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ANNUAL REPORT FOR 1973 TO THE
USAEC DIVISION OF BIOMEDICAL
AND ENVIRONMENTAL RESEARCH

PART 4 PHYSICAL AND ANALYTICAL
SCIENCES

Battelle

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PACIFIC NORTHWEST LABORATORY
ANNUAL REPORT FOR 1973
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 1974

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

PREFACE

The Annual Report for 1973 to the US Atomic Energy Commission's Division of Biomedical and Environmental Research represents a change from previous annual reports. For the past 22 years, its composition has reflected our organizational structure--each part of the report was the responsibility of the appropriate research department. In the past several years, research performed for DBER has become more interdisciplinary and more interdepartmental until now only a few projects are conducted wholly within one department. To reflect this change, this report is organized by major program categories according to our schedule-189 submissions. Each part of the Annual Report is comprised of project reports authored by scientists from several research departments. The Annual Report consists of four parts:

Part 1	Biomedical Sciences	Coordinator: R. C. Thompson Editor: J. L. Simmons
Part 2	Ecological Sciences	Coordinator: B. E. Vaughan Editor: J. L. Engstrom
Part 3	Atmospheric Sciences	Coordinator: C. L. Simpson C. E. Elderkin Editor: J. A. Powell
Part 4	Physical and Analytical Sciences	Coordinator: J. M. Nielsen Editor: L. L. Lahart

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1957	HW-53500
1958	HW-59500
1959	HW-63824, HW-65500
1960	HW-69500, HW-70050
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1962	HW-76000, HW-77609
1963	HW-80500, HW-81746
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1971	BNWL-1650, Vol. 1, PT 1-2, BNWL-1651, Vol. 2, PT 1-2
1972	BNWL-1750, Vol. 1, PT 1-2, BNWL-1751, Vol. 2, PT 1-2
1973	BNWL-1850, Vol. 1, PT 1-4

W. J. Bair
Program Director, Life Sciences

FOREWORD

Part 4 of the Pacific Northwest Laboratories Annual Report for 1973 to the USAEC Division of Biomedical and Environmental Research 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.

Many programs formerly included under the subheading Radiological Sciences appear in other parts of this Report. Studies of the uranium mine aerosols and their inhalation are in Part 1, Biomedical Sciences. Part 2, Ecological Sciences, contains reports on the studies of radioanalytical procedure development, fate and effects of radionuclides in the environment in Alaska, ^{55}Fe in the world population, biogeochemical cycling in the Columbia River, studies related to the Geochemical Ocean Section Study (GEOSECS), and physical and radiological chemical studies of ocean solutions. Part 3, Atmospheric Sciences, contains reports on studies of precipitation scavenging, tracer studies in the atmosphere (METROMEX), radioactive fallout rates and mechanisms, and studies performed at a survey station on the Olympic Peninsula to determine radioactive and chemical pollutant levels in air entering the United States after a long trajectory over the Pacific Ocean.

Julian M. Nielsen, Manager
Radiological Sciences Department

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INSTRUMENTATION TECHNOLOGY

- **RADIATION INSTRUMENTATION - RADIOLOGICAL CHEMISTRY**
- **NUCLEAR TECHNIQUES IN MEDICAL SCIENCE**

has an efficiency for very small samples as high as 15%, but this falls rapidly to about 0.2% for a 10-in.-diameter source. The background for this detector is in the range of about 0.1 counts/min/peak area, similar to the multidimensional spectrometer.

These factors indicate that for coincidence gamma-ray emitters (which include about three-fourths of the gamma emitters of interest in the environment), the multidimensional

analyzer would be the system of choice. This system is also the best for large samples. The detector of choice for a point source of single gamma-ray-emitting radionuclides would be the Ge (Li) detector. In practice both of these systems are often used to measure as many as 20 to 25 radionuclides in environmental samples without chemical separation. In more complex mixtures chemical separations may be required before instrumental analysis.

Neg 736710-1

FIGURE 1.1. Gamma-Ray Spectra of ^{60}Co Plus ^{137}Cs

A NaI(Tl) anticoincidence shield serves to increase the peak to Compton ratio of a large Ge(Li) detector system by a factor of 65 for ^{60}Co gamma emissions from a thick sample. This provides an overall peak to Compton ratio of greater than 1000 to 1 with a 20% efficient Ge(Li) diode. This development allows many single gamma-ray emitters such as ^{137}Cs to be easily detected in the anticoincidence spectra from a sample matrix containing much higher levels of a variety of coincidence gamma-ray emitters.

Instrumental Neutron Activation Analysis for Determination of Neutron Sputtering of Candidate Materials for Controlled Thermonuclear Reactor Containment Vessels

R. L. Brodzinski and L. A. Rancitelli

Fossil fuel reserves are diminishing rapidly. Fission power reactor construction will not be adequate to fill energy requirements of the future and new energy sources are therefore being critically evaluated. The controlled thermonuclear reactor (CTR) appears very promising as a long-term, high-efficiency, economical power plant.

One engineering problem to be considered early in the development of the CTR is fast-neutron sputtering of atoms from the wall of the primary containment vessel. This process, analogous to atomic sputtering of materials, has not been examined experimentally, although theoretical calculations indicate it could be sufficiently severe to render the current concept of the CTR inoperable. If the sputtering rate is too high, fast neutrons produced by the fusion reaction might remove containment vessel material so rapidly that the vessel burnup rate would be prohibitive and/or the fusion reaction would be quenched by the sputtered material.

Instrumental neutron activation analysis techniques developed for environmental and biological research programs will be adapted to experimentally determine the fast-neutron

sputtering rates of various CTR containment-vessel candidate materials. The large crystal NaI(Tl) gamma-gamma and beta-gamma-gamma multidimensional spectrometers, the Ge(Li) anticoincidence shielded diodes developed earlier, and a gas-flow proportional counter or Si(Li) diode system will be used to measure the activation products of the sputtered materials. A target-catcher arrangement will be irradiated with known fluences of fast neutrons. The atoms sputtered from the surface of the targets will adhere to the catchers and subsequent thermal neutron activation analysis of these atoms will yield the sputtering rates.

Principal candidate materials for CTR containment vessels are niobium, molybdenum, titanium, vanadium, and stainless steel. High-purity silicon and aluminum foils are being evaluated for use as catcher foils. Initial investigations will concentrate on niobium targets where the principal activation radiations are X-rays from ^{94m}Nb decay. Rapid chemical separation techniques will be developed and the niobium activity will be measured with a gas-flow proportional counter or Si(Li) diode at the reactor site. Other reactions and detection systems will be used for the various target materials according to Table 1.1 which gives the sensitivity of the technique in minimum number of measurable sputtered atoms.

TABLE 1.1. Sensitivities of Various Detectors for Neutron Activation Analysis of Candidate Materials for Controlled Thermonuclear Reactor Containment Vessels

ELEMENT	DETECTION REACTION	$t_{1/2}$	γ TRANSITION(S) (MeV)	DETECTOR ^(a)	COUNTING TIME (min)	NUMBER OF ATOMS NECESSARY FOR					
						$\pm 10\%$ PRECISION ^(b)				$\pm 50\%$ PRECISION ^(b)	
						TRIGA	IRRAD. TIME (hr)	TRIGA	IRRAD. TIME (hr)	TRIGA	N REACTOR
Ti	$^{50}\text{Ti}(n,\gamma)^{51}\text{Ti}$	5.8 min	0.320	MDA Ge(Li)	12	1.5×10^{13} 5.5×10^{14}	0.3	1.5×10^{12} 5.5×10^{13}	0.3	2.5×10^{12} 9.8×10^{13}	2.5×10^{11} 9.8×10^{12}
Ti	$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	83.9 d	1.121 x 0.889	MDA	1000	3.3×10^{13}	6	8.3×10^{11}	24	4.9×10^{12}	1.2×10^{11}
Ti	$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	1.83 d	1.040 x 0.983	MDA	1000	5.8×10^{11}	6	1.7×10^{10}	24	8.6×10^{10}	2.5×10^9
V	$^{51}\text{V}(n,\gamma)^{52}\text{V}$	3.75 min	1.435	MDA Ge(Li)	8	1.4×10^{10} 8.9×10^{11}	0.2	1.4×10^9 8.9×10^{10}	0.2	1.8×10^9 3.6×10^{10}	1.8×10^8 3.6×10^9
Cr	$^{50}\text{Cr}(n,\gamma)^{51}\text{Cr}$	27.8 d	0.320	MDA	1000	7.1×10^{11}	6	1.7×10^{10}	24	1.1×10^{11}	2.6×10^9
Mn	$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	2.58 hr	1.811×0.847 0.847	MDA Ge(Li)	360	3.6×10^9 2.2×10^{11}	6	3.6×10^8 2.2×10^{10}	6	3.6×10^8 4.4×10^{10}	3.6×10^7 4.4×10^9
Fe	$^{54}\text{Fe}(n,\gamma)^{55}\text{Fe}$	2.6 yr	Mn X - RAYS	GFPC	1000	4.8×10^{14}	6	1.2×10^{13}	24	6.2×10^{13}	1.6×10^{12}
Fe	$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	45.6 d	1.095 x 0.193	MDA	1000	5.8×10^{15}	6	1.4×10^{14}	24	8.6×10^{14}	2.2×10^{13}
Co	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	5.26 yr	1.173 x 1.332	MDA	1000	9.1×10^{11}	6	2.2×10^{10}	24	1.3×10^{11}	3.2×10^9
Ni	$^{58}\text{Ni}(n,p)^{58m}\text{Co}$	9.2 hr	Co X - RAYS	GFPC	1000	3.2×10^{11}	6	1.4×10^{10}	24	4.0×10^{10}	2.5×10^9
Ni	$^{64}\text{Ni}(n,\gamma)^{65}\text{Ni}$	2.56 hr	1.115×0.368 1.481	MDA Ge(Li)	360	6.9×10^{12} 1.5×10^{13}	6	6.9×10^{11} 1.5×10^{12}	6	1.0×10^{12} 2.4×10^{12}	1.0×10^{11} 2.4×10^{11}
Cu	$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	12.8 hr	1.340	MDA Ge(Li)	1000	8.7×10^9 4.6×10^{10}	6	3.0×10^8 1.5×10^9	24	1.7×10^9 1.8×10^9	5.8×10^7 6.1×10^7
Nb	$^{93}\text{Nb}(n,\gamma)^{94m}\text{Nb}$	6.3 min	Nb X - RAYS	GFPC	13	7.7×10^{11}	0.3	7.7×10^{10}	0.3	7.5×10^{10}	7.5×10^9
Nb ^(c)	^{94}Nb	2×10^4 yr	0.871 x 0.702	MDA	10,000	2.1×10^{11}	—	—	—	3.8×10^{10}	—
Zr	$^{94}\text{Zr}(n,\gamma)^{95}\text{Zr}$	65.5 d	0.756 or 0.724	MDA Ge(Li)	1000	2.9×10^{14} 2.6×10^{16}	6	7.2×10^{12} 6.4×10^{14}	24	5.6×10^{13} 5.2×10^{15}	1.4×10^{12} 1.3×10^{14}
Mo	$^{98}\text{Mo}(n,\gamma)^{99}\text{Mo}$	66.7 h	0.410 x 0.372	MDA	1000	1.3×10^{12}	6	3.5×10^{10}	24	2.0×10^{11}	5.3×10^9
Au	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	2.7 d	0.412	MDA Ge(Li)	1000	1.8×10^9 2.7×10^{10}	6	4.9×10^7 7.7×10^8	24	3.5×10^8 7.0×10^9	9.6×10^6 2.0×10^8

(a) MDA - MULTIDIMENSIONAL ANALYZER - NaI (TI), Ge(Li) - DIODE, GFPC - GAS FLOW PROPORTIONAL COUNTER

(b) PRECISION IS BASED UPON COUNTING STATISTICS ONLY, AND CALCULATIONS ASSUME NO DECAY PERIOD BETWEEN TIME OUT OF REACTOR AND START OF COUNTING.

(c) PRODUCED BY IRRADIATION OF PURE Nb AT 4×10^{15} n/cm²/sec FOR 60 DAYS

Laboratory Computer Processing of Ge
(Li) and Coincidence NaI(Tl) Spectra
J. R. Kosorok

A laboratory computer has been developed for effective and accurate quantitative analysis of gamma-ray spectra from environmental samples and neutron activated specimens.

The laboratory facilities serviced by the computer include Ge(Li) detectors and their associated spectrometers as well as single and multidimensional NaI(Tl) spectrometers. Data are transferred to the computer with DECTape or with special purpose hardware and programs developed at Battelle-Northwest (BNW).

For quantitative analysis, spectra are transferred into the system on one of the spectrum input channels, reports are printed on a fast printer, and the spectra examined on a display. Spectra can enter the analysis instrument through the spectrometer memory, pulse-height-to-digital-converter interfaces, or magnetic tape. A Teletype keyboard/printer is used as the instrument controller because the instrument and the user can interact to provide all the auxiliary data required for complete spectrum analysis.

Program development uses magnetic tape to store the completed programs, the disc to store the parts of the programs that will be loaded into the main computer memory, the keyboard/printer to edit the programs, and the fast printer to list the programs.

The large number of gamma-ray spectra from environmental radioactivity obtained from the NaI(Tl) spectrometers are the impetus for

developing computer programs for multidimensional analysis. A small control computer transfers data from the NaI(Tl) spectrometer memories to magnetic tape. The tapes are then carried to the computer for processing. Having spectra available on tape allowed the development of the analysis programs for the direct transfer channel which will eventually be used. The standard spectra from the NaI(Tl) spectrometers are not accumulated with sample spectra but are collected during the calibration of the spectrometer. The calibration results are used to prepare the environmental radionuclide (ER) libraries which contain peak locations, background information and matrices for Compton and efficiency corrections. Hence, a number of libraries for different geometries and collections of radionuclides will be available. The computer program for ER samples calculates disintegrations per min per unit volume or weight taking into account uncertainties in the background, the original sample, the Compton correction matrix and the efficiency factors. The report also contains information about the quality of the analysis and allows a user who may question the results to check manually the computer analysis.

Neutron activation analysis (NAA) spectra are either transferred from spectrometer memories or are collected in the computer memory directly from a pulse-height-to-digital-converter (PHDC). Since a direct PHDC to computer interface occupied too much computer time for counting intervals longer than 10 min, a direct transfer from the analyzer

memory to the computer was developed. When a spectrum is stored on magnetic tape or listed on the printer, enough identification data accompany it to make quantitative analysis possible. The following example is for NAA spectra, but similar data are required for ER spectra.

- Each spectrum is given an identification code and processed with a specific reference standard.
- The weight is given to compute a ppm value for each element.
- The time irradiated and the time counted are listed to correct for decay.
- The counting interval must be recorded to normalize all spectra and to correct for decay of radionuclides with short half-lives.
- A spectrum must be identified as a standard or a sample to allow automatic data analysis.

A standard of known composition is selected for a specific group of elements common to both standard and sample. In any single library up to a maximum of 40 isotopes may be chosen. For every spectrum, the data in the standards library are constant.

With the computer, any hardwired spectrometer memory can be used that signals when the contents of a channel are ready for reading and that advances to the next channel when signaled that the data have been read. Each memory that is connected to the computer has part of the computer interface located near it. When the computer reads a spectrum, it first reads eight digits which identify the particular memory, the

number of channels to be read (512, 1024, 2048, etc.), and an identification number for the particular spectrum. Then the spectrum itself is read. Locating part of the interface near the spectrometer has two advantages: (1) differences in the manufacturer's memories can be normalized so the main interface at the computer sees a standard data form and (2) individual memories can be isolated electrically from the computer and from each other so electrical noise in signal grounds will not degrade the detector resolution.

The data channel interface located at the computer has capacity for eight different analyzer memories. It selects the first memory requesting access to the computer and locks out the others. The computer requires only a few seconds to transfer a 2048 channel spectrum and to convert from the binary coded decimal data of the analyzer to the binary data that are computer compatible. When the data have been transferred, the interface reexamines all eight memories.

Natural Background in Gamma-Ray Spectrometer Systems

N. A. Wogman

A variety of radionuclides in our environment is presently being studied at very low levels to more carefully define the radiation exposure of man. To measure these low-level concentrations, the development of low-background spectrometer systems is required. Cooper⁽¹⁾ has discussed the critical parameters which govern

the sensitivity limits of gamma-ray spectrometers. The two most important factors in his discussion are the detector efficiency and the magnitude of the background. For most high-efficiency, low-level gamma-ray spectrometer systems little can be done to improve the efficiency. However, much can be done to lower the background, which lowers the minimum detectable disintegration rate for a specific isotope.

The radioactive chains of ^{232}Th and ^{238}U plus the ^{40}K are present to some extent in the aluminum housing material of NaI(Tl) or Ge(Li) detector systems and in the NaI reflective materials and the glass phototubes. The simplest method of reducing the background from the ^{40}K in the phototube glass is by use of a NaI light pipe. A ten-fold reduction is possible with a 4-in. NaI light pipe and a 100-fold reduction can be achieved with an 8-in. NaI light pipe. Since Ti metal has been found to be very pure, methods are being studied to fabricate a Ti housing for a 12-in.-diameter by 8-in.-thick NaI(Tl) detector. The Ti housing should have a lower background than the aluminum housing presently used because of lower U, Th and ^{40}K impurities. The selection of Ti or other specialized construction materials should allow a substantial improvement in what are already the world's lowest level NaI(Tl) and Ge(Li) gamma-ray spectrometer systems. Improvements in these systems will make possible many meteorological, oceanographic and other environmental research

experiments which require accurate, low-level radioactivity measurements.

High-Purity MgO for Low-Level NaI(Tl) Crystal Systems

W. C. Weimer, N. A. Wogman and
L. A. Rancitelli

In the analysis of samples for their low-level gamma-ray-emitting radionuclides, various types of spectrometers are utilized, but the principal low-level spectrometer systems use NaI(Tl) crystals as detectors. As in all counting systems, the lower limit of detectability is governed by the natural background of the detector system. The residual background in these NaI(Tl) low-background systems originates from activity within the detector or its assembly and from cosmic-ray interactions within the assembly or its shield. A major source of the daughter isotopes of ^{238}U and ^{232}Th in these systems is the MgO used as a light reflector partially surrounding the NaI(Tl) crystals. We have determined the U and Th contents of MgO from several suppliers to select materials which will reduce the background in future multidimensional analyzer systems.

The ^{232}Th and ^{238}U impurity concentrations in some of these samples have been determined by direct counting of the MgO with a multidimensional system. However, since the direct method requires a counting period of approximately one week per

A practical limit to the sensitivity and selectivity of the system depends on the efficiency of the beta gates and their insensitivity to gamma-radiation. For example, if the sample has large amounts of non-beta-gamma-coincident isotopes, such as ^{137}Cs , and the beta detectors are frequently triggered by gamma-rays, then coincident, single, and Compton scattered gamma-rays which are in random coincidence with the pseudo-beta event will be stored in the beta-gated memory. The net effect is an increase in the background of the beta-gamma-gamma spectrum.

Two additional detectors have been evaluated for use as beta gates in an effort to reduce the gamma sensitivity of the beta detectors without significant losses of beta efficiency. ORTEC gold-surface-barrier silicon diodes with a surface area of 100 mm^2 and a depletion depth of 100μ were coupled to ORTEC 125 preamplifiers and the output signals used as beta gates. These small solid-state detectors are less sensitive to gamma-radiation by a factor of 20 than the large gas-flow counters but they are also less sensitive to beta particles, although by a smaller factor which is somewhat dependent on the energy spectrum. However, due to their small size, the solid-state beta gates only provide an overall sensitivity increase for point source samples and are not applicable to large samples.

The second detector type evaluated for gamma insensitivity is a BNW laboratory prototype gas-flow counter. Since gamma interaction probabilities are roughly proportional to the mass

of material present, counters were constructed of very thin-walled plexiglas coated with an electrical conducting silver paint; this is in direct contrast to the large 5-in. commercial detectors which have heavy copper bodies. In addition, the large commercial counters only operate effectively with P-10 gas (90% Ar, 10% CH_4) or pure CH_4 . If suitable lightweight gas counters can be designed to operate on Q-gas (98.7% He, 1.3% C_4H_{10}), further reduction in gamma sensitivity should be possible due to the reduced nuclear interaction probability of the counting gas. Work is continuing on their design, and it is expected that the overall sensitivity and selectivity of the beta-gamma system will be increased by a factor of 10 and of the beta-gamma-gamma system by a factor of 100.

Development of a Gas Proportional Counter for Measuring Noble Gas Radioisotopes at Atmospheric Levels
R. L. Brodzinski

A large-volume, high-pressure, internally anticoincidence-shielded proportional counter is being constructed for determination of noble gas radioisotopes, especially atmospheric ^{37}Ar . Argon-37 is a naturally occurring cosmogenic radionuclide which can be used to monitor the flow of air masses, interchanges of stratospheric and tropospheric air, and a variety of other atmospheric and climatic phenomena. It is also produced by nuclear detonations, primarily by

(n, α) reactions on ^{40}Ca in underground nuclear explosions, and as such can be used to trace the escape of gases from nuclear excavations and underground tests. Krypton-85, ^{133}Xe , and ^{135}Xe are also fission-product radionuclides, and the presence of these isotopes in the atmosphere from nuclear tests, nuclear reactors, or nuclear fuel reprocessing plants is of biological and political interest.

Since the rare gases are not ordinarily subject to physical or chemical combination, they are unperturbed tracers available for studies involving releases to and motions of the atmosphere. Argon-37 presents a special problem since the normal concentrations of the 35.1-day electron capture isotope are too low to be measured by ordinary radioanalytical techniques. To increase the sensitivity for this and other gaseous radioisotopes, investigators have variously increased the active volume of their counters, reduced their background, or discriminated against unwanted pulses depending on their respective applications. A gas proportional counter is being developed which will incorporate the best of all these innovations and will be applicable to the determination of all radioactive gases at essentially all levels. However, it will be of limited use for extremely small sample sizes.

The counter is of the Oeschger internal anticoincidence design. A sample counting chamber approximately 40 in. in length and 5 in. in diameter utilizes a 0.0005-in. stainless steel

anode with a cathodic wall of 0.010-in. stainless steel wires. A surrounding ring of alternating cathodes and anodes divides the outer annular area into 18 anticoincidence counters of about 1 in. in diameter. The pressure vessel, which also serves as the outer cathode for the anticoincidence guard ring, is of 3/8-in. steel. Design specifications call for operating pressures up to 20 atmospheres or more which allow for a sample counting capacity of ~ 360 liters. The counter will be housed in a 40-in.-diameter plastic scintillation anticoincidence shield and the entire assembly will be mass shielded with 4-in.-thick lead and 4-in.-thick paraffin containing 5 wt% boron for further background reduction. When applied to the determination of ^{37}Ar , pulse rise time discrimination will be employed to distinguish true X-ray events from other isoenergy pulses. Design criteria of the counter are for a background of less than 1 count per min with essentially 100% efficiency.

Thin NaI(Tl) Detectors for Measuring ^{129}I at Sub-Picocurie Levels

C. W. Thomas

A volatile fission product, ^{129}I (1.6×10^7 yr), is released from nuclear reactors, fuel element separation plants, nuclear weapons testing, and natural processes including cosmic-ray spallation of atmospheric xenon and spontaneous fission of uranium. With the increased use of nuclear power and the more stringent

controls in the release of hazardous radionuclides, it has been necessary to develop analytical methods and appropriate detectors and instrumentation to measure the radionuclides at sub-picocurie levels in environmental samples.

For each 100 disintegrations of ^{129}I , 7.52 gamma-rays (39.58 keV), 57.37 $\text{K}\alpha$ X-rays (29.5 keV), and 12.83 (34 keV) $\text{K}\beta$ X-rays are emitted. Conversion electrons of 5 and 34 keV energies are also emitted. The counting efficiencies and backgrounds as well as the monetary cost and detector size were considered in selecting a detector system suitable for measuring ^{129}I down to 0.2 pCi per liter following separation from 4 liters of milk. It was determined that backgrounds near 1 cpm and efficiencies near 50% would be required to meet this level. Liquid scintillation spectrometry was inadequate due to excessive background counting rates (>25 cpm). The counting efficiencies for beta detectors Si(Li), Ge(Li), and Ge intrinsic diodes were all less than 5% and were subsequently ruled out. NaI(Tl) thin crystal detectors had low background and relatively high efficiencies for measuring the low-energy photons from ^{129}I . In addition, the detectors could be purchased in sufficient sizes to use liberal sample mounts for counting.

The size of detectors necessary to measure ^{129}I in environmental samples was based on the supposition that iodine would be chemically isolated and mounted on an area of about 1 sq in. The thickness of detector necessary

to stop a photon of 35 keV was 1 mm. Based on these conditions, a detector size of 1-1/2 in. diameter by 1 mm thick was selected. By sandwiching samples between two detectors, counting configurations approaching 4π geometry were possible. The detectors, covered with 5-mil beryllium windows, were ordered coupled to 1/2-in.-thick quartz light pipes and low-background multiplier phototubes. Each multiplier phototube was supplied with a potentiometer for adjusting the gain of the tube. This allowed the individual systems to be balanced prior to coupling the output signals for routing to common preamplifier-amplifier units. Taking into consideration the detection limit necessary to obtain sub-picocurie levels, a photopeak area equivalent to about 45% with a corresponding background of about 1.6 cpm was chosen for measuring separated ^{129}I concentrations of 0.4 dpm/liter with a standard deviation of better than $\pm 10\%$. By using the common 3σ method of reporting detection limits, ^{129}I could be detected at levels of 0.07 dpm/liter.

A spectra of ^{129}I from a pair of coupled NaI(Tl) crystals is shown in Figure 1.3. Interposed is a spectra from a germanium diode showing the relative efficiency of the systems for the $\text{K}\alpha$ X-rays, $\text{K}\beta$ X-rays, and gamma-ray photons. The skewness of the photopeak near the base due to the multi-energy components is quite apparent. The efficiency of the system for measuring ^{129}I is greater with the NaI(Tl) system; although the resolution is far better with the Ge(Li) detector.

FIGURE 1.3. Spectra from Coupled Signals of 2 NaI(Tl) Detectors 1 1/2 In. Diameter by 1 mm Thick with 1/2-In. Quartz Light Pipe

Radionuclide Standards

L. A. Rancitelli and N. A. Wogman

The quantitative measurement of gamma-ray-emitting radionuclides in complex mixtures requires instrumentation such as the multiparameter gamma-ray spectrometer which has been calibrated with precisely known radioactive standards. In the procedure to measure minor components of a radionuclide mixture, it is generally required that spectra reduction is performed using a standard library spectra which are assumed to be free of other radioactive contaminants. While the commercially available radioisotopes from which standards are

prepared meet the requirements necessary to achieve accuracies on the order of 5%, trace contaminants in the standards prevent further reduction of these errors. An example is an ^{26}Al solution available from International Chemical, Nuclear Chemical and Radioisotopes Division which contains trace impurities of ^{22}Na , ^{56}Co , ^{57}Co , ^{58}Co and ^{60}Co .

These impurities arise in the production of ^{26}Al from the proton bombardment of aluminum foils. The Co isotopes are produced from Ni and Fe impurities in foil while ^{22}Na is produced from a direct proton reaction on Al. The levels of the impurities present in these standards are on the order of 1000 dpm ^{22}Na and ^{60}Co per 100,000 dpm of ^{26}Al .

To achieve a pure ^{26}Al standard, further purification was required with several chemical separations before it could be used as a primary standard. Sodium-22 was removed quantitatively by acidifying the standard to 8 N HCl and passing the solution through a hydrated antimony pentoxide resin. The Co isotopes were removed following the Na separation by taking the sample to dryness, adding a few ml of 0.1 N HCl, 10 mg of Co carrier and precipitating the Co in a potassium ferrocyanide solution. The resultant ^{26}Al standard contained less than 1 dpm of ^{22}Na and Co isotopes which provided a radiochemically pure standard for future spectral stripping permitting the assay of minor constituents in ^{26}Al samples to an accuracy of better than 2%.

In Situ Analysis of the Major and
Minor Elements in Manganese Fields

N. A. Wogman, H. G. Rieck, Jr. and
H. L. Nielson

The potential utilization of our seabed resources has recently become important as a means to help fill the increasing demand for raw materials. At the present time ocean mineral utilization is limited to continental shelf areas; however, much of the mineral needs of the future may have to be met from the use of deposits on the deep ocean floor. These include manganese nodules and encrustations.

A particularly promising application of ^{252}Cf neutron activation analysis is in situ sea floor mineral analysis. During the past two years technology and prototype instrumentation have been developed for the in situ quantitative analysis of several elements at depths comparable to continental shelf areas. The prototype nuclear probe incorporating this technology was recently evaluated and also employed in shallow water continental shelf areas.

The seabed probe included a 0.2-mg ^{252}Cf neutron source, a NaI(Tl) detector or a 15% efficiency Ge(Li) diode detector, and a nuclear data 4410 analyzer which served as the central processing unit for data collection and analysis. The Ge(Li) detector unit, which is presently designed to operate at depths to 1000 feet, is cooled with a solid cryogen, a freon-propane mixture which permits up to 40 hr of continuous subsurface operation. Since one in. of steel will not significantly reduce the neutron flux

and will only reduce by a few-fold the resultant gamma-ray intensities of the neutron activation products (four-fold for 1.33 MeV line of ^{60}mCo), the system can quite easily be designed to withstand 18,000-foot ocean depths.

The nuclear probe was evaluated for analysis of simulated manganese nodule fields in the open ocean about eight kilometers off the Florida Keys. The simulated deposits were made from manganese nodules placed in water depths ranging to 20 meters. In addition to the manganese nodules, test samples of basalt, sea floor shell, dunite sand, and other seabed material were positioned on the sea floor by scuba divers. For the measurements the probe was placed over the samples which were irradiated for two min, allowed to decay for 40 sec, and then counted for two min.

Our evaluation indicated that when the instrument was used on a shelf sediment composed primarily of CaCO_3 , as off the Florida Keys, a NaI(Tl) detector and a small 200- μg ^{252}Cf source were sufficient for the analysis of the manganese nodule elements, Mn and Al, but not of V, Cu or Co. When the instrument was used to investigate the concentrations of nodules with a dunite-sand-like substrate, the elements Al, Mg, Mn and sometimes V, which are normally found in dunite, produced some interference. However, this did not seriously interfere with the analysis of the Mn in the nodules. The poor resolution of NaI(Tl) crystal would not allow the analysis of Cu and Co, the elements of major interest. The use of the nuclear

probe with its Ge(Li) diode detector readily allowed the detection of Mn, Al and V in the simulated nodule fields on both the CaCO_3 shell and on the dunite substrates. The Ge(Li) spectra of the manganese nodules obtained from this study were comparable to those obtained in laboratory investigations in which Co and Cu were detected. The similarity of the spectra indicated that with a five-fold larger ^{252}Cf source or with more sensitive detectors, it would be possible to measure Co and Cu in addition to those elements which were seen with the 200- μg source.

Review of a Workshop on X-Ray
Fluorescence Analysis of Aerosols
J. A. Cooper

The utility of X-ray fluorescence analysis will be determined by its applicability to current problems and its analytical merits relative to other possible methods of analysis. Aerosol analysis is a current problem area where X-ray fluorescence is clearly applicable. However, the capabilities and limitations of general X-ray fluorescence analysis have not been well established for this particular application and the relative merits of energy-dispersive X-ray fluorescence analysis are just beginning to be understood.

Twenty-eight participants attended and contributed to a three-day workshop on "X-Ray Fluorescence Analysis of Aerosols" at the Battelle

Seattle Research Center. Aerosol sampling techniques, methods of X-ray excitation and analysis, quantitative procedures, and other independent methods of analysis were discussed.

It was generally agreed that of the energy-dispersive X-ray fluorescence techniques, mono-energetic photon and proton excitation offered the best detectabilities. Proton excitation appeared to offer greater sensitivity for the analysis of small quantities of material on thin backings while photon excitation appeared more versatile and applicable to thicker samples. Wavelength-dispersive X-ray fluorescence seemed to have better detectability for low-atomic-number elements and was generally applicable to aerosol analysis.

It was also generally accepted that X-ray fluorescence analysis has demonstrated an ability to provide definitive quantitative analysis in urban aerosols for up to 18 elements (Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br, Sr, Zr, Mo, Cd, Sn, Ba and Pb) and semi-quantitative analysis for up to 9 low-atomic-number elements (Na, Mg, Al, Si, P, S, Cl, K and Ca). Membrane filters were generally preferred for X-ray fluorescence analysis. Standards and correction factors used were considered adequate for most elements but had to be established for elements below Ti. Current computer programs were considered adequate for the more abundant elements, but further work is required for effective analysis of low-intensity and

complex peaks. It was also concluded that the publication of additional quantitative results and quantitative intercomparison studies would greatly advance the general acceptance of X-ray fluorescence as a quantitative tool for aerosol analysis.

X-Ray Fluorescence Analysis -
Results of a First Round Inter-
comparison Study⁽²⁾

J. A. Cooper, D. C. Camp* and
J. R. Rhodes*

The analysis of aerosol particulate samples for trace elements is of growing concern to local, regional and national governments. Energy-dispersive X-ray spectrometry is a relatively new technique which is clearly applicable to this work and capable of simultaneous quantitative multielement analysis. However, its range of elemental capability is just beginning to be defined and the accuracy limitations of the method have not yet been well established. This discussion summarizes the results of the first round of an intercomparison study designed to help define the capabilities, precision, and accuracy possible with this technique. Eleven investigators using five different analytical techniques participated.

The five analytical techniques proposed for use in the intercomparison study were energy-dispersive photon-excited X-ray spectrometry, energy-dispersive particle-excited X-ray

spectrometry, wavelength-dispersive X-ray spectrometry, neutron activation analysis, and atomic absorption spectrophotometry. Results from atomic absorption were not submitted in time for inclusion in this report.

The intercomparison study demonstrated that energy-dispersive X-ray fluorescence can yield an accurate, quantitative multielement analysis of solution standards which are uniformly deposited on a light-element substrate such as filter paper. For elements K and above, accuracies within 10% and precision approaching 1% were obtained with all five energy-dispersive X-ray fluorescence techniques, some of which excited sample areas as small as 0.5 cm^2 , others at areas as large as 5 cm^2 . Some of the samples were run in vacuum, others were run in air. Some substrates were cut up and others were not.

A second objective was to test the methods used for correcting for substrate absorption. In this study, the substrate absorption of the Whatman 41 filters was about 8 mg/cm^2 and that of the Millipore filters was 5 mg/cm^2 , less than a factor of two difference. However, even this difference was sufficient to give rise to about a 20% correction between the two filters for the 3.3 keV potassium K X-rays. Those who used particle-excited X-ray fluorescence must have made reasonably accurate corrections for matrix mass absorption and partial penetration of the incident particles because their results were consistent with the photon fluorescence results.

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The third objective was to analyze particle size and absorption effects but because of variable mass loss from the particulate samples, no meaningful conclusions could be drawn. The weight percents reported were all significantly lower than the USGS values for BCR-1. However, it was interesting to note that if the weight percent results reported for the major elements and filter types for all of the investigators were averaged by element and average weight percent ratios for adjacent elements were calculated (i.e., Fe/Mn, Mn/Ti, Ca/K), then these ratios were in excellent agreement with the corresponding ratios for BCR-1.

These conclusions, gleaned from the first round of a small inter-laboratory comparison, suggested the need for a second round. In the next round, only Millipore filters will be used. Duplicate solution standards will be made; duplicate micronized particulate "standards" will be made and possibly covered with a very thin film to minimize particle loss; and finally, an effort will be made to provide one or two real aerosol samples collected in the field. This new set, then, will consist of six or seven samples. Also about a dozen more investigators will participate and a sixth analytical technique, emission spectroscopy, will be included. Hopefully, more definitive comparisons can be drawn at that time between X-ray fluorescence analysis and the more established "referee" techniques such as atomic absorption and emission spectroscopy.

A ^{252}Cf Neutron Multiplier Irradiation Facility

H. G. Rieck, Jr. and N. A. Wogman

An irradiation facility is being designed utilizing existing critical mass expertise wherein a high flux of neutrons will be produced by fission multiplication of neutrons from a ^{252}Cf source. A 100-mg ^{252}Cf source will be used in a 95% ^{235}U enriched uranium core to produce a thermal flux on the order of 10^{10} neutrons/cm²-sec. The neutron multiplier assembly (NMA) will be a subcritical system located at the bottom of a pool of water.

The total facility will have supportive equipment consisting of a water circulation pump, filter, ion exchanger, electric hoist, miscellaneous control equipment and laboratory hoods and benches. Air conditioning, heat, sanitary water, compressed air, electrical power and fire protection will also be provided.

A hatch located directly over the multiplier pool will facilitate loading the ^{252}Cf source into the pool. Ventilation will be a once-through system with exhaust through a single-stage HEPA filter.

The NMA will be located in the bottom of a 20-foot-deep by 5-foot-diameter water-tight standard density concrete pit. Design of the pit will incorporate protection from earthquake ground motions. The pit will have a 6-in. concrete curb around its perimeter and will be covered with a steel grate.

A 50-gallon-per-min (gpm) pump with intake 18 in. below the pool surface will circulate the pool water through a filter and ion-exchange resin bed for cleanup and return to the discharge near the bottom of the pool. Water flow meter, temperature, and conductivity probes will monitor system components and water condition.

The NMA will be turned "on" and "off" by moving the ^{252}Cf source into and out of the uranium core region by a source drive mechanism controlled from the top of the pool. The source will be remotely positioned into the core by an arm extending down near the bottom of the pool.

The ^{252}Cf source strength affects the shielding required; up to 300-mg ^{252}Cf sources could be used if experimental verification demonstrates that shielding is adequate. Contamination potential from the ^{252}Cf source will be reduced by its double encapsulation in stainless steel.

Uranium-aluminum alloy fuel rods will be 93.14% ^{235}U enriched. The fuel pin will be 0.3 in. in diameter by 20.6 in. in length. The fuel pin cladding will be 1100 aluminum seamless tubing with a 40-mil wall thickness; ends will be sealed by welded plugs.

The fuel region of the NMA will be an annular ring of 2-1/2 in. ID and 12 in. OD. The fuel rods will be fixed in a hexagonal lattice in four fuel elements which will comprise the fuel region. Fuel pins will be separated by approximately 200 mils of water. The four fuel elements will

be supported and aligned in the annular shape by a bottom grid plate attached to a support structure and a top grid plate fastened above the fuel elements.

A central irradiation tube will be a wet 2-in-diameter aluminum tube with a wall thickness of 35 mils. The position will provide the highest available thermal flux. Six irradiation tubes will be provided along the outer lateral surface of the NMA. Four tubes will be 2 in. in diameter and two will be 4 in. in diameter. The tubes will extend from 5 in. above the top of the steel grating to the bottom of the core and will be positioned at the bottom end. These external radiation tubes will be dry and will be bent to reduce radiation streaming.

Safety Considerations in a ^{252}Cf Neutron Multiplier Facility (NMF) H. G. Rieck, Jr.

The basic components of a neutron multiplier assembly (NMA) will be the neutron source, ^{252}Cf , and a subcritical arrangement of fissile material which surrounds the source and multiplies the source neutrons by the fission reaction. If the fissile material assembly should accidentally become critical, a severe hazard to operating personnel and the public could result. The NMA must, therefore, be designed to remain subcritical under all normal and all credibly abnormal conditions. Radiation exposure can also result from unloading irradiated samples from the NMA and handling the ^{252}Cf source.

Safety in the NMF will be established and maintained by accepted methods of good mathematical design and analysis, which have been confirmed by experimental analysis followed by firm administrative control. Design of the NMA is evolving as follows:

- A mathematical design of the NMA followed by a safety analysis is being performed.
- A detailed fueled mockup of the NMA is being constructed.
- The final operational parameters and safety aspects are being experimentally determined and confirmed at the BNW Critical Mass Laboratory.
- The NMA will be moved to the building facility and will be incrementally loaded in place to confirm the subcritical core parameters, such as total reactivity, multiplication factor and temperature effects in situ.
- A final safety analysis report based on these parameters will be prepared.

Firm administrative control will be necessary to maintain a high degree of safety in the operation of the NMA. Safety of personnel, equipment and the public will be a dominant factor in developing and implementing these controls.

To minimize the problem of accidental or intentional damage, the NMF will be maintained as a restricted area. Routine entry will be through locked doors and access will be limited to qualified operators and others who have work responsibility in the NMF.

Irradiation procedures will be developed and the impact of the samples on the operation of the NMA will be evaluated. Sample flow into the NMF will be screened to prevent introduction of additional fissile material which would decrease the safety margin of operation. The sample history will be important to establish absence of fissile materials. A sample log will be maintained to control the sample traffic through the NMF.

Standard radiation work procedures will be developed for safe handling of exposed samples. Adequate monitoring procedures will be used to prevent accidental personnel exposure to radiation from irradiated samples.

Routine instruction programs will be implemented to insure that all supporting equipment is operating properly. Periodic radiation surveys will be made to insure proper contamination control.

A training program will be organized and maintained to insure qualification of operators. Each person will be made aware of the possible consequences which may result from failure to observe all established operational procedures.

Activation Analysis with a ^{252}Cf Neutron Multiplier Facility

N. A. Wogman and L. A. Rancitelli

Preliminary experiments have shown the practicality of utilizing the subcritical multiplier of ^{252}Cf neutrons

for activation analysis of the elements contained in a variety of environmental materials. Since the subcritical assembly uses ^{252}Cf (a neutron-emitting radioisotope) with a half-life of 2.63 yr (2.34×10^9 n/sec/mg), its flux is extremely constant--varying only as the half-life. This uniform flux allows samples and standards to be irradiated at different times in exactly the same geometrical position with respect to the energies of the neutrons. This latter condition creates a distinct advantage for neutron activation in the multiplier rather than in a typical reactor (TRIGA).

The sensitivities for detection of seventeen elements are considered in Table 1.3 using several typical irradiation, decay, and counting periods. The elements F, Al, Sc, Ge, Dy, and Er have about equal detection sensitivities (listed as $\gamma\text{dpm/g}$) in a reactor with a thermal flux of 10^{12} n/sec/cm² and a subcritical assembly with a thermal flux of 5×10^{10} n/sec/cm². This is due either to the production of a shorter half-life radioisotope used for detection of the elements or to the shorter decay of the sample from the subcritical assembly. This short delay between irradiation

TABLE 1.3. Activation Analysis Sensitivities
Utilizing a Subcritical Neutron Multiplier (SNM)

SNM ^(a) $\gamma\text{-dpm/g}$	$T_{(1/2)}^{\text{Sec}}$	Nuclide	Element	Nuclide	$T_{(1/2)}^{\text{Min}}$	TRIGA Reactor ^(b) $\gamma\text{-dpm/g}$
7.4×10^8	11.6	^{20}F	F	^{20}F	0.2	3.7×10^8 (1)
1.4×10^{10}	138	^{28}Al	Al	^{28}Al	2.3	1.3×10^{10} (2)
5.0×10^6	0.74	^{38}Cl	Cl	^{38}Cl	33.7	2.8×10^9 (3)
1.5×10^8	20	^{46}Sc	Sc	^{46}Sc	120960	8.5×10^{10} (5)
5.3×10^8	48	^{75}Ge	Ge	^{75}Ge	82	5.6×10^8 (4)
1.8×10^{10}	18	^{75}Se	Se	^{75}Se	172800	3.6×10^8 (5)
3.8×10^9	13	^{81}Kr	Kr	^{81}Kr	76	3.3×10^8 (4)
1.2×10^{10}	42	^{104}Rh	Rh	^{104}Rh	4.4	3.0×10^{11} (1)
1.1×10^9	22	^{109}Pd	Pd	^{109}Pd	810	6.1×10^7 (5)
2.7×10^{10}	24	^{110}Ag	Ag	^{110}Ag	2.4	4.8×10^9 (2)
6.4×10^9	14	^{116}In	In	^{116}In	54	1.6×10^{12} (4)
1.6×10^{11}	75	^{165}Dy	Dy	^{165}Dy	139	1.2×10^{11} (4)
8.9×10^9	2.3	^{167}Er	Er	^{171}Er	451	1.6×10^{10} (5)
1.6×10^9	4.3	^{178}Hf	Hf	^{175}Hf	100800	1.1×10^9 (5)
1.0×10^{11}	18	^{179}Hf	Hf	^{181}Hf	61920	8.0×10^9 (5)
1.6×10^8	5.1	^{183}W	W	^{187}W	1440	1.3×10^{11} (5)
1.4×10^7	14	^{199}Pt	Pt	^{197}Pt	1200	1.8×10^9 (5)
6.9×10^6	0.74	^{207}Pb	Pb	--	--	--

(a) Irradiation time = 3 half-lives of radionuclide
Decay time - 2 seconds - transit time from SNM to counter
Neutron flux = 5×10^{10}

(b) Irradiation time = decay time
(1) 1 min (3) 120 min (5) 1440 min
(2) 10 min (4) 240 min Neutron flux = 1×10^{12}

and counting is most easily accomplished with the subcritical assembly since it is constructed close to the counting systems used. The elements Cl, Rh, In, W, and Pt are more sensitively analyzed with the greater neutron flux of a reactor. The elements Se, Kr, Pd, Ag and Hf are more sensitively analyzed in the subcritical neutron system in close proximity to the detectors. Lead is one element which can be detected only with a short half-life specie, ^{207m}Pb , and thus can be analyzed with the subcritical assembly. One additional advantage with the elemental analysis technique using short half-life radioactive species is that the same material can be cycled many times to enhance the sensitivity; thus, the irradiation-decay-counting cycle can be repeated until the long half-life components of the matrix start to interfere with the analysis.

For the analysis of many environmental samples, the multiplier assembly allows, in general, a more reliable and sensitive technique than a reactor since a kilogram sample can be irradiated. This procedure eliminates the inherent error in subsampling which is typical of reactor analyses and compensates for the 10^2 to 10^4 difference in neutron flux with the 10^3 to 10^6 greater sample size.

Inert Tracers for Sand Movement

N. A. Wogman and L. A. Rancitelli

Harbors and estuaries as well as the beaches of our world's waterways

are continuously eroded or modified through wave and tidal action. This often necessitates the elimination of a beach for pleasure or the removal of vast quantities of harbor sands and muds to keep the necessary deep water channels open for shipping. Techniques which accurately measure the drift of these materials and thus minimize their cost of removal are of great interest. One of these methods is in situ neutron activation analysis.

In recent studies of neutron activation methods for identification of pollutants or economically important metals on our continental shelf, some of the elements which could be detected with high sensitivity were not found in the normal sands and muds of beaches and harbors. Ocean muds and sea sand samples were neutron irradiated in situ and analyzed (using a seabed probe designed at BNW) for S, Ca, Cl, Mo, Mg, Al, F, K, Na, In, Ga, Nd, Ni, Co, Cu, Ba, Sn, V, I, Eu, Ti, Au, Se, Nb, W, Sb, Dy, Ag, Os, Rh, Pt, La, Ir and As. The analysis scheme utilized a 2-min irradiation with a 1-mg ^{252}Cf source, a 1-min decay and successive 2-min and 3-min counts with a 70-cc Ge(Li) gamma-ray analysis unit.

The elements Dy, Ag, Au, Se and In were of low concentrations in these samples. For example, the concentration of In was equivalent to only 2 kg spread over one square kilometer within the top 1 cm of sediment, 4 kg of In would thus triple the amount of In present in these sands, as shown in Table 1.4. Not all these elements have the same

TABLE 1.4. Inert Tracers for Sand Movement Studies
Utilizing the Battelle In Situ Seabed Mineral Probe
with a 1-mg ^{252}Cf Source and a 15% Ge(Li) Detector

Element	No. of kg Required to Triple Background in 1 km ² to a Depth of 1 cm	Minimum Detectable Amount kg/km ²
Dy	180	2.0
Ag	4.0	120
Au	0.1	80
Se	1.8	46
In	4.1	770

detectabilities with the in situ neutron activation technique because they possess different nuclear properties such as neutron capture cross sections and decay schemes. As shown in Table 1.4, the element Dy has the lowest minimum detectability. For example, Dy can be detected at a concentration of 2.0 kg/km²; the other low-concentration elements must be at least 20-fold higher in amount to be detected. Since the short-lived $^{165\text{m}}\text{Dy}$ (74 sec) could not be detected in the sand background and yet has a good detectability limit for the required short irradiation and counting procedures, it is of practical value in tracer studies of sand movement. Dysprosium can be added to sand as a rare-earth ore such as gadolinite and the seabed probe used to activate and follow its movement. In view of its low-background concentrations in ocean sediment, its excellent detectability, and relatively low cost, Dy

appears to be an ideal tracer of sand movement, an important but little understood geologic process, in littoral zones and on the continental shelf.

Feasibility Study of In Situ Sediment Analysis by X-Ray Fluorescence

J. A. Cooper, H. L. Nielson,
N. A. Wogman and R. W. Perkins

Man is releasing many hazardous and potentially toxic elements to the aquatic environment at an increasing rate. Spatial and temporal distribution measurements of potential pollutant elements in sediments of coastal marine, estuarine, river and lake systems are required to evaluate the relative hazards of this action and the potential hazards associated with sediment drift and dredging operations. However, mapping the surface

composition of sediments based on grab samples may be misleading due to unknown sampling depth. Also this method does not provide the information required to make on-the-spot decisions to study a particular region in greater detail or to change the course of an investigation. Mapping of the elemental composition of a water basin floor and studies of the movement of sediments could be greatly simplified by a rapid, reliable in situ multi-element analysis technique.

An investigation of the feasibility of in situ analysis by energy-dispersive X-ray fluorescence has established that energy-dispersive X-ray fluorescence analysis can provide in situ sediment detectabilities which approach those attainable in the laboratory and can be used to map the concentration of many elemental pollutants in lakes, rivers, and estuaries. The method involves excitation with a ^{109}Cd radioisotopic source and analysis of the X-rays with a Si(Li) detector housed in a solid-cryogen cryostat with a 40-mil-thick Be window. The X-ray probe can be operated from either a submersible vehicle as shown in Figure 1.4 or from a surface vessel.

FIGURE 1.4. Schematic Representation of a Possible In Situ X-Ray Fluorescence Survey System--a Probe Controlled by a Submersible

Energy-dispersive X-ray fluorescence analysis, with available technology, is capable of providing definitive analyses for about 13 elements (Cr, Mn, Fe, Ni, Cu, Zn, As, Br, Rb, Sr, Y, Zr and Pb)(see Figure 1.5) at their typical concentration levels in some polluted areas and can provide less than values in the 5 to 20 ppm range for Au, Hg, Se, Ge, and Ga in analysis times of about 4 min. A system using advanced excitation techniques should be capable of providing low ppm detectabilities in analysis times of 2 to 3 min. The concentration of Cd can be determined at levels of about 20 ppm, but it would require a special excitation source.

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FIGURE 1.5. X-Ray Spectra of the Elements Contained in Dry Hanford Sand Spiked up to 100 ppm Pb and Au as Analyzed With a Normal Laboratory and a Simulated In Situ Seafloor X-Ray Fluorescence Probe: (A) Normal detector-source window arrangement, (B) Source inside 40-mil-thick Be window, (C) Source outside 40-mil-thick Be window which illustrates that the use of a thick, high-pressure Be window will decrease the sensitivity for Pb and ANL lines minimally.

Application of the ^{85}Kr Monitoring
System to Meteorological Diffusion
Experiments

J. D. Ludwick and P. W. Nickola

Continuing applications of the krypton-85 field grid to atmospheric diffusion studies have been made during the year. Improvements, expansions and completion of work reported in the Annual Report for 1972 have been made. A new data processing procedure is now complete for transferring the experimental information to either of two readout methods. Data collected by the two 4096 Packard memories in sequence are transferred to the DEC-type tape transports. These tapes are then reformulated by a PDP-15 computer for delivery to a larger, more versatile computer. Programs for defining certain meteorological parameters can then be written directly into this central computer. Up to this time the PDP-15 has been used to give direct digital readouts of the data. Several field tests during this year have been read out in this manner, and subsequent aerosol concentration plots have been made from these readouts.

Field tests were conducted during day- and night-time conditions under moderate wind conditions and fairly stable atmospheric temperature gradients. The ^{85}Kr tracer was released

from an aloft position 85 ft above ground level in coordination with other particulate tracer materials.

To assure the precise dispensing of radiokrypton gas, a special mass flow controller was incorporated into the emission system. The controller which responds to the transfer of thermal energy along the gas path is calibrated for flow with Argon carrier gas. Typically, a 30-min continuous source was desired. The generated thermal conductivity current controls the opening of an electrical solenoid valve. Hence, flow can be preset by selecting the current values desired and trace source cylinders can be emptied in any predetermined time interval. This dispersal method was found to be insensitive to changes in gas cylinder pressure due to outside temperature fluctuations during a 30-min experiment with a continuous source. A second on-off solenoid valve was inserted between the cell and the source so the desired flow rate can be preset and the source can be turned on or off from a remote location such as the data collection center. Thus, one person can operate the krypton system when the meteorological conditions are appropriate for dispersal.

The present series of aloft releases have been completed and the

krypton grid system is being modified for stagnation-type conditions. An additional 20 sensors are being added to the network to monitor approximately 150 field locations. This test series will utilize the continuous releases (rather than puff or instantaneous) of ^{85}Kr gas over 15 to 30

min intervals. Diffusion data derived from measurement of the air concentration of the krypton will be useful in predicting the personnel exposure problems associated with continuous releases of hazardous gases into rather stable atmospheres.

• NUCLEAR TECHNIQUES IN MEDICAL SCIENCE

The purpose of this study is to perform research in the application of nuclear technology and radioisotopes to medicine and biology by combining the advanced knowledge, techniques, and instrumentation of the nuclear field with the talent and knowledge of the medical profession to improve and to increase the direct medical benefits to mankind. Advanced instrumentation concepts, detectors, and circuitry are also being developed for use in health physics environmental studies, particularly in measuring in vivo plutonium and other alpha emitters. These studies usually culminate in a cooperative effort with a medical institution for their practical application.

A Parallel-Plate Gas Scintillation Proportional Counter for Improved Resolution of Low-Energy Photons

H. E. Palmer and L. A. Braby

A very significant improvement in resolution of low-energy photons was recently obtained in gas scintillation proportional counters by replacing the center wire anode with a spherical anode.⁽³⁾ When this type of counter is used with pure xenon gas and an electric field, which is slightly lower than that required for secondary ionization, the energy resolution of the light pulses is much better than the resolution obtained from an ordinary center wire proportional counter. With good light collection efficiency, the light obtained per unit of absorbed x-ray energy is about 100 times that from NaI(Tl) detectors; a full width at half maximum of 500 eV (8.5%) for the 5.9-keV photopeak has been obtained.⁽³⁾

From the experiments with larger ball anodes it became obvious that a 2.5-cm-diameter ball anode operating inside a 11-cm-diameter cathode begins to approach the conditions of a parallel-plate chamber. This suggested that a parallel-plate counter with transparent grid anode and cathode might provide an improved counter due to the uniformity of the voltage between the parallel plates, the unlimited size of this type of counter, and the more favorable counting geometry which could be obtained.

Figure 1.6 shows a sketch of a 5-cm-diameter counter which was constructed to test the use of parallel plates for gas scintillation proportional counting. The walls of the counter were constructed from ordinary laboratory glass tubing which was coated on the inside with magnesium oxide. A $\leq 100\text{-}\mu\text{g}/\text{cm}^2$ layer of p-quarterphenyl wave length shifter was vacuum vaporized onto the magnesium oxide surface. Nickel mesh was

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FIGURE 1.6. Sketch of 5-cm Diameter Parallel-Plate Gas Scintillation Proportional Counter

stretched onto both ends of section B of the counter wall. This mesh had a wire thickness of 0.0137 mm, 7.87 wires per cm, and 97% transparency. The three sections of the counter wall were then cemented together with a low vapor pressure epoxy. A quartz window coated with $\leq 100 \mu\text{g}/\text{cm}^2$ p-quarterphenyl was cemented to one end of the tube and a brass plate containing a small beryllium window and tubes leading to a hot calcium purifier was cemented to the opposite end. The inside of the brass plate was also coated with magnesium oxide and p-quarterphenyl. The counter is filled to a pressure of 1 atmosphere with 99.995% pure xenon. The convection-flow calcium purifier is operated at 400°C.

The thickness of gas in section C is more than enough to absorb essentially all 5.9 keV x-rays which enter the small beryllium window. For the best results, the potential of the grid between sections B and C should be such that all primary electrons will drift to the grid without gaining enough energy to cause excitation in section C. The grid between sections A and B is operated at a significantly higher potential but just below the point where secondary ionization occurs. If secondary ionization occurs the resolution of the counter reverts to that of an ordinary proportional counter. The optimum grid voltages for the best resolution for this counter were found to be at 1900 and 8500 volts

which produced an electrical field of 600 V/cm in the photon absorption and electron drift region of section C and 3473 V/cm in the excitation and light producing region of section B.

Figure 1.7 shows the pulse height spectra of an ^{55}Fe source measured with this counter. The resolution of 8.4% is slightly better than that obtained with the spherical anode counters and more than a factor of two better than the resolution of 18% obtained from an ordinary center wire proportional counter.

Even better resolution is expected from parallel-plate gas scintillation proportional counters as they are further developed. They probably can be constructed with rather large detection areas and will be useful in those studies where better resolution is required than that obtained from a center wire proportional counter and where a larger area detector is needed than is available from Si(Li) and Ge(Li) detectors. Applications for this type of detection will be in medicine, in vivo plutonium counting, and x-ray fluorescence studies.

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FIGURE 1.7. Pulse Height Spectra of a ^{55}Fe Source from Parallel-Plate Gas Scintillation Proportional Counter

A Study of the Use of ^{37}Ar in
Expired Air After Neutron Irradia-
tion as a Measure of Total Body
Calcium in Humans

H. E. Palmer, T. K. Lewellan,*
W. P. Nelp* and G. M. Hinn*

The use of ^{37}Ar in the expired air of rats and dogs after neutron irradiation as a measure of total body calcium has been described previously.⁽⁴⁾

The amount of ^{37}Ar in expired air during the first 1 1/2 hr after neutron irradiation was found to be reproducible in repeated measurements and proportional to the total body calcium content in rats.⁽⁵⁾ Argon-37 studies also were performed at the University of Washington School of Medicine on several patients who were irradiated with neutrons to determine total body calcium by the whole body counting of the induced ^{49}Ca . The ^{37}Ar content of expired air fractions up to 16 hr after irradiation indicates that the excretion rate of ^{37}Ar from bone is much slower in humans than in rats and dogs. Even after 16 hr a significant fraction of the ^{37}Ar appears to remain in the body. It also appears that the excretion rate becomes slower with the age of the patient; this agrees with the results of radon excretion from animal bone found by Mays, et al.,⁽⁶⁾ where the excretion rate became slower as the animal became older. This effect was attributed to the formation of larger bone mineral crystals with increasing age.

* University of Washington School of Medicine

These preliminary results indicate that a quantitative measure of total body calcium in humans is not feasible through the determination of expired ^{37}Ar ; however, excretion rate might be used as a measure of bone quality.

Radiation Instrumentation

K. L. Swinth

Efforts with avalanche diode probes for detection of plutonium in the tracheobronchial lymph nodes revealed the lack of sensitivity and reliability needed for a useful probe. Since previous efforts indicated that this approach had potential for producing a useful probe if photomultiplier noise problems could be reduced, attention was directed toward development of a fiber optic coupled scintillator.

The use of a flexible fiber optic coupling to transmit light from a scintillation crystal to a photomultiplier causes serious light losses. Most of this loss is due to the limited acceptance angle of the fiber optics with transmission losses playing a secondary role. Total losses for NaI(Tl) scintillations through our fiber optics (60 cm) amounted to about 85%. With such large light losses, variations in the light collection efficiency of crystals ($\pm 18\%$ standard deviation for seven crystals) and the transmission of fiber optics ($\sim 10\%$ variation) become important. Successful detectors were constructed by selection of crystals and fiber optics to optimize the light collection efficiency; see Figure 1.8. For the

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FIGURE 1.8. Photograph Showing Esophageal Probe Connected to Photomultiplier Housing. Two wound counting detectors are shown alongside the esophageal probe.

17-keV average energy photons from ^{239}Pu decay, a mean of 8 photoelectrons from the photocathode of a high sensitivity photomultiplier is obtained.

In the region, 10 to 40 keV (3.5 to 18 photoelectron equivalents), used to count the L x-rays of ^{239}Pu , noise due to Cerenkov radiation, faceplate scintillations and recoil electrons is important. The

background in this energy range was 513 ± 3 counts/min for our best photomultiplier, an RCA 8850.

This is an unacceptably high background. An evaluation was made of magnetically limiting the photocathode, pulse shape discrimination, photomultiplier cooling and lead shielding individually and then in combination for reducing the photomultiplier background. The lowest

background, 10.0 ± 0.5 counts/min, was obtained using magnetic limitation, pulse shape discrimination, and shielding; see Figure 1.9. It was found that photomultiplier cooling had no noticeable affect in our energy region.

In another technique, a bifurcated light pipe and two photomulti-

pliers viewing the same scintillation crystal were used. This produced the lowest backgrounds with about 1.6 ± 0.28 counts/min from 6.5 to 40 keV. Based on these experiments a probe was built using the pulse-shape discrimination technique since it was easier to implement at the early stages.

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FIGURE 1.9. Background Spectra of Phototube With and Without Reduction of Background Noise. Magnetic limitation of the photocathode area and pulse-shape discrimination were used to lower the background count rate.

Tests with the human thorax phantom indicated a minimum detectable amount of 3.0 nCi for a 20-min count when only the L x-ray counts were measured for material located in the tracheobronchial lymph nodes. Table 1.5 shows the results for the fiber optics probe and compares these results with previous avalanche diode probes. Plutonium activity located in the lungs does contribute to the observed count rate and must be corrected for by counts taken above and below the lymph node region. The esophageal probe, when centrally located in the lungs to measure ^{241}Am gamma-rays associated with plutonium, can be used to estimate lung deposition. Using the gamma-ray counts, a minimum detectable amount of 16 to

60 nCi based on calculated sensitivities for lung deposited material can be estimated.

The probe was tested in dogs which had alveolar depositions of plutonium and were about two years past exposure. The estimated sensitivity for lymph node burdens was 1 cpm/nCi based on extrapolations from initial lung burdens and extrapolation of mean translocation rates observed on sacrificed animals. The sensitivity of the probe to plutonium in the lymph nodes of a dog depends strongly on the position of the detector with respect to that of the probe. Figure 1.10 shows the relative count rate of the probe as it approaches and then goes past the radioactive lymph node area.

TABLE 1.5. Comparison of Detectable Amounts for Plutonium in Human Tracheobronchial Lymph Nodes. Measurements were made with a phantom representing the human thorax.

PROBE	BACKGROUND, $B^{(a)}$ (CPM)	SENSITIVITY, S (CPM/nCi)	MINIMUM DETECTABLE AMOUNT (nCi) $^{(b)}$
AVALANCHE 1	5.05	0.025	52
AVALANCHE 2	5.56	0.039	34
FIBER OPTICS:			
X-RAY ENERGIES (17 keV)	18.4 (9.20) $^{(a)}$	0.807	3.0
γ -RAY ENERGIES (60 keV)	38.7 (3.06) $^{(a)}$	1.22	2.9

(a) - Values in parenthesis are for shielded detector

(b) - Calculated at 90% confidence level with a 20-min counting period

$$\left(\text{i.e. } \text{MDA} = \frac{2.56}{S} \sqrt{\frac{B}{20}} \right)$$

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FIGURE 1.10. Count Rate Versus Insertion Distance For a Dog With Inhaled Plutonium which has Translocated To the Tracheobronchial Lymph Nodes.

The benefit-risk aspects of using the esophageal probe were reviewed in detail. The risk was estimated assuming a similarity between a gastroscop and the probe. Based on the literature, the rate of pharynx or esophageal perforation is estimated to be less than 1 in 2000 examinations. Accident rate varies with the clinician; with experience and care the potential accident rate should be much lower.

Based on work with fiber optics, prototype wound probes were fabricated and evaluated. The minimum

detectable amount based on a one-min count was 1.9 nCi which compares with 67 nCi for a similar sized avalanche diode.

The effort on intercalibration for low-energy photon measurements progressed in two directions. Additional data from the source intercalibration effort were collected and analyzed. Following this, a manuscript covering the intercalibration effort was prepared and published. With the exception of one laboratory, the intercalibration indicated good instrumental control.

Normalized sensitivities were in good agreement with major differences probably due to calibration standards. The largest discrepancy was in background count rate from 12 to 24 keV; it varied from 0.0157 to 1.12 cpm/cm². This is a factor of 8 difference in minimum detectable amount.

Data from a dog phantom were studied to indicate the possible usefulness of a properly constructed human phantom for plutonium counting. In one experiment with 12 dogs, the average body burdens determined by phantom calibration were +17% and +11% higher for the 17 keV x-ray and 59.6 keV gamma energies, respectively, than the measured burdens determined by alpha counting of tissues. This included two isotopic compositions; corrections for tissue distribution and body differences were not performed. In another experiment involving a more detailed calibration and correction for tissue distribution, the average difference was +3.88% for the x-ray energies compared to alpha counting of tissues. This suggests a suitable phantom can be fabricated.

The general implications of radiation deposition in the tracheobronchial lymph nodes were studied in relation to environmental nuclides. Autopsy samples of lymph nodes were gathered and counted as pooled samples in low-level counters at the Laboratory for Nuclear Medicine and Radiation Biology at UCLA, Battelle-Northwest, and Lawrence Livermore Laboratory (LLL). No environmental gamma-emitting nuclides were detected. This is not surprising, since calculations indicated that at present environmental levels it would take on the order of 50 people inhaling the material for a number of years before measurable levels would be obtained in the lymph nodes. X-ray fluorescence measurements made at BNW and LLL indicated a large number of stable elements; however, the absolute levels and concentration differences between lungs and lymph nodes have not been determined. It is expected that many of these materials will correlate with known environmental pollutants. If the lymph nodes play an important mitigating role in pulmonary cancer, the implications of material (radioactive and stable) could be very important.

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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 atoms and molecules bombarded by protons is studied to provide information on the 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, the mechanism of energy transport and transformation from the energetic initial processes into chemical species is studied using time resolved luminescence. Electron emission cross sections have been improved for the very important low-energy electrons ejected from several gas targets. Theoretical studies of the angular distribution of ejected electrons have continued with a marked improvement for the lower energy electrons. Monte Carlo calculations of track structure and microdosimetric parameters have been started and the results are proving useful in formulating better initial conditions for the distribution of active species. This will permit development of more realistic models for radioluminescence.

Low-Energy Electron Emission

L. H. Toburen and W. E. Wilson

A time-of-flight electron energy analyzer has been developed for the study of low-energy electrons ejected in fast proton-gas target collisions. This system uses our pulsed proton beam and delayed coincidence electronics to determine electron energies by measurement of the time of arrival of electrons at the detector following the beam pulse. A pulse width of approximately 0.7 nsec, with repetition rate of 3 MHz, is obtained by r-f chopping of the proton beam from our 2-MV Van de Graaff accelerator. Magnetic fields are reduced to less than a few milligauss in the collision region by magnetic shielding supplemented with field

nullification coils wrapped around the shielding. By comparing spectra obtained for electron drift lengths varying from 3.8 to 15.2 cm, reliable spectral shapes could be obtained for electron energies as low as 0.8 eV. For electron energies lower than 0.8 eV, the spectra changes considerably as a function of various experimental parameters; more refinement of the technique will be necessary to investigate electron spectra in that energy range. There is, however, evidence of structure in spectra of N_2 at electron energies from 0.2 to 0.6 eV and in xenon spectra for electron energies from 0.4 to 1.2 eV. Pressing the technique to these low energies may provide new information related to the atomic and molecular binding effects which occur

during ionizing collisions.

The primary incentive for the development of the time-of-flight system of electron energy analysis arose from the discrepancies between spectra measured at three laboratories for 0.3-MeV protons on nitrogen gas. Figure 2.1 illustrates the results reported by Stolterfoht,⁽¹⁾ Crooks and Rudd,⁽²⁾ and BNW from electrostatic and time-of-flight analysis. Our time-of-flight analysis is in excellent agreement with Crooks and Rudd. When the integral over electron energy and emission angle is performed, it provides excellent agreement with total ionization cross

sections measured by Hooper et al.⁽³⁾ The comparison with total ionization measurements is a sensitive test of the accuracy of low-energy electron measurements since it is the low-energy portion of the spectrum which contributes the major part to the total integral which is performed to obtain the total ionization cross sections.

Measurements of low-energy electron spectra for several gas targets are shown in Figure 2.2. These spectra were all normalized to the same value at 100 eV to observe the relative variations in spectral shape as a function of the type of target atom

FIGURE 2.1. Double Differential Electron Emission Cross Sections for 0.3-MeV Protons on Nitrogen. Previous electrostatic measurements are from Stolterfoht⁽¹⁾ and Crooks and Rudd.⁽²⁾

FIGURE 2.2. Low-energy Electron Spectra are Shown for Several Target Gases and 1-MeV Proton Impact. The spectra were normalized at 100 eV. The photoionization cross sections are from calculations of Cooper and Manson.⁽⁴⁾

or molecule. Large differences are observed in the low-energy electron spectra; this is most evident in the comparison of neon and hydrogen data, where the cross sections at 1 eV differ by nearly two orders of magnitude. Other features which can be seen in Figure 2.2 include evidence of the Cooper minimum associated with the 3p- ϵ d ionization channel in the ionization of argon.⁽⁴⁾ This effect was first observed in photoionization spectra and appears here as a broad dip in the region of 50-60 eV. The results presented for SF₆ do not show any large variations from simpler targets which might be expected as a consequence of the potential barrier associated with the electronegative fluorine atoms. The existence of "inner-well" states⁽⁵⁾

appears to be masked by the large number of electrons ejected from the "outer-well" states; the shape of the electron emission spectra is very similar to that of neon. There is some low-energy structure in the SF₆ spectra but it is not clear whether this is due to autoionization spectra superimposed on the continuum or whether this may be due to the "inner-well" and "outer-well" states discussed by Dehmer.⁽⁵⁾

Electron Emission from Molecular Targets

L. H. Toburen and L. Porter

A comprehensive study is in progress to study the electron emission cross sections using both electrostatic and time-of-flight energy

analysis for protons on SF_6 and TeF_6 . The combination of these two analyzers provides reliable electron emission cross sections for electron energies from 0.8 to 5000 eV and electron emission angles from 20 to 125 degrees. A comparison of cross sections for these two molecules with our earlier data on simpler molecules is expected to provide more definitive information on the effect of the electronegative potential barrier in symmetric molecules such as SF_6 . The analysis of these data for proton energies from 0.3 to 2.0 MeV is in progress.

Theoretical Electron Emission Cross Sections

L. H. Toburen and S. T. Manson

Electron emission cross sections, differential in electron energy and emission angles have been calculated for incident protons on neon and helium. These calculations are based on the Born approximation with both bound and continuum wave functions derived from Hartree-Slater atomic potentials. These results are a significant improvement over hydrogenic Born calculations previously performed. Preliminary results of this work are shown in Figure 2.3 and illustrate the unreliability of the hydrogenic Born approximation which is commonly used for atomic collision calculations. By using better wave functions, such as those derived from the Hartree-Slater (H-S) potential, it appears that reliable results may be obtained within the framework of

the Born approximation. The calculations for helium have been extended to proton energies investigated at other laboratories and to electron impact to more fully investigate the region of applicability of the H-S Born calculations. The results have been compared to the electron impact measurements of Crooks⁽⁶⁾ and Opal et al.⁽⁷⁾ as well as to available proton impact data from BNW, Stolterfoht⁽¹⁾ and Rudd, et al.⁽²⁾ The calculated values are in excellent agreement with measured cross sections with the exception of the enhancement of the cross sections in the forward direction by long-range proton-electron interactions. The effect of this long-range interaction appears to be negligible in the high-energy (5.0 MeV) data of Stolterfoht⁽¹⁾ with increasing importance as the proton velocity is reduced. The details of this calculation and comparison with available helium data are presently being compiled and will be published soon.

Electron Ejection From Carbon Foils

L. H. Toburen and H. G. Paretzke

Preliminary data have been obtained for the energy and angular distributions of electrons ejected from carbon foils. Foil thicknesses of 3.0 and 20.0 $\mu\text{g}/\text{cm}^2$ were investigated. It is expected that these data will provide a sensitive test of the Monte-Carlo codes developed by Dr. H. Paretzke of the Institut für Strahlenschutz in Munich. By using

FIGURE 2.3. Electron Emission Cross Sections for 2.0-MeV Protons on Neon. The experimental results are normalized to theory at 30 eV.

gas phase source terms in the calculations and comparing them to foil measurements, we hope to gain some insight into the effects of the physical state of the target in which the energy is absorbed.

Computer Simulation Methods for Track Structure

W. E. Wilson

A comprehensive picture of the processes occurring in a charged particle track during the physical stage of energy deposition is of crucial importance to understanding radiation effects of fast ions and neutrons. Direct observations of primary track structure in condensed media is, however, extremely difficult and the experimental information presently

available is limited in quantity and is generally for gaseous phase. Therefore, calculation of features of the primary track structure, which might be correlated with the radiation effect under consideration, takes on a new importance. Specific track structure properties of interest include radial distribution of energy deposition, spatial distribution of ionization, and event size distributions in small volumes. We are collaborating with H. G. Paretzke* and M. J. Berger+ in developing the necessary capabilities. Computer

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codes, which model the primary interactions of charged particles with matter in various approximations, have been written by these researchers. Some codes use a continuous slowing-down-approximation (CSDA), some use only Monte-Carlo (MC) techniques and some are hybrid, using both CSDA and MC. CSDA codes have the advantage of speed, and hence, economy, but the disadvantage that they calculate only mean values. The MC codes are required to obtain distributions; however, MC techniques are inherently inefficient and, hence, expensive. The hybrid codes are attempts to use the advantages of both for certain types of calculations.

We have borrowed the codes developed by Paretzke and are adapting them to our needs. Our first effort has been to use a full MC code to calculate energy deposition distributions for comparison with the wall-less proportional counter measurements of Glass and Roesch.⁽⁸⁾ In that experiment, they measured pulse height distributions produced in a cylinder of 125-nm effective radius in unit density material for protons passing through or by the volume at various distances. The calculated mean energies of the deposition events are compared in Figure 2.4 with the observed mean energies. The calculated and experimental values

agree quite well for proton paths within the proportional counter. Agreement is not as good for paths near the edge and outside the

FIGURE 2.4. Comparison of Calculated and Experimental Results for the Mean Energy Deposited by Protons in a Cylinder of 125-nm Radius as a Function of the Beam Position from the Center. Open symbols are Monte-Carlo calculations, closed symbols are experimental data.⁽⁸⁾ Solid lines are calculated from proton stopping power and beam cord length.

counter. This disagreement may arise from an imprecisely defined counter boundary due to the inevitable fringing of electric field lines. For comparison, the mean energies given by the product of proton stopping power with cord length are also shown in Figure 2.4.

Direct Calculation of Energy Straggling Parameters

W. E. Wilson, L. H. Toburen
and W. A. Glass

The variance of an energy straggling distribution for thin absorbers is characterized by the cross section for energy loss in single collisions. This variance divided by the mean energy loss, δ_2 , is independent of the thickness of the absorber; it is also numerically equal to the quotient of the second by the first moment of the single collision spectra,

$$\delta_2 = \frac{\text{var.}(E)}{\bar{E}} = \frac{\int_0^\infty E^2 \sigma(E) dE}{\int_0^\infty E \sigma(E) dE}, \quad (1)$$

where $\sigma(E)$ is the cross section for a total energy loss E in a single collision.

It has been shown^(9,10) that Eq. 1 can be expressed $\delta_2 = m_2/m_1$ where

$$m_2 = \int d\epsilon \epsilon^2 \sigma(\epsilon) + 2[\alpha_K I_K + \alpha_L I_L] \int d\epsilon \epsilon \sigma(\epsilon) + [\alpha_K I_K^2 + \alpha_L I_L^2] \int d\epsilon \sigma(\epsilon) + \sum_n \sigma_n \xi_n^2 \quad (2)$$

and

$$m_1 = \int d\epsilon \epsilon \sigma(\epsilon) + [\alpha_K I_K + \alpha_L I_L] \int d\epsilon \sigma(\epsilon) + \sum_n \sigma_n \xi_n. \quad (3)$$

In Eqs. 2 and 3, $\sigma(\epsilon)$ is the cross section for ejection of secondary electrons of energy ϵ ; α_K and α_L are the fractions of ions created by respective K-shell or L-shell ionization, for which the binding energies are I_K and I_L . The terms containing $\sigma_n \xi_n$ and $\sigma_n \xi_n^2$ account for the inelastic collisions which produce no ionization.

We have calculated δ_2 using Eq. 2 and 3 and experimental cross sections for $\sigma(\epsilon)$ and $\alpha_K = \alpha_K/\alpha_{\text{ion}}$ and in Figure 2.5 have compared the results with the simple free electron model for which

$$\delta_2 = E_{\text{max}}/2 \ln(E_{\text{max}}/I). \quad (4)$$

In Eq. 4, $E_{\text{max}} = (4/1836)E_{\text{ion}}$ and I is the mean excitation potential of the target.

Generally, values for δ_2 , calculated from Eqs. 2 and 3 are 10 to 20% larger than predicted by the free electron model. If Eqs. 1, 2 and 3 are used to evaluate the energy deposited in a small absorbing site, it is implicitly assumed that all energy transferred from the proton to the matter within the site remains there and, in particular, that the site is large enough so all ejected electrons are absorbed. In many cases of radiological concern, the site or target of interest is too small for this assumption to be valid. This is particularly true when the absorbing site is $<1 \mu$ or smaller in tissue; then the more energetic ejected electrons have a high probability for escape from the absorbing region. In such cases, the secondary electron track will not contribute in its entirety to the energy deposited within the absorbing site.

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FIGURE 2.5. Quotient of the Variance by the Mean Energy Loss for Energetic Protons in Several Gases; Experimental Values are Obtained from Measured Single Collision Cross Sections.

One approximation which can be made to account for energy transport out of the volume of interest by energetic electrons is to exclude the contribution of any secondary electron whose initial energy is greater than an arbitrary limit. This is accomplished simply by setting a finite energy limit, Δ , on the integrals in Eqs. 2 and 3. If we do this we have

$$\delta_2(\Delta) = m_2(\Delta)/m_1(\Delta) \quad (5)$$

where

$$m_2(\Delta) = \int_0^\Delta E^2 \sigma(E) dE \quad (6)$$

and

$$m_1(\Delta) = \int_0^\Delta E \sigma(E) dE. \quad (7)$$

The quantity $m_1(\Delta)$, defined by Eq. 7, is closely related to energy restricted LET.

Values of the parameter, $\delta_2(\Delta)$, calculated for nitrogen from Eqs. 5, 6 and 7 are presented in Figure 2.6 for proton energies between 0.3 and 1.7 MeV. The marks on the abscissa indicate the classical kinematic maximum energy that a secondary electron can have for the corresponding proton energy, ($E_{\max} = 4E_p/1836$). The secondary electron energy distributions decrease rapidly for electron energies above this limit. Therefore, the kernels of the integrals in Eq. 2 and 3, and hence, the ratio of the moments, δ_2 , change little for larger values of Δ . This is why the curves in Figure 2.6 for a given E_p level off at the kinematic maximum electron energy. Below this point the curves are remarkably independent of proton energy. This follows since the basic shape of the secondary electron energy distributions does not change rapidly with proton energy.

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FIGURE 2.6. Restricted Values of δ_2 Obtained by Employing an Energy Cutoff Analogous to Energy Restricted LET

This independence of $\delta_2(\Delta)$ on E_p was anticipated by Glass and Roesch.⁽⁸⁾ They measured the ionization distributions produced by protons of various energies and observed a constant value for δ_2 of approximately 400 eV for proton tracks passing through and near a tissue equivalent site with a radius $12.5 \mu\text{g}/\text{cm}^2$. This value agrees quite well with the results reported here. According to Figure 2.6, a δ_2 value of 400 eV implies an energy cutoff of about 2.5 keV; a 2.5-keV electron has a range in air of $\sim 20 \mu\text{g}/\text{cm}^2$, which is consistent with the dimensions of the absorbing site employed by Glass and Roesch.

Differential cross sections for electron emission can be used to calculate the quotient of the variance of an energy straggling distribution by its mean energy loss. This ratio is an important quantity in microdosimetry because it is to a first approximation independent of the thickness of the absorber. It also indicates the relative importance of straggling in the observed variance of the microdosimetric quantity *lineal energy density*, y . Restricted ratios, obtained by invoking an electron-energy cutoff, are not sensitive to proton energy and are in quantitative agreement with measurements of ionization distributions.

Pulsed Radioluminescence

M. L. West

Investigations continue on the transport and transformation of energy in organic media following its

deposition by ionizing and non-ionizing radiation. These early stages of radiation action and subsequent energy transfer are conveniently probed by the pulsed radioluminescence technique using a 2-MeV Van de Graaff accelerator for pulsed proton excitation and a TRW pulsed light source for ultra-violet excitation.

Fluorescence response curves are measured by the single photon counting technique. Recent detailed measurements on benzene vapor fluorescence have enabled us to compare intramolecular energy transfer processes in the gaseous and liquid phase. Figure 2.7 shows an Arrhenius plot of the observed decay rate over the temperature range 5 to 50°C for

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FIGURE 2.7. Decay Rate of Benzene Monomer as a Function of Temperature

dilute solutions of benzene in cyclohexane excited by ultra-violet radiation. The solid line is a fit to the data with the functional form

$$k = k_0 + k_1 e^{-W/KT}.$$

Extrapolating to the gaseous isolated molecule condition, k_0 would be the sum of radiative and nonradiative intramolecular processes that are temperature insensitive; $k_1 e^{-W/KT}$ is a thermally activated quenching process with activation energy, W . Spears and Rice⁽¹¹⁾ recently measured lifetimes and decay rates of numerous vibrational levels of benzene vapor at 0.1 Torr pressure and concluded that a nonradiative channel began to appear at energies 2900 cm^{-1} above the zero vibrational level. This energy, obtained for the collision free condition, agrees quite well with our activation energy of $W = 2800 \text{ cm}^{-1}$ for the liquid phase measurements. The exact nature of this nonradiative channel is not known but several authors have suggested a photochemical process leading to isomerization of the benzene molecule. The medium pressure gas phase experiments of Ludwig⁽¹²⁾ (10-20 Torr) yield a pressure independent quenching rate of $11.5 \times 10^6 \text{ sec}^{-1}$. This compares to our value of $11.2 \times 10^6 \text{ sec}^{-1}$.

An attempt has been made to calculate the expected decay rate for an initial Boltzman distribution of vibrational levels. Unfortunately, there are several levels observed in the low-pressure gas phase data whose lifetimes have not been measured. Some of these levels correspond to normal mode vibrations not easily

reached by photon excitation but which can easily be populated by collisional activation. Our present interpretation is that radiative and nonradiative intramolecular processes are essentially unaffected by the perturbing medium and that cyclohexane merely acts as an inert solvent, furnishing a rapid vibrational relaxation of the initially populated levels.

A more detailed analysis has been made of the nonexponential decay previously reported⁽¹³⁾ for proton excitation of organic scintillators. The diffusional quenching model, based on track structure which considers radical-radical and radical-excited state interactions, has been critically reviewed and shown to be consistent with our present physical and chemical knowledge of charged particle behavior in the gas and liquid phase. The assumption of rapid recombination ($\sim 10^{-12} \text{ sec}$) of electron-ion pairs to form initial distributions of radical and excited states is in qualitative agreement with measured electron distributions produced by 1-MeV protons. The initial high density of low-energy electrons leads to rapid recombination through small electron-ion pair separation and through enhanced prethermalization recombination in densely charged regions.

Figure 2.8 illustrates a recent fit to some of the data. For this fit we used a radical-excited state interaction of $10^{10} \text{ l/mole-sec}$ which is approximately the upper limit for a diffusion-controlled reaction in cyclohexane. In the radiolysis of

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FIGURE 2.8. Non-Exponential Decay of Benzene Fluorescence Following Proton Excitation

cyclohexane, the radical yield is 4.8 to 5.5 rad/100 eV; of this radical yield, cyclohexyl (C_6H_{11}) has a G value of 4 to 4.5. Using this value and an integrated track length, we computed an initial radical concentration of approximately 0.3 rad/Å. The radical-radical interaction rate of 2.5×10^9 l/mole-sec is the experimental value of cyclohexyl radical-radical recombination. The initial track radius was varied from 10 to 30 Å. The calculated decay curves are sensitive to variation in initial track radius only in the subnanosecond region of decay. The important parameters are the ratios of

radical-radical and radical-excited interaction rates. A pulsing and detection system capable of probing this subnanosecond time range is being investigated as it could help determine the initial interaction radius and the energy transfer mechanism.

Computer simulated track structure in gases and solids, using experimental cross-section data as input to Monte-Carlo calculations, has been used to give the spatial distribution of events around an impinging ion path for comparisons to the initial boundary conditions of the previously formulated diffusion model. Details of this calculation are given by Wilson in "Computer Simulation Methods for Track Structure," pp. 44-46 of this report. Figure 2.9 displays a portion of a simulated proton track.

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FIGURE 2.9. Computer Simulated Track Structure For 1-MeV Proton

The points represent ionization events, both primary (caused by the passing protons) and secondary ionizations (resulting from energetic ejected electrons). The total track in material of unit density is about 200 times the length displayed in the figure. On this scale, the track is well simulated by a continuous core. For a 1-MeV proton, 75% of the ions have nearest neighbors less than 10 Å and 50% have nearest neighbors less than 5 Å. The proton loses about 25% of its energy in creating the primary ionizations along the track, 25% is lost in direct excitation along the proton track, and the other 50% goes into kinetic energy of the first generation of secondary electrons. Classical calculations, based on Bohr theory, show that these primary excitation and ionizations occur within molecular dimensions of the passing proton, i.e., within 5 to 10 Å of the incident particle. Secondary electrons, of course, spread this primary energy distribution. Our next attempt will be to use this Monte-Carlo generated track as the initial boundary conditions for the transport and transfer of energy. This should allow us to consider more accurately the physical processes of diffusion, thermalization, recombination, etc.

A dual pulsing system, capable of observing decay in the microsecond range, has been used to investigate long-lived fluorescence observed in solutions of aromatic hydrocarbons and alkanes when excited by ionizing radiation. Quenching studies with electron scavengers indicate that these fluorescence result from excited state formation via recombina-

tion of long-lived ions. The postulation of negative ion formation through capture of thermal electrons, followed by charge neutralization, also explains the absence of delayed fluorescence on the benzene cyclohexane decay curves obtained for proton excitation. Benzene has a negative thermal electron affinity, thus, inhibiting long-lived negative ion formation.

Data Buffer Interface for a PDP8 Computer

C. A. Ratcliffe

In many kinds of experiments where large volumes of data are collected, the efficiency and reproducibility of the tests are increased by adding some automation. This interface has been designed for application to cross-section measurements made in our laboratory. Its function is to collect counting information from two sources and relay it to a PDP8 computer under specified conditions. Another function is to receive new instrument settings from the PDP8 and relay these on to the experimental equipment. Thus, desired counting statistics and experimental conditions can be obtained by switch settings and software program entries. The counting information is generated by a channeltron electron detector, with a wide-band amplifier, and a voltage-to-frequency converter which effectively measures the accumulating proton beam charge.

The experimental equipment controlled by the PDP8 through the interface is one of several digitally

controlled voltage sources applying potential to an energy analyzing system.

Data collection is accomplished by two variable length scalers and a data buffer. A choice may be made whether to count for preset beam charge or preset pulse count (constant statistics). Both scalers have a range of preset values. When the selected scaler overflows, the contents of the other scaler are transferred to the data buffer and presented as digital information to the PDP8. This overflow starts a timing sequence which sends an interrupt signal to the PDP8. The PDP8

then ingests the data and sends a new voltage source setting (with a strobe) to be stored in a register in the interface. The register data is available at all times to the digital-to-analog converter in the variable voltage source.

The register is also capable of accepting manual data entry via a set of switches or acting as a triangle generator for sweeping the voltage source back and forth across a selected range. Here the upper and lower limits, as well as the sweep frequency, are operator adjustable.

• RADIATION DOSIMETRY

The development of an understanding of radiation effects on living systems at the cellular level continues to be the objective of the Radiation Dosimetry program. The emphasis is on the theoretical development and experimental realization and measurement of those physical parameters that appear appropriate for describing radiation effects. The major theoretical thrust has been on expanding the statistical theory to 1) more adequately incorporate dose rate and recovery effects, 2) examine the applicability of microdosimetric parameters, and 3) accommodate sublethal effects. This theory is then applied to a variety of published experimental radiobiological data to demonstrate its effectiveness in correlating the data. The microscopic distribution of energy in an irradiated absorber continues as an active topic of theoretical and experimental investigation. The major experimental effort, however, has been directed toward bringing to fruition a wide range of irradiation facilities for use in the Radiation Biophysics program. Facilities now provide the capability of irradiating cell monolayers and tissue culture with pulsed, monoenergetic protons, electrons, and filtered 330 kVp x rays. A wide-range of dose rate, LET, and dose fractionation are now available with well-documented dosimetric parameters.

Hit Theory: Dose Rate and Energy Density Effects

W. C. Roesch

The low-LET hit theory^(14,15) predicts that the survival, S , of a population in which sublethal damage may accumulate but in which it may also disappear without doing harm is given by

$$S = \exp [-CD - 1/2yAD^2]. \quad (1)$$

C and A are constants for a given radiation, D is the absorbed dose, and y is a function of the quotient of the absorbed dose by the dose rate. Ordinarily (see below) y is nearly 1.0 when the quotient is small and when the quotient is large it is inversely proportional to the dose.

This article discusses additions to the theory and illustrates its application to experimental data.

Experimental data is conveniently analyzed by plotting the quotient of the negative natural logarithm of the survival by the dose versus the dose. From Eq. (1)

$$-\ln(S)/D = C + 1/2yAD. \quad (2)$$

Figure 2.10 illustrates the possible results. The value at zero dose equals the parameter C . At intermediate dose rates the curve, a , rises monotonically from this value and approaches another constant value. For very high dose rates (y nearly 1.0), the curve, b , is a straight line continuing the initial slope of curve, a ; this slope equals $1/2A$.

FIGURE 2.10. Theoretical Predictions for the Quotient of the Negative Natural Logarithm of the Survival by the Absorbed Dose for an Intermediate Dose Rate, a, All Very High Dose Rates, b, and All Very Low Dose Rates, c. The mean recovery time can be determined from the dotted lines; see text.

For very low dose rates (γ nearly 0.0), the curve, c, is a horizontal line at the value C.

The mean recovery time of the damage can be determined from the dose, D_a , at which the high-dose-rate curve, b, attains the same value of $-\ln(S)/D$ reached by the intermediate dose-rate curve, a, at high doses; see the dotted lines in Figure 2.10. The relation is

$$\text{mean recovery time} = 1/2 D_a / \dot{D}, \quad (3)$$

where $\dot{D}$ is the dose rate.

The classical hit theories, for example one-hit, two-target or two-hit, single-target, give curves similar to curve, a. They can, however, be distinguished from the low-LET hit theory, hereafter referred to as the present theory. In the classical theories the value of $-\ln(S)/D$ at high doses is independent of dose rate; in the present theory the limiting value is related to the dose

rate by Eq. (3). In the classical theories the limiting value is related to the initial slope. If the square of the high-dose value is greater than the slope, classical theories do not apply.

Dose-Effect Relations. Any radiation effect that can be characterized as either present or absent in each member of the population (for example, a mutation) can be treated by a theory similar to that for lethality. The fraction, E, of the population showing the effect is 1.0 minus the fraction not showing the effect, i.e. $1-S$ for lethality. Assuming that the fraction E is small, then

$$E/D = C + 1/2\gamma AD \quad (4)$$

for effects in which preliminary damage may accumulate and disappear. Equations (2) and (4) are identical in form; therefore, the theory and analysis of survival and of effects are similar.

The assumption that E is small is necessary not only to permit a series expansion that leads to Eq. (4), but, more important, it means that instances of multiple initiations of the effect in individual members of the population can be neglected.

A particular effect may be produced by the radiation but then be rendered unobservable by some other effect of the radiation. Usually the given effect is not detected in a member of the population that dies, since normally only survivors are examined for it. Therefore, no theoretical correction is necessary provided the effect and death are stochastically independent phenomena. However, the interfering effect may

not be lethal. For example, a mutation may be produced but not expressed because of non-lethal damage elsewhere on the genome. The theoretical treatment of the non-appearance of such interferences is analogous to that for survival; the probability of non-appearance will be denoted by S^* and called the "effect survival." Where preliminary damage can accumulate and disappear, Eq. (4) should be modified to

$$E/D = (C + 1/2yAD) S^*. \quad (5)$$

In the analysis of experimental data it simplifies the work to represent the effect survival as either a simple exponential in the dose, e^{-kD} , or (except for low doses) as an exponential with an extrapolation number, $N e^{-kD}$. In the latter case, only the products NC and NA can be extracted from the data.

Unsynchronized Populations. The theory above assumes that all members of the population have the same values for the probabilities of the different responses they can make to the radiation. In general this will not be true. It might be true, for example, for a synchronized cell population; but it would probably not be true for an exponentially growing population, because cells in different phases of the mitotic cycle might have different properties. The results above have to be averaged over the subpopulations that have different properties.

Dose-Rate Dependence. In principle, the form of the function, y , above depends on how sublethal or

preliminary damage is assumed to disappear, i.e. how the cell recovers. In practice, the dependence is insensitive to exactly how the damage disappears. Hence, analysis of experimental data cannot disclose much beyond the mean rate of recovery, see Eq. (3). This insensitivity, on the other hand, means that experimental data can be analyzed without worrying about the details of recovery mechanisms. The assumption of exponential recovery made earlier^(14,15) is both convenient and sufficient for practical purposes.

An exception to the preceding conclusions would occur if sublethal or preliminary damage had to pass through some steps, taking some time, before they could combine to produce the observed effect. Then y would be zero at low doses or high dose rates instead of 1.0 as above, where no time delay was assumed. As an example of the further effects of a delay, at low doses or high dose rates, the effect E would become $CD + BD^3$, where B is another constant, rather than $CD + 1/2AD^2$ (Eq. (4)); in other words, the effect would depend on the cube of the dose rather than the square. No experimental data have been found that show this behavior.

In the case of exponential recovery

$$y = 2 [x^{-1} - (1 - e^{-x}) x^{-2}], \quad (6)$$

where $x = bD/\bar{D}$ and b is the inverse of the mean recovery time given by Eq. (3). For x less than about 3, a useful approximation is

$$y \approx e^{-0.28 x}. \quad (7)$$

Different Radiations. The above equations were derived on the assumption that the mean number of hits was large--as it is for low-LET radiation. Below it is shown that C at first increases and A remains unchanged as the LET increases; thus the C-term in the exponent in Eq. (1) begins to predominate as the LET increases. Quite separate arguments, applicable when the mean number of hits is small, show that for high-LET radiation the survival is the simple exponential

$$S = e^{-CD}. \quad (8)$$

Thus Eq. (1) is applicable at all LET's, with the A-term in the exponent of negligible importance at high LET.

The parameters C and A depend on the energies of the charged particles of the radiation; the exact connection is not known. The quantities employed in microdosimetry reflect both the density and the stochastics of energy deposition by radiation. The theory already employs them to relate the absorbed dose and the mean number of hits received. It appears natural to use them as a first approximation in establishing a connection between different radiations. When this is done, it is found that for radiations of low and intermediate LET the parameter A does not depend on the choice of radiation and C is a linear function,

$$C = c_1 + c_2 \xi, \quad (9)$$

of the energy-mean, ξ , of the specific energy density in single energy deposition events, that is $\xi = \langle z^2 \rangle_1 / \langle z \rangle_1$, where $\langle z^2 \rangle_1$ is the mean-square and $\langle z \rangle_1$ is the mean of the

specific energy density in single events. For very high LET radiation, the behavior of A is not known, and C equals the inverse of the mean specific energy

$$C = \langle z \rangle_1^{-1}. \quad (10)$$

Kellerer and Rossi⁽¹⁶⁾ introduced a "dual-action" theory, which is related to the present one⁽¹⁵⁾ by the equations

$$c_1 = 0 \quad \text{and} \quad c_2 = 1/2A. \quad (11)$$

In the analysis of experimental data it is simplest not to assume the Eqs. (11) but to test them at a late stage of the analysis. A frequently used analytical procedure is first to relate C to the (mean) stopping power of a radiation, L. This relation is then converted to relations with ξ and $\langle z \rangle_1$ by use of known⁽¹⁶⁾ and approximate relations between the latter and L; these relations involve the size of the site hit. Finally, the Eqs. (11) can be tested, if all the needed information is available.

The analyses reported below were chosen to illustrate or to contrast with the theoretical work described above.

Human Kidney Cells. Several analyses^(17,18) have been made of the LET-dependence of survival of human kidney T1 cells found by Barendsen⁽¹⁹⁾ and by Todd.⁽²⁰⁾ Figure 2.11 shows another similar analysis made to put the results in terms of the theory above and to provide comparisons for the analyses below. The experiments were conducted with unsynchronized cells; therefore, the values of C in the figure are averages over an exponentially growing population of cells.

FIGURE 2.11. Average Over the Cell Cycle of the Probability of Killing T1 Cells in Single Energy Deposition Events. Solid line, fit to experimental data; (19,20) o, normalized *Chlamydomonas* results; x, normalized *Neurospora* results

Figure 2.11 shows C for T1 cells as a function of stopping power, L . Between 0 and about $60 \text{ keV } \mu\text{m}^{-1}$, C is a linear function of L ; above $200 \text{ keV } \mu\text{m}^{-1}$ (not shown), C is inversely proportional to L . This behavior accords with Eqs. (9) and (10), because both ξ and $\langle z \rangle_1$ are approximately proportional to L . Also, A for the lower LET's is found to be independent of L , and at higher LET's the survival curves are so near to simple exponentials that A cannot be determined--again, in agreement with the theory.

Chlamydomonas. Figure 2.12 shows survival data obtained by Gruber and Nachtwey⁽²¹⁾ for the green algae,

FIGURE 2.12. Survival of *Chlamydomonas* in ^{60}Co Gamma Rays.⁽²¹⁾ □, $1.27 \text{ rad sec}^{-1}$ (plus four points outside the figure); Δ, 33 rad sec^{-1} ; --, asymptote and equivalent dose for 3.3 rad sec^{-1} ; (22) . . . , survival for 1.5-MeV protons

Chlamydomonas reinhardtii, in ^{60}Co gamma rays. A dose rate of $1.27 \text{ rad sec}^{-1}$ gave the horizontal line and thus a value for C of $4.0 \times 10^{-5} \text{ rad}^{-1}$. A dose rate of 33 rad sec^{-1} gave points that were fitted by the straight line with the same value of C and $A = 3.3 \times 10^{-8} \text{ rad}^{-2}$. Bryant's⁽²²⁾ data for 5-MeV electrons at 330 rad sec^{-1} , a dose rate ten times higher, agree well with this second line, thus, confirming that it is the high-dose-rate limit. Bryant also obtained data for electrons at 3.3 rad sec^{-1} ; they approached asymptotically the value $1.2 \times 10^{-4} \text{ rad}^{-1}$ shown by the dashed line. The high-dose-rate line reaches this value at a dose of $5 \times 10^3 \text{ rad}$. Equation (3) thus gives a mean recovery time of 750 sec, i.e. 12 min.

Figure 2.13 illustrates the preliminary data of Braby and Nachtwey⁽²³⁾ for survival of Chlamydomonas when exposed to 104 rad sec^{-1} of 1.5-MeV protons at different times after the end of the dark cycle. While the data have considerable statistical scatter, they do not appear to differ for the first ten hr. The straight line fitted to the data for the first ten hr has $A = 5.7 \times 10^{-8} \text{ rad}^{-2}$, a value almost twice that for ^{60}Co gamma rays. This line is plotted as the dotted line in Figure 2.12. By $12 \frac{1}{4} \text{ hr}$ the sensitivity to protons has started to increase; A is $8.8 \times 10^{-8} \text{ rad}^{-2}$.

The value of C chosen for the two proton lines of Figure 2.13 is the same as the value for ^{60}Co gamma rays. Although the scatter of the data would have permitted choice of a different value, it could not have been

very much different. Thus, apparently, the value of C is about the same for ^{60}Co gamma rays and 1.5-MeV protons, while the value of A is significantly different. This is contrary to the prediction of the theory above, which predicts that the C 's should be different for the two radiations and the A 's the same.

FIGURE 2.13. Survival of Chlamydomonas in 1.5-MeV Protons.⁽²³⁾ x, 3 hr after the end of the dark cycle; o, $4 \frac{3}{4}$ to 5 hr; Δ $7 \frac{3}{4}$ hr; $\square$, 10 hr, +, $12 \frac{1}{4}$ hr

Figure 2.14 shows the preliminary data of Braby and Nachtwey⁽²³⁾ for a mutation in Chlamydomonas that expresses itself as a reduced colony size. High and low dose rates, 33 and $1.27 \text{ rad sec}^{-1}$, of ^{60}Co gamma rays give $C = 5.0 \times 10^{-6} \text{ rad}^{-1}$ and $A = 3.0 \times 10^{-9} \text{ rad}^{-2}$; both are about one-tenth the values of those for survival. Only high-dose-rate data were

FIGURE 2.14. A Mutation of *Chlamydomonas* in Gamma Rays⁽²¹⁾ and in Protons.⁽²³⁾ $\square$, for 1.27 rad sec⁻¹ (plus four points outside the figure) and Δ for 33 rad sec⁻¹ from ⁶⁰Co; x for 1.5-MeV protons

available for protons. While the scatter in the data prevent a definitive judgment, the proton data appear to have the same value for A as the ⁶⁰Co data, but the value for C is considerably larger, $2.0 \times 10^{-5} \text{ rad}^{-1}$. These results, an increase in C with LET but no change in A, are in accord with the theory above. Further, the ratio of the two C's is not too much different from that for mammalian cells. This is shown in Figure 2.11, where both C's have been multiplied by a factor that makes the ⁶⁰Co value lie on the mammalian cell curve. The proton value is then too high; but not radically different.

Tradescantia. The x's in Figure 2.15 are the observed rates per unit dose of pink mutant cells per

FIGURE 2.15. Pink Mutations in *Tradescantia* Clone 02.⁽²⁴⁾ x, observed rates; o, rates corrected for event survival; 250-kV x rays and 0.43-MeV neutrons

stamen hair found by Sparrow et al.⁽²⁴⁾ in *Tradescantia* clone 02. The x rays were 250 kV at a rate of 1.6×10^{-2} to $10^{-1} \text{ rad sec}^{-1}$; the neutrons were 0.43 MeV. The yield in both cases declines at high doses, which indicates a loss of mutants. Event survivals of the simple exponential form, e^{-kD} , were sought that transformed the observed data into straight lines. The o's in the figure are the transformed data for $k \approx 1.1 \times 10^{-2} \text{ rad}^{-1}$ for x rays and $3.4 \times 10^{-2} \text{ rad}^{-1}$ for neutrons. One x-ray point, at 43 rad did not fit its line. The lines give $C = 5.6 \times 10^{-4} \text{ rad}^{-1}$ and $A = 1.0 \times 10^{-4} \text{ rad}^{-2}$ for x rays; $C = 3.0 \times 10^{-2} \text{ rad}^{-1}$ and $A = 0.0$ for neutrons.

Sparrow et al.⁽²⁴⁾ observed the very large RBE at low doses (the

ratio of the C's, $3.0/0.056 = 54$) and explained it by the "dual-action" theory. The present analysis does not affect their argument.

Mutations in Mice. Figure 2.16 illustrates the mutations per locus per gamete per unit dose that Russell⁽²⁵⁾ observed in mice following single exposures to 250 kV x rays at 1.5 rad sec^{-1} . The solid points are the observed rates; the open points are those rates after correction for a simple, exponential, e^{-kD} , effect survival. Data for males and females are indicated by the conventional signs. The observed data for males and females appear to be unrelated, but, when corrected for event survival, they fall on a common straight line. The parameters of this line are $C = 1.0 \times 10^{-7} \text{ rad}^{-1}$ and $A = 4.6 \times 10^{-10} \text{ rad}^{-2}$.

The sensitivities, the k's, for event survival are $1.6 \times 10^{-3} \text{ rad}^{-1}$ for females and $3.2 \times 10^{-3} \text{ rad}^{-1}$ for males; within the accuracy of the data, males are twice as sensitive as females. W. A. Glass suggested to me that this might be connected with the male gamete being haploid and the female diploid at the time of exposure.

Russell also obtained data at low dose rates and for fractionated exposures. The low-dose-rate data indicate a mean recovery time of 30 to 40 min. The fractionation data for females is consistent with the present event-survival model. That for males is not. Russell⁽²⁶⁾ observed that there is something peculiar about the fractionation data for males and suggested that different mechanisms are at work.

FIGURE 2.16. Mutations in Male and Female Mice⁽²⁵⁾ for 250-kV X Rays at a Rate of 1.5 rad sec^{-1} . Solid circles are the observed rates; the open circles are the rates corrected for event survival; males and females denoted by the conventional signs

Mutations in Neurospora. Figure 2.17 shows the mutations per unit dose in the ad-3 region of Neurospora crassa found by de Serres, Webber, and Lyman⁽²⁷⁾ for x rays and for helium and carbon ions. The solid points are the observed rates; the open points are those rates after correction for a simple-exponential, e^{-kD} , effect survival.

These data are of interest, because different mechanisms are at work than those considered in the sublethal-damage model discussed above. Here many of the mutations are deletions and must be analyzed with Wolff's⁽²⁸⁾ modification of the one-hit, two-target model. Webber

FIGURE 2.17. Mutations per Unit Dose in Neurospora Crassa.⁽²⁷⁾ The rates are for all the mutations observed at the ad-3 region. Solid points are observed rates; open points are the rates corrected for event survival; Δ , 250-kV x rays; $\square$, 18.5 keV μm^{-1} helium ions; o, 200 keV μm^{-1} carbon ions

and de Serres⁽²⁹⁾ presented a breakdown by type of mutation for x rays. Analysis of these more detailed data justifies lumping all mutations together as is done in the present theory to study the LET effect. After correction for effect survival, the data lie on three straight lines with a common intercept: $C = 6.0 \times 10^{-9} \text{rad}^{-1}$. The slopes are quite different: $A = 2.3 \times 10^{-12} \text{rad}^{-2}$ for

x rays, $1.0 \times 10^{-11} \text{rad}^{-2}$ for helium, and $8.8 \times 10^{-11} \text{rad}^{-2}$ for carbon.

As for Chlamydomonas discussed earlier, the C does not change with LET but the A does. For Neurospora, however, the reason for this difference in behavior from the sublethal-damage theory is explained by Wolff's theory. He showed that the quantity expressed as $1/2A$ in this paper is the square of a quantity similar to C,

i.e. a probability of producing an effect in a single energy deposition event. The quantity $(1/2A)^{1/2}$ is $1.1 \times 10^{-6} \text{ rad}^{-1}$ for x rays, $2.3 \times 10^{-6} \text{ rad}^{-1}$ for helium, and $6.6 \times 10^{-6} \text{ rad}^{-1}$ for carbon. To compare them with mammalian cell probabilities, these probabilities are plotted (see the x's in Figure 2.11) after they have been normalized so the x-ray point lies on the curve. The Neurospora probabilities are very similar (proportional to) those for mammalian cells. The variation with LET implies that "sub-mutational" effects occur that combine to produce the observed mutation; c.f. Eq. (9).

The quantity denoted here by C is, according to Wolff's theory, the probability of a single charged particle inactivating both targets. This probability is one-tenth of a percent or less of the probability just discussed of a single particle inactivating a single target. The lack of LET dependence of C indicates sub-mutational damage is not involved.

Models for Inactivation by Interaction of Radiation Products

W. C. Roesch

In the development of the low-LET hit theory a specific assumption was made about how the probability of surviving interactions between the products of two different energy deposition events varied with the time between them. It was assumed that the time dependence was exponential; the probability of surviving two hits was taken to be

$$S_1^2 (1 - a e^{-bt}). \quad (12)$$

The a and b are constants, t is the time between the hits, and S_1 is the probability of surviving each hit individually when the time between them is so great that there is no influence of one on the other. Simple models of time-dependent behavior, on the other hand, predict multi-exponential functions for this probability of the form

$$C_0 + C_1 e^{-b_1 t} + C_2 e^{-b_2 t} + \dots + C_n e^{-b_n t}. \quad (13)$$

For a number of different models for the rate of interaction between radiation products, the present work showed that the terms in the series of Eq. (13) after the second are negligible for physically reasonable values of the parameters in the models. Since the first two terms of this series are easily transformed into the form of Eq. (12), the assumption made in the low-LET theory is justified.

The models studied were stochastic, Markov, discrete-state, continuous-time models. The "state" was the number of one type of radiation product in a member of the population. Radiation was simulated by one or more instantaneous redistributions in state; LET was simulated by the manner of redistribution. Between hits radiation products could disappear innocuously, could cause death, or could combine with another product to cause death. The modes of interaction studied included those in which: a) each pair of products had the same chance of interaction, b) only the N nearest neighbors could

interact, c) the probability of interaction decreases by the same factor for each intervening neighbor, d) the probability of interaction decreased exponentially with the distance between two products and the average distance between products decreased with their number. The last assumption is the most realistic; however, none of the models considered led to any markedly different conclusions.

Energy Deposition Theory

W. C. Roesch

Calculation of Compound-Poisson Processes. An important, basic problem in energy deposition theory is the calculation of the probability density, $f(E)$, of a quantity, E , that is a sum of a random (Poisson distributed) number of contributions, each of which is drawn from the one basic distribution, $f(E:1)$. Kellerer⁽³⁰⁾ and Roesch⁽³¹⁾ developed general calculation methods based on the convolution integral. The Fourier transforms, denoted by F , of the two distributions are related by

$$F(f(E)) = \exp [-r(1 - F(f(E:1)))], \quad (14)$$

where r is the mean number of contributions to E . The desired distribution can be calculated by producing and inverting the transforms by the fast Fourier transform method.⁽³²⁾

The transform can also be inverted using the well known method of steepest descents; see reference 33. The solution is an asymptotic series. It requires inversion and taking the

square root of another series to evaluate the solution. The solution is algebraically quite complicated, but it is easily calculated by computer. The solution obtained by steepest descents is in a form that algebraically becomes the normal (Gaussian) distribution in the limit of large r . General theory⁽³⁴⁾ indicates that it should, but other methods of calculation do not show by their functional form that it does.

Each method of calculation works over a limited range of r . The convolution method works over the largest range. For typical energy deposition distributions, the convolution method becomes impracticable for r greater than about 10^3 ; above this value the resulting distribution is so narrow that too fine a computing grid is needed. The discrete Fourier transform method works only up to an r of about 10 for a reasonable number of Fourier components; however, it takes about one-tenth the computing time of the convolution method. The method of steepest descents works for r greater than about 100; for smaller r the convergence of the asymptotic series is poor. The method thus supplements the convolution method for large r . It also saves about a factor of ten in computing time. There are at present no fast methods of calculation for r between about 10 and 100.

Irradiation Facilities and Dosimetry

L. A. Braby

To accommodate the varied needs of the radiation biophysics, physics,

and dosimetry programs, increased flexibility in our irradiation equipment was required. Existing equipment had to be modified and acquired to produce x-ray, electron, and proton radiations of varying pulse lengths and repetition rates, as well as continuous beams.

In the biophysics program pulses about one minute in length separated by minutes to hours are necessary to study gross cellular responses, sub-microsecond pulses separated by up to hundreds of microseconds are required to apply the oxygen depletion technique developed by Epp et al.,⁽³⁵⁾ and subnanosecond pulses to simulate high-LET effects with low-LET radiation. In the dosimetry program very small doses of electrons and x rays must be delivered in submicrosecond pulses to measure microdosimetric distributions for low-LET radiation. In the physics program nanosecond or shorter pulses are needed to study the initial, fast, non-exponential luminescence of organic scintillators in aliphatic hydrocarbons excited by low-LET radiation. Submicrosecond pulses are required for characterization of radiation-induced free radical reactions through pulsed radiolysis experiments.

To meet these varied requirements, the Van de Graaff electron accelerator was modified so it could emit radiation in pulses from a few nanoseconds to many microseconds in length as single pulses or long trains of pulses. Its ability to produce DC beams was retained. The cell irradiation apparatus at the

Van de Graaff proton accelerator was changed to produce lower dose rates and preset doses and apparatus for x-ray exposures was prepared.

Electron Van de Graaff. To accomplish these major changes a new high-voltage terminal and a new accelerator tube and tank extension were added to the electron Van de Graaff accelerator. These changes do not interfere with the continued use of the machine to calibrate health-physics instruments. The different modes of operation of the accelerator are produced by different biasing of the cathode relative to the grid which is grounded. The cathode and related parts of the electron injector were purchased from a Van de Graaff manufacturer.⁽³⁶⁾ The cathode is the end of a 50-ohm coaxial cable, which is terminated by resistors very near the cathode. The choice of mode of operation is made at the operating console. A rotary solenoid at the base of the accelerator transmits this choice to a switch in the terminal by turning a polystyrene rod that connects the two.

For steady currents, the coaxial cable and hence the cathode are biased with a negative voltage sufficient to secure the desired current. For pulsed currents, the cathode is biased positive to prevent a beam. At the console a push button for single pulses or a variable-rate pulser for pulse trains actuates a light-emitting diode attached to the grounded end of one of the lucite tension rods that supports the accelerator column and terminal. Light is

transmitted up the rod and detected by a photomultiplier tube in the terminal. An amplified signal from the photomultiplier triggers one of two pulsers that drives the cathode negative for the duration of the pulse. The beam current achieved is controlled by changing the pulser output voltage or the static positive bias voltage.

The length of short pulses is determined by discharging an open-ended coaxial cable. This pulse-forming cable is charged up to as much as 3000 V negative by a power supply in the terminal. On the signal from the photomultiplier, a one-shot circuit closes a mercury-wetted relay that connects the pulse-forming cable to the cathode. The beam is turned on during the resulting negative pulse. The shortest pulse length that could be achieved was one nanosecond. Actually, no pulse-forming cable was required since the components connected to the mercury relay perform its function. For 10-nsec pulses, a length of RG-8 cable is connected to the mercury relay by a coaxial switch. The longer pulses--50, 150, 500, 1500, and 5000 nsec--are produced similarly but by using three-section, lumped-constant delay lines instead of coaxial cables for pulse formation. The lumped-constant circuits are individually connected by miniature relays to a thyatron that fires on signal from the photomultiplier. The thyatron can operate at up to 1000 pulses per second.

Figure 2.18 illustrates a measurement of the accelerator beam current during a 10-nsec pulse. The beam was collected on a funnel-shaped 50-ohm

FIGURE 2.18. Sampling Scope Display of Nominal 10-nsec Electron Pulse

Faraday cup which was terminated in a 50-ohm transmission line connected to a high-speed sampling oscilloscope. Figure 2.19 shows measurements of the 50- and 5000-nsec pulses with an ordinary fast oscilloscope. The maximum currents obtained were approximately 0.5 A; other results indicate 1.0 A will usually be possible. The maximum current is limited by the condition and temperature of the cathode as well as by the bias and pulsing voltages. The condition of the cathode may be limiting for long pulses and for pulse trains of high repetition rate; this needs more study to be fully understood. Figure 2.20 lists the pulse lengths and currents available and shows corresponding doses superimposed on the ranges of pulse lengths and doses over which some of the phenomena of interest in this program are observed. The dose achieved in a pulse is proportional to the current density. For Figure 2.20 it was assumed that the density was 1 A cm^{-2} . Much higher (lower) doses can be achieved in specimens of smaller (larger) cross sections.

FIGURE 2.19. Oscilloscope Traces of Single 50- and 5000-nsec Electron Pulses

FIGURE 2.20. Relationship of Electron Pulse Lengths and Amplitudes to some Chemical and Biological Effects

Proton Van de Graaff. The apparatus for proton irradiation of cells used by Braby and Nachtwey⁽³⁷⁾ gave very high dose rates, e.g. 10^4 rad sec^{-1} . Much lower rates are needed to determine the low-dose-rate limit for survival curves. Also, irradiations were made for selected times; the resulting doses showed undesirable variation because of short-period fluctuations in accelerator output.

The dose rate was reduced by introducing a second scattering foil into the apparatus and covering it with one of two interchangeable masks, 0.034 and 0.140 cm in diameter. At low dose rates, timing is not so critical, so the electrostatic deflector used for this purpose was replaced by a mechanical shutter. This made it possible to increase the distance between the scattering foil and the cells to further reduce the dose

rate. Dose rates of 3 to 30 rad sec^{-1} can now be produced.

Measurement of dose rate and beam profile at the low rates required a surface-barrier detector masked down to a small area and moved across the beam. This was combined with an improved Faraday cup for the higher dose rates. The dose falls to about 87% of the value at the center at the edges of a 3/8-in. disc.

Reproducible doses are now obtained by automatically terminating the exposure when a preset count is

obtained from the surface-barrier detector used to monitor the dose delivered.

X Rays. The x-ray machine used in calibrating health-physics instruments is now available for cell irradiation studies. This 330-kVp industrial x-ray machine is used with 4.75 mm of Al filtration resulting in a half-value layer of 1.5 mm of Cu. A tissue culture flask is held by a jig at a distance that results in a dose rate of 200 rad min^{-1} in water with less than 10% variation over the flask.

• RADIATION BIOPHYSICS

*In an effort to understand the effects of radiation on biological systems at the cellular level, we have chosen two systems to study at sublethal radiation levels. The CHO is a mammalian cell line amenable to detailed investigation of radiation-induced division delay and radiosensitivity changes near the end of the cell cycle. Theoretical effort has been on mathematically describing the progression of cultured cells through the division cycle and formulating plausible models to account for the biochemical processes that are perturbed by radiation. The experimental emphasis has been in carefully selecting and characterizing the cell line to be used throughout the course of these collaborative experiments. Tissue culture techniques have also been perfected that will allow irradiation under the physically well-defined beams of monoenergetic protons and electrons. A second biological system, the single celled organism *Chlamydomonas reinhardtii*, is being studied with particular attention being directed to its unique mutagenic behavior. Close coordination of experimental design, including dosimetric and biological considerations, and theoretical interpretation continue to be of central importance in this program.*

Radiation Sensitivity in the Cell Cycle

W. C. Roesch

The growth and division of cells is a cyclic process. Radiation affects different stages of this cycle differently. Irradiated cells move through the stages of the cycle at changed rates. Understanding the physical and chemical antecedents of radiobiological effects requires knowledge of these phenomena.

Ideally, the stages of the cell cycle would be identified by microscopic examination or by biological or biochemical tests. Then time would be used as one variable with which to make correlations. Some known characteristics of the sort

needed are mitotic phenomena, uptake of radioactive precursors, biochemical effects, fluorescence, and size. However, there are not now enough of them known to give sufficient discrimination. In practice time itself is used to characterize the stages of the cell cycle. The stage has the characteristics that normal cells treated in a standard manner have on the average at the given time after division. When cells are irradiated or given some other treatment, one asks at what time during the normal cell cycle he would have found the cells he now finds at some given time as a result of the irradiation. The form of the question is simplified by calling time in the normal cycle by a different name,

"phase" or "biological time," and denoting it by a different symbol, x instead of t . Now one asks for the distribution in x at time t . Of course, providing a name does not solve the problem, one needs some test for determining that the cells would have been in phase x .

Since stage of the cycle is defined by time in the cycle, the time rate of movement through the cycle is by definition uniform. As a result, the cells are known⁽³⁸⁾ to be exponentially distributed in x when the total number of cells is increasing exponentially with time.

Radiation Sensitivity. Sinclair and Morton⁽³⁹⁾ determined survival curves for Chinese hamster V79 cells at several phases. From these and other data they prepared curves of survival versus phase for two different doses. Since there are two curves and since there are two unknowns, C and A , in the high-dose-rate survival equation,

$$S = \exp [-CD - 1/2yAD^2] \quad (1)$$

(with $y = 1$), it was possible to calculate C and A as functions of phase. Figure 2.21 gives the results. The results between 8 and 10 hr are highly uncertain because of uncertainties in the survival curves. Elsewhere the accuracy is not high, but it is sufficient for a preliminary appraisal. Figure 2.21 shows that C , which is related to the probability for inactivation as a result of a single energy deposition event, decreases steadily from division until about the end of the S-phase when it returns rapidly to its value at division. A , which is related to the

probability of inactivation as a result of interactions between the products of two different energy deposition events, increases from division until about the same time that C starts to rise, at that time it starts to decrease. The calculation has more uncertainty in the values of A than in those of C ; this, rather than real changes in sensitivity, is probably responsible for some of the small fluctuations in A .

FIGURE 2.21. Survival Parameters C and A Deduced for Chinese Hamster V79 Cells

The biochemical steps that a cell goes through may be thought of as a large complex web of reaction pathways. If there are rate-limiting steps in this web, the values of C and A before those steps will dominate the curves illustrated in Figure 2.21. The populations of cells in states following synchronization should follow ordinary chemical kinetic laws; in simple cases they will change exponentially. The C -curve was analyzed to 7 or 8 hr into three exponential functions of mean lives 20 min, 1 hr, and 3 hr. The same exponentials fit the A -curve, provided they are followed by a slight delay

and then a very slow reaction. The analysis does not yield the sequence in which the reactions occur, but four of the six possibilities for three reactions can be ruled out because they give negative values for C or A. The most promising of the remaining possibilities, because of the results discussed below, has the three in the sequence: 20 min, 3 hr, 1 hr. The calculated populations preceding these reactions and the very slow one are shown in Figure 2.22. The relatively low population in the third stage results from its being fed by a slow reaction and drained by a fast one. Because of its low population, it has large values of C and A--larger than any of the values in Figure 2.21.

FIGURE 2.22. Populations of the First Four Rate-Limiting States in the Chemical-Kinetic Model for Chinese Hamster V79 Cells

That both the C- and the A-curves can be fitted by the same reaction kinetics suggests that the above analysis may be more than ad hoc

curve fitting. Other evidence is accumulating. Hydroxyurea blocks cells at the end of G_1 and produces a decrease in survival.⁽⁴⁰⁾ The above chemical-kinetic model predicts correctly the change in survival for the first several hours provided the block is of the 1-hr reaction. Then the survival increases again for reasons not now in the model; the value of survival finally reached is that for the second state. When n-ethylmaleimide is added with the hydroxyurea, the late increase in survival does not occur, i.e. the survival continues to drop. The model correctly predicts the decrease over the entire period measured so far. (This is a step toward determining the x of cells observed at t.) These results suggest that not only the normal response of cells to radiation but the effects of radiation-modifying drugs and other treatments may be strongly influenced by cell population changes. In addition, cyclohexamide⁽⁴¹⁾ and puromycin⁽⁴²⁾ blockages observed in non-radiation experiments have been explained by a 3-hr mean-life phenomenon that is in the right phase to be the one found above. Obviously much more theoretical and experimental work is needed to develop the chemical-kinetic model.

Mitotic Selection. Some cells become less tightly bound to their substratum for a short time just prior to division. Schneiderman et al.⁽⁴³⁾ developed a technique for periodically collecting these loosely bound cells for long times. The collection period was shorter than the interval of loose binding. The technique is

useful in studying the properties of cells near the end of the cycle. We are interested in using it to study radiation sensitivity at the end of the cycle and radiation-induced division delay.

Leeper, Schneiderman, and Dewey⁽⁴⁴⁾ measured the amount of radiation-induced division delay as a function of phase for Chinese hamster ovary cells. It is small (an hour or two, depending on the dose) immediately after division, falls slightly and then increases steadily until about the end of S. In late S and early G₂ it is constant at about twice the initial value. After a point in late G₂, termed the x-marker, there is no delay. The results of the mitotic selection procedure depend on where in phase the cells spend their extra time. A theoretical study, suggested by Schneiderman, was made of different hypotheses about the extra time. For this purpose, the phase scale was divided into a large number of small steps. Untreated cells were assumed to take steps at random; after a time they would be in a Poisson distribution beyond their origin. Figure 2.23 shows the results for the following two hypotheses. Cells remain in the phase in which they were irradiated; see Figure 2.23 solid line. The cells progress normally until they reach the x-marker where they spend any remaining delay time; see Figure 2.23 dashed line. The first cells collected are those that were already past the x-marker when they were irradiated; they come through unaffected in both cases. Those delayed at the x-marker produce a large peak after the release of the

longest-delayed cells. Such a peak appears in experimental data but it is smaller. Other hypotheses considered were that the cells accumulate at the x-marker, as above, but before they are released they are randomly distributed through G₂ and that the cells do not stop anywhere but are simply slowed down--dx/dt changes (uniformly in x).

FIGURE 2.23. Theoretical Yield Curves for the Mitotic Selection Technique. Cells spend their delay in the phase in which they were irradiated; solid line. Cells move normally to the x-marker, where they spend the rest of their delay period; dashed line.

The calculations showed that the delays measured by conventional techniques⁽⁴⁴⁾ were about two-thirds of those observed by mitotic selection.⁽⁴³⁾ We attribute this to differences in pH resulting from differences in the two techniques. Also, preliminary values for the initial yields of the cells we are now using were about one-half those calculated for similar cells used elsewhere. We decided to postpone further theoretical work until the properties of our

cells could be measured; measurements are described in "Chinese Hamster Ovary Tissue Culture," pp. 72-74 of this report.

Chinese Hamster Ovary
Tissue Culture

M. H. Schneiderman

The equipment, instruments, supplies and biological materials necessary for supporting growth of tissue culture cells in the biophysics laboratory have been assembled and Chinese hamster ovary (CHO) cells are being cultured. Synchronous mitotic cells have been selected from monolayer cultures using a portable mitotic selection apparatus (PMSA). The PMSA was designed to facilitate tissue culture experiments at remote radiation facilities, such as Van de Graaff accelerators, with a minimum perturbation of the cultured cells. The PMSA contains a heater and a carbon dioxide purge system to maintain temperature and pH control, respectively. Since the mitotic cells are selectively removed from a monolayer culture by creating a shear force sufficient to remove tenuously attached mitotic cells, a mechanical shaker was installed to control the amplitude and frequency of the selection force. Because some experiments require large numbers of mitotic cells, a cold temperature storage compartment was incorporated into the system to hold the temperature between 0 to 4°C which prevents cell progression. The entire support system is portable and can be moved from building to building.

A microspectrophotometer will be used in data acquisition to determine the source and amount of cells exhibiting a given intensity of fluorescent emission and a cytofluorograf (flow microfluorimeter) to determine the number. Since techniques are now available for specifically staining cellular constituents with fluorescent dyes, measurements can, for example, be made of the distribution of DNA in the CHO cell population. Figure 2.24 is a cytofluorogram showing the frequency distribution of DNA in CHO cells. The cells were fixed in formaldehyde and stained with acriflavine by the fluorescent Feulgen technique. The first peak represents cells which have not yet duplicated their DNA (G_1 phase cells) and have half the relative fluorescence of the second peak. The second peak represents cells which have duplicated

FIGURE 2.24. Cytofluorogram illustrating the DNA Distribution of an Asynchronously Growing Population of CHO-K1 Cells Stained with Acriflavine Using the Fluorescent Feulgen Technique. The fluorescence is from specifically stained DNA; the two peaks represent the DNA content of the G_1 and G_2+M populations. The interpeak portion represents the S-phase cells.

their DNA but have not yet divided (G_2 +M phase cells). The cells in the inter-peak region represent cells actively engaged in DNA synthesis (S-phase cells).

A clone of CHO cells, designated CHO-K1, was obtained from the American Type Culture collection. This is a certified cell line and is listed as CCL-61. The doubling time (Figure 2.25) was determined by counting replicate fields of cells at the indicated times. The initial count, relative time 0, was not taken until 24 hours after plating. This insured that the lag in growth due to handling of cells did not interfere with

FIGURE 2.25. Doubling Time (T_D) of Asynchronously Growing CHO-K1 Cells

the growth curve. The doubling time was 13 hr. Figure 2.26 shows the differential mitotic index and the time in minutes of the different phases of mitosis. The duration of the phases of mitosis were determined by the formula:

$$N_M = 2^{T_M/T_D} - 1.$$

N_M is the number of cells (fraction of total population) in phase, T_M the time in phase, and T_D the doubling time, 13 hr; see Figure 2.22. The T_D was determined earlier (Figure 2.25); the fraction of cells in each phase of mitosis, N_M , was determined by scoring 25000 asynchronously growing cells for the differential mitotic index (see Table 2.1). Our initial results with this cell line indicate that CHO-K1 can be selected in mitosis with a high mitotic index (>90%) and that after ionizing radiation (100 rad, x ray) CHO-K1 cells exhibit kinetics similar to those observed earlier. (43)

To facilitate the understanding of radiation effects on living cells, a model expressing the growth of mammalian tissue culture cells was developed by W. C. Roesch and is described in the "Radiation Sensitivity in the Cell Cycle," pp. 68-72 of this report. This model is particularly adapted to interpreting data gained by the mitotic selection technique and can be used to predict the number of mitotic cells entering the mitotic selection window as a function of time. The model can also be used to predict kinetics of cell progression after radiation challenge resulting in a specified delay or cell killing.

The collaborative nature of the Biophysics Program established during this first full year will continue

with the design and execution of experiments to study the interaction of radiation with CHO cells.

FIGURE 2.26. Length of Mitotic Phases in Minutes Before Division and Differential Mitotic Index (% of the mitotic cells in each phase of mitosis)

TABLE 2.1. Differential Mitotic Index of CHO-K1 Cells

	Interphase	Prophase	Metaphase	Anaphase and Telophase	Total
Number	24481	133	276	110	25,000
Length of Phase (min)	757	6	12	5	23 ^(a)

(a) Mitotic time

Lethal and Mutagenic Responses of *Chlamydomonas Reinhardi*

L. A. Braby and D. S. Nachtwey

In these experiments, we have attempted to minimize physical and biological variables which normally contribute to uncertainty in radiobiological results. We have used a monoenergetic source of protons and a cell population synchronized by controlling the light-dark culture conditions for the test organism, *Chlamydomonas reinhardi*. The irradiation facility used has been previously described by Braby and Nachtwey⁽⁴⁵⁾ and in "Irradiation Facilities and Dosimetry," pp. 63-67 of this report.

Dishes of cells grown on minimal media agar were irradiated with 1.5-MeV protons; the central area of each dish was then photographed at about 20X magnification to identify the positions of individual cells. The fraction of cells surviving irradiation was determined by making an overlay showing the position of all cells on the original photograph and marking it with the position of survivors found on photographs made four days after irradiation. This technique produces good statistical results in the shoulder portions of the survival curves because it eliminates any uncertainty in the total number of cells at risk. For the mutation frequency experiments, cells were irradiated as a monolayer on a 1/4-in.-diameter millipore filter. After irradiation, a majority of the cells washed off the filter when it was placed in medium and these cells were then plated and checked for mutations.

When irradiated by monoenergetic protons ($dE/dX = \sim 20 \text{ keV}/\mu\text{m}$) at a rate of approximately 10^3 rad/sec , *C. reinhardi* show a sigmoidal survival curve similar to that resulting from conventional x-ray irradiation. However, as indicated in Figure 2.27, there is a substantial change, primarily in the shoulder width, in the shape of the curve as a function of time in the cell cycle.

Within the time span investigated, from about three to twelve hr following the onset of the light cycle, the LD50(4) reaches a maximum of about $5.3 \times 10^3 \text{ rad}$ near the fifth hour and decreases to about $3.4 \times 10^3 \text{ rad}$ at the twelfth hour. The shape of these curves, of course, suggests that multiple events are required for inactivation and that a repair mechanism may also be active. If we take the 7 3/4-hr curve as typical, we see the LD50(4) is about 5 krad; at this dose approximately 47 protons traverse a three-square micron site, the size of

FIGURE 2.27. Survival Curves for *Chlamydomonas Reinhardi* Irradiated at 7 3/4 and 12 1/4 hr into the Daily 12-hr Light Period

a cell nucleus. Split dose experiments, in which approximately 8×10^3 rad were given in two separate nearly equal fractions, verified that there is a repair mechanism which appears to reach its maximum effectiveness, reducing lethality a factor of two, in approximately 20 min. Bryant,⁽⁴⁶⁾ using 22.5×10^3 rad of 5-MeV electrons, showed recovery of this organism with a similar time course, but with a survival ratio of several thousand. Since neither low-dose-rate proton experiments nor very-high-dose-rate x-ray or cobalt-60 experiments are yet completed, it is not possible to give a good RBE value for proton irradiation. However, it appears that protons are approximately twice as effective at the LD50(4) level as are x rays when the x-ray exposures take several minutes.⁽⁴⁷⁾

C. reinhardi show an easily detectable radiation-induced mutant phenotype which can be called "slow growth." These cells produce colonies only 1/8 to 1/3 the diameter of wild type colonies at five days. This phenotype occurs with significant frequency, even at very high survival rates, as indicated in Figure 2.28, probably because mutations in many different genes lead to the same phenotypic expression. Statistics of the experiments have not allowed observation of any variation in

mutation frequencies (if there are any) with time of cell cycle; thus, data shown in Figure 2.28 was pooled from several experiments performed at different times in the cell cycle. As expected, mutation frequency increases with decreasing survival, but it reaches the surprisingly high levels of approximately 25% at a surviving fraction of 5%. Gruber and Nachtwey⁽⁴⁷⁾ have investigated the mutation frequency of C. reinhardi with x-irradiation and have found that at all survival levels, it is somewhat lower than observed in these experiments with proton irradiation. The same has been found⁽⁴⁸⁾ for cobalt-60 irradiation. However, since experiments at comparable dose rates have not yet been performed, direct comparisons are not possible.

FIGURE 2.28. Mutation Frequencies and a Typical Survival Curve for Proton Irradiated Chlamydomonas

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NUCLEAR MEDICINE TECHNOLOGY AND OTHER HEALTH APPLICATIONS

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

• NUCLEAR POWERED PROSTHESES

A 50-watt intrathoracic $^{238}\text{Pu}^{16}\text{O}_2$ heat source in a miniature swine produced no clinical signs of untoward effect over a period of almost 10 months, in spite of aortic occlusion and burnup some time prior to sacrifice. Retroperitoneal abdominal implant experiments indicate surface heat flux tolerance limits of less than 0.01 watts/cm^2 immediately postoperatively, increasing to nearly 0.04 watts/cm^2 within one month, exploiting surface tissue ingrowth techniques. A miniature swine continues on experiment with a 29-watt $^{238}\text{Pu}^{16}\text{O}_2$ heat source implanted retroperitoneally in a discoid aluminum container with a velour fabric coat.

Biological Effects of Intracorporeal Radioisotope Heat Sources

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The objectives of this program are to study in miniature swine the short- and long-term effects of heat and radiation from intracorporeal radioisotope heat sources similar to those envisioned for use in totally implanted cardiac assist/replacement systems and to obtain basic information on bulk tissue heat conductivity in the vicinity of large, tissue-ingrown radioactive implants as required to establish engineering design criteria for prototype systems.

Intrathoracic (intraaortic) and abdominal (retroperitoneal) heat exchangers with electrical (for surface temperature versus heat flux studies) and encapsulated $^{238}\text{Pu}^{16}\text{O}_2$ heat sources (29- and 50-watt), the latter for long-term study of biological effects of their heat and radiation,

are implanted in female miniature swine having a body mass similar to an adult man. We will implant smaller, planar devices in various locations to obtain basic information on tissue heat conductivity and its variation with type of tissue and with time following implantation, using heat generating surfaces which encourage tissue ingrowth.

A thermally insulated, Silastic coated graphite heat exchanger containing a 50-watt $^{238}\text{Pu}^{16}\text{O}_2$ heat source was implanted in the descending thoracic aorta of a female miniature swine on February 8, 1973, using aortic blood flowing through the device as a coolant. Except for a few days of lethargy and anorexia in September, we noted no evidence (physical examination, periodic hematology, serum chemistry and sternal marrow biopsy) of untoward sequelae until late November. On November 27 anorexia, lethargy, mild dyspnea, relative neutrophilia and elevated lactic acid dehydrogenase prompted radiographic examination (Figure 3.1)

(a)

(b)

(c)

(d)

FIGURE 3.1. Lateral Radiograms of Animal No. 4442 Following Implantation of an Intrathoracic Aortic Heat Exchanger Containing a 50-watt Plutonium-238 Heat Source. (a) Immediately following implantation, (b,c,d) ca. 42 weeks after implantation. (b) Flat film revealing large, dense scar tissue capsule surrounding implant; (c) aortogram showing total occlusion of aorta anterior to the implant, hypertrophied collateral arterial circulation (especially the internal thoracic arteries) and dense matrix of arteries in the scar tissue capsule; (d) pulmonary arteriogram showing its considerable contribution to the arterial supply in the scar tissue capsule.

which confirmed device occlusion. Except for the obliteration of a portion of the thoracic aorta, the presence of a large and thick vascular scar tissue capsule (with a charred lining), and cooked (literally) vertebral bodies and intervertebral discs over the implant site, there were no remarkable gross necropsy findings. Numerous special and routine tissue specimens are now in the process of histopathological evaluation. Three control animals (same implant but a lead slug in place of the heat source) continue on experiment without incident.

Spherical (5 in. diam) and discoidal (8-in.-diam x 3-in.-center thickness) implants containing electrical heaters and internal and surface thermistors were implanted retroperitoneally in the ventrolateral abdomen of miniature swine. We first established that surface heat fluxes, reported elsewhere as tolerable,⁽¹⁾ parboils tissues and produces death within a few days under such conditions. We estimate that, immediately following implantation of such massive implants, local surgical shock and anatomic disruption limit tolerable surface heat flux to something less than 0.01 watts/cm^2 . We also established, however, that surface heat flux tolerance increases markedly with time due to restoration of

normal local blood perfusion and surface ingrowth of vascularized scar tissue.

On December 6, 1973, three weeks following implantation, we surgically removed the electrical heater from a discoidal implant and replaced it with a 29-watt $^{238}\text{Pu}^{16}\text{O}_2$ heat source. This produces a surface heat flux of 0.036 watts/cm^2 and an interface temperature of 108°F or less (Figure 3.2). Based on the preceding three weeks of experimentation with electrical heating and favorable observations at surgery, this animal should continue on experiment indefinitely.

FIGURE 3.2. Tissue Temperature ($^\circ\text{F}$) at the Surface of a Velour Covered Retroperitoneal Discoid Implant Versus Applied Internal Heater Power (watts) Three Weeks Following Implantation Surgery; surface heat flux = $(\text{power}/810) \text{ watts/cm}^2$

Estimates of the surface area represented by the viable tissue shell of the scar capsule enclosing the thoracic source (not tissue-ingrown) suggest that, given adequate time for proper capsule development and vascularization, surface heat fluxes of up to 0.06 watts/cm^2 or more may be eventually tolerable. Abdominal experiments, however, suggest a practical tolerance limit (after 3 weeks of

healing, exploiting surface ingrowth of tissue) of about half that. The low tolerance limit immediately after source implantation (less than 0.01 watts/cm^2) means that existing subsystem prototypes may have to be cooled temporarily by means other than direct conduction during the postoperative healing period.

• MEDICAL USES OF ISOTOPES

An inexpensive portable blood irradiator for clinical use in suppressing circulating lymphocytes and organ graft rejection and a generator for Nitrogen-13 ($T_{1/2} = 10$ min) suitable for clinical diagnosis are two applications for isotopes being investigated under this program.

A new approach to blood irradiation has been developed using Thulium-170 cast as an integral part of a vitreous carbon body. Production of the irradiator does not involve personnel exposures since the ^{169}Tm used is neutron activated after manufacture. Reactivation is anticipated as units lose effectiveness with the 125-day half-life of ^{170}Tm . Total weight of units applicable to clinical practice is anticipated to be less than 1 kg. An irradiator using Strontium-90-Yttrium-90 was also constructed and tested in a dog.

Alpha irradiation of Boron-10 is the basic route being investigated for the ^{13}N hospital generator. Both $^{13}\text{N}_2$ and $^{13}\text{NH}_3$ have potential value as diagnostic pharmaceuticals. Nitrogen-13 has been produced in proof-of-principle tests using the $^{10}\text{B}(\alpha, n)^{13}\text{N}$ reaction with Americium-241 as the alpha source. Significant quantities of ^{13}N were measured in the B powder and trace quantities of ^{13}N were detected in a sweep gas. Use of BN is expected to release more gaseous products. Methods are currently under study for producing $^{13}\text{NH}_3$, principal among these is the use of KOH to release $^{13}\text{NH}_3$ from irradiated powder. Primary criteria are cost, simplicity and short process time.

Development of a Blood Irradiator

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Blood irradiation to suppress tissue and organ rejection and to control some blood diseases has been shown to be successful. It has also been demonstrated that chronic irradiation may be superior to repetitive acute exposures. This work is directed toward developing small inexpensive irradiators for clinical use and evaluating their effectiveness in animals for suppressing the cellular

immune reaction under different dose rate regimes.

$^{90}\text{Sr}^{90}\text{Y}$ irradiators, reported in the literature, have linearized the source material and thus have required distributed and relatively heavy shielding. To minimize the shielding, we contained 2 Ci of $^{90}\text{Sr}^{90}\text{Y}$ in a cylindrical zone 1 cm long within an approximately 25-mm-thick outer layer of Hevimet shielding. A 0.025-mm-thick layer of titanium covered the $^{90}\text{Sr}^{90}\text{Y}$; see Figure 3.3. Thin-walled Silastic tubing, 2.64 mm ID and 3.66 mm OD, served to transport the circulating

FIGURE 3.3. Schematic of the Mark XIV $^{90}\text{Sr}/^{90}\text{Y}$ Irradiator (units in millimeters)

blood through the irradiator. The blood dose was estimated by passing Fricke dosimeter solution through the Silastic tubing at known flow rates and measuring optical density change. The transit dose to blood was estimated to be 40 rad at a flow rate of 100 ml/min. Total weight of the irradiator (Figure 3.4) was slightly less than 2 kg. When the arterio-venous shunt was installed in the dog, the blood flowed through the irradiator more than 8 hr but less than 12 hr before clotting occurred and the shunt was removed. Following this dose of 2000 to 3000 rad, the levels of small lymphocytes fell and remained at approximately 2/3 of their pre-exposure level for 6 weeks, well after other blood constituents had returned to pre-implant levels.

Embrittlement of the Silastic tube was observed which stimulated the

consideration of alternative materials for the blood interface and body of the irradiator. Carbon is little affected by radiation, even at intensities in the core of reactors. As pyrolytic graphite, it is also essentially nonthrombogenic. It is a low Z material and produces minimum bremsstrahlung. We selected a recently developed technique of polymerizing polyfurfuryl alcohol with subsequent vacuum high temperature carbonizing that produces a vitreous carbon. This casting technique also permits inclusion of the source material as an integral part of the carbon body. Thulium-170 was chosen to be the radiation source because of its 125-day half-life and its maximum energy beta of 0.96 MeV. All the carbon casting can be done with non-radioactive ^{169}Tm and neutron activation of the encapsulated ^{169}Tm to

Neg 731930-2CN

FIGURE 3.4. Photograph of the Mark XIV Irradiator Ready For Insertion of the Silastic Blood Transport Tube

form ^{170}Tm . Furthermore, the unit can be repeatedly reactivated as its effective strength declines below useful levels. Figure 3.5 is a section through the wall of the vitreous carbon showing the 0.05-mm inner barrier of carbon, the 0.15-mm layer of $^{169}\text{Tm}_2\text{O}_3$ and the 2.3-mm covering layer of carbon. Note the micro as well

as macro containment of the source material. Total weight of the vitreous carbon unit following carbonizing is 2 g; with a containment layer of 6.4 mm of lead, the weight is 160 g. Figure 3.6 illustrates the components of the first prototype and Figure 3.7 shows them assembled.

FIGURE 3.5. Longitudinal Section Through the Wall of the Vitreous Body of the Thulium Irradiator (X180)

Neg 735804-4CN

FIGURE 3.6. Prototype Thulium Irradiator Components: the Pyrolytic Graphite Blood Tube, 2.8 mm ID, 3.3 mm OD, and 51 mm long, the 2-gram Vitreous Carbon Irradiator Body, and the Lead Shield

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FIGURE 3.7. Prototype Thulium Irradiator Assembled with Silastic Tubing to Connect to Shunt Cannulas; Total wt 160 g

In this initial ^{170}Tm irradiator, a pyrolytic graphite tube⁽²⁾ having a 0.025-mm wall thickness is used as the blood interface because of its non-thrombogenic character. Silastic tubing connects this graphite tube to the arterial and venous cannulas. External dose rates were less than 50 mR/hr at the surface and a transit dose of 21 rad (at a flow rate of 100 ml/min) was measured by Fricke dosimetry. This unit is being tested on a miniature goat for effectiveness in suppressing lymphocyte levels and extending skin allograft rejection times.

Calculations using the Unidentified Flowing Objects (UFO) code⁽³⁾ indicate the possibility of obtaining several-fold higher transit doses if the vitreous carbon can serve as the blood interface. Figure 3.8 shows the dose profile calculated for a

typical unit and for a comparable unit without a liner. Samples of vitreous carbon are now being tested for their thrombogenicity but no data are yet available.

The combined use of ^{170}Tm and vitreous carbon offers excellent potential for developing inexpensive, safe and lightweight blood irradiators for testing and for appropriate clinical application. Until the development of this irradiator, testing for effects from chronic irradiation had not been possible; now broader use can be expected. The greatest initial use is expected to be in suppressing early rejection of kidney transplants. Also some forms of leukemia are reported to respond favorably to blood irradiation. Radiation safety during production and use of the units appears to be very favorable.

FIGURE 3.8. Calculated Cross-Section Dose Rates to Blood from a 1-Ci ^{170}Tm Source

Hospital Generator for Nitrogen-13

D. H. Lester, W. F. Riernath and
J. A. Partridge

Nitrogen-13 is a positron emitter with a half-life of 10 min. Tagged N_2 gas provides a good diagnostic tool for lung function because the shortness of its half-life and low absorption in the lungs lead to a low body burden. ^{13}N has been used successfully as a tracer to locate and size myocardial infarctions in experimental animals in the form of $^{13}\text{NH}_4^+$ in solution and to detect tumors in animals.⁽⁴⁾ Studies of

kidney and liver function have also been suggested for $^{13}\text{NH}_3$ or $^{13}\text{NH}_4^+$ solutions. Many other diagnostic uses are also possible because of the basic role of NH_3 in body function.

The disadvantage of the use of ^{13}N is that it must be generated at the time and place of its use. To date experimenters have used high-energy accelerators available at the site where the biological testing was performed. The most popular route is deuteron bombardment of methane, $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$. This reaction requires 8 to 10 MeV deuterons which must come from an accelerator. Since the deuteron reaction requires special techniques, staff and equipment, widespread use is not practical. An effective ^{13}N generator must be economical, easy to operate, safe to use, and readily accessible.

Early in the program a number of candidate reactions were screened for possible use. To date, the most promising is $^{10}\text{B}(\alpha,\text{n})^{13}\text{N}$. The boron may be used in a pure form or in a compound. Whatever process is used, the time from irradiation to output must be short due to the isotope's short half-life.

Proof-of-principle tests have been performed. The generation system consisted of a high-intensity ^{241}Am source of alpha particles adjacent to ^{10}B in a container separated from the alpha source by Mylar film. This geometry was found necessary to eliminate background interference during counting to test for the presence of ^{13}N . The ^{13}N production was verified by gamma-ray spectrometric measurements made on the assembly containing the ^{10}B . Counting peaks

were observed in the energy channels appropriate for the annihilation gammas formed by positron decay of the ^{13}N . Peak height decay rates were in good agreement with the reported 10-min half-life of ^{13}N .

Supplementary experiments were conducted to determine the extent of ^{13}N release from the ^{10}B powder. Similar irradiation geometry was used for a smaller chamber containing ^{10}B . This chamber was connected, as shown in Figure 3.9, to a circuit which allowed the atmosphere of the ^{10}B chamber to be swept out, passed through two filters in series (each capable of removing particles 3 microns or larger in size), and counted in an external, separate chamber. In two experiments of this type, 13 and 18 counts of ^{13}N were recorded for a 5-min counting period. In comparison, approximately 170,000 counts of ^{13}N were recorded for a 5-min counting period in the first experiments in which the entire ^{10}B assembly was counted.

These results indicate that for these approximate STP conditions, most of the ^{13}N formed was retained in the ^{10}B . Preliminary indications

FIGURE 3.9. Schematic Diagram of Apparatus Used to Test ^{13}N Release

from thermodynamic calculations are that boron nitride formation is in fact favored in these circumstances. Thus, it appears that a practical ^{13}N generator design will require conditions more favorable for ^{13}N release.

Work is underway to explore use of ^{10}BN powder to promote ^{13}N release as a constituent of $^{13}\text{N}^{14}\text{N}$ molecules. Boron nitride can be heated to high temperatures to aid nitrogen release.

Also under investigation are methods to produce $^{13}\text{NH}_3$. One direct route is alpha irradiation of ^{10}BN followed by treatment with KOH which will liberate NH_3 gas, some of which is tagged with ^{13}N . Early identification of the existence of ^{13}N was made using such a method.⁽⁵⁾ Another possibility is

One advantage of this route is that NH_3 should be the only gaseous product. The ammonia could be recovered in a bubbler (as is done in the $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ route) and used as a $^{13}\text{NH}_4^+$ solution. It could also be possible to obtain ^{13}N gas by treating the NH_3 to liberate N_2 .

Other boride compounds have been studied as possible candidates but many are unstable and too hazardous for use.

In addition to technical evaluations, economic evaluations are needed. At present, the quantity of ^{241}Am required is cost prohibitive. Studies are underway to determine how the cost of alpha irradiation can be reduced.

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ARTIFICIAL HEART PROGRAM

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

• **MEDICAL GRADE PLUTONIUM-238 FROM AMERICIUM-241**

The objective of this program is to compare and evaluate the relative economics and technical practicability of various approaches to producing both high-quality (low Pu-236 content) and high-purity (low dilution from other plutonium isotopes) Pu-238 by irradiating Am-241 recovered from spent power reactor fuel.

The scope of the program includes (1) projecting the availability of Am-241, (2) identifying the process facilities required for americium recovery, and (3) identifying the optimum target irradiation and processing cycle.

Medical Grade Pu-238 from Am-241

R. W. McKee

Up to the present time, the principal source of Pu-238 has been neutron irradiation of Np-237 in AEC reactors and subsequent chemical separation of the Pu-238 that was formed by neutron capture. A small amount of Pu-236 is also formed and contributes a disproportionately high radiation component to the product because of its intensely radioactive daughters. In the near future, power reactors will be discharging spent fuel containing significant quantities of Am-241 which, if separated, could be irradiated to produce a Pu-238 product free of Pu-236. The objective of this program is to compare and evaluate the relative economics and technical practicability of various approaches to producing both high-quality (low Pu-236 content) and high-purity (low dilution from other plutonium isotopes) Pu-238 by irradiating Am-241 recovered from spent power reactor fuel. The Am-241

target irradiation would most likely be performed in power reactors but irradiation in AEC reactors is also being considered.

The scope of the program includes (1) projecting the availability of Am-241, (2) identifying the process facilities required for americium recovery, and (3) identifying the optimum target irradiation and processing cycle. In developing availability projections, both current and probable future power reactor fuel cycles are to be examined. The primary parameters influencing availability include (a) nuclear power growth projections, (b) extent of plutonium recycle, (c) startup and equilibrium fuel exposure, and (d) neutron cross sections of the americium isotopes and their precursors. Both capital and operating cost estimates are to be developed for the process and facilities required for recovery of Am-241/Am-243. Possible americium target capsule configurations and compositions are to be defined and burnup

calculations carried out to develop relationships between irradiation time, initial Am-241/Am-243 ratios, and product compositions. Since Am-241 and Am-243 do not burn out at the same rate, the target cycle for efficient utilization of the Am-241 through recycle of unburned americium becomes complex and the feasibility of full utilization of the Am-241 depends to some extent on a possible coproduct market for the Cm-244 produced from the Am-243.

Because of the branching decay of 16-hour Am-242 during irradiation that produces both Cm-242 and Pu-242, the Pu-238 (from 163-day Cm-242 decay) is diluted with Pu-242. Possibly up to one-half of the Pu-238

could be recovered free of any Pu-242 if the undecayed Cm-242 could be separated from the plutonium immediately after target discharge. However, this rapid separation is difficult and costly because of the high radioactivity level. A computer model of the americium recovery and Pu-238 production cycle is to be developed to calculate optimized production costs for various conditions and various production requirements. This model may, for example, be used to prepare correlations of optimized production costs as a function of Pu-238 purity, production rate, and market price for Cm-244.

• RECIPIENT RADIATION EXPOSURE

The objectives of this study are to determine the dose from radioisotopic heat sources suitable for circulatory support systems and to develop radioisotopic heat source holders and shams for use in an animal study to assess the combined effects of heat and radiation from implanted plutonium heat sources.

Improved input data and an ancillary code developed for specification of source strength and spectra have been used in parametric computations of dose rates to organ systems and whole body. Conceptual designs of heat source holders for abdominal implantation of heat sources have been completed.

Circulatory Support Systems

F. T. Cross

The major objective of this study is to determine the dose from radioisotopic heat sources suitable for circulatory support systems. This is being accomplished by measuring and calculating, with suitable codes, the dose rates within and surrounding various tissue-equivalent phantoms. Another portion of the program concerns the development of radioisotopic heat source holders and shams for use in an animal study aimed at assessing the combined effects of heat and radiation in miniature swine implanted with plutonium heat sources.

Because of personnel change, some effort was required to clarify the parameters and data used as input to the modified QAD-P5A code. Some of the earlier experimental-calculational correlations were rerun.

When satisfied the code is being used properly, computations of dose rates to organ systems and whole body from prototypic heat sources will be continued. Improved data and a code developed in another 1830 project for specification of source strength and spectra (including the daughter products) as a function of aging are being used in computations.

Conceptual designs of noncirculatory heat exchangers for abdominal implantation of heat sources were completed. Work continues on the design and fabrication of both thoracic and abdominal circulatory and noncirculatory implant devices incorporating telemetry. Eventual guidance on the dose rates received by the animals with implanted heat sources will be provided either in the form of measurements or calculations for correlation with any observed biologic effect.

• POPULATION RADIATION EXPOSURE

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

At the beginning of 1973, preliminary calculations indicated the most significant component of dose was that transmitted to spouses. Because the age and sex of persons exposed are critical factors in developing estimates of possible genetic or somatic effects, the objectives of the study were broadened to develop greater definition of the age and sex of persons potentially exposed. An extensive survey to collect required interpersonal distance data was carried out in the Salt Lake City area. To obtain required detailed descriptions of the household characteristics of potential artificial heart recipients, copies of the data tapes from the March 1972 Current Population Survey program were obtained from the Census Bureau, and a special computer program was written to process these tapes. At year end, the principal effort was being devoted to the computer program to carry out the detailed dose calculations and summations.

Population Radiation Exposure

R. W. McKee

Use of Pu-238 fueled artificial hearts may expose persons in the vicinity of the user to small amounts of radiation. The objective of this project is to develop reliable estimates of the radiation dose to the population that would result from successful development and routine use of Pu-238 fueled artificial hearts. The results will be used to identify the physiological implications of the artificial heart to others besides the recipients of the device. The approach taken in this study is to subdivide the potential artificial heart population into identifiable subgroups small enough

and sufficiently well defined such that their activities and daily relationships with others can be reasonably identified using sample survey techniques. Dose calculations can then be made for these activities and a summation of all doses for all activities and all groups can then provide 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 the population dose.

	<u>Total Annual Dose 1000's of rem</u>	<u>Average Annual Dose rem/Person</u>
Dose to Spouse		
Sleeping in Double Bed	1460	12
Sleeping in Twin Beds	120	1
Dose to Other Family Members	62	0.3
Dose to Work Associates	116	0.07
Dose to Others and the General Public	250	0.001

The study had progressed at the beginning of 1973 to the point where preliminary calculations could be made. The above table summarizes these initial results based on the year 2000 and a medium projected artificial heart use of 188,000. The most significant component of dose was shown to be that transmitted to spouses. This could be substantially reduced if spouses were to sleep in twin beds.

These initial results were based on interpersonal distance surveys in the local area. Additional survey data collected later in the Seattle area produced comparable results.

These initial radiation dose estimates indicated significant doses would be incurred by persons closely associated with artificial heart recipients. Because the age and sex of persons exposed are critical factors in developing estimates of possible genetic or somatic effects, the objectives of the study were broadened to develop greater definition of the age and sex of persons potentially exposed. This substantially expanded the data matrix required for the calculations.

To satisfy the additional interpersonal distance data requirements, an extensive survey was carried out in the Salt Lake City area with assistance from Dr. Irwin Altman, Chairman of the Psychology Department of the University of Utah. Ten graduate students were temporarily employed to collect the survey data. The data were obtained through a combination of interviews and direct measurement of the environment. For example, a subject would identify his location with respect to other household members during specific activities such as eating or TV viewing and the interviewer would then measure the distances separating the subject and the other persons present. This technique for data collection had been evolved and tested during the previous smaller scale surveys and works quite well. For public activities a different technique was used to collect data. This technique is based on observing average people-densities and uses a computer program employing Monte Carlo techniques to calculate probable interpersonal distance relationships.

Although the Census Bureau produces extensive tabulations of various kinds of population characteristics, none of these tabulations provide the kind of detail required in this study. To obtain the detail needed, copies of the data tapes from the March 1972 Current Population Survey program were obtained from the Census Bureau. These tapes contain records on approximately 45,000 households interviewed in March 1972. A special computer program was written to process these tapes to obtain detailed descriptions of the household characteristics of potential artificial heart

recipients (PHR). The resulting data bank can provide details such as the number, sex, and age of other household members as a function of sex, age, and marital status of a PHR.

At year end, the Salt Lake City survey data had been processed and analyzed, the processing of the census data had been completed and work was going forward to expand the main computer program, REPREV, to carry out the detailed dose calculations and summations.

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