionized. The temperature of the ribbon can be varied, and at high temperatures each particle produces a burst of ions by which particles could be counted. Lower temperatures on the ribbon average the concentration, which is recorded on a continuous basis. Greatly improved sensitivity can be achieved by collection of particles on a cold filament and then suddenly vaporizing the impacted material. Solutions could be analyzed by ionizing the solute in droplets formed by aspiration. Many inorganic species, including uranium and plutonium, can be detected with high ionization efficiencies. The technique offers many advantages in

mass spectrometric analyses of organic compounds over conventional ionization sources, since the fragmentation patterns are simpler due to the low ionizing energy of the Re ribbon.

In order to take full advantage of air injection mass spectrometry there are several areas which require further study. A straightforward calibration procedure is needed which can be easily and quickly performed. An investigation of the vaporization efficiencies for elements of interest should be undertaken. For example, elements in silicate material may be incompletely vaporized compared to elements in a marine aerosol, so compensating methods must be found to obtain valid quantitative numbers.

The potential usefulness of this technique lies in the continuous analyses of composition and number of air particulates, including particles with diameters less than $0.1 \mu\text{m}$. If problems of vaporization efficiency and calibration can be solved, this instrumental technique could be of great value in the real-time monitoring of a variety of airborne particulates.

• FAST REAL-TIME MEASUREMENT OF EXTREMELY LOW-LEVEL AIRBORNE PLUTONIUM

A small mass spectrometer with a direct air-inlet to the ion source region is being developed to detect and measure individual submicrometer plutonium-bearing particles in ambient air. The instrument will be capable of measuring plutonium concentrations below the MPC level. Development of the inlet and the readout system for the spectrometer is in progress.

Direct-Inlet Mass Spectrometer Development

J. J. Stoffels, C. R. Lagergren,
and A. L. Franklin

A mass spectrometric technique is being developed for monitoring low-level airborne plutonium on a real-time basis. Current techniques for monitoring airborne plutonium are limited by the rate of radioactive decay which, for ^{239}Pu , amounts to only one disintegration per sec for every 10^{12} atoms present. The new technique will measure one ion count for every 10^2 atoms of plutonium present in an individual particle and do this in a burst lasting about 1 sec.

The instrument being developed is a small mass spectrometer with a direct air-inlet to the ion source region. Particulate material entrained in the stream of air sampled will strike a hot filament in the source and produce a burst of ions. The ions will be mass separated in the magnetic field of the spectrometer and an ion detector will monitor the mass position of interest (in this case, mass 271 corresponding to $^{239}\text{PuO}_2$).

The output signal from the mass spectrometer will be proportional to the amount of plutonium in each particle. For those cases in which the

plutonium particles exist as pure compounds, the output signals may be related by appropriate calibrations to the particle sizes. In addition, the number of ion bursts per unit time will be a measure of the particle concentration in air.

With this instrument it will be possible to detect and measure individual submicrometer plutonium-bearing particles in ambient air. Since collection of particles will not be required and since the measurements will be accomplished very rapidly, information on the concentration of plutonium in air at any given time will be immediately available. Plutonium concentrations well below the MPC level will be measurable on this real-time basis.

The major technical limitation of the direct-inlet mass spectrometric method is the air volume sampling rate. Because the ion source of the mass spectrometer must operate under vacuum (about 10^{-5} Torr), it is necessary to separate the carrier gas (air) from the particulate material by differential pumping. Since the volume of air sampled dictates the size of pumps required to achieve the necessary pressure reduction, a compromise must be made between a useful sampling capacity and instrument

size and cost. Preliminary measurements of pressure versus flow through a capillary tube indicate that an inlet system with a sampling capacity of 10 to 20 cc's per second will result in an instrument which is roughly desk-sized.

Procurement and fabrication of equipment for the development of such an inlet is in progress. An aerosol generator is also being built for testing the inlet and the entire spectrometer (when completed) with particulate aerosols of known parameters.

Development of a readout system for the direct-inlet mass spectrometer is also in progress. Both digital and analog methods of signal processing are being investigated as

means of presenting the data generated in the most meaningful form. For the spectrometer to be capable of giving quantitative information on particles in the size range of 0.01-10 μm , the detector and readout system must have a dynamic range of 10^9 . Since no single detector and readout device can provide such a wide dynamic range, it will be necessary to use two different measurement techniques--one for particles larger than about 0.25 micrometer diameter and another for particles smaller than that. Means will also have to be provided for the system to automatically accommodate whatever size particle is encountered. Current work is focusing on integration of the total current in ion-burst events due to the larger particles.

• ANALYTICAL TECHNIQUES FOR MEASUREMENT OF ^{99}Tc IN ENVIRONMENTAL SAMPLES

Mass spectrometry promises greater sensitivity for measurement of ^{99}Tc in the environment than any method previously reported. During the first 5 months of this study $^{99}\text{Tc}^+$ ion beam intensities have been obtained which indicate that picogram and sub-picogram measurements may indeed be feasible. Further studies of ionization source design, operating parameters, and other factors affecting measurement sensitivity are being conducted.

Ultrasensitive Measurement of ^{99}Tc by Mass Spectrometry J. H. Kaye

Assessment of potential hazards due to release of ^{99}Tc to the environment should be made before large numbers of nuclear power plants become operational. Technetium-99 is of concern because it is produced in relatively high fission yield, has a long half-life (2.13×10^5 years), may be concentrated in biological systems, and may be released to the environment in significant quantities as the volatile heptoxide, Tc_2O_7 , or as pertechnetate, TcO_4^- . Measurement of this nuclide by direct counting methods has been difficult and has not provided adequate sensitivity due to the long half-life and lack of gamma radiation accompanying its decay. Thermal neutron activation analysis techniques have been reported but serious problems have been encountered in measurement of the short lived (16-sec) ^{100}Tc activation product. Thermal ionization mass spectrometry appears to be more promising than either of these measurement methods and this approach is therefore being pursued.

In this method an aqueous sample of about 1 μliter volume is placed

onto a thin rhenium filament and dried. The filament temperature is then raised by resistive heating to a high value. This causes a useful fraction of the ^{99}Tc atoms to leave the filament surface as positive ions, which can be accelerated and subsequently detected.

In recent, highly encouraging experiments $^{99}\text{Tc}^+$ ion counting rates of 10^6 counts/sec $^{-1}$ have been achieved with 10 ng samples, and this counting rate has been maintained for periods of about 2 min. Attempts to obtain even higher ion counting rates by further increases in filament temperature have not been fruitful, since the samples vaporize so rapidly that the ion beam, though initially relatively intense, decreases rapidly within a few seconds. However, by extrapolation from the studies with nanogram samples, it appears that sensitivity should be sufficient for measurement of picogram or sub-picogram levels of ^{99}Tc .

It is anticipated that isotopic dilution techniques will be employed in order to obtain reliable quantitative ^{99}Tc measurements. This will involve addition of a known amount of ^{97}Tc to each sample prior to the chemical separation steps. The ^{99}Tc

will then be estimated from mass spectrometric measurement of the $^{97}\text{Tc}/^{99}\text{Tc}$ ratio. This method eliminates having to determine the chemical yield and efficiency of the mass spectrometer for each sample. Unfortunately it has been found that trace levels of

molybdenum in the rhenium filaments cause a serious interference at mass 97. This problem is presently being studied. In addition, further studies of ion source configuration and other parameters affecting measurement sensitivity are in progress.

- **NUCLEAR TECHNIQUES IN MEDICINE**

Nuclear technology and radioisotope techniques are developed in this program for use in the clinical diagnosis of disease and in medical and biological research. The application of these developments is done in cooperation with medical institutions and the Pacific Northwest Laboratories Biology Department. The development of new and improved instrumentation and techniques for measuring in-vivo plutonium and other transuranic elements is also provided in this program.

Methods for Medical Research

Nuclear technology developments are providing new and better methods for use in the medical field. Important contributions have been and are continuing to be made under this program in the area of body composition studies as related to various disease states.

The Determination of Total Body Nitrogen in Animals by Measuring ^{11}C in Expired Air After Neutron Irradiation H. E. Palmer

In addition to the ^{37}Ar found in the expired air of animals and humans after neutron irradiation,¹ several other gaseous radioisotopes have recently been identified which might be related to elements of body composition. One of the most abundant of these is ^{11}C . Carbon-11 is a positron emitter, has a half-life of 20.4 min and emits 0.51 annihilation photons which are easily measured. The ^{11}C appears to be produced by a (p, α) reaction on stable ^{14}N in the body.

The two major compounds of the expired ^{11}C have been identified as ^{11}CO and $^{11}\text{CO}_2$. Preliminary attempts to identify $^{11}\text{CH}_4$ were unsuccessful but low concentrations are probably present. The ^{11}CO and $^{11}\text{CO}_2$ are easily separated from the expired air of small animals and measured. During and after irradiation the gas containing the expired air is passed through CaSO_4 to remove water vapor and small quantities of nitric and/or nitrous oxides containing ^{13}N . The gases then flow through a soda lime trap where the $^{11}\text{CO}_2$ is collected. The ^{11}CO and other gases then flow through hot copper oxide which converts CO to CO_2 and the $^{11}\text{CO}_2$ formed there from the ^{11}CO is collected on a second soda lime trap. The ^{11}C in the soda lime traps is then measured between two large NaI(Tl) detectors operated in coincidence so that essentially only the 0.51 MeV annihilation photons are counted. The soda lime traps are constructed in a circular configuration so that the ^{11}C is counted with the same geometry regardless of where it is deposited within the circular tube.

Most of the ^{11}C is expired as ^{11}CO from a rat. The ratio of ^{11}CO to $^{11}\text{CO}_2$ is about two and appears to be constant over a variety of environmental conditions. The $^{11}\text{CO}_2$ is expired about twice as rapidly as the ^{11}CO when the rat breathes a gas mixture containing 20% O_2 . The slower excretion of the ^{11}CO is due to its strong attachment to the hemoglobin of the blood. Since most of the ^{11}C is in the form of ^{11}CO and because the decay half-life is only 20.4 min it is important that the ^{11}CO be rapidly excreted from the body before it decays away. Several methods were tried in an attempt to increase the excretion rate of the ^{11}CO . Both exercise and cold environment were used to increase the metabolic activity of the animal and both of these stresses resulted in at least a doubling of the excretion rate. However, the most significant increase occurred when the rat breathed pure O_2 which resulted in an excretion rate 4.5 times that obtained with 20% O_2 , no exercise and room temperature. The results of these studies are shown in Figure 1.10 and indicate that essentially all the ^{11}CO is expired in time for it to be measured before it decays. The use of pure O_2 as the breathing gas does not change the amount or the excretion rate of the $^{11}\text{CO}_2$. Its only effect seems to be that of enhancing the release of the ^{11}CO from the hemoglobin so that it can be released through the lungs. This is indicated by the fact that the oxygen tension of the blood is increased by a factor of five when changing from 20% O_2 to 100% O_2 . This results in a change of a factor of 4.5 in the excretion rate of ^{11}CO .

The interest in ^{11}C measurements in expired air is its possible use in determining the total body nitrogen which is directly related to the

FIGURE 1.10 ^{11}Co Excretion in Expired Air from a Rat After 14 MeV Neutron Irradiation (30 min Sample Collections)

total body protein content. This measurement could become important in nutritional studies and in the study of certain diseases. The ^{11}C is produced only from the nitrogen in the body, but it has not been directly correlated by making the corresponding chemical analysis of total body nitrogen. The ^{11}CO extracted from various concentrations of nitrogen solutions in water has been found to be exactly proportional to the nitrogen content. Future studies will compare the expired ^{11}CO from small animals with the actual total body nitrogen content and will determine if this technique can be extended to humans. The lower ventilation rates per unit of body surface area of the human compared to that of the rat may limit the use of this technique with humans due to a possible much slower excretion rate of the ^{11}CO and $^{11}\text{CO}_2$.

Instrumentation for Measuring In Vivo Plutonium

A major problem in radiation protection in nuclear industries is the assessment of location and amount of plutonium in humans after accidental exposure. Instrumentation development and techniques for more sensitive detection of plutonium are in progress under this program.

Progress in Gas Scintillation Proportional Counters H. E. Palmer

The excellent resolution of gas scintillation proportional counters suggests that improved counting sensitivity for in vivo plutonium could be realized if large-volume, low-background counters could be developed. Three different counter configurations were studied for use as large volume counters. These include the spherical anode inside a short cylindrical cathode, such as that used by Policarpo, et al;² a series of transparent parallel plates;³ and a rod anode inside a long cylindrical cathode.

A xenon filled counter having a rod anode inside a cylindrical cathode appears to provide the best configuration for providing a uniform field and the best resolution over the total volume of the counters. The use of a center rod in a cylindrical shaped counter with collimated radiation provides the same excellent resolution as that obtained with the spherical anode or parallel grid counters. The spectrum of a beam of radiation from an ^{241}Am source collimated into the counter midway between the two ends is shown in Figure 1.11. The counter chamber is 5 cm in diameter and 7.5 cm long and has a 0.3-mm-diameter polished aluminum rod anode. The resolution of the 14 and 17.8 keV photopeaks are 6.4% and 7.6%, respectively, which compares to 11.8% and 10.9% obtained by Yaniv, et al,⁴ in

a regular proportional counter chamber which had dimensions of 5.08 x 5.08 x 35.6 cm and was filled with a 90% xenon - 10% methane gas mixture.

Rod anodes of both 3 mm and 6 mm diameter have been used and both resulted in the same degree of resolution. The 3 mm rod gives the best ratio of absorption volume to light reproduction volume and, therefore, is preferred for the higher energy radiation.

Due to the nonuniform field at the ends of the counter, poor energy resolution is obtained for a broad beam of radiation. Efforts are currently being made to eliminate the end effects of the counter by using field shaping rings similar to that described recently by Horstman, et al.⁵ These field shaping rings were incorporated into a large 12.8-cm-diameter by 17.8-cm-long counter with a large 12.8 x 12.8 cm beryllium window. The counter with the attached hot calcium gas purifier is shown in Figure 1.12. This counter needs further development but it does have a uniform electric field over the total length of the counter which allows the entire volume of the counter to be used for detection. With further development, this counter should be useful in measuring in vivo plutonium and in many other applications which require better resolution than an ordinary proportional counter and a larger detection area than the Ge or Si solid state detectors.

FIGURE 1.11 Spectrum from ^{241}Am Source Counted on a 5 cm Diameter, 7.5 cm Long, Rod Anode Gas Scintillation Proportional Counter (Anode Diameter = 3.2 mm)

FIGURE 1.12. Large Rod Anode Gas Scintillation Proportional Counter with Large Beryllium Window

The Determination of ^{239}Pu in Animals by Measuring Fission Gases in Expired Air After Neutron Irradiation
H. E. Palmer and V. H. Smith*

The techniques which have been recently developed for measuring gaseous radionuclides in expired air of animals after neutron irradiation are being applied to the measurement of fission gases from metabolized plutonium in animals after neutron irradiation. The results from measurements of fission gases from solutions of uranium containing the natural amount of ^{235}U indicate that 10 μCi of ^{239}Pu in an animal should be easily measured and much lesser amounts will be detectable as more sensitive methods for measuring the radioactive krypton and xenon are developed. Measurements on animals containing known quantities of ^{239}Pu will be made during the next year. The application of this technique to human use appears unlikely at the present time due to the neutron radiation dose required, but the development of extremely sensitive methods for fission

* Biology Department

gas measurement might allow application of this technique in the future.

Intraesophageal Probe K. L. Swinth

Following experimental development of the fiber-optic coupled probe concept, three probe assemblies were fabricated to human use standards. Along with two photomultiplier assemblies, enough redundancy was obtained to rapidly cope with equipment emergencies during medical testing. Tests of the angular and linear sensitivity of the intraesophageal probe indicated the response was uniform within $\pm 4\%$. This compares with variations in angular sensitivity of $\pm 35\%$ and linear sensitivity of $\pm 60\%$ for the avalanche diode probes.⁶

By varying the source geometry in the tracheal phantom, calibration tests were performed which indicated a standard deviation of $\pm 40\%$ in the calibration at x-ray energies and $\pm 30\%$ at γ -ray energies (60 keV- ^{241}Am). This represents the variations expected over a range of possible tracheobronchial lymph node geometries. If the geometry and variations were more accurately known, the standard deviation of the calibrations would be reduced. Calibrations were also performed for interlobar and paratracheal nodes.

Previous calculations have shown that count rates from the intraesophageal probe should be corrected

for contributions from lung-deposited material. These contributions will cause an error in the amount of activity assigned to the lymph nodes. Measurements were performed with plutonium loaded phantom lungs to accurately determine the correction factor. The counter sensitivity was 25 cpm/ μCi at x-ray energies and 72 cpm/nCi at gamma ray energies (^{241}Am); these values are within $\pm 25\%$ of calculated values.

Following approval of human use committees at Battelle and Los Alamos Scientific Laboratory (LASL), the probes were transported to LASL and evaluated on two human volunteers; one of whom acted as a control. The control subject had no known exposure to plutonium while the other individual had been exposed to plutonium dioxide 14 years previous to our counts. The exposed subject tolerated the procedure quite well and provided important information showing a large amount of material in the lymph nodes. Results are shown in Table 1.12 where error limits represent precision, not the accuracy, with values estimated to be within a factor of two of true organ contents. Corrections were applied for contribution of lung deposited material and for isotopic composition of the material.

The x-ray procedure used to position the probe causes phosphorescence of the NaI thus introducing noise in the energy region of the x rays. Phosphorescence components with half-lives of 2.5 min and 12 min have been

TABLE 1.12 Results from Intraesophageal Probe Counts³

Position	Net Counts (per 10 min)		Approximate Activity (nCi) ¹	
	X-Ray (12-40 keV)	γ -Ray (41-95 KeV)	X-Ray	γ -Ray
1a	88 $\pm$ 37	80 $\pm$ 17	11 $\pm$ 4	7.7 $\pm$ 1.9
1b	63 $\pm$ 28	53 $\pm$ 16	8 $\pm$ 3	5.1 $\pm$ 1.8
2	---	81 $\pm$ 11	---	49 $\pm$ 15 ²
3	155 $\pm$ 26	204 $\pm$ 21	18 $\pm$ 3	17 $\pm$ 2

¹ Based on phantom calibration with ^{239}Pu for activity in simulated lymph node geometry.

² Activity in "lungs" based on lung calibration using phantom and corrected for contribution from lymph nodes.

³ Errors represent statistical errors only.

observed with these probes; however, other traps are reported in the literature.⁷ This "phosphorescence noise" raises the lower detection limit and techniques to eliminate this problem are under study.

Ten dogs exposed by inhalation of $^{239}\text{PuO}_2$ and one exposed to $^{238}\text{PuO}_2$ were counted in two separate experiments. When count rates were high enough, scans were taken down the esophagus which in some cases showed the presence of activity at the location of the mediastinal lymph nodes in addition to activity in the tracheobronchial nodes. Postmortem counts have not been obtained on these dogs; however, observed counts correlate closely with lymph node burdens expected from extrapolation of measured lung burdens.

The fiber-optic coupled scintillator was compared to other biomedical probes and found to have a sensitivity 90 times that of previously described solid-state probes.⁶ This comparison is based on the count rate in standard solutions of $^{99\text{m}}\text{Tc}$. In support of a bronchial carcinoma detection scheme, a 25 mm³ detector was tested with mock tumors in a lung phantom.* These preliminary tests indicated a

* Tests performed in collaboration with G. C. Huth at Laboratory of Nuclear Medicine and Radiation Biology, UCLA.

4-mm-diameter tumor could be barely detected in a 1 min counting period. Indium-111 was used in these trials because of its favorable x-ray energies (23 to 26 keV); a target-to-nontarget ratio of 5:1 was used in these measurements.

Plutonium Counting In Vivo and Interlaboratory Calibration

K. L. Swinth

Interlaboratory calibration efforts centered on a review of whole-body counting data from cooperating laboratories and a review of phantom data in conjunction with a calibration phantom to be built in the future. It was found that the shape and size of typical phantom lungs is not representative of tissues observed at autopsy. Lungs cast in vivo at Lovelace were selected as realistic models. Another shortcoming is the lack of lymph node and lung dimensions and masses.

An extensive review of the plutonium inhalation problem showed the need for a simple technique for rapid assessment of early plutonium deposition and techniques for assessment of lymph node burdens. The present intraesophageal probe answers the latter need and calculations show that an adaptation of the probe could answer the former need. Calculations suggest that it might be possible to detect lung burdens of 0.5 nCi.

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RADIOLOGICAL AND CHEMICAL PHYSICS

- **RADIATION PHYSICS**
- **RADIATION DOSIMETRY**
- **RADIATION BIOPHYSICS**

• RADIATION PHYSICS

The radiation physics program addresses three major research problems associated with understanding the effects of radiation on materials of biological significance. Electron emission from molecules bombarded by protons is studied to provide information on primary collision processes. This information is then used to formulate descriptions of track structure and the interaction of ionizing tracks with volumes of biological and biochemical importance. Finally, mechanisms of energy transport and transformation from the energetic initial processes into chemical species is studied. Significant progress has been made this year in extending electron emission cross section measurements to lower energies and to more complex molecules. Measured cross sections have been compared with various theoretical calculations for electron energies from 0.5 eV; such comparisons provide insight into the relative importance of various basic mechanisms and methods for extending theories to complex molecular systems. Empirical analytical expressions for collision cross sections are being developed to facilitate introduction of these data into track structure codes. Track structure parameters are being incorporated, as initial conditions, into kinetic models describing fluorescence life time and nonlinear decay of proton excitation in the condensed phase.

Low-Energy Electron Emission

L. H. Toburen and W. E. Wilson

By carefully accounting for contact potentials, the time-of-flight (TOF) electron energy analysis system can now be used to measure electron energy distributions as low as 0.3 eV. The energy distributions, which cover an energy range of 0.3-200 eV and an angular range of 30-130°, can be converted to absolute cross sections by normalization to cross sections measured with our electrostatic analyzer for ejected electron energies greater

than about 30 eV. The TOF system is now routinely used to provide reliable results at low energies for all measurements made with our electrostatic analysis system and, as such, improves the overall quality of the measurements discussed in the following sections.

The TOF measurement of low-energy electron distributions has several advantages over electrostatic analysis. First, no electrostatic or magnetic deflection fields are needed and thus there are no distortions due

to fringing fields. Second, short flight paths can be chosen to minimize the effects of the earth's magnetic field. In the present measurements a flight path of 7.5 cm is routinely used and residual magnetic fields are reduced to ± 2 mG by a combination of magnetic shielding and electric coils. A further advantage of TOF is that the system inherently produces the best energy resolution and most closely spaced data points at the lowest electron energies.

The capabilities of the TOF system are illustrated in Figure 2.1 where the energy spectrum of electrons ejected from xenon by 1 MeV proton impact is shown. The spectrum, which has been corrected for contact potentials, exhibits structure at low energies resulting from autoionizing transitions which occur at energies in excellent agreement with photoabsorption data. The photoabsorption

results shown as an insert in Figure 2.1 were taken from the work of Comes, et al.¹ The N00 Auger lines are also evident in the energy range from 8 to 34 eV, however, the energy resolution is not sufficient at these energies for resolution of the individual lines and the lines are partially hidden in the large continuum background. The structure in the N00 Auger spectrum is somewhat more evident in the original TOF spectrum shown in Figure 2.2; still, it will be necessary to improve the time resolution in order to resolve the individual Auger lines. The Auger line positions shown in Figures 2.1 and 2.2 are taken from the work of Seigbahn, et al.² In addition to the Auger and autoionizing transitions shown in Figures 2.1 and 2.2, a number of autoionization lines are expected in the energy spectrum between 7 and 12 eV.² These lines are

FIGURE 2.1. The Time-of-Flight Electron Energy Spectrum Resulting from 1 MeV Proton Ionization of Xenon. The observation angle is 130° . The photoabsorption spectrum is from Ref. 1 and the N-Auger line energies are taken from Seigbahn, et al.²

FIGURE 2.2. The Time-of-Flight Electron Spectrum Resulting from 1 MeV Proton Ionization of Xenon. The observation angle is 130° . The N-Auger line energies are from Seigbahn.²

not separable from the large continuum background present in our measurements. We are currently in the process of replacing our electron detector with a faster large-surface-area detector, which should enable measurements with much better counting statistics and with much better energy resolution.

Electron Cross Sections from Molecular Targets

L. H. Toburen, W. E. Wilson,
L. E. Porter,* and D. L. Lynch**

Data from molecular targets are important in building an understanding of the effects of molecular binding, in determining the consequences

of phase changes, and in assessing the scalability of simple molecules to more complex systems. In addition, these cross sections are necessary in making realistic calculations of energy deposition in biological systems since presently no theoretical methods appear capable of accurately predicting these quantities.

During the past year we have continued our analysis of data from SF_6 , TeF_6 and numerous hydrocarbons, as well as acquired new data on ammonia (NH_3), monomethylamine (CH_3NH_2), dimethylamine [$(\text{CH}_3)_2\text{NH}$], molecular oxygen (O_2), and water vapor (H_2O). The results for methane and ammonia will be compared with CH_3NH_2 to investigate additivity; and comparison of CH_3NH_2 data with $(\text{CH}_3)_2\text{NH}$ will provide information on the effects of chemical substitution. A similar check on additivity can be performed using the data of hydrogen, oxygen and water vapor. Preliminary analysis of this data indicates that the

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amines and ammonia follow very closely the scalability by the number of weakly bound electrons which was observed for the larger hydrocarbon molecules.³ The methane results appear more hydrogen-like than the larger molecules, i.e., the angular distributions are somewhat more peaked and there are relatively fewer electrons ejected at higher electron energies. A striking feature of the results obtained for the more complex molecules is the isotropy of the low-energy electron-ejection cross sections. This is illustrated in Figure 2.3 for 1.8 MeV proton ionization of SF₆. The results for electron energies below 40 eV were obtained from TOF spectra normalized to the electrostatic measurements in the 50-100 eV region of the electron spectra. The TOF results were obtained for an angular range of 50-125°. These results indicate complete isotropy for electron energies below 5 eV or possible evidence of cross sections actually increasing in the backward angles at 1 eV. We intend to try to extend these measurements to lower energies and to a larger range of angles to determine more accurately the actual angular distributions of the low-energy ejected electrons.

In addition to the measurement and analysis of emission cross sections, a major advance in instrumentation was implemented during the last year. A computer interface was designed to place our measurements using the electrostatic analyzer under computer control. With this device we can select any voltage region within a 0-6 kV range and record data at any step size, linearly or nonlinearly, under computer software control. Although we are continuing to manually select the angle at which data is obtained, it is possible to add the capability of software-controlled angle manipulation at a later date with minor modifications to the present interface. This interface, coupled with our PDP-8 computer and a programmable voltage supply, has added versatility, reliability and a greater degree of precision to our cross section measurements.

Theoretical Investigation of Electron Emission Cross Sections

L. H. Toburen and S. T. Manson*

Evaluation and explanation of the effects of radiation on matter depends

* Georgia State University, Consultant.

FIGURE 2.3. Angular Distributions of Electrons of Selected Energies Ejected from SF₆ by 1.8 MeV Protons. Cross sections for electron energies below 40 eV are obtained from time-of-flight measurements.

on knowledge of the basic interaction mechanisms. A careful comparison of experimental data with theoretical results provides the basis for determination of the importance of various interaction processes. The comparison of double-differential cross sections for electron ejection by charged particles has led to detailed information on the ionization process which is the major means by which fast charged particles lose energy.

In order to understand more complex systems, a foundation of information on the simpler systems is essential. Therefore, we have begun our detailed theoretical-experimental comparison of double-differential electron emission cross sections with a comprehensive comparison of data for ionization of helium by electron and proton

impact. In a paper currently being prepared in collaboration with Dr. D. H. Madison of Drake University and Dr. N. Stolterfoht of Hahn Meitner Institute of Berlin, Germany, we have compared 2 keV electron impact and 100-5000 keV proton impact data with calculated double-differential cross sections obtained using the Born approximation with initial discrete and final continuum wave functions derived from a Hartree-Fock-Slater (H-S) potential. Using these realistic wave functions we find that the theoretical cross sections are in excellent agreement with electron impact results and with 5 MeV proton impact results. At lower proton energies the calculated cross sections are increasingly too small at small angles of electron emission, i.e., for 100 keV protons the cross section for ejection of a 160 eV electron at 15° is a factor of 6 larger than the calculated value.

In contrast to this discrepancy at smaller angles the H-S Born calculation is in excellent agreement with measurements for angles greater than

about 60° . It should be noted that previous calculations using hydrogenic wave functions underestimated large-angle cross sections as well as those at small angles. The success of the H-S calculation at large angles results from a proper consideration of the interference of the various partial waves which enter into the calculation. The discrepancies at small angles of electron emission result from the effects of a mechanism for ionization which is not an inherent part of the Born approximation. The comparison of measured and calculated cross sections, therefore, provides detailed information on this process which was referred to previously as "charge-transfer to the continuum."⁴ The relative importance of the charge-transfer mechanism for ionization is illustrated in Figure 2.4, as a function of proton energy. The relative importance of the continuum charge-transfer process is found to peak the protons of about 0.5 MeV and to be most strongly peaked in the forward direction. If the process is truly a charge-transfer mechanism then one would expect the proton energy dependence to be similar to

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FIGURE 2.4. Relative Cross Sections For Charge-Transfer to the Continuum as a Function of Electron Emission Angle and Incident Proton Energy.

charge-transfer to bound states. An estimate of the cross section for continuum charge-transfer can be obtained by subtracting the calculated cross sections from the measured values; these are plotted in Figure 2.5. The slope of the high-energy portion of the curves in Figure 2.5 was drawn to have the slope determined in bound state charge-transfer;⁵ insufficient numbers of data points were available for a direct determination of the slope of this portion of the curve. This analysis implies that the ionization mechanism referred to as resulting from continuum charge transfer is certainly compatible with previous knowledge of the charge transfer process. A more complete discussion of the process as well as the comparison of experimental and theoretical double-differential cross sections for proton and electron impact ionization of helium will be presented in a forthcoming paper.

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FIGURE 2.5. Cross Sections for the Contribution Attributed to Continuum Charge Transfer as a Function of Electron Emission Angle and Proton Energy.

In addition to the detailed investigation of helium discussed previously, preliminary work has begun for comparison of experimental and theoretical double-differential electron-emission cross sections for proton ionization of neon and argon. Argon is a particularly interesting atom in that the generalized oscillator strength for the 3p- ϵ d ionization channel passes through a minimum. In order to expect accurate results from the Born calculation of double-differential electron-emission cross sections for this channel, a full Hartree-Fock calculation is needed. Both Hartree-Fock and Hartree-Fock-Slater calculations are being carried out and the results will be compared with recent measurements for 1 MeV Proton impact ionization of Argon.

In a recent paper, Kim described a method of analyzing experimental single-differential electron-emission cross sections which provides criterion for assessing the reliability and consistency of the measured values.⁶ This method relies on the similarity between fast charged-particle and photoionization cross sections for low-energy electron ejection and on the applicability of simple binary collision theory for high-energy electron ejection. This method was used to a large degree of success by Kim to analyze data from electron impact ionization. We are presently working in collaboration with Y. K. Kim of Argonne National Laboratory in the investigation of this analytic method for proton impact. The results for 1 MeV proton ionization of helium are shown in Figure 2.6. The function $Y(E)$ plotted in Figure 2.6 is the ratio of the measured differential cross section divided by the Rutherford cross section. At large values of E/R the curve for $Y(E)$ should converge to the number of electrons in the target which contribute to the interaction. As expected, for helium the magnitude of $Y(E)$ approaches 2 at high energies. Also shown are calculations using binary encounter theory and the H-S Born approximation. Perhaps coincidentally, binary encounter theory is in somewhat better agreement with measured cross sections at low energies than is the H-S calculation. The H-S calculation appears to overestimate the very-low-energy portion of the spectrum by about 20%.

It should be pointed out that the measured cross sections shown in Figure 2.6 are derived from both TOF and electrostatic energy analysis and should be accurate to $\pm 20\%$ in absolute value for the electron energy range of 0.5-6000 eV; and from a comparison to the Rutherford cross section at the higher energies, it appears that the absolute value of the cross sections may be about 10% too large. The importance of this type of analysis is that one can accurately assess the uncertainties in the cross sections. The 20% uncertainty quoted above is an estimate based on propagation of the uncertainties in each of the experimental parameters and may

be considered a conservative estimate. The Rutherford cross section, on the other hand, should provide an accurate estimate of the cross section for emission of fast electrons from helium by 1 MeV protons. From this comparison we feel that the uncertainty in the measured helium cross sections is more nearly 10% than our earlier 20% estimate. Further evidence to support this conclusion is that the total ionization cross section obtained by integration of the single differential cross sections gives $\sigma = 1.94 \times 10^{-17}$ compared to 2.0×10^{-17} obtained by Hooper, et al.⁷ and 1.7×10^{-17} measured by Solovev, et al.⁸

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FIGURE 2.6. The Ratio of Measured Single-Differential Cross Sections to the Rutherford Cross Section as a Function of Energy Loss in 1 MeV Proton Ionization of Helium. The electron impact results are from Ref. 9, the oscillator strength distributions are from Ref. 6,* the calculation of zero energy electron emission by Kim is from Ref. 11, and the measured zero energy cross section is from Ref. 10.

* Work of Berkowitz quoted by Kim in Ref. 6.

An insert in Figure 2.6 illustrates the low-energy portion of the $Y(E)$ curve and indicates that although similarities exist there are basic differences between the oscillator strength distributions obtained from photoionization and the low-energy emission cross sections. The proton results do not fall off nearly so rapidly with increased electron energy as is observed for electron impact and photoionization. This is partially due to additional angular momentum states being important for proton ionization in contrast to the dipole term relevant to photoionization and partially due to the enhancement in the emission cross section resulting from the continuum charge-transfer process which is not relevant in either photoionization or electron impact ionization. An extrapolation of the measured spectrum for proton impact to zero ejected electron energy appears to be in very good agreement with the value measured for equivalent velocity electron impact by Grisson, et al.¹⁰ and with the calculated cross section of Kim, et al.¹¹ This comparison lends further confidence to the accuracy of the measured cross sections. This type of analysis is now being continued in collaboration with Dr. Kim to include neon and argon cross sections. These results will be described in detail in a forthcoming journal article.

Analytical Expressions for Cross Section Data

W. E. Wilson and L. H. Toburen

In order to more accurately represent the proton ionization process in our Monte Carlo track structure calculations, we are developing a semi-empirical phenomenological representation of the cross sections for electron ejection, especially from polyatomic molecules. Since the MC codes require probability distributions for emission of electrons as a function of angle as well as a function of energy, we are trying to develop a parameterized analytic expression for the double-differential cross sections.

By scaling the cross sections to remove the major dependence on proton and electron energies one is left with angular distributions, which appear relatively simple and which we had hoped could be represented by a

finite (small) number of Legendre polynomials; that is we have represented the DDCS in the following way. We have scaled the cross sections and then have tried to fit the $S(\epsilon, \theta)$ with a finite series of the form

$$S(\epsilon, \theta) = \sum_{i=0}^N A_i(\epsilon) P_i(\cos\theta), \quad (1)$$

where

$$S(\epsilon, \theta) = T/R \frac{[(\epsilon+I)^2/R]}{4\pi A_0^2} \sigma(\epsilon, \theta), \quad (2)$$

where $P_i(\cos\theta)$ are Legendre polynomials, $T = E_p/1836 =$ the energy of an electron with velocity equivalent to the proton, $R = 13.6 \text{ eV} =$ Rydberg energy, $A_0 =$ Bohr orbit radius. Figure 2.7 shows angular distributions scaled in this way for ejection of electrons from helium by 1 MeV protons, for several electron energies. The distributions are relatively simple, well-behaved functions and do not cover a large range of ordinal values. We have determined the coefficients $A_i(\epsilon)$ in Eq. (1) by least-squares fitting with successively higher order terms in Eq. (1). A typical example is shown in Figure 2.8, for $(\epsilon+I)/R = 20$ and for polynomials fits with P_5 , P_7 , and P_9 terms. For this example, one can see that a minimum of nine polynomials are required to properly represent the angular distribution, especially in the backward direction. Still more terms are needed to fit the distributions for higher electron energies.

The Legendre coefficients for the nine polynomial fit to the 1 MeV helium data are plotted in Figure 2.9. The magnitude of the coefficients changes abruptly for energy losses between 10 and 15 Rydbergs. Below 10 Rydbergs the coefficients are largely independent of energy loss. (The theoretical implication of this change in coefficients is that different mechanisms of interaction in the proton-helium atom collisions are changing their relative importance).

From our attempts to fit the angular distributions with Legendre polynomials, we have learned that a large number of coefficients are required

FIGURE 2.7. Angular Distributions for Electron Ejection from Helium by 1.0 MeV Protons. Cross sections are scaled according to Eq. (2).

FIGURE 2.8. Least-Squares Fit of Legendre Polynomials to the Angular Distribution for Energy Loss of 20 Rydbergs. Curves are the fits, respectively, with maximum polynomial order of 5, 7 and 9.

FIGURE 2.9. Coefficients for Least-Squares Fit of Legendre Polynomials (Through 9th Order) to Angular Distributions for Electron Ejection from Helium by 1 MeV Protons.

for a proper fit and that they are not simply related to electron energy. These two facts make the Legendre polynomial formulation unattractive for modeling the cross sections for use in our Monte Carlo simulation of track structure. For this reason we have started to look for formulations other than Eq. (1) for describing the DDCS. Nevertheless, the Legendre fitting scheme can provide important information for theoretical analysis of the interaction mechanisms.

Early Physical-Chemical Effects and Luminescence M. L. West

The study of fluorescence from excited molecules in condensed systems provides a useful technique for probing the early physical-chemical effects following deposition of ionizing radiation. Excited molecules are extremely sensitive to their surrounding environment via energy transfer and other quenching mechanisms. Many of these processes occur on every collision as compared to chemical reactions with typical reaction rates of 10^9 sec^{-1} (radical-radical reaction) corresponding to one reaction in every 10 collisions. In some cases, energy transfer involving excited states occur over large distances and are not diffusion limited.

Single photon techniques are used to measure small populations by time averaging over many separate events and the nanosecond time scale is easily probed. Although this technique provides direct kinetic information on the early stages of radiation action, a major disadvantage is its inability to detect nonfluorescing species. Consequently, direct observation of many of the early products of irradiated systems (ions, electrons, radicals, triplet states) is impossible and their presence must be inferred from indirect evidence based on detailed analysis of fluorescence formation and decay. Most pulsed luminescence studies have been undertaken with lightly ionizing β and γ radiation. Our investigations are concerned with densely ionizing proton radiation where most of the energy is initially deposited very close to the path of the incident particle. Proton pulses of less than 0.3 nsec FWHM are available from a

2 MeV Van de Graaff accelerator and a TRW nanosecond pulsed light source is used for ultra-violet excitation.

Proton Irradiation

Differences in fluorescence response for lightly ionizing and densely ionizing radiation have been the subject of numerous experimental investigations. Fluorescence yields (α/β ratios) and some lifetimes have been measured but no quantitative model has been developed to explain these differences for liquid systems. Blanc, et al.¹² has formulated a general theory of fluorescence quenching in organic crystals based on exciton diffusion and bimolecular quenching of excited states. Such a model predicts an initial rapid quenching of fluorescence and has been experimentally¹³ supported for α bombardment of anthracene crystals. No such model exists for dilute solutions of aromatics.

Fluorescence response curves have been obtained for dilute solutions of aromatics in numerous organic solvents. Nonexponential decay for some of these systems is explained by a long-lived fluorescing component. Quenching studies with electron scavengers indicate that this component originates from delayed excited state formation via negative ion formation and recombination. For other systems, an initial rapid quenching is followed by a decay rate approaching that observed for ultra-violet excitation. Fluorescence response curves for benzene in ethanol, cyclohexane and hexane, show that benzene excited states exhibit an initial rapid quenching and do not exhibit the long-lived delay component. This is consistent with the observation that benzene has a negative thermal electron affinity, thus inhibiting long-lived negative ion formation.

A detailed analysis has been made of this nonexponential decay for dilute solution of benzene in cyclohexane and a fluorescence quenching model has been developed based on the nonhomogenous deposition of energy which considers radical-radical and radical-excited state interactions. The simultaneous effects of quenching and diffusion are mathematically described by the coupled equations,

$$\frac{\partial S^*(r,t)}{\partial t} = D \nabla^2 S^*(r,t) - k_{uv} S^*(r,t) - k_{RS} C(r,t) S^*(r,t) \quad (1)$$

$$\frac{\partial C(r,t)}{\partial t} = D \nabla^2 C(r,t) - k_{RR} C^2(r,t) \quad (2)$$

where $S^*(r,t)$ and $C(r,t)$ are the space time distribution of excited states and radicals, respectively, D is the diffusion coefficient, k_{uv} is the sum of radiative and nonradiative rate constants observed for ultra-violet excitation where no radical quenching is present, k_{RS} is the rate of excited state radical interaction, k_{RR} is the bimolecular radical quenching rate, and ∇^2 is the Laplacian operator. Eq. (1) and (2) can be solved to yield the excited state population,

$$S^*(t) = S_0 e^{-k_{uv}t} e^{-k_{RS} \int_0^t R(t) dt} \quad (3)$$

where $R(t)$ is determined by the initial excited state and radical distribution. This solution assumes that excited states and radicals are created in times short compared to diffusion interactions. For excited states and radicals created at later times, a source term must be added to Eq. (1) and (2).

Boundary conditions appropriate for solution of Eq. (3) have been considered in detail:

- Computer simulated track structure of 2 MeV protons using experimental cross sections as input to Monte Carlo programs¹⁴ show that initial distributions of ionization and excitation are well represented by a continuous core (where approximately 75% of the total energy is deposited) with sparsely spaced δ -ray tracks of high LET. A track picture based on a core plus an averaged secondary electron energy density does not accurately describe bimolecular interactions. Such a picture presents a high-density core surrounded by a low density of δ -ray distribution. Although the total energy deposited at large radius is small, the

local energy density is quite large. This is extremely important for processes such as fluorescence quenching that depend on the local density of radicals.

- The possible role of plasmon formation and decay in determining the initial distribution of excited states has been investigated. Calculations based on energy losses through plasmon oscillations show that for a 2 MeV proton the energy is deposited within molecular distance of the particle path. Only for very energetic protons or for very low frequency plasmons can the energy be deposited at large radial distances.
- The assumption of rapid ion recombination to form excited states has not been verified experimentally. However, Ko and Mozumder¹⁵ have shown that for densely ionized regions such as found along the path of keV electrons, the probability of prethermalization neutralization is extremely high (>60%). Such electrons are rapidly captured by a positive ion prior to thermalization and are not available for electron scavenging.
- The role of energy transfer from excited states of cyclohexane and possible quenching of these states by radicals is being considered. Such processes have to be included in explaining the initial rise of the fluorescence response curves. At present, detailed analysis of fluorescence formation is detector limited. The present model is valid for the region of observation (>1 nsec) if excited states of cyclohexane are also quenched by radicals. Fluorescence emission from cyclohexane has been observed in photolysis, radiolysis, and x-ray irradiation; and this excited state, which has a lifetime of about 0.3 nsec, has been suggested as the precursor of excited benzene molecules in solution. Emission from this excited state has been observed for proton irradiation. Two decay components are observed, a very fast component and a slower component that is sensitive to electron scavenging. This slow component has not been previously reported

and may explain differences reported in lifetimes and transfer rates.

Alternate explanations of nonexponential decay of proton irradiation have been considered:

- Decay from an initial homogenous distribution of excited states and quenchers will be nonexponential if diffusion is not fast enough to re-establish equilibrium after each quenching process. However, if such a quencher were present, the observed decay would not approach that observed for ultraviolet excitation but would show a reduced lifetime characteristic of the quencher concentration and reaction rate. Experiments using homogenous concentrations of excited state quenchers (oxygen, etc.) must consider this phenomena.
- Decay from an initial homogenous distribution of radicals and excited states where the radical disappear through bimolecular quenching also predicts nonexponential decay. This reaction scheme could perhaps give qualitative fits to experimental decay. However, decay from such an assumed distribution is very sensitive to the proton beam intensity. No change was observed in decay shape for two orders of magnitude change in proton-beam current.
- A sum of exponentials is not consistent with the observed decay of solutions of benzene in cyclohexane measured over the temperature range 5-50°C.
- Because of the extreme sensitivity of fluorescence lifetime to solvent temperature, the possibility of a thermal spike model was considered. It can be shown that if the proton energy is assumed to be initially deposited as thermal energy along a proton track of 3.5 nm radius from which it then flows by macroscopic heat conduction, the temperature will have dropped to less than a degree above ambient temperature for times greater than a few nanoseconds. This model can thus be ruled out.

The nonexponential decay of benzene fluorescence following proton irradiation has been shown to be consistent with a diffusion model based on radical quenching of excited states. We have obtained an initial track radius of 3.5 nm which sets an upper limit on the distribution of excited states at times when diffusion processes become important ($>10^{-12}$ sec). Radical quenching of excited states is very efficient and approaches the diffusion controlled limit of a reaction on every encounter. For initial distributions of excited states created by lightly ionizing radiation, $R(t)$ (see Eq. 3) will have a different form from that for proton irradiation, and will therefore lead to different pulse shapes and fluorescent yields. If a spur-type distribution is considered to be correct, then we have diffusion from spherical entities. Figure 2.10 illustrates the decay for a spherical spur of radius 3.0 nm with 4 radicals per spur, also shown are the predicted decay for proton irradiation and for decay where there is no radical quenching. For lightly ionizing radiation the initial densities are low enough and the diffusion fast enough that nonexponential decay is observed only on a subnanosecond time scale. Areas under the curves represent expected differences in fluorescence yields for protons and γ -rays. They give a qualitative explanation of α/β ratios based on radical quenching of excited states. Before detailed comparison can be made we must include a more realistic distribution of initial energy deposition for lightly ionizing radiation. Also, the rate of excited states in the solvent and a detailed analysis of ion recombinations must be included.

U.V. Excitation

Investigations are continuing on the intramolecular and intermolecular processes in organic liquids. Our recent studies of the temperature dependence of benzene fluorescence suggest that the radiative and nonradiative intramolecular processes are essentially unaffected by environment and the solvent furnishes a rapid vibrational relaxation of the

FIGURE 2.10. Fluorescence Decay for Excitation by Protons and Fast Electrons.

initially populated levels. Decay rates in solution are adequately described by

$$k = k_0 + k_1 e^{-W/KT} \quad (4)$$

where k is the sum of radiative and nonradiative intramolecular processes and $k_1 e^{-W/KT}$ is a thermally activated quenching process of activation energy W .

At extremely low temperatures, intramolecular processes dominate and the decay approaches that of the isolated molecule. A comparison of gas phase measurements suggest that W is related to intramolecular nonradiative channels. In order to test this hypothesis we have begun to study benzene in numerous solvents with the intent of extending these measurements over a wide range of temperatures.

• **RADIATION DOSIMETRY**
AND
• **RADIATION BIOPHYSICS**

The joint purpose of these two closely integrated programs is to explore the connections between the primary physical events produced by radiation and their biological consequences in cellular systems. The radiation dosimetry program includes theoretical description of the primary events and their connection with the observable effects and experimental realization and measurement of the physical parameters needed by the theory. The radiation biophysics program tests or makes use of the theoretical developments and gathers information needed for the theories by experiments on cellular systems. This year, theoretical modeling and experimental testing on Chinese hamster (CH) cells were directed at the fundamentally interesting and an observationally distracting problem of the variation of radiosensitivity during the mitotic cycle. Also, theoretical work on time-dependent effects continued; theoretical analysis and experimental measurements for split-doses on C. reinhardi indicated that oxygen and LET both act on the primary radiation damage mechanisms but do not affect recovery processes. In a search for phenomena that have simple primary physical mechanisms, an experimental search for stochastic effects in the radiation-induced division delay of CH cells was begun. Theoretical development of the theory of LET effects encompassed the disparate behavior of CH cells and C. reinhardi into a single broad pattern consistent with our time-dependent theory. Work on microdosimetry included experimental measurements for fast electrons and Monte Carlo calculations for protons.

Radiation Sensitivity During the
Cell Cycle
W. C. Roesch

A cell's radiosensitivity changes as it moves through its mitotic cycle. These changes affect cell observations; therefore, they have to

be known so that observations may be interpreted to obtain the parameters that are determined by the primary physico-chemical phenomena. Experimental information about these changes is difficult to obtain and thus will always be fragmentary; the gaps in the data have to be filled

by modeling. Modeling¹⁶ the changes in radiosensitivity of Chinese hamster (V79) cells was begun last year. The modeling was based on the idea that rate-limiting steps in the sequence of chemical reactions constituting the mitotic cycle of the cell would make the radiosensitivity appear to be the consequence of simple, chemical-kinetic transitions between a few chemical states. (This idea is an explanation and generalization of the G_0 concept.) The study was continued to expand understanding of the basic concept and refine its application.

A fundamental advance in understanding occurred when the initial G_0 concept was distinguished from a related, many-target concept which seems more suited to radiobiological studies. The G_0 concept is that cells exist in the G_0 state until released at random to proceed through their mitotic cycle; in this model, if a population of cells that had been synchronized at some earlier time was sampled, it would be found to contain a mixture of cells--some in G_0 and some in the subsequent stages of the cell cycle. The type of mathematical averaging necessary to give the properties of a mixture of cells turns out not to be the kind needed to describe the radiosensitivity of Chinese hamster cells. The kind needed is that for a population of cells, all of which are essentially identical, but each of which contains a number of parts whose radiosensitivity may change individually. But this accords very well with the idea¹⁷ that a cell may contain a number (probably large) of targets any one of whose inactivation leads to cell death (not to be confused with the multi-target theory, in which all of a number of targets must be inactivated to cause cell death). This leads to consideration of the theory of rate-limiting steps to apply to the targets within the cells rather than to the cells themselves. Although this is an important change in interpretation, it did not affect the numerical part of the modeling.

Last year, the age-response data of Sinclair and Morton¹⁸ was used for the modeling. Continued study convinced us that it would be more accurate to start from our own analysis of the individual survival curves

that they measured. The points in Figure 2.11 show the one-event, C, and the two-event, A, factors we found for their data. The solid lines show the fit to these points of

$$C/\text{rad} = 7.5 \times 10^{-3} e^{-t/3.1 \text{ hr}} \quad (1)$$

$$A/\text{rad}^2 = 15$$

$$\times 10^{-6} (t/3.1 \text{ hr}) e^{-t/3.1 \text{ hr}}. \quad (2)$$

These are equations of the simple, chemical-kinetic type. They represent a rate-limiting step early in the mitotic cycle in which the targets are inactivated only by one-event processes and a second rate-limiting step following it in which the targets are inactivated only by two-event processes. (Processes of the two-event type can be produced in single events; there is a small correction to the data for C for this.) The mean residence times in the two steps are equal, or nearly so, at 3.1 hr. No attempt is made in this simple model to account for the rapid rise of the radiosensitivity at the end of the cycle.

These two states do not resemble the conventional G_1 -S- G_2 -M model of the mitotic cycle. Perhaps there does not have to be a relation between the two models: the cell is a very complex system and the two models may simply represent different parts of the system, namely the part sensitive

FIGURE 2.11. One-Event (o) and Two-Event (Δ) Factors for Chinese Hamster Cells Synchronized at Mitosis Fitted by the Two-State Model.

to radiation and the part that takes up tritiated thymidine during DNA replication.

In the two-state model presented here, drugs that interfere with the normal progression of the targets after the cells have been synchronized can produce what appear to be changes in radiosensitivity without actually altering the targets. Figure 2.12 shows the consequences of a number of possible ways of interfering with target progression. The points show experimental data for hydroxyurea.¹⁹ The intent is not to claim that hydroxyurea blocks target progression in any of these ways; it is to show that blockage can produce changes similar to those found experimentally and that, therefore, the possibility of blockage should be considered in drug experimentation.

Radiation Sensitivity and DNA Synthesis

M. H. Schneiderman

The preceding paper ("Radiation Sensitivity During the Cell Cycle") reported the development of a two-state model which describes the change in radiation sensitivity of Chinese hamster cells during the cell cycle. The model does not resemble the conventional G_1 -S- G_2 -M model of the cell cycle; this disparity suggests that the two models represent different parts of the cellular system and involve different metabolic pathways. If this is so, it might be possible to demonstrate the two pathways experimentally by using a drug that blocks progression along one pathway but not the other. We began experiments aimed at finding such a differential blocking.

In work some years ago we showed²⁰ that addition of cycloheximide (CHM) for a suitable interval stops DNA synthesis and seems to move mitotically synchronized cells backwards in their cycle to a point early in G_1 ; when the CHM is removed, they have to pass through G_1 again before starting DNA synthesis. Cells treated in this manner and untreated cells will be irradiated with x rays. If the CHM treatment does not also block the pathway responsible for radiation sensitivity, the CHM-treated cells may achieve the radioresistance they normally reach in S before they start DNA synthesis.

FIGURE 2.12. Comparison of Population-Change Effects with Drug [Hydroxyurea (Δ)] Effects for Chinese Hamster Cells Synchronized at Mitosis with Drug Added 1 hr Later. o - no population change; 1S - no exit from first state; 2S - no exit from second state; 12S - no exit from either state. Sites within the cells can progress normally, but the radiosensitive entity blocked: 1B - entity of first state blocked; 2B - entity of second state blocked (same as 2S); 12B - both entities blocked.

Cycloheximide itself kills some of the cells to which it is applied; this killing effect must be known and corrected for. Figure 2.13 shows the first results in this experiment: the survival of Chinese hamster ovary cells after various length treatments with CHM. The survival for G_1 cells is between 80 and 90% of the controls; this is high enough that it will not interfere with the experiment.

Split-Dose Theory W. C. Roesch

The time-dependent hit theory we developed²¹ included an equation for the survival for a split-dose, doses D_1 and D_2 delivered with an interval t between them. (The effect of changes in radiosensitivity associated with progression in the mitotic cycle was not included in this part of the theory.) This result was put into a new form that is more useful in analyzing experimental data.

FIGURE 2.13. The Effect of Cycloheximide (CHM) Treatment During G_1 on the Survival of Chinese Hamster Ovary Cells. Synchronous mitotic cells were plated into medium containing $10 \mu\text{g/ml}$ CHM and left for up to 5 hr; then the CHM was washed off and the cells allowed to grow for 10 days, when they were stained and scored for survival. Similar measurements were made on cells treated with CHM only in the intervals 2-4 hr and 2-6 hr after plating.

The new form substitutes quantities that are ordinarily measured in a split-dose experiment for parameters of the theory which are not known. These quantities are: 1) the survival, S_0 , for a single dose $D_1 + D_2$, 2) the survival, S_1 , for the dose D_1 alone, and 3) the survival, S_2 , for the dose D_2 alone. In terms of these quantities, the survival, S , for the split dose is contained in

$$\ln(S_1 S_2 / S) = \ln(S_1 S_2 / S_0) e^{-t/\tau} \quad (1),$$

where τ is a constant, the mean recovery time. The product $S_1 S_2$ equals the maximum value of the survival, S_m , expected in a split-dose experiment; S_m is the survival when enough time separates the two doses to permit complete recovery.

This equation suggests that a useful way to analyze experimental data is to plot either $\ln(S_1 S_2 / S)$ or $\ln(S_m / S)$, quantities containing only experimental data, versus the interval t on semi-log paper. If the result is a straight line, the line's slope gives the mean recovery time.

Figures 2.14 and 2.15 show this method applied to examples taken from the work of Bryant.^{22,23} Bryant used the green alga, *Chlamydomonas reinhardtii*, which is a good choice for split-dose experiments because it has a long period, some 10-12 hr, during which its radiosensitivity does not change even though it is progressing through its mitotic cycle. The figures show results for different conditions of exposure in air and nitrogen. Although there is considerable scatter in the points, it was possible to draw a straight line through each set and to draw all the lines with the same slope. Since the slope gives the mean recovery time, all the curves having the same slope-hence the same mean recovery time-means that the recovery mechanism seems to be practically uninfluenced by the ambient gas. The mean recovery time is about 35 min under all conditions.

The theory also gives a relation between the first factor on the right of Eq. (1) and one of the parameters of the theory, A , the two-event factor:

$$\ln(S_1 S_2 / S_0) = \ln(S_m / S_0) = A D_1 D_2 \quad (2).$$

FIGURE 2.14. Split-Dose Survival, S , for *C. reinhardi* for Time, t , Between Doses; S_m is the Maximum Survival Reached. (○, ●) 10.7 and 11.8 krad from 5-MeV electrons, in air; (Δ) 27.3 and 30.1 krad in nitrogen; (▲) 32 and 26.8 krad in nitrogen. Lines drawn for 35 min mean recovery time.

This quantity is easily determined, because it is the intercept on the scale of ordinates of the semi-log plot; from it, the two-event factor can be found. The two-event factors found from the data in Figures 2.14 and 2.15 for irradiation in air show some relatively small differences that can probably be attributed to differences in culturing conditions. However, the two-event factors for cells irradiated in nitrogen are about 5-10% of those irradiated in air. It seems to make little difference what gas was present between the irradiations. Thus, it appears that the ambient gas affects the amount of a certain type of radiation damage that is produced but has no influence on the rate of recovery of that damage.

The simplicity and usefulness of Eq. (1) led to examination of other theories for similar expressions. The Swann-del Rosario^{24, 25, 26} two-state model gives

$$S_1 S_2 - S = (S_1 S_2 - S_0) e^{-t/\tau} \quad (3).$$

This result differs from Eq. (1), but the difference may not be distin-

FIGURE 2.15. Split-Dose Survival, S , for *C. reinhardi* for Time, t , Between Doses; S_m is the Maximum Survival Reached. (○) 10 and 11 krad in air, recovery in air; (▲) 30 and 25 krad in nitrogen, recovery in air; (Δ) 10 and 11 krad in air, recovery in nitrogen; (■) 30 and 25 krad in nitrogen, recovery in nitrogen. Lines drawn for 35 min mean recovery time.

guishable in experimental data unless $S_1 S_2 / S_0$ is fairly large. Figure 2.16 compares split-dose curves from Eq. (1) with ones in which S was calculated from the two-state equation, Eq. (3). On the semi-log plot, the two-state curves are not straight lines, but, when $S_1 S_2 / S_0$ is small, data with the scatter normal for split-dose experiments could probably be fitted by either theory equally well. When $S_1 S_2 / S_0$ is large, there is a pronounced curvature for times that are about equal to the mean recovery time.

FIGURE 2.16. Comparison of Two Theories for Split-Dose Experiments: Hit Theory, Eq. (1), and Two-State Theory, Eq. (3).

The data of Figures 2.14 and 2.15 have very large values of S_1S_2/S_0 , but none of them show such a curvature; therefore, the two-state model does not appear applicable to them.

LET and Recovery in *Chlamydomonas*
D. S. Nachtwey and L. A. Braby

Many things, such as the surrounding gas, drugs, temperature, LET, etc.²⁷ are known to affect experiments that reflect the ability of cells to recover from radiation damage. Progress beyond the qualitative knowledge that an effect is produced can now be made by using the theory of the preceding report ("Split-Dose Theory") to analyze split-dose experiments and quantitate the effects of the agents on the parameters of the organism. Such quantitative data are needed before the effect can be related to the basic physical mechanism. In addition, however, (as in the preceding report) the quantitative analysis may give an immediate guide to new experiments by indicating whether it is the amount of the initial radiation damage or the properties of the subsequent recovery process that are affected by the agent. We are studying the quantitative effects of agents on the survival of *Chlamydomonas reinhardi* and here report on the effects of LET.

Survival curves presented last year²⁸ for *C. reinhardi* showed that 1.55-MeV protons (stopping power 20 keV/ μ m) have a two-event factor about twice that of the recoil electrons from ^{60}Co gamma rays (0.3 keV/ μ m). The same equipment and techniques have now been used to measure the split-dose survival for the protons. Two 5 krad doses delivered at 1.5 krad/s and separated by times up to 1 hr were used. Figure 2.17 shows the results along with the results of Bryant²³ for ^{60}Co gamma rays. The data are plotted as suggested in the preceding article. Both sets of data can be fitted with straight lines and those lines can be drawn with the same slope. The equality of the slopes means that the mean recovery time for the protons is the same as for the less-densely ionizing recoil electrons, namely about 35 min.

The effect of increasing the LET of the radiation is similar to that

FIGURE 2.17. Split-Dose Survival, S , for *C. reinhardi* for Two Equal Doses a Time, t , apart; S_m is the Square of the Survival to Just One of the Doses. (Δ) 10 krad from ^{60}Co gamma rays; (O) 5 krad from 1.55-MeV protons.

of substituting air for nitrogen over *C. reinhardi* (see preceding article): the two-event factor is increased and the mean recovery time is not visibly affected. Both, then, affect the initial production of damage rather than the process by which recovery from the damage occurs. The increase in LET probably increases the number of radiation products formed in the cell; the oxygen from the air probably increases the number of products that react with the cell in ways that become lethal.

Colony Survival Versus Single-Cell Survival

W. C. Roesch

In cellular radiobiology experiments, the experimenter observes whether cells can grow into colonies of a certain size. If the cells are not initially completely separated, but some of them are clumped together, the experimenter measures a probability of survival (often called the colony survival), S_0 , which is larger than the true value, S , for a single cell. Research has shown that if x_n is the probability of there being n cells in a clump and $\langle n \rangle$ is the mean of this distribution, then approximately

$$S \approx 1 - (1 - S_0)^{1/\langle n \rangle} \quad (1).$$

Many authors have assumed this equation to be true and used it to analyze data. The present work provides a theoretical basis for the formula. It also provides a means of estimating the degree of approximation of Eq. (1): the difference between $(1 - S_0)$ and $(1 - S)^{\langle n \rangle}$ is less than $[(\langle n^2 \rangle_0 - \langle n \rangle^2)/2] S^2$. Also, higher degrees of approximation can be obtained if needed.

Division Delay at Low Doses

M. H. Schneiderman, L. A. Braby, and W. C. Roesch

It seems that the killing of cells by radiation may occur in many different ways, starting from different primary physical (and biological) phenomena. There is interest in other radiation effects that may be simpler in origin and therefore be easier to analyze. We have chosen to study radiation-induced division delay in

the hope that it will turn out to be one of these simpler phenomena.

The investigation of cell killing has profited by starting with survival curves, in which the stochastic nature of the primary physical events is reflected in the killing of some cells but not others. Radiation-induced delay is found to be proportional to the absorbed dose, and there may be a stochastic explanation for this; but there have been no measurements that reflect where some cells are delayed while others are not. We began the experiment that is described here in the hope of providing at least some preliminary information of this character.

The experiment is based on the properties of cells between two particular points in the mitotic cycle; cells that have not yet passed the first point (the CHM transition point) are prevented from reaching mitosis by the presence of cycloheximide (CHM);²⁰ cells that have passed the second (the x-ray transition point) do not suffer radiation-induced delay.²⁹ Thus, in the mitotic selection technique,³⁰ the removal and counting of mitotic cells from a monolayer culture, the difference between the mitotic selection curve for CHM-treated cells and a curve for cells irradiated with a large* dose is due to cells that in the initial culture were between the two transition points.

If CHM is present in the cells during the irradiation, the mitotic selection curve is the same as that for a large dose of x rays alone;²⁰ this indicates that the cells between the two transition points were prevented from reaching mitosis. It is assumed that what stopped them was interference by CHM with the synthesis of proteins needed for their repair.

The experiment consists of irradiating cells, in the presence of CHM,

* The dose, and the resulting division delay, have to be large enough to separate the arrival at mitosis of the cells that were before and after the x-ray transition point when irradiated; any dose above about 100 rad will do this.

to small doses of radiation and comparing the mitotic selection curve with that for a high dose (with or without CHM) and with that for CHM only. If the result agrees with the former, most of the cells between the two transition points contained the delay-producing damage; if it agrees with the latter, they do not. If the result lies between the two, some cells contain the damage and some do not. In other words, they are in the dose range being sought, that in which stochastics in the primary physical events are reflected in the biological effect.

Figure 2.18 shows the results of the initial experiment. The variance of the points from their anticipated positions was greater than expected; a few points show very large deviations. This is now being studied (see "The Mitotic Selection Technique"); it may be due to incomplete collection of mitotic cells. Nevertheless, the results indicate that already at 25 rad, in the presence of CHM, the mitotic selection curve lies between that for x rays and that for CHM only. When the variance has been reduced, this result will be confirmed

FIGURE 2.18. The Effect of a Large Dose of X Rays Alone, a Small Dose of X Rays Alone, Cycloheximide Alone, and a Small Dose of X Rays Plus Cycloheximide on the Mitotic Selection Yield of Chinese Hamster Ovary Cells.

and extended to other doses to determine over what range the transition from one limiting curve to the other takes place.

The Mitotic Selection Technique M. H. Schneiderman

The technique of periodically collecting loosely bound mitotic cells by shaking them from a monolayer culture was developed sometime ago.³⁰ This mitotic selection technique is a promising way to study changes in cell progression during the mitotic cycle; plans are to use it to study radiation sensitivity near the end of the cycle and to study radiation-induced division delay. Last year, a method by which the expected yield for the shaking technique could be calculated was reported.³¹ The curve in Figure 2.19 was calculated this way for cells having a 12-hr doubling time. It was also assumed that all cells reaching mitosis, and only those cells, were shaken off and that the cells shaken off were not returned to the culture. Our early experimental yields were about half the calculated values. Changing a number of environmental factors and shaking conditions gave little increase in yield. It was then decided to establish a line of cells that had been selected for easy removal during mitosis. The crosses in Figure 2.20 show the results of shaking over a period of nearly eight hr for this cell line. Although there is still some difference from the calculated values, the results are promising and we can account for almost all of the mitotic cells.

The Lajtha-Oliver Theory W. C. Roesch

Alternative radiobiological theories frequently fit experimental data with about equal accuracy. When this happens, further development of the theories is necessary if they are to be distinguished. Extensions and conclusions regarding a dose-rate and fractionation theory that has been influential in British radiological circles are reported here.

Lajtha and Oliver³² theorized that the decrease in survival produced by an increment in dose depends only on the amount of radiation damage remaining when the increment is delivered. Further, they assumed that radiation damage decreases exponentially with

FIGURE 2.19. The Theoretical Mitotic Selection Yield of Cells from a Monolayer Culture. The population was assumed to be in exponential growth at the start of selection, to have a doubling time of 12 hr, and to have a dispersion in cell doubling times of about 20%. As cells are selected, they are removed from the population.

FIGURE 2.20. The Mitotic Selection Yield for Chinese Hamster Ovary Cells (Kla Line) During a Period of Nearly 8 hr. The crosses are measured data; the circles are the theoretical yield based on 5.8×10^6 starting cells. The initial high point probably represents loosely attached interphase cells and early G_1 cells that have not reattached after floating free in mitosis.

time after it is produced. As one consequence of these assumptions, a survival curve obtained at a very high dose rate contains the connection between radiosensitivity and radiation damage needed by their theory. Thus, each different high-rate survival curve that one assumes gives a different model based on the theory.

Oliver³³ presented an algorithm by which, given a high-rate survival curve, one can calculate survival curves for lower dose rates and survival for split doses according to their theory. We derived a formula from Oliver's algorithm:

$$S = \exp \left[\int_0^{D_E} (1/S_h) (dS_h/dD) (1 - \mu D/d)^{-1} dD \right]. \quad (1)$$

It gives the survival, S , in terms of the high-dose-rate survival, S_h , as a function of dose, D . D_E is Lajtha and Oliver's "equivalent dose,"

$$D_E = (d/\mu)(1 - e^{-\mu T}), \quad (2)$$

where d is the dose rate and μ is the decay constant of the radiation damage; here T is the length of the irradiation period. Having an analytical expression for the theory permits easier comparison with other models and theories.

The Lajtha-Oliver theory can be related to the theory that we reported²¹ by substituting

$$S_h = \exp(-CD - AD^2/2), \quad (3)$$

the high-rate survival that we obtained, into Eq. (1). The result,

$$S = \exp \left[-(C + Ad/\mu)D + A(d/\mu)^2 \left(1 - e^{-\mu D/d} \right) \right], \quad (4)$$

is the same as we obtained for the survival. Of course, Eq. (3) and Eq. (4) were conclusions of our theory while it is an ad hoc assumption for the Lajtha-Oliver theory.

It is not possible, however, to substitute the high-rate expression from any theory into Eq. (3) and obtain the low-rate result for that theory. For example, if the high-rate survival is given by the one-target/two-hit equation,

$$S_h = (1 + kD) e^{-kD}, \quad (5)$$

the Lajtha-Oliver theory gives for the survival

$$S = \left[1 + (kd/\mu) \left(1 - e^{-\mu D/d} \right) \right] \left[kd/(kd + \mu) \right] \exp[-k^2 Dd/(kd + \mu)]. \quad (6)$$

On the other hand, Swann and del Rosario²⁴ and a number of other authors^{25,26} showed that a simple, two-state model led to

$$S = (r_1 e^{-r_2 D} - r_2 e^{-r_1 D}) / (r_1 - r_2) \quad (7)$$

for the survival (where r_1 and r_2 are known functions of k , d , and μ) and that Eq. (5) is the high-rate limit of this expression. Swann and del Rosario's equation, Eq. (7), differs from Lajtha and Oliver's, Eq. (6),

even though the two have the same high-rate expression, Eq. (5). Models having two (or more) states in general lead to low-rate equations that differ from what Lajtha and Oliver predict for the same high-rate survival.

The formula we derived for the survival to a split dose according to the Lajtha-Oliver theory is

$$S = \exp \left[\int_0^{D_{E1}} (1/S_h) (dS_h/dD) \right. \\ \left. (1 - \mu D/d)^{-1} dD + \int_{D_{E2}}^{D_{E3}} (1/S_h) (dS_h/dD) \right. \\ \left. (1 - \mu D/d)^{-1} dD \right] \quad (8)$$

where

$$D_{E1} = (d/\mu) (1 - e^{-\mu D_1/d}) \\ D_{E2} = D_{E1} e^{-\mu t} \\ D_{E3} = D_{E2} e^{-\mu D_2/d} + (d/\mu) (1 - e^{-\mu D_2/d}) \quad (9)$$

and where D_1 and D_2 are the two doses and t is the time between them. If our high-rate survival, Eq. (3), is substituted in these equations, the result is the same as given by our split-dose theory (see the preceding report "Split-Dose Theory").

Theory of LET Effects W. C. Roesch

Different radiations produce different degrees of biological effect. The differences are correlated with the stopping powers (LET, or LET_∞) of the charged particles in the radiation field. They are a principal source of information for relating effects to primary physical mechanisms. Our theoretical program began with time-dependent effects; last year²¹ the emphasis began to shift to LET effects. An elementary theory was presented that agreed with the observations for mammalian cells, but data for *Chlamydomonas reinhardtii* indicated a different LET dependence. This year the theory was enlarged to include both types of behavior.

Figures 2.21 and 2.22 indicate the two types of LET effect found experimentally for low and intermediate LET

FIGURE 2.21. Survival Data for T_1 Cells from Barendsen³⁴ for Low and Intermediate LET Radiations.

FIGURE 2.22. Survival Data for *C. reinhardi* for Low and Intermediate LET Radiations. Data at 20 keV/μm from Braby and Nachtwey³⁵; the rest from Davies.³⁶

radiations. In both figures the ordinate is the quotient of the natural logarithm of the survival by the dose. This exploits the theoretical expression for the survival at high-dose rates:

$$S = \exp(-CD - AD^2/2), \quad (1)$$

which gives

$$-\ln(S)/D = C + AD/2, \quad (2)$$

the equation of a straight line. The intercept on the vertical axis equals the one-event factor, C , and twice the slope equals the two-event factor, A . Figure 2.21 shows results for mammalian cells; the one-event factor shows a linear increase with the stopping power, L , while the two-event factor is the same for all L shown. Figure 2.22 shows results for *C. reinhardi*; the one-event factor is the same for all L shown while the two-event factor increases rapidly with L .

The theory presented in last year's report was based on chemical-kinetic arguments: If time-dependent behavior arises from reactions requiring two radiation products, the probability of a lethal reaction is proportional to the product of their concentrations, each of which we take to be proportional to L . The two-event factor is equal to this probability divided by a quantity, proportional to L^2 , introduced to change from number of energy-deposition events to absorbed dose. Thus, the two-event factor is independent of L . The one-event factor can arise both from the preceding, two-product mechanism and from one-product mechanisms; its divisor to convert from events to dose is proportional to the first power of L . Thus, the one-event factor is a linear function of L :

$$C = C_I + C_{II}L \quad (3)$$

$$A = \text{independent of } L, \quad (4)$$

where C_I and C_{II} are constants. This is what is observed for mammalian cells.

Work on the time-dependent theory began from a chemical-kinetic basis as a conscious attempt to move away from the site-oriented (target-oriented) approach of most previous authors. Alternative types of LET dependence were accommodated when it was realized that a modified, site-oriented model could give rise to the same kind of time-dependence we had studied.

For example, suppose that two sites have to be inactivated to kill a cell (more specifically, the sites might be on the two strands of a DNA molecule). In contrast to the usual two-target theory, however, suppose that it is not two particular sites that have to be inactivated but any two of a group (or groups) of them (cf the similar idea in the preceding report "Radiosensitivity During the Cell Cycle"). It can be shown that the time-dependence of this model is the same as that of our earlier one and that, hence, it is characterized by one-event and two-event factors. The LET dependence is different, however. In the previous argument the probability of a time-dependent lethal event was proportional to the product of two concentrations; here it is proportional to the product of two probabilities, the probabilities of inactivating individual sites. There is no a priori way of determining how these probabilities depend on L ; however, with an eye to explaining the *C. reinhardi* data, let us assume that site inactivation is by fast reactions. Then each probability is given by an expression like that leading to Eq. (3). Since two-site inactivation might also occur in a single event, the result is

$$C = C'_I + C'_{II} L \quad (5)$$

$$A = (C''_I + C''_{II} L)^2, \quad (6)$$

where the primes indicate different constants. This is roughly what is observed for *C. reinhardi*. The experimental one-event factor does not depend on L , thus C'_{II} may be negligible. The two-event factors are not accurate enough to be sure that they are quadratic in L , but they are compatible with a quadratic.

A by-product of the above developments is an idea of where to look for radiobiological phenomena characterized by short reaction times such as radiation chemists observe for primary reactions, rather than by the times of tens of minutes usually observed for dose-rate and fractionation effects. It seems unlikely that the fast processes can be observed in the presence of the slower ones; therefore, one should consider cells for which the two-event factor as ordinarily observed is zero--in other words, cells with ex-

ponential survival curves. If such cells still show a change of survival with LET, it must be due to the C'_{II} constant. Since this constant cannot come from slow reactions, which presumably do not exist, it must come from reactions that are too fast to be observed by ordinary methods. At high enough dose rates or short enough inter-fraction times, these cells should show dose-rate and fractionation effects similar to those for slower reactions. We are studying this possibility further with the intention of doing such experiments with the pulsing capability we added to our electron Van de Graaff last year.

Microdosimetry

L. A. Braby

Some radiobiological models employ measurements of the energy deposited in small volumes, a quantity used in microdosimetry, to predict the effects of different radiations. The predictions are subject to error, particularly for low-LET radiations, because the instruments that are used in the measurements distort the results for small energy depositions. The distortion is produced by thermal noise in the electronic equipment and by signals from background radiation in the proportional counter. The background signals and some of the noise can be measured separately and corrected for; but the residual noise must be corrected for by an extrapolation. The work reported here, done in the course of obtaining microdosimetry data for our other studies, shows that the extrapolation is accurate enough for the microdosimetry of fast electrons.

The data employed in the study were obtained with a "wall-less" proportional counter³⁷ that simulated a cylindrical site $3 \mu\text{g}/\text{mm}^2$ in height and in diameter. The electrons came from a 2 MV Van de Graaff, which was operated in the pulsed mode. The Van de Graaff pulses were 50 nanoseconds long, which is much shorter than the rise time of the counter, hence, each counter signal was due to all of the electrons that entered the counter during the pulse and there was just one signal for each pulse. This method of operation adds less noise than combining the signals from the individual electrons in a multichannel analyzer³⁸ does, because noise is added to the signal only once instead of for each

electron. Background and noise not associated with the Van de Graaff pulses were eliminated by using a coincidence circuit to analyze only signals coincident with pulses. The coincident background and noise is added to the size of the analyzed signals rather than to their number; hence, it had to be removed by a Fourier-transform technique rather than by simple subtraction as in non-pulse measurements. Figure 2.23 shows a typical result; the residual noise produced the rise below 0.1 keV and the dashed line is the type of extrapolation usually made to correct for it.

To show the adequacy of the extrapolation, we used a mathematical property of energy-deposition distributions: the distribution for one dose can be obtained from that for another dose by taking the Fourier transform of the latter, raising it to the power equal to the ratio of the doses, and taking the Fourier inverse of the result. Figure 2.24 illustrates that this is an accurate calculation for distributions that have negligible distortion at small energy depositions. The open circles are the distribution

FIGURE 2.23. Typical Energy-Deposition Data Showing Noise Below About 100 eV. Dashed line indicates the approximation commonly used for the energy deposition in the noise region.

FIGURE 2.24. Energy Deposition in a $3 \mu\text{g}/\text{mm}^2$ Site When the Dose is Large Enough that the Noise is not a Serious Problem. (●) 1.7 rad, (x) 14 rad; (o) calculated from the 1.7 rad distribution and the ratio of the Van de Graaff pulse sizes.

calculated from the one marked by the solid circles to fit the one marked by the x's.

Figures 2.25 and 2.26 are similar data for doses that are small enough that distortion at small energy depositions is serious. Figure 2.25 shows the original data; the peak at 0.03 keV in the low-dose curve is due to the residual noise. The calculated curve does not fit the experimental result for the high-dose curve. In Figure 2.26 the peak is replaced by an extrapolation. The calculated and experimental results now agree, which shows that the extrapolation is accurate enough for use in microdosimetry.

Microdosimetry by Monte Carlo Calculations

W. E. Wilson

A detailed picture of the physical processes which constitute a fast ion track is important in understanding the radiation effects of such physical processes. Direct observations of

FIGURE 2.25. Energy Deposition in a $3 \mu\text{g}/\text{mm}^2$ Site When Noise is a Problem and No Correction is Made for it. (●) Over 50% of the Van de Graaff pulses deposited no energy, (x) 0.48 rad, (o) calculated from the low-dose curve and the ratio of the Van de Graaff pulses.

FIGURE 2.26. The Same Energy Deposition Data Shown in Figure 2.15 with Indicated Corrections.

track structure in condensed media is possible only in special cases; most of the available data are for gaseous phase. We are, therefore, developing the ability to calculate otherwise unavailable properties of fast ion (proton) tracks in liquid and solid phase, using computer simulation of the stochastics of the particle interactions. Quantities of interest are: spatial distributions of ionization and energy deposition, distribution of the distance to the nearest neighboring ion, and electron slowing down spectra. As with an experimental procedure, in order to develop and maintain confidence in the computer simulations it is essential to compare the calculations with actual measurements whenever possible.

We are using Monte Carlo (M-C) techniques to calculate ionization distributions in small absorbing sites for comparison with measurements made by Glass and Roesch.³⁷ In their experiment they measured pulse height distributions that protons produced in a cylindrical proportional counter filled with tissue-equivalent gas and having an effective radius in unit density material of 125 nm. The calculations of the distributions were done for a water medium rather than for tissue-equivalent gas; therefore, in order to obtain distributions for the same mean energy deposition, it was necessary to calculate the distributions for a cylinder of radius $R_{\text{H}_2\text{O}} = (S_{\text{TE}}/S_{\text{H}_2\text{O}}) R_{\text{TE}} = 135 \text{ nm}$, where S_{TE} and $S_{\text{H}_2\text{O}}$ are the stopping powers of the tissue-equivalent gas and water, respectively.

Figures 2.27 and 2.28 compare the calculated distributions with the experimentally measured ones for proton tracks passing through the site. The location of the track within the site is measured in terms of its distance from the axis of the site and is expressed as a fraction of the site radius. Figure 2.27 is for proton tracks passing through the site about 30% of the radius out from the center. Figure 2.28 shows the distributions for tracks 80% of the radial distance outward. The shapes of the respective distributions agree well in view of the large uncertainties involved in both the measurements and the calculations. Conversion of the calculated

FIGURE 2.27. Monte-Carlo Calculation of the Ionization Deposition for 3 MeV Protons Passing Through a Site of Radius 125 nm. The site is a right cylinder of height equal to its diameter. This distribution is for a proton track passing through the cylinder in its midplane and 30% of the radius away from the center.

FIGURE 2.28. Monte-Carlo Calculation of the Ionization Deposition Distribution for 3-MeV Protons Passing Through a Site of Radius 125 nm. This distribution is for a proton track in the midplane of the cylinder and 80% of the radius away from the center.

distributions from number of ionizations to units of energy was accomplished through an effective W value. We have used $W = 34$ eV for Figures 2.27 and 2.28. This value was arrived at by calculating ionization deposition distributions for 1.0-MeV protons and assuming that there was a negligible amount of delta-ray escape from the site; this allows one to equate the mean number of ionizations calculated with the product of stopping power and path length in the site.

The mean ionization produced by tracks crossing the site are shown in Figure 2.29 for three different proton energies. The open symbols indicate experimental measurements, the closed symbols are results of M-C calculations and the solid lines are the product of stopping power and path length. One can see that for 1.7 and 3.0-MeV protons the experimental and

M-C calculated results are in reasonable agreement; at 4.0 MeV, however, the M-C results are too low. We presently suspect that this shortcoming of the M-C code arises from using a form for the proton ionization cross section that is proportional to E^{-1} rather than $E^{-1}(\ln E)$; the latter is more accurate at high energy.

The quotient of the variance (σ^2) by the mean ($\bar{\Delta}$) of the ionization distributions is independent of the number of interactions; hence, this quotient is an important parameter in describing the interactions making up the observed effect. The ratios calculated with the M-C code are shown in Figure 2.30, where they are compared with the measured ones from the Glass-Roesch experiment. The rather large variation with proton energy is believed to be due to poor statistics; not enough proton tracks were averaged.

FIGURE 2.29. Comparison of the Mean Ionization Produced in a Right Cylinder of 125 nm Radius by 1.7, 3.0, and 4.0-MeV Protons. Solid symbols are the calculated results using Monte Carlo simulation of proton tracks, open symbols are experimentally measured results using a gas proportional counter, and the solid lines are the product of proton stopping power and path length within the counter.

FIGURE 2.30. Ratio of Variance (σ^2) to Mean Ionization ($\bar{\Delta}$) for the Ionization Distributions for Proton Tracks at Different Distances from the Center of the Cylindrical Site. For tracks just outside the site, the ratio increases abruptly, reflecting the transition from peaked distributions of ionization to monotonically decreasing distributions. Closed symbols are Monte Carlo calculations and open symbols are from experimental measurements.³⁷ The Monte Carlo codes predict the same abrupt change in $\sigma^2/\bar{\Delta}$ at the site boundary.

However, the overall behavior is well represented. The abrupt rise in $\sigma^2/\bar{\Delta}$ reflects a transition from peaked distributions for tracks passing through the site to monotonically decreasing distributions for tracks passing near the site.

We conclude that the M-C codes do predict the significant features of the Glass-Roesch experiment to within the accuracy of the experiment. Improvements will be made in the codes to achieve better agreement for high-energy protons.

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ANALYSIS AND EVALUATION

- **TOXIC SUBSTANCES IN NUCLEAR FUELS**
- **DEVELOPMENT OF A METHODOLOGY FOR THE ASSESSMENT OF SOCIAL VALUES**

• TOXIC SUBSTANCES IN NUCLEAR FUELS

The long-term objective of this program is to characterize toxic materials present in the nuclear fuel cycle and to provide quantitative projections of the controlled accumulation and routine release of these materials. Detailed characterization of the amounts and the chemical, physical and radiological properties of toxic effluents is required to give guidance to biomedical and environmental research and development programs. Quantities and characteristics of pollutants associated with existing and future nuclear energy systems will form a basis for guiding these research programs and help to assure that source terms used are consistent with AEC expectations of the growth of nuclear power.

This program was initiated in fiscal year 1975. The principal effort has emphasized characterizing toxic effluents from the light water reactor UO_2 fuel cycle.

Toxic Effluents from the UO_2 Fuel Cycle

R. M. Fleischman and R. C. Liikala

The implementation of nuclear energy systems poses potentially serious questions about the ecological and health effects of routine effluents and the potential release of materials from large accumulations of toxic materials. Measuring the health and ecological effects of pollutants released to the environment requires collecting data over long periods of time. Therefore, it is important that potential source terms of toxic materials in the growing nuclear industry be identified as early as possible to give guidance to these experimental programs.

This is a new program, and considerable attention has been given to establishing a technical approach that

will provide results that can be used effectively by the biomedical and environmental research community. The scope of the task for the first year has been restricted to concentrating on the most viable short-term nuclear option, i.e., the light water reactor UO_2 fuel cycle. Within that restriction, emphasis has been placed on the characterization of toxic effluents, and secondary importance has been placed on characterizing the controlled accumulation of toxic materials and projection methodology. Little or no attention will be given to developing a dynamic fuel cycle model for the current studies.

The effort to characterize toxic effluents has been initiated utilizing multi-disciplinary expertise from several available Battelle-Northwest departments. For each fuel cycle component (i.e., mining through waste

disposal) toxic effluents are being identified and characterized by their radiological, chemical and physical properties:

Radiological Properties

- Isotope
- Decay reactions to stable condition
- Half-life of each decay

Chemical Properties

- Chemical composition
- pH of liquid effluents

Physical Properties

- Physical form
- Suspended solids in liquid effluents

- Aggregates for solid effluents
- Size range of particulates in air effluents

The rate of effluent release for each fuel cycle component will be modeled based on available information. These models will be used to generate a pure UO_2 fuel cycle release projection consistent with AEC forecasts of the growth of nuclear power. Output will be in the form of Ci/year and kg/year for radiologically and chemically toxic substances released from the present to approximately 1985. Subsequent efforts will incorporate plutonium recycle in the light water reactor fuel cycle and investigations of the HTGR and fast breeder reactor cycles. As data are generated for more advanced fuel cycle concepts, the release projections will be extended to the year 2000 and beyond.

• DEVELOPMENT OF A METHODOLOGY FOR THE ASSESSMENT OF SOCIAL VALUES

The objective of this project is to develop a technique for the combination of techno-economic data with social values for decision-making concerning design and siting options for nuclear power plants. Specifically, quantified technical information is weighted by social values and subsequently summed to differentiate among alternatives. The current phase of the project is designed to test the feasibility of such a technique in a community already experienced with a nuclear power plant. The ultimate objective is to develop a standard procedure for inputting community values into the decision matrix prior to a final site/design selection.

Public Values in Nuclear Plant Siting J. B. Burnham and J. A. Hébert

The feasibility of weighting techno-economic data by social values has been demonstrated in a previous phase of this project sponsored by the Office of the Director of Regulation [Contract AT(45-1)-1830 BNWL Report 1787]. In that phase, a multiplicative model was proposed in which technical values (established by experts) were multiplied or weighted by social values (established by representative citizens).

A reanalysis of the first phase results has indicated that the social values are not independent of the technical information. This is undesirable with respect to the weighted sum model, since the multiplicative relationship requires such independence. A revision of the questionnaire as well as a revision in the sequencing of information are in progress in order to alleviate the problem.

The selection of a community will shortly be made via liaison with the Electric Power Research Institute (EPRI). A community experienced with a nuclear power plant will allow for a retrospective test of the methodology. Considerable effort has been devoted to developing guidelines for selecting a sample of involved citizens. It was considered that a random sample of citizens in a community would be insensitive to the actual dynamics of the process of selecting a site. Accordingly, three somewhat overlapping approaches will be used to identify citizens likely to be active in the selection process.

The first, the positional approach, will identify citizens and groups by virtue of formal titles (e.g., mayor, Chamber of Commerce, etc.). The second, decisional method will identify those groups and citizens active in past major decisions. Third, the reputational approach will tap those groups and citizens who are deemed

active and influential by reputation in the community. As a contrast, a small random sample of citizens can be taken. These methods, taken together, should provide a good balanced sample of active citizenry.

It is the goal of this phase to progress up to the point where a full-dress test of the technique can be

made. A complete test will require the technical quantification of the dimensions of impacts identified in the first phase of the study: aesthetics, health and safety, cultural/recreational, water quality, air quality, economics, and animal/plant life. To date, only the aesthetics dimension has been quantified.

NUCLEAR MEDICINE TECHNOLOGY AND OTHER HEALTH APPLICATIONS

- **NUCLEAR POWERED PROSTHESES**
- **MEDICAL USES OF ISOTOPES**

• NUCLEAR POWERED PROSTHESES

The totally implantable artificial heart will dissipate waste heat. We have implanted $^{238}\text{Pu}^{16}\text{O}_2$ heat sources in the thorax (50-watt) and abdomen (29-watt) of miniature swine, one of the latter being temporarily cooled by an external percutaneous coolant loop to control surface temperatures during the tissue ingrowth period. We have designed, fabricated and implanted large discoid and smaller right parallelopiped (flat plate) devices for the study of tissue heat conductance and tolerance at various surface heat flux levels. It appears that device-tissue interface temperatures of 108°F and fluxes of at least 30 mW/cm^2 are tolerable once tissue ingrowth has occurred. At least one month following implantation is required for this state to be reached.

Biological Effects of Intracorporeal Radioisotope Heat Sources

M. F. Gillis, R. D. Carmichael,
J. R. Decker, J. M. Creer,
E. G. Kuffel and A. J. Clary

The objectives of this project are: 1) to study the long-term effects of heat and radiation from radioisotope heat sources typical of those intended for use in completely implantable cardiac assist and replacement systems, 2) determine the surface temperature (or heat flux) limits for tissue-ingrown devices, and 3) work in collaboration with other AEC artificial heart program contractors as required to further program progress. We have abandoned the concept of intrathoracic (intraaortic) heat source implants, having studied one such preparation and its surgical control, and devoted the past year to: a) developing suitable devices which will contain 29-watt $^{238}\text{Pu}^{16}\text{O}_2$ heat sources and can be implanted in the abdomen of miniature swine for indefinite

periods, and b) defining surface temperature limits for such devices which will permit intimate and permanent tissue ingrowth for long-term fixation and acceptance.

Thoracic Implants. In our last annual report we described the findings on animal #4442, in which an intrathoracic (intraaortic) 50-watt heat source had been in place for about 42 weeks. Occlusion and burnup resulted. Animal #4444, a surgical control having an identical implant (minus the heat source), was sacrificed approximately the same period after implantation and the findings were unremarkable. Total thrombus occlusion of the device had occurred, and the implant was surrounded by a scar tissue capsule 1 cm thick at its thickest, considerably less thick and vascular than the capsule found surrounding the hot implant in #4442. Work on such preparations was then discontinued in favor of concentrating on abdominally located sources.

Abdominal Discoid Implants. In our previous annual report we described a swine preparation in which a discoid (8 in. diameter, 3 in. center thickness, 810 cm² surface area) having a central electrical heater was implanted retroperitoneally and, three weeks later, the heater surgically replaced with a 29-watt ²³⁸Pu¹⁶O₂ heat source. Surface temperature just before surgery, with 30 watts electrical power input, was 108°F. Telemetric measurements immediately following surgical insertion of the heat source was 112°F; this dropped to a low of 110°F in 4 days, then rose to a maximum of 116°F over the subsequent 30-day period. The animal was then sacrificed. Wherever tissue ingrowth had not taken place (over the source cavity cap and around nonveloured thermistor lead sites), the tissue was grossly necrotic. The device was enveloped by a 2-cm thick scar capsule comprised of dense, grossly healthy tissue. Histologically, the basal layer (0.5 mm) of tissue immediately against the device was necrotic, however viable granulation and fibrous tissue were found in the velour nap and beyond. It is possible that 108°F is a tolerable interface temperature, and in this case it is likely that postoperative hyperthermia caused the surface temperature to rise to a point sufficient to cause surface tissue necrosis, decreasing effective heat conductance and producing the surface temperature rise subsequently recorded.

Postoperative hyperthermia, local or generalized, is common and is an important factor in implant heat tolerance. We have commonly observed temporary hyperthermic excursions following surgery or simply anesthesia in our swine (Figure 4.1), and we have no satisfactory explanation for this. It seems to be less likely if we avoid using a physiological saline drip during surgery, though injections of this same solution in rats produced no hyperthermia. Postoperative treatment with an antipyretic (Pyrilgin, a brand of dipyrone) had no detectable effect. Since device surface temperature is directly related to inflowing blood temperature, it is important to somehow control body temperature of hot device recipients throughout the period during which tissue ingrowth is taking place; in any event, the fact that periodic py-

rexia may occur in any recipient may place a practical upper limit on device surface heat flux to something below that observed in euthermic individuals.

The importance of intimate ingrowth of viable tissue at the surface of a hot implant was dramatically, if accidentally, displayed by another discoid implant experiment in which the discoid consisted of fiberglass with a flat plate heater in the center of one side. Plate heater power was adjusted automatically to produce a surface temperature of 106°F. As shown in Figure 4.2, effective tissue heat conductance rose postoperatively (as measured by the amount of power required to produce a 106°F surface temperature) until about the fourth week, when a downward trend began. At necropsy, we found that the fabric had come loose from the discoid (and plate) surface and the intervening space had filled with purulent exudate. It can be safely concluded that the downward trend in tissue heat conductance (Figure 4.2) signalled the separation of tissue-ingrown fabric and the heater surface.

A discoid implant was fabricated containing a 29-watt ²³⁸Pu¹⁶O₂ heat source and internal cooling coils. This was implanted between layers of abdominal musculature in a miniature swine (#4397) with percutaneous tubing carrying coolant (water) tubes and electrical leads to thermistors inside the implant to record surface temperature. A feedback network (Figure 4.3) monitors surface temperature and automatically turns on water to the cooling coils whenever this temperature exceeds a preset level. Contralateral retroperitoneal temperature is continuously monitored by a telemetric implant. The surface temperature was maintained at 104°F for three weeks postoperatively, then increased to 106°F. This animal continues on experiment at this time without external evidence of implant site abnormality. The presumption is that eventually the coolant will be unnecessary and all percutaneous tubes can be severed below skin level and the animal released for long-term (years) study of this abdominally-located heat source. It is hoped that in time the animal's tissues will maintain the device surface temperature

FIGURE 4.1. Hyperthermic Excursions Following Anesthesia in Miniature Swine. All temperatures were measured telemetrically from the retroperitoneal space. The anesthetics used are indicated in parentheses; in one case no surgery at all was performed. No explanation for this phenomenon has been found, though it appears to be related to the use of IV physiological saline drip during anesthesia (see text).

FIGURE 4.2. Variation of Effective Heat Conductance K with Time Following Retroperitoneal Implantation of a Fiberglass Discoid Containing a Flat Electrical Plate Heater on the Muscle Side. The upward trend indicates tissue ingrowth; the downward trend beginning at about 4 weeks signals separation of the velour covering from the heater surface and subsequent abscessation. Plate heater power was automatically regulated to produce a tissue interface temperature of 106°F .

FIGURE 4.3. A Schematic Diagram of the Control System Used to Provide Water Coolant to a Radioisotope Heat Source in a Retroperitoneal Discoid Implant, Maintaining the Surface-Tissue Interface Temperature at a Preset Value.

at something less than 108°F , our best guess to date as to a maximum tolerable interface temperature.

This last preparation should prove whether the current device design is sufficient for long-term implantation of 29-watt heat sources, the primary objective of this program.

Flat Plate Heaters. Flat plate electrical heaters (Figure 4.4) were fabricated so that applied heat is dissipated from one side; the heat dissipation surface is a flat square 2×2 in., and the entire device is covered with nylon velour fabric. One was implanted retroperitoneally with the hot surface facing laterally against overlying muscle (animal #4879), and a second was implanted similarly but with the hot plate facing the peritoneum (animal #5193). Effective heat conductivity $K = \dot{Q}(T_s - T_b)$ was monitored and recorded, where $\dot{Q}$ is the surface heat flux in watts/cm², T_s is the temperature of the surface-tissue interface, and T_b is the temperature of the contralateral retroperitoneal space. The latter was measured telemetrically and is assumed to represent the temperature of blood flowing into the area of the heater implants.

In animal #4879, K ranged from 4.5 to $9.7 \text{ mW/cm}^2\text{-}^{\circ}\text{F}$, and in animal #5193

K ranged from 9.7 to $19.0 \text{ mW/cm}^2\text{-}^{\circ}\text{F}$, suggesting that the peritoneal direction affords better heat conductance during the early periods following implantation. These values should be compared with immediate postoperative K values of $1\text{-}2 \text{ mW/cm}^2\text{-}^{\circ}\text{F}$, increasing to about $9 \text{ mW/cm}^2\text{-}^{\circ}\text{F}$ in four weeks, in the case of the large fiberglass discoid having a plate heater in one surface (facing musculature), as shown in Figure 4.2. This supports two hypotheses: 1) that large implants result in lower initial effective heat conductance than small implants, 2) but with time and tissue ingrowth the differences become insignificant.

Large day-to-day differences in K are observed in addition to long-term trends upward. These variations are almost certainly due to variations in $(T_s - T_b)$, which enters into the calculation of K in a critical way when $(T_s - T_b)$ is small. Although increases in T_b will produce increases in T_s for a fixed surface heat flux $\dot{Q}$, the relationship is apparently not a simple one and "scatter" in K values during any experiment is the rule. Future experiments with flat plate heaters having automatically controlled surface interface temperatures are planned in an effort to define the maximum tolerable interface temperature and heat flux for retroperitoneal or abdominal intermuscular implants.

FIGURE 4.4. An Illustration of a Flat Electrical Plate Heater and Temperature Sensors (Located in the Surface Plate) Used to Study Variations in Retroperitoneal Tissue Heat Conductance with Time (and Tissue Ingrowth) and to Define Maximum Tolerable Interface Temperatures for Permanent Ingrowth of Healthy Fibrous Tissue.

• MEDICAL USES OF ISOTOPES

Initial tests in goats of the newly developed vitreous carbon-Tm (VCTm) portable blood irradiator indicate successful and persistent reduction in numbers of circulating lymphocytes. In addition, the cellular immune response, as measured by skin allograft rejection times, is reduced. Changes in design and fabrication techniques are in progress to facilitate further testing and to establish a basis for ultimate clinical use.

A study was done to assess the feasibility of developing an economical ^{13}N hospital generator. The evaluation was done on the basis of $^{10}\text{B}(\alpha, n)^{13}\text{N}$ route using $^{241}\text{AmO}_2$ as an α source. Lung scanning was considered the goal use. Cost of developing the first two units, licensing and initial check-out was estimated at \$1,283,000. Requirements for a $^{13}\text{NH}_3$ generating device cannot be assessed without further research into usable chemical/nuclear schemes.

Blood Irradiation for Medical Applications

F. P. Hungate, L. R. Bunnell,
D. H. Lester, W. F. Riemath,
M. T. Karagianes

The objectives of this program are to develop a fully portable inexpensive blood irradiator, adaptable to clinical uses, and to conduct sufficient studies of its efficacy, safety and functional characteristics so that its potential for clinical applications can be evaluated. These objectives are being approached through the combined efforts of materials, design, radiochemistry and biology specialists with animal testing of prototype units coordinated with design changes and materials modifications. Interactions with physicians who will ultimately use the devices serve to maintain a clinical orientation in this development program.

More than 30 VCTm irradiators have been constructed during the year with most being used for quality evaluation leading to improved fabrication techniques. The basic design is essentially as described in the 1973 Annual Report (BNWL-1850-PT4) with $^{169}\text{TmO}_2$ powder cast within polyfurfuryl alcohol which is subsequently converted to vitreous carbon by heating to 1000°C . Present techniques involve 12 basic steps ending with neutron activation of the ^{169}Tm to ^{170}Tm , a beta emitter (0.96 MeV max) having a 129-day half-life. The large neutron capture cross section of ^{169}Tm (125 barns) is very favorable for such activation. All machining and manipulations are done before activation so that no radiation exposure occurs during fabrication. Since the carbonization involves a 23% linear shrinkage, all dimensions are calculated to allow for this. The body of the finished VCTm unit is a

tube about 4 cm long and weighing approximately 2 g. Animal testing is done in goats having standard carotid artery-jugular vein (A-V) shunts.

Evidence of slight amounts of ^{170}Tm leakage through the inner barrier layer of early units caused a careful evaluation of this layer. Inspection with the scanning electron microscope revealed the occasional presence of extremely thin points in the inner 0.05-mm-thick barrier formed in the last step of production by flowing catalyzed polyfurfuryl alcohol through the tube lumen. The presence of these very thin spots was traced to a number of water droplets entrapped in the wax mandrel on which the active source layer was deposited, hot water having been used to heat the tool used for sizing the wax mandrel. An electrically heated sizing tool and careful polishing of the sized wax mandrel has eliminated this leakage problem. The resulting pour-through containment layer through which the blood flows is extremely smooth and appears to be nonthrombogenic. Figure 4.5 compares the surfaces of the vitreous

carbon and pyrolytic carbon tubes, the latter obtained from Gulf General Atomic for use as liner tubes in earlier VCTm units. The smoothness of the vitreous carbon has special importance in view of recent findings that smooth surfaces are minimally thrombogenic.

To optimize the blood dose from VCTm units and to minimize radiation exposures while inserting the devices into the arteriovenous (A-V) shunts, a new means of connecting to shunt tubing was required. Following consultation with physicians who will ultimately be users of the devices, a new design was developed with teflon connectors butt-joined to a square shoulder drilled in each end of the device. Because of the extreme hardness of the vitreous carbon, this drilling is done prior to carbonization. The silastic A-V shunt tubing slips over the teflon connectors when the unit is inserted into the shunt. In addition to permitting easier insertion, these new units also will yield higher blood doses with less ^{170}Tm since an inner lining tube, used in

a

8,000X

b

8,000X

FIGURE 4.5. (a) A Vitreous Carbon Blood Interface Surface as Observed with a Scanning Electron Microscope. (b) A Pyrolytic Graphite (Gulf General Atomics) Blood Interface Surface as Observed with a Scanning Electron Microscope.

earlier prototypes, is eliminated. These new devices are just becoming available so no animal tests have yet been done.

Analysis of amounts of ^{170}Tm leachable from faulty units and from purposefully broken coupons is in progress to acquire data needed in safety analyses. Leaching is done in 6N HCl with sequential samples drawn to evaluate rates of removal. While the first phase is completed, no data are as yet available. Intact units have consistently shown no indication of ^{170}Tm loss. When a metallographic sample of ^{169}Tm in vitreous carbon was examined with a scanning probe, Tm levels were at background within 5 μm of the TmO_2 layer, indicating that no diffusion occurs during carbonization.

The VCTm devices are tested in standard carotid artery-jugular vein shunts using materials common in clinical practice, i.e., teflon cannulas and silastic tubing. Since the shunts are expected to remain patent for several weeks, the tubing is stabilized to surrounding tissues near each cannula and at points of exit from the skin by dacron velour. On recovery from anesthesia the animals show no ill effects from this partial loss of vasculature to the head. Typically, the irradiator is not inserted into the shunt for several days to permit shunt stabilization and some restoration of cell pattern in circulating blood.

Animal testing has suffered from A-V shunt occlusion, typically at the venous cannula. The principal difficulty appears to be the disparity between cannula (2.6 mm) and jugular vein (12-16 mm) size in larger animals. This disparity in size is being overcome by increasing the tubing and irradiator diameter to increase flow and minimize thrombus growth. There has been no evidence of thrombus formation in the irradiation units tested so far.

Two VCTm units have been successfully used in animals. The Mark XV irradiator was tested on a male miniature 17-kg goat. Lymphocyte response is shown in Figure 4.6. At the start of testing the VCTm delivered a transit dose of 16 rads (assuming a flow rate of 100 ml/min through the

carotid-jugular shunt), giving a daily dose to blood (1170 ml) of nearly 2000 rads.

Circulating lymphocyte levels fell rapidly to less than 20% of the pretreatment level by the seventh day and then a slow rise commenced even before the exposure was terminated by shunt occlusion on the 12th day. Post-exposure circulating lymphocyte levels continued to slowly rise, but had attained only about 2/3 of the pre-exposure level during 4 months postexposure.

Reciprocal full-thickness skin allografts 2 x 2 cm were placed on the lateral body wall of both the irradiated and an unrelated male goat on the 12th day, i.e., the day the shunt closed and irradiation treatment ceased. Graft survival on the irradiated animal was 24 days while on the nonirradiated it survived only 12 days.

Packed cell volumes fell during the irradiation and, since the total exposure was over 20,000 rads, some damage to red cells may have occurred. However, 5000 units of heparin were being administered by subcutaneous injection daily to suppress clot formation in the shunt and nematodiasis (the stomach worm, *Haemonchus contortus*) was discovered after the experiment was started, so that increased abomasal blood loss due to heparinization may have accounted for much of the lowered packed cell volume.

A similar irradiator (Mark XVII, having a transit dose of 28 rads at 100 ml/min flow) was tested on a 55 kg goat with similar evidence of reduced lymphocytes in circulating blood. Because of the larger size of the animal, total blood received daily exposures of only slightly over 1000 rads. Also, due to periodic shunt occlusion, irradiation was sporadic. Circulating lymphocyte numbers fell more slowly than in the smaller goat, suggesting that daily blood dose is more important in suppressing lymphocyte numbers than is the transit dose. There was also some evidence of a correlation between periods of irradiator function and levels of serum creatine phosphokinase with decreases during irradiation and rise in level

FIGURE 4.6. Lymphocyte Response in Goat 709 Exposed to Extracorporeal Blood Irradiation from the Mark XV VCTm Irradiator.

between exposures. There is no apparent explanation of this observation and confirmation will be sought in future testing. No significant change in packed cell volume was noted during the exposure period.

Northwest physicians in kidney transplant and leukemia centers have indicated strong interest in the success of this program and continuing interaction with such ultimate users is an integral part of this program.

No effort was devoted this year toward completing the prototype semi-portable irradiator designed during the last fiscal year because of the intense interest in the portable units for potential clinical applications.

Hospital Isotope Generator

D. H. Lester, R. B. Dakan and T. E. Divine

The major effort in this program was directed to a feasibility study to determine whether continued work on the ^{13}N generator was justified by a reasonable chance of eventual suc-

cess. The goal of the work is to develop an economical system to generate ^{13}N in the hospital at the time and place of need. ^{13}N generation at the time of use is necessary due to the short half-life of approximately 10 min.

Three general areas were studied: 1) determine dose size required to carry out useful isotope scanning; 2) determine theoretical minimum for ^{241}Am source size to produce required ^{13}N dose; 3) apply efficiency factors connected with a conceptual design and do an overall cost estimate.

Previous work has established that the only viable nuclear technique other than a cyclotron was $^{10}\text{B}(\alpha, n)^{13}\text{N}$ using an α emitter as a source. ^{241}Am is one of the best of the few good α emitters; many of the others are very rare or have a poor energy spectrum for the application. This was established in earlier years of the program. Proof-of-principle tests last year indicated that the $^{10}\text{B}(\alpha, n)^{13}\text{N}$ could be made to produce countable ^{13}N .

Dr. Gordon L. Brownell of the Massachusetts General Hospital has been working on ^{13}N scanning especially for location and sizing of myocardium infarctions. He was contacted to obtain an estimate of probable dose size. Their positron camera requires 15 counts/sec for good scanning results. To meet this requirement a 10 mC per diagnosis is needed for heart or lung scanning. Much larger doses would be needed for more ambitious uses such as kidney function tests and tumor imaging. The 10 mC was used for the feasibility study since feasible use for both lung and heart imaging would be enough to justify the development work.

Theoretical calculations indicated that an intimate mixture of $^{241}\text{AmO}_2$ and ^{10}B (i.e., no attenuation losses) would give a yield of 5.2×10^{-6} ^{13}N atoms/ α .

The conceptual design considered is a chamber containing stacks of plates. Each plate consists of a thin substrate with an $^{241}\text{AmO}_2$ layer on one side and a ^{10}BN layer on the AmO_2 layer. ^{10}BN would be used to produce $^{13}\text{N}^{14}\text{N}$ which should be more easily evolved as a gas than $^{13}\text{N}_2$ since the probability for

is much lower than for

Some heating of the substrate plates was envisioned to aid the liberation of the gas.

Nitrogen gas from such a unit was considered for use in lung imaging. 10 mC of ^{13}N represents 5×10^{-13} moles of ^{13}NN . A sweep gas would be circulated through the chamber to collect the active gas. The vital capacity (i.e., the volume which can be forcibly inhaled and exhaled) in an adult human lung is about 4.2 liters so that a sweep gas volume of 3-4 liters would permit inhalation of the 10 mC dose. The chamber should therefore have a void space of 3-4 liters.

The following basis was used in evaluating the AmO_2 quantity required:

- 5×10^{-6} (α, n)/ α yield
- 50% α attenuation in substrate
- 20% α attenuation by 5 micron layer of AmO_2
- no loss due to attenuation in an AmO_2 containment material
- 100% collection of all ^{13}NN generated in a 17 micron-thick layer of BN.

The total amount of AmO_2 needed is 1.66×10^3 g. If deposited as a 5-microns-thick layer (perhaps by RF sputtering), the total surface area would be 2.85×10^5 cm^2 . The minimum volume feasible to contain plates 1 mm apart is about 30 liters. A one-turnover sweep would result in 10 times the lung capacity (on a factor of 10 dilution). Constant operation of the generator causes build-up of ^{13}N which would help overcome this problem. However, this could result in long regeneration periods (on the order of hours) between uses of the generator.

No good understanding of the requirements for an activated ammonia generator, necessary for heart studies and other uses, has been obtained. The chemical routes considered to date are catalytic synthesis of NH_3 from ^{13}NN

and

where the $\text{Mg}_2^{13}\text{N}_3$ is obtained from

The first method involves high temperatures and pressures, the second is very uncertain. Experiments would be needed since the chemical/nuclear reactions are not well understood.

Table 4.1 summarizes a cost estimate for two developmental units to produce activated gas for lung imaging.

TABLE 4.1. ^{13}N Generator Estimated Development Cost Summary

	Total for Two Units (K)
1.0 <u>R&D Engineering Phase</u>	
1.1 Engineering Development	\$ 80
1.2 Laboratory Development	160
SUBTOTAL	\$ 240
2.0 <u>Prototype Fabrication Phase</u>	
2.1 Material Cost	\$ 600
2.2 Fabrication Labor	120
2.3 Instrumentation and Controls	30
2.4 Power and Controls	20
2.5 Engineering Design	60
2.6 AEC Licensing and Inspection	10
SUBTOTAL	\$ 840
3.0 <u>Other Development Costs</u>	
3.1 Technical Direction	\$ 30
3.2 BNW Management/Human Subjects, Committee Review	10
3.3 Hospital Testing	60
SUBTOTAL	\$ 100
4.0 <u>Contingency Factor (15%)</u>	<u>\$ 170</u>
TOTAL	\$1350

The conclusions of the study are as follows:

- Based on the present information, the development looks marginally feasible. The program needed would be costly with a long time to pay off.
- Further laboratory scale testing is needed to determine some of the materials and design requirements. There are many potential

stop points where the program might be terminated due to prohibitive problems.

- The understanding of possible routes to $^{13}\text{NH}_3$ is insufficient to make a good evaluation of feasibility. A substantial theoretical and experimental program is needed to provide enough information for evaluation. Such a program is not likely to be justified at this time.

PUBLICATIONS AND PRESENTATIONSPublication

Hungate, F. P., W. F. Riemath, L. R. Bunnell and M. F. Gillis, "Chronic Irradiation of Blood with a Portable Irradiator," Rad. Res. vol. 59, p. 169, (Abstract), 1974.

Presentation

Hungate, F. P., W. F. Riemath, L. R. Bunnell and M. F. Gillis, "Chronic Blood Irradiation: A New Approach," Presented at 14th Annual Hanford Biological Symposium, September 30 - October 2, 1974, Richland, WA, (Also in press in the Proceedings.)

ARTIFICIAL HEART PROGRAM

- **MEDICAL GRADE PLUTONIUM-238 FROM AMERICIUM-241**
- **RECIPIENT RADIATION**
- **POPULATION RADIATION EXPOSURE**

• MEDICAL GRADE PLUTONIUM-238 FROM AMERICIUM-241

The objective of this program is to compare and evaluate the economics and the technical practicability of producing medical grade ^{238}Pu by irradiating ^{241}Am recovered from spent power reactor fuel. The ^{241}Am target irradiation could be performed in power reactors.

During 1974 the ALTHAEA computer code was calibrated to match experimental data for americium and curium compositions; irradiation calculations were carried out with the ALTHAEA code to define compositions of spent nuclear fuel and americium target elements; work progressed on development of a computer program for the ^{241}Am irradiation cycle analysis and cost optimization calculations; and, preliminary estimates of ^{241}Am recovery costs from spent power reactor fuel were developed.

Medical Grade ^{238}Pu from ^{241}Am R. W. McKee

For applications in medical devices such as pacemakers and artificial hearts, a supply of ^{238}Pu with less than 0.3 ppm of ^{236}Pu is needed because of the disproportionately high radiation emitted by the ^{236}Pu daughters. Up to the present time, the principal source of ^{238}Pu has been through neutron irradiation of ^{237}Np in AEC reactors and subsequent chemical separation of the ^{238}Pu that was formed by neutron capture. However, a small amount of ^{236}Pu is also formed. Power reactors will be discharging spent fuel containing significant quantities of ^{241}Am which, if separated, could be irradiated to produce a ^{238}Pu product free of ^{236}Pu . The objective of this program is to evaluate the economics and technical practicability of producing medical grade ^{238}Pu by irradiating ^{241}Am recovered from spent power reactor fuel. The ^{241}Am target irradiation could be performed in power reactors.

The scope of the program includes: 1) projecting the availability of ^{241}Am , 2) identifying the facilities and cost of americium recovery, and 3) identifying the optimum target irradiation and processing cycle and cost of producing ^{238}Pu .

During 1974 work on this project consisted of: a) calibrating the physical constants in the ALTHAEA computer code to produce irradiation calculations that match experimental data for americium and curium compositions, b) irradiation calculations for spent fuel and americium target compositions, c) development of the AMRAD computer program for analyses of the ^{241}Am target irradiation cycle, and d) analyses and preliminary estimates of the cost for recovering ^{241}Am from spent power-reactor fuel.

Analysis of the feasibility of producing ^{238}Pu from americium recovered from spent power reactor fuel is dependent on irradiation burnup calculations at two points: 1) defining the

quantities of the americium isotopes in the spent fuel and 2) defining the composition of the americium target elements after target irradiation. The ALTHAEA¹ computer code has been used at PNL to carry out these types of calculations. The physical constants in the code have been calibrated to match analytical data for spent fuel compositions of power reactor fuels and results are quite reliable for the isotopic compositions up through the plutonium isotopes. However, the physical constants related to the production of the higher actinides including americium and curium have not been as well calibrated. For this reason an effort was made to improve the calibrations related to the americium and curium calculations and to establish the magnitude of the uncertainty in these calculations.

The resulting differences between calculated and experimental compositions were still relatively large for these isotopes and point out the desirability of further improving the calibration. Results indicate an uncertainty of 25 to 35% in the calculated americium and curium compositions. Calculated americium and curium compositions for PWR fuel are shown in Table 5.1.

A series of ALTHAEA cases were run to develop target irradiation data over the range of conditions believed to be practical. The three primary variables are irradiation time, americium isotopic composition and ameri-

cium density in the targets. Results for one case that is believed to be well within feasible limits are shown in Table 5.2.

In addition to the transmutation reactions, there will also be some fission reactions taking place in the target elements. To evaluate the feasibility of early reprocessing of the target elements, knowledge of the fission product content of discharged targets is also essential. The ORIGEN computer code² is being used for this purpose. This code provides a complete list of all of the fission products as well as the transuranium isotopes present at selected times following discharge. Physical constants in this code were adjusted to produce results that coincide with the ALTHAEA results.

The principal nuclear reactions involved in americium target irradiations are illustrated in Figure 5.1. The entire americium cycle for production of ²³⁸Pu is illustrated in Figure 5.2. A computer program based on this cycle and called AMRAD is being developed to calculate production rates, product compositions, production costs, etc. It will be used

TABLE 5.1. Americium and Curium Content of PWR Fuel at 32,000 MWd/MT Exposure.

Isotope	U Enriched Fuel g/MT		1st Pu Recycle g/MT	
	150-Day Cooling	365-Day Cooling	150-Day Cooling	365-Day Cooling
²⁴¹ Am	64	97	484	626
^{242m} Am	0.6	0.6	8	8
²⁴³ Am	79	79	872	872
²⁴² Cm	8.2	3.3	71	29
²⁴³ Cm	0.1	0.1	1.3	1.3
²⁴⁴ Cm	17.1	17.3	391	382

TABLE 5.2. Americium Irradiation Products.

Irradiation Conditions:

11-Month PWR Irradiation

Target is 33% ²⁴¹Am/67% ²⁴³Am

Target Density is 0.3 g Am/cm³

Product from First Reprocessing After 3 Months Decay:

²³⁸Pu Yield = 9 g/100 g Americium

²³⁸Pu Purity = 72%

²³⁶Pu Content = 4 Parts Per Billion*

Product from Second Reprocessing 18 Months After First Reprocessing:

²³⁸Pu Yield = 5 g/100 g Americium

²³⁸Pu Purity = 90%

²³⁶Pu Content = 0

*From neutron capture by ²³⁷Np from alpha decay of 433-year ²⁴¹Am.

FIGURE 5.1. Americium Irradiation Reactions.

FIGURE 5.2. Americium Irradiation Cycle.

for optimizing the cycle and evaluating the various alternatives illustrated in Figure 5.2. One option not illustrated in Figure 5.2 is the number of times the americium is to be recycled. One recycle appears almost certainly desirable. At some point, however, the ${}^{241}\text{Am}$ will be too depleted to be useful for recycle.

A recent study³ by the General Atomic Company of ${}^{244}\text{Cm}$ and ${}^{237}\text{Np}$ recovery from spent-power-reactor fuels for AEC's Space Nuclear Systems Division appears to provide most of

the information needed to develop costs for recovery of americium from reactor fuel reprocessing waste. The process flow sheet developed for this study provides for processing all of the high-level liquid waste from the Barnwell Nuclear Fuel Plant, after a 3-year storage period. The reference flowsheet for curium recovery consists of a solvent extraction step and an ion exchange step. The americium follows the curium in the process and is not separated until the final elution step. A pure americium fraction

could be collected in place of or together with the curium at no additional cost. Thus the costs developed for curium recovery also apply to americium recovery. If both elements were recovered, the cost could be shared.

Preliminary price estimates for ^{241}Am recovery were developed based on limited details from the above study and will be revised when additional details are obtained. The principal conclusion that can be drawn is that when artificial heart

use is sufficient to require near capacity recovery of ^{241}Am (around 1990), it should be available at \$50 to \$100/g (1974 dollar values with actual escalated cost possibly several times larger). The low end of this price range would require co-product recovery and sale of ^{244}Cm . These ^{241}Am prices should result in acceptable ^{238}Pu production costs. The ^{241}Am may cost substantially more during the introduction period of artificial hearts when production rates are low.

• RECIPIENT RADIATION EXPOSURE

The objective of this study is to determine the dose from radioisotopic heat sources suitable for circulatory support systems. Computations and measurements were undertaken to form a basis for population dose calculations. The data are presented in terms of average dose rates to a bystander as a function of distance from a recipient.

Circulatory Support Systems F. T. Cross

For the dose rate calculations, two heterogeneous mathematical phantoms as described in MIRD Pamphlet No. 5, *J. Nucl. Med.*, 10, Supplement 3, 1969 (see also Figure 5.3) were used to represent the artificial heart recipient (Phantom 1) and the bystander (Phantom 2). A Rando® female trunk phantom was used to measure the bystander dose rates and is shown in Figure 5.4. Thermoluminescent dosimeters were placed on the surfaces of this phantom, along the centerline and various other internal locations in order to determine average photon dose rates as a function of source-phantom separations. The neutron dose rates were measured with a small tissue-equivalent proportional counter which was incorporated into the Rando phantom by use of a specially constructed slab drilled to insert the dosimeter from the posterior side. The recipient phantom was built-up from tissue-equivalent material of varying frontal thickness to determine the effect of depth of burial of the artificial heart power source.

A ^{252}Cf source was used as a stand-in for plutonium in order to increase the range of neutron dose measurements and to decrease the experimental time. The californium neutron dose rates were normalized to plutonium on the basis of relative neutron emission rates cross-checked by a few relative dose rate measurements. A

nominal 50 W electrorefined $^{238}\text{PuO}_2$ source (80.14 wt% ^{238}Pu and 0.64 ppm ^{236}Pu impurity) was used to represent the artificial heart power source. The measurements were corrected to beginning of life or 0 years post-electrorefining. The measurements were further corrected for experimental variabilities in order to compare to the computations which were for a nominal 33 W source containing 0.3 ppm ^{236}Pu and located on the recipient centerline at 22 cm above the plane separating the trunk from the legs. This size source is more representative of that projected for powering the artificial heart.

Figure 5.5 shows the computations normalized to the measurements for a 33 W ^{238}Pu source containing 0.3 ppm ^{236}Pu . The dose rates, with an estimated accuracy of $\pm 25\%$, are shown at beginning of life or 0 years post-electrorefining. The integrated dose rates in gm-mrem/hr can be determined by multiplying the values in Figure 5.5 by the masses of the various regions which are given in the MIRD Pamphlet as 43,982 gms for the trunk, 70,036 gms for the total body, 5,278 gms for the head and 20,776 gms for the legs. As the mean design life of the artificial heart is projected at 10 years, the photon dose rates in Figure 5.5 should be multiplied by 1.9 to account for the ^{236}Pu impurity at 10 years and by 1.4 to determine the mean photon dose rates over the 10-year period (for extrapolation purposes, the mean photon dose rates

Neg 747021-2

FIGURE 5.3. Dimensions and Coordinate System of Adult Human Phantom.

Neg 747773-1

FIGURE 5.5. Average Dose Rates to Phantom 2.

Neg 747021-2

FIGURE 5.4. Average Woman Rando Trunk Phantom.

would increase by a factor of 2.0 for 0.6 ppm ^{236}Pu impurity). The mean neutron dose rates are about 4% lower than the values in Figure 5.5 for the 10-year period.

As demonstrated in Figure 5.5, the regional neutron and photon dose rates converge upon one another as the phantom separations increase. With even as close as a 4 ft separation, the values are within 20%. For source locations other than (0,0,22) the dose rates to Phantom 2 can be adjusted by application of the usual inverse-square of the distance and the exponential change in dose rate with tissue thickness. The tissue interaction coefficient is approximately 0.031 cm^{-1} for the photons and 0.121 cm^{-1} for the neutrons.

• POPULATION RADIATION EXPOSURE

The objective of this project is to develop comprehensive estimates of the radiation dose to the population that would result from successful development and routine use of Pu-238 fueled artificial hearts.

The REPRIEVE computer program designed to carry out the calculations and summations necessary to identify the population dose by detailed classifications of heart device recipients and radiation exposure subjects was completed.

The basis for projecting future heart disease death rates was revised and the "best estimate" projection now tracks statistical data remarkably well. A series of REPRIEVE cases were prepared that covered the range of uncertainties in future artificial heart use. Population doses were substantially less than the preliminary estimates reported in the 1973 annual report. Total population dose estimates range from 60,000 to 500,000 rem/yr in the year 2000.

Population Radiation Exposure R. W. McKee and L. L. Clark

Successful development and routine use of Pu-238 fueled artificial heart devices would expose persons in the vicinity of these devices to small amounts of radiation. The objective of this study is to develop comprehensive estimates of these low-level radiation doses so that the physiological implications to others than the device users can be evaluated.

The method used in this study to develop the estimates was to subdivide the potential artificial heart population into small identifiable groups whose daily activities could be identified employing sample survey techniques. Dose calculations for all of the activities were summed up for all user groups to estimate the population dose. Essential components of the study include: 1) projections of potential artificial

heart users by age, sex, household characteristics, etc., 2) surveys to develop time-distance relationships of potential recipients with others, 3) calculations of radiation dose factors, and 4) development of a computer program to sum up and classify the population dose.

During 1974 the REPRIEVE computer program expansion was completed, dose rate calculations were revised, new projections of heart disease death rates were developed and a series of REPRIEVE cases were prepared that encompass the range of uncertainty in the principal parameters of the population dose estimates. An analysis of potential population dose from Pu-238 powered cardiac pacemakers was also prepared and a report published.⁴

The REPRIEVE computer program was revised to provide a more comprehensive dose estimate and a detailed breakdown of the population dose.

The calculation requires 23,000 input items and the final population dose matrix includes 240,000 components. The population dose can be identified in terms of age, sex and relationship to the device recipients; age, sex and household type of the recipient and by major activity classifications. However, a portion of the dose (about 35%) cannot be identified other than to the general populace.

The dose versus distance relationships were recalculated using an improved version of the QADP5A computer code. This resulted in significantly lower dose rates than previous calculations. The calculations showed good agreement with phantom experiments.

One of the important parameters affecting potential demand for an artificial heart is the future incidence of fatal heart disease. From an analysis of statistical data for other fatal diseases now under control, a learning curve relationship had previously been derived that appears to predict fatality incidence. Use of the relationship requires identification of a peak in death rate statistics. Recent data pointed up some inconsistencies in the previous application of the relationship to fatal heart disease and the analysis was redone based primarily on ischemic (formerly arteriosclerotic) heart disease. The agreement between the resulting projection and the available statistical data is remarkable. For example, starting with the 1965 death rate for total heart disease, an age-adjusted (1940 basis) death rate of 242 deaths/100,000 was projected for 1973 compared to 243 deaths/100,000 obtained from provisional vital statistics.

In addition to the above "best estimate" projection of fatal heart disease incidence, both a maximum and minimum incidence were projected. The maximum projection assumes that crude heart disease death rates will remain at the average rate since 1965. (The trend appears clearly downward, however.) The minimum projection assumes that heart disease will de-

cline at a rate characterized by other fatal diseases now controlled (this is a quite optimistic assumption).

Combining these projections with estimates of eligible candidates, death rates and device failure rates, and assuming general use could begin in 1985, the estimates of potential artificial heart use shown in Figure 5.6 were developed. The number of devices in use and population dose estimates for the year 2000 are shown in Table 5.3. A typical distribution of the dose is shown in Table 5.4. Other cases have also been calculated taking into account uncertainty ranges in eligible candidates, death rates after implant and device failure rates. These expand the range of population dose estimates in the year 2000 to 60,000 to 500,000 rem/yr. Results will be published in a topical report in preparation at year end.

FIGURE 5.6. Estimates of Potential Artificial Heart Use.

TABLE 5.3. Estimated Population Dose From Artificial Hearts in the Year 2000.

	Number of Devices in Use	Total Annual Population Dose, Manrem
Best Estimate	210,000	360,000
Maximum	260,000	450,000
Minimum	100,000	190,000

TABLE 5.4. Distribution of Population Dose.

	Percent of Population Dose	Average Dose Per Person rem/yr
Spouses		
Under Age 50	10	0.9
Age 50 and Over	25	0.8
Other Household Members		
Age 0 - 17	6	0.2
18 - 49	3	0.2
50 +	1	0.2
Non-Work Associates	9	0.007
Work Associates	11	0.02
General Populace	35	0.0003 to 0.0006

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