27
11-20-81
24 to 215

(2)

38455

UCRL-53208
Thesis

Gamma-Ray Spectroscopy Following High-Flux 14-MeV Neutron Activation

MASTER

Robert Eugene Williams
(Ph.D Thesis)

October 12, 1981

Lawrence
Livermore
National
Laboratory

Gamma-Ray Spectroscopy Following High-Flux 14-MeV Neutron Activation

Robert Eugene Williams
(Ph.D Thesis)

Manuscript date: October 12, 1981

DISCLAIMER

This document contains information that is classified as "Confidential" under the Atomic Energy Act of 1954, as amended, and the Atomic Energy Regulations, 10 CFR 835.21. The information is being furnished to you for your information only and is not to be distributed outside your organization. The information is being furnished to you for your information only and is not to be distributed outside your organization. The information is being furnished to you for your information only and is not to be distributed outside your organization.

LAWRENCE LIVERMORE LABORATORY
University of California • Livermore, California • 94550

Available from: National Technical Information Service • U.S. Department of Commerce
5285 Port Royal Road • Springfield, VA 22161 • \$12.00 per copy • (Microfiche \$3.50)

UCRL-53208

219

GAMMA-RAY SPECTROSCOPY FOLLOWING
HIGH-FLUX 14-MeV NEUTRON ACTIVATION

Robert Eugene Williams
Nuclear Engineering Program
University of Illinois at Urbana-Champaign, 1981

Dr. Philip K. Hopke
Director of Thesis Research
Institute for Environmental Studies
University of Illinois at Urbana-Champaign

Dr. Richard A. Meyer
Site Advisor
Nuclear Chemistry Division
Lawrence Livermore National Laboratory

The Rotating Target Neutron Source (RTNS-I), a high-intensity source of 14-MeV neutrons at the Lawrence Livermore National Laboratory (LLNL), has been used for applications in activation analysis, inertial-confinement-fusion diagnostic development, and fission decay-heat studies. The fast-neutron flux from the RTNS-I is at least 50 times the maximum fluxes available from typical neutron generators, making these applications possible. Facilities and procedures necessary for gamma-ray spectroscopy of samples irradiated at the RTNS-I were developed.

Standard reference materials (fly ash, orchard leaves, and bovine liver) from the National Bureau of Standards were analyzed for 29 elements using high-flux, multielement fast-neutron activation analysis (FNAA). From gamma-ray spectra collected after the 14-MeV neutron irradiations of these materials, over 80 radioisotopes were observed from 37 elements. Procedures were developed that avoided problems due

to the complicated irradiation geometry and minimized interferences from multiple reactions. The results were the most complete analyses of any samples to date by 14-MeV neutron activation, and demonstrated these procedures to be reliable and accurate.

In support of efforts by the Laser Fusion Program at LLNL to use 14-MeV neutron-activation as a fuel-density diagnostic of inertial-confinement-fusion (ICF) targets, various materials were irradiated to examine their suitability for advanced diagnostics. Enhanced positron-annihilation radiation was observed in gamma-ray spectra from trace amounts of Br mixed with target material, indicating Br is a viable candidate to be used as a seed gas added to the fuel to achieve a direct fuel-density measurement. The cross section for the $^{79}\text{Br}(n,2n)^{78}\text{Br}$ reaction was measured.

The final application of high-flux, fast-neutron irradiations was the production of ^{148}Pr , a radioisotope of interest in fission decay-heat studies, from ^{148}Nd . From gamma-ray spectra obtained from a single detector, the energies and intensities of gamma rays in the decay of ^{148}Pr have been tabulated. The success of these irradiations has demonstrated the feasibility of production of shorter-lived radioisotopes, such as ^{100}Nb and ^{62}As , for similar decay scheme studies.

TABLE OF CONTENTS

Chapter	Page
I. INTRODUCTION	1
II. FAST-NEUTRON INTERACTIONS WITH NUCLEI	3
A. Systematics of 14-MeV Neutron Reactions	3
B. Radioisotope Production by 14-MeV Neutron Irradiation	9
C. Fast-Neutron Attenuation	12
III. PRODUCTION OF 14-MeV NEUTRONS	15
A. The DT Reaction	15
B. Neutron Generators	18
C. The Rotating Target Neutron Source, RTNS-I	23
IV. ACTIVATION ANALYSIS	32
A. Current Status of Neutron Activation Analysis	32
1. Thermal-Neutron Activation Analysis	33
2. Fast-Neutron Activation Analysis	35
B. Quantitative Analysis Methods	38
C. Limitations of Activation analysis	40
D. Errors in Activation Analysis	45
1. Irradiation-Related Errors	45
2. Flux Monitoring	47
3. Sources of Error in Gamma-Ray Spectroscopy	48
4. Additional Sources of Error	51
E. Procedures for Multi-element FNAA	51
1. Sample and Standard Preparation	52
2. Irradiations	52

Chapter	Page
3. Gamma-Ray Spectroscopy	55
F. Results and Discussion	57
1. Radioisotopes Observed in Standard Reference Materials	57
2. Preliminary Results	69
3. Analysis of Standard Reference Materials	71
G. Conclusions and Recommendations	76
V. DENSITY DIAGNOSTICS OF INERTIAL-CONFINEMENT-FUSION TARGETS ..	78
A. Density Measurements by 14-MeV Neutron Activation	78
B. Direct Measurements of the Fuel Areal Density	85
C. Measurement of the $^{79}\text{Br}(n,2n)^{78}\text{Br}$ Cross Section	89
D. Conclusions	92
VI. FISSION DECAY-HEAT STUDIES	94
A. Fast-Neutron Irradiation of ^{148}Nd to Produce ^{148}Pr	95
1. Background	96
2. Irradiations of ^{148}Nd at the RTNS-I	97
B. Results and Discussion	99
VII. SUMMARY	113
REFERENCES	115
APPENDIX	124
VITA	166

TABLE OF TABLES

Table	Page
III-1 RTNS-I Source Parameters	25
IV-1 Constituents of the Standards Used to Analyze NBS Fly Ash Following Short Irradiations	53
IV-2 Radioisotopes with Half Lives Less than 1 Hour Observed in 14-MeV Neutron Activation	59
IV-3 Radioisotopes with Half Lives Between 1 Hour and 3 Days Observed in 14-MeV Neutron Activation	60
IV-4 Radioisotopes with Half Lives Greater than 3 Days Observed in 14-MeV Neutron Activation	61
IV-5 Interferences in Multielement FNAF	62
IV-6 Summary of Elemental Concentrations in NBS Fly Ash Determined by FNAF	72
IV-7 Summary of Elemental Concentrations in NBS Orchard Leaves Determined by FNAF	73
IV-8 Summary of Elemental Concentrations in NBS Bovine Liver Determined by FNAF	74
V-1 Tracer Candidates for Use in a Direct Measurement of Fuel Areal Density of ICF Targets	87
V-2 Nuclear Properties of Br, Fe, and Al Used in the Determination of the $^{79}\text{Br}(n,2n)^{78}\text{Br}$ Cross Section	91
VI-1 Energies and Relative Intensities of Gamma Rays in the Decay of ^{148}Pr Produced in High-Flux 14-MeV Neutron Irradiations of ^{148}Nd	107
VI-2 Comparisons of Relative Gamma-Ray Intensities of ^{148}Pr with those Reported by Taylor and Singh, and Ikeda <u>et al.</u> .	109
VI-3 High-Energy Gamma Rays in the Decay of ^{148}Pr that Support the Existence of Three New Energy Levels in ^{148}Nd	111

Table	Page
A-1 Valve Control Commands	145
A-2 Trouble-Shooting	146-147
A-3 Rabbit System Programs and their Constituent Procedures and Functions	150

TABLE OF FIGURES

Figure	Page
II-1 Fast-Neutron Cross Section Trends for the (n,2n), (n,p), and (n, α) Reactions	7
III-1 Neutron Energy vs. Angle of Emission for Deuteron Energies of 100-500 keV	17
III-2 DT-Neutron Energy Spectrum at 0 ^o and 90 ^o for a Deuteron Energy of 600 keV	19
III-3 RTNS-I Machine Room	26
III-4 Rotating Tritium Target Assembly at the RTNS-I	27
III-5 Neutron Flux at Points along the Beam Axis vs. Distance from the Target	30
III-6 Neutron Flux at Points off the Beam Axis 0.6 and 1.1 cm from the Target	31
IV-1 Gamma-Ray Spectrum from NBS Fly Ash Following a Short 14-MeV Neutron Irradiation	63
IV-2 Gamma-Ray Spectrum from NBS Fly Ash Following an Intermediate 14-MeV Neutron Irradiation	64
IV-3 Gamma-Ray Spectrum from NBS Fly Ash 7 Days after an Intermediate 14-MeV Neutron Irradiation	65-66
IV-4 Gamma-Ray Spectrum from NBS Orchard Leaves Following an Intermediate 14-MeV Neutron Irradiation	67
IV-5 Gamma-Ray Spectrum from NBS Bovine Liver Following an Intermediate 14-MeV Neutron Irradiation	68
V-1 Ablative Inertial-Confinement-Fusion Target Used at the Shiva Laser Facility	80
V-2 Apparatus for Collection of Target Debris	82
V-3 Coincidence Detector Used for 14-MeV Neutron Activation Diagnostic of ICF Target Pusher Areal Density	83

Figure	Page
VI-1 Decay Scheme of ^{148}Pr Proposed by Ikeda <u>et al.</u>	98
VI-2 Gamma-Ray Spectrum Following 14-MeV Neutron Irradiation of ^{148}Nd	100-105
A-1 Rabbit Loader and Plunger	130
A-2 The Irradiation Station Adjacent to the RTNS-I Target Assembly	131
A-3 Schematic of the Rabbit System	132
A-4 Rabbit Body, Sample Capsule, and Snap-In Cap	134
A-5 Details of the 14-MeV Neutron Irradiation Geometry Utilizing the RTNS-I Rabbit System	136

I. INTRODUCTION

For well over 20 years, small accelerators have been able to produce 14-MeV neutrons for use in nuclear physics and activation analysis. These neutron generators were inexpensive and relatively easy to maintain, and as a result there has been a great deal of research utilizing 14-MeV neutrons. In recent years, the need to study fast-neutron-induced radiation damage to potential fusion-reactor first-wall materials has lead to the development of much stronger sources of 14-MeV neutrons.

Until 1979, the Rotating Target Neutron Source, RTNS-I, at the Lawrence Livermore National Laboratory (LLNL) was the most intense source of 14-MeV neutrons in the world. When the RTNS-II came on line, at LLNL, and was dedicated to fusion-materials research, the RTNS-I became available for more experiments. Although the RTNS-I has a source strength an order of magnitude less than the RTNS-II, the peak flux from the RTNS-I is 50-1000 times greater than fluxes typically available from small neutron generators. Thus, this unique facility can be used for applications which are impossible using smaller sources.

The application central to the present work is fast-neutron activation analysis (FNAA). Quantitative analysis by 14-MeV neutron activation has been used for twenty years, but has generally been limited to a few single-element analyses because of the flux limitations of neutron generators. In 1977, preliminary experiments indicated the great potential of utilizing high-flux, fast-neutron activation at the RTNS-I for multielement analysis. One of the major goals of this

research has been the realization of that potential through the development of procedures for high-flux, multielement FNAA.

Additional applications of high-flux, 14-MeV neutron activation, using the same basic techniques as FNAA, that is fast-neutron irradiations followed by gamma-ray spectroscopy, were also being considered at LLNL. The Laser-Fusion Program, having developed a fuel-density diagnostic using 14-MeV neutron activation, wished to test materials of interest for advanced inertial-confinement-fusion diagnostics, and to measure fast-neutron reaction cross sections of these materials. The Nuclear Structure Group in the Nuclear Chemistry Division wished to produce certain radioisotopes of interest in fission decay-heat studies using fast-neutron irradiations of stable isotopes, avoiding the need of complicated chemical separations of mixed fission products.

All three of these applications were hampered by the inability to irradiate samples at the RTNS-I and quickly retrieve them for counting short-lived activity. A pneumatic sample-transfer system, in place since 1968, had become unreliable, making the required irradiations nearly impossible. Refurbishing this rabbit system in 1980 enabled the completion of the studies described in this work.

At first glance, the applications of high-flux, fast-neutron activation presented in this thesis may appear to be a potpourri of subjects. However, the facilities and the procedures developed for these three series of experiments are nearly identical. Hopefully, the work described herein will serve as a foundation for additional applications in the future.

II. FAST-NEUTRON INTERACTIONS WITH NUCLEI

A description of fast-neutron reactions is required in order to appreciate procedures used in 14-MeV neutron irradiations. (Throughout this work, the term "fast neutron" will be synonymous with 14-MeV neutron, contrary to the usual nuclear engineering usage of the term to mean neutrons with energies greater than about 100 keV.) The first section contains a summary of 14-MeV reaction systematics and a review of the current status of cross-section data. Radioisotope production by neutron activation is described in Section B. Fast-neutron attenuation, which can be a major problem in FNAA, is discussed in the final section.

A. Systematics of 14-MeV Neutron Reactions

The total cross section for a neutron interaction with a nucleus is the sum of the elastic scattering cross section and the non-elastic cross section. Elastic scattering may occur with or without the formation of a compound nucleus (shape elastic or compound elastic). It is the non-elastic interactions, inelastic neutron scattering, neutron absorption, and neutron reactions such as $(n,2n)$, (n,p) , and (n,α) , that are of primary interest to this work. Most non-elastic neutron interactions proceed through the formation of a compound nucleus. A brief discussion of the nuclear models that best describe fast-neutron reactions is appropriate before an empirical description of reaction systematics.

In efforts to predict neutron cross sections, the optical model is used to calculate the cross section for compound-nucleus formation. Transmission coefficients, needed for cross-section calculations, are

obtained from the numerical solution of the Schrodinger equation using an optical model potential. Such potentials all feature real and imaginary parts as well as a spin-orbit term.⁽¹⁾ The exact form of the potential may vary, however, and usually global⁽²⁾ or local⁽³⁾ (for target elements of interest) parameters in the expression for the potential are adjusted to fit empirical data. Frequently, the compound-nucleus-formation cross section so calculated will underpredict measurements at 14-MeV because a small fraction of interactions occur through direct reactions; a hybrid model⁽⁴⁾ is needed to fully describe these cases.

De-excitation of the compound nucleus into the available channels is predicted by statistical theory for all but the lightest nuclei. The major assumption in the compound-nucleus model is that the decay of the compound nucleus is independent of the mode of formation (within the limitations imposed by conservation laws). Compound nuclei formed in fast-neutron reactions are in highly excited states where the density of states can be characterized by the Fermi gas model.⁽⁵⁾ Cross sections for the (n,n') , (n,γ) , $(n,2n)$, (n,p) , and (n,α) reactions are then calculated using Hauser-Feshbach cross-section theory.

Any further discussion of nuclear theory of fast-neutron reactions is beyond the scope of this work. Application of the above models to predict cross sections requires complicated computer codes and the fitting of nuclear-potential and level-density parameters to empirical data. It should be noted, however, optical model calculations of compound-nucleus-formation cross sections, followed by Hauser-Feshbach reaction cross-section calculations have been very

successful in predicting 14-MeV neutron cross sections and both the energy and angular distributions of emitted particles.^(5,6)

For most activation analysis applications, it is desirable to eliminate the need for accurate cross-section data. In order to plan activation experiments, however, and to correct for interferences in FNAA, 14-MeV neutron cross sections are required. For these reasons, a discussion of fast-neutron reaction systematics is needed. The reactions of most importance in the 14-MeV neutron irradiations included in the present work are the $(n,2n)$, (n,p) , and (n,α) reactions.

The most thorough compilation of 14-MeV neutron cross sections currently available is in the IAEA Handbook of Nuclear Activation Cross Sections, in which Bormann et al.⁽⁷⁾ recommend values based on hundreds of reported references. For many reactions, there is widespread disagreement among the measured cross sections, resulting in large uncertainties in recommended values, or no recommendation at all. Much of this disagreement is due to varied methods of cross-section measurements. Although most measurements are made using the activation method, variations in irradiation geometries (influencing neutron energy), flux monitoring, and counting procedures, are responsible for the spread in measured values. In general, more recent measurements using high-resolution semiconductor detectors^(8,9) are considered to be the most reliable, along with cross sections deduced from measurements of emitted particles.^(6,10) Details of an activation measurement of the $^{79}\text{Br}(n,2n)^{78}\text{Br}$ reaction cross section, performed in connection with laser fusion diagnostics, will be presented in Chapter V. Cross sections used throughout the present work will be

taken from Bormann et al. unless otherwise noted.

The general behavior of $(n,2n)$, (n,p) , and (n,α) reaction cross sections for 14-MeV neutrons is shown in Figure II-1.⁽¹¹⁾

Individual cross sections may vary greatly from the general trends sketched in Figure II-1, so the literature must always be consulted for a particular value. In general, fast-neutron cross sections are lower than thermal-neutron cross sections. For low-Z elements, the charged-particle-emission reactions dominate as $(n,2n)$ cross sections are small. As Z increases past 20, the $(n,2n)$ cross sections become much larger than the (n,p) and (n,α) cross sections, which drop to a few millibarns at high values of Z.

Very often, a fast-neutron cross section varies significantly between 13 and 15 MeV. Bormann et al. have tabulated cross sections at 14.1, 14.5, and 14.9 MeV so the correct value can be used depending upon irradiation conditions. In addition, excitation functions are plotted for those isotopes for which reliable data is available, in order to indicate the nature of the energy dependence. If a sufficient fraction of neutrons suffer energy loss in a sample, errors may occur in FNAA from varying activities because of changing cross sections.

The $(n,2n)$ reaction is always endoergic with a Q-value equal in magnitude to the neutron separation energy of the target nucleus, but of opposite sign. The incident neutron must have a center of mass energy greater than or equal to $-Q$, and in the lab system

$$E_{th} = -\frac{A+1}{A} Q \quad (II-1)$$

For many low-Z nuclei, $E_{th} > 14$ MeV and the $(n,2n)$ reaction cannot

Figure II-1. Fast-Neutron Cross Section Trends for the $(n, 2n)$, (n, p) , and (n, α) Reactions. Individual values may vary greatly from the curves depicted.

occur. Qaim⁽⁹⁾ and Lu and Fink⁽¹²⁾ describe (n,2n)-reaction systematics in terms of the asymmetry parameter (N-Z)/A. For asymmetry parameter between zero and 0.1, there is a rapid increase in (n,2n) cross sections, whereas for (N-Z)/A greater than 0.1, the cross sections rise very slowly from about 1 to 2 barns. Lu and Fink⁽¹²⁾ give an empirical equation for quick estimation of (n,2n) cross sections for Z=28 to Z=82:

$$\sigma(n,2n) = 61.6(A^{1/3}+1)^2 \left[1 - 1.319 \exp\left[-8.744 \frac{(N-Z)}{A}\right] \right] \text{ mb} \quad (\text{II-2})$$

Except for a few cases, usually the lightest of several stable isotopes of an element, such as ⁶⁴Zn or ⁹²Mo, Eq. II-2 reproduces (n,2n) cross section measurements within 20%.

Reactions resulting in charged-particle emission may be exoergic or endoergic. The apparent threshold energy for these reactions is not given by Eq. II-1, however, because of the Coulomb barrier, B, given by

$$B = \frac{zZe^2}{R} \quad (\text{II-3})$$

where z and Z are the proton number of the charged particle and the residual nucleus, respectively, and R is the radius at which the nuclear force becomes negligible. As the Coulomb barrier increases in heavier nuclei, the cross sections for charged-particle emission decrease. In general, (n,p) cross sections are larger than (n, α) cross sections for the same reason. The systematics of (n,p) cross sections are discussed by Lu and Fink⁽¹²⁾ and more recently by Molla and Qaim.⁽¹³⁾ There is little correlation in the cross sections with Z or (N-Z)/A for

low- Z elements. For isotopes with asymmetry parameter greater than about 0.5, there is a steady, exponential decrease in (n,p) cross sections with increasing $(N-Z)/A$. An empirical relationship proposed by Levkovskii⁽¹⁴⁾ agrees with measured values within 20% for $Z > 20$:

$$\sigma(n,p) = 45.2(A^{1/3}+1)^2 \exp \{-33(N-Z)/A\} \text{ mb} \quad (\text{II-4})$$

As with $(n,2n)$ reactions, exceptions to Eq. II-4 are usually lightest stable isotopes of elements with several isotopes. These cases have unusually large (n,p) cross sections due to a proton separation energy that is relatively lower than the other stable isotopes in the region.⁽¹³⁾ Systematics for (n,α) reactions follow the (n,p) cross sections closely, but with lower values.

Equations II-2 and II-4, and Fig. II-1 are presented only to represent gross behavior in fast-neutron cross sections, and are no substitute for measured values whenever they can be obtained. It is best to consult several literature values of a cross section, or a thorough compilation because of the widely separated values reported for many cross sections.

B. Radioisotope Production by 14-MeV Neutron Irradiation

Throughout this work, samples were irradiated with fast neutrons to produce radioactive nuclei for gamma-ray spectroscopy subsequent to irradiations. If n_i target nuclei are placed in a neutron flux, ϕ , to produce an activation product with a decay constant, λ , the rate of production of the radioisotope is

$$\frac{dN(t)}{dt} = \phi \sigma n_i - \lambda N(t) \quad (\text{II-5})$$

where, $N(t)$ = the number of radioisotopes at any time, t ,

σ = the cross section for the reaction (cm^2), and

ϕ = the neutron flux ($\text{n/cm}^2\text{-s}$).

The solution of Eq. II-5 for the number of radioisotopes produced in an irradiation of length, t_i , is

$$N(t_i) = \frac{\phi \sigma n_i}{\lambda} (1 - e^{-\lambda t_i}) \quad (\text{II-6})$$

The expression in parenthesis is called the saturation factor which approaches unity for irradiations much longer than the half life, corresponding to the case of an equilibrium in the production and the decay of the activation product. The activity produced, in units of disintegrations per unit time, is λN .

For the purpose of gamma-ray spectroscopy, the measure of radioisotope production is the count rate in the full-energy gamma-ray peak. The gamma-ray count rate, $A(t)$, at a time t after an irradiation of an element i is

$$A(t) = b \epsilon \phi \sigma \frac{m_i f L_o}{M_i} (1 - e^{-\lambda t_i}) e^{-\lambda t} \quad (\text{II-7})$$

where, b = gamma-ray branching ratio in the activation product decay,

ϵ = detector efficiency,

m_i = mass of element i ,

f = isotopic abundance of the target nucleus,

L_0 = Avogadro's Number, and

M_i = Atomic weight of element i .

The detector efficiency is the product of the absolute, intrinsic efficiency of the detector for a given gamma-ray energy and the geometrical factor. The fast-neutron flux is the average flux at a particular energy, such as 14.9 MeV (the average neutron energy for most of the irradiations in the present work) and cross sections should be the correct value at that energy.

A sample may be irradiated and counted repeatedly to build a cumulative spectrum. The number of counts, D_1 , collected in the first counting interval of t_c is obtained by integrating Eq. II-7 over the counting interval:

$$D_1 = b\epsilon\phi\sigma \frac{m_i f L_0}{\lambda M_i} (1 - e^{-\lambda t_i}) e^{-\lambda t} (1 - e^{-\lambda t_c}) \quad (\text{II-8})$$

Following notation of Givens et al.⁽¹⁵⁾, the cumulative detector response, D_{tot} , for n cycles of total cycle period $T > t_i + t_c + t$, is given by

$$D_{\text{tot}} = D_1 \left\{ \frac{n}{(1 - e^{-\lambda T})} - \frac{e^{-\lambda T} (1 - e^{-n\lambda T})}{(1 - e^{-\lambda T})^2} \right\} \quad (\text{II-9})$$

From Eq. II-8, the optimum irradiation and counting times would be

$t_i = t_c = (1/2)T$, i.e. no decay time. In reality, such factors as additional activities in the sample and cost efficiency may dictate the irradiation parameters.

C. Fast-Neutron Attenuation

The flux of fast neutrons is reduced as neutrons interact with the sample, with the attenuation dependent upon sample composition and thickness. In FNAA, the magnitude of the attenuation may change from sample to sample resulting in errors in the analysis due to different irradiation conditions. For this reason, fast-neutron attenuation will be examined in order to develop corrections.

Assume the fast-neutron flux decreases exponentially with distance, d , in the sample:

$$\phi(d) = \phi_0 \exp [-\Sigma d] \quad (\text{II-10})$$

where, ϕ_0 = neutron flux with no sample present, and

Σ = macroscopic cross section.

Although fast-neutron cross sections are energy dependent, Eq. II-10 is found empirically to be true with a judicious choice of Σ . For a sample containing n elements, the total macroscopic cross section is

$$\Sigma_T = \frac{L_0}{V} \sum_{i=1}^n \frac{\sigma_{Ti}^m}{M_i} \quad (\text{II-11})$$

where, V = sample volume (cm^3), and

σ_T = microscopic total cross section.

The total macroscopic cross section does not, however, represent a judicious choice of Σ , because for $A > 10$, elastically scattered 14-MeV neutrons lose very little energy and are scattered through very small angles. (16)

A specific example at this point can illustrate the role of elastic scattering. From the neutron slowing-down equations in reactor theory, the number of elastic collisions, N , needed to slow a neutron of energy E_0 , to an energy below an effective threshold energy, E_{th} , is given by

$$N = \frac{1}{\xi} \ln \frac{E_0}{E_{th}} \quad (\text{II-12})$$

where ξ is the average logarithmic energy decrement, given by

$$\xi = 1 + \frac{(A-1)^2}{2A} \ln \frac{A-1}{A+1} \quad (\text{II-13})$$

Following the arguments of Nargolwalla and Przybylowicz⁽¹⁷⁾ (who, incidentally have incorrectly reported ξ for Al), one can calculate ξ and N for a threshold energy of 11 MeV, a value below which many fast-neutron reactions cannot occur. For Al and polyethylene (taken to be CH_2), the average logarithmic energy decrements are 0.007 and 0.58, respectively. In the case of Al, a major component of neutron generator target assemblies, an average of 33 elastic collisions is required to reduce a neutron's energy from 14 to 11 MeV. For polyethylene, the material used to construct the rabbits and sample capsules, only one collision is needed to drop the neutron energy below 11 MeV. Obviously, use of the total cross section to calculate the macroscopic cross section of Al would over predict attenuation. A fast-neutron removal cross section is needed.

The removal cross section, σ_R , is equal to the nonelastic cross section for all but the lightest of nuclei. Eqs. II-11 and II-10

are replaced by

$$\Sigma_R = \frac{L_o}{V} \sum_{i=1}^n \frac{\sigma_{Ri} m_i}{M_i} \quad (\text{II-14})$$

and

$$\phi(d) = \phi_o \exp [-\Sigma_R d] \quad (\text{II-15})$$

A table of microscopic removal cross sections for most elements appears in Nargolwalla and Przybylowicz, and the use of σ_R results in much better agreement with experiment than the use of total cross sections.

The macroscopic removal cross sections for Al and polyethylene are 0.060 and 0.070 cm^{-1} , respectively. Because one collision in polyethylene is sufficient to slow a neutron to less than 11 MeV, it may appear polyethylene is a poor choice for rabbits and sample capsules. However, the frequency of such collisions must also be considered. The removal mean free path, the reciprocal of σ_R , is the average distance traveled by a neutron before undergoing a reaction removing it from the fast-neutron group. For polyethylene, the removal mean free path is 14 cm, so there is negligible attenuation due to the 0.2 cm of polyethylene around the sample.

The flux given by Eq. II-15 can be integrated over the sample thickness to obtain the average flux through a sample,

$$\phi_{AV} = \frac{\phi_o}{\Sigma_R d} (1 - \exp [-\Sigma_R d]) \quad (\text{II-16})$$

In the event fast-neutron attenuation becomes a problem in FNAA, ϕ_{AV} must replace the value obtained from the flux monitor foil irradiated with the sample.

III. PRODUCTION OF 14-MeV NEUTRONS

The most common method used to produce 14-MeV neutrons is the reaction between deuterium and tritium nuclei,

In this chapter, the deuterium-tritium (DT) reaction will be examined and a description of a typical neutron generator will demonstrate how the reaction is used to produce neutrons. Finally, details of the high-flux facility at LLNL, the RTNS-I, are presented.

A. The DT Reaction

The highly exoergic DT reaction releases 17.86 MeV of kinetic energy to be partitioned between the neutron and the alpha particle, the neutron receiving an average of 14 MeV and the alpha particle the remainder. To produce neutrons, deuterons are generally accelerated into a target containing tritium. Enough energy must be supplied to the deuteron to overcome the Coulomb repulsion of the hydrogen nuclei. Although the height of the Coulomb barrier is about 500 keV, through barrier penetration, the onset of DT reactions occurs at a deuteron energy of only 30 keV. The cross section has been measured over a wide range of deuteron energies revealing a distinct maximum of 5 b at 107 keV.⁽¹⁸⁾ The relatively small accelerating voltage needed to overcome the Coulomb barrier allows the use of the DT reaction in small accelerators to produce 14-MeV neutrons.

The actual energy of the DT neutron is a function of the emission angle relative to the deuteron beam. Benveniste and

Zenger⁽¹⁹⁾ have presented the equations for the kinetics of the DT reaction and calculated the energy of a neutron emitted at any angle. Fowler and Bromley⁽²⁰⁾ have compiled tables of neutron energy versus angle, in the relativistic case, for deuteron energies ranging from 100 keV to 25 MeV. A portion of their results is plotted in Fig. III-1. At 400 keV, for example, the neutron energy at 0° is 15.6 MeV and drops to 12.9 MeV at 180° .

Three factors tend to degrade the neutron spectrum from monochromatic 15.6-MeV neutrons one expects from Fig. III-1. First, deuterons may slow considerably in the target before inducing a DT reaction.⁽¹⁹⁾ In this case, the energy of a neutron emitted at 0° would be much less than 15.6 MeV. The second factor is that the materials used in the target assembly will act to moderate the neutron energy. Often several millimeters of water and Al are between the source and the irradiation position, resulting in elastic scattering as discussed in the previous chapter. Finally, the geometry of the irradiation influences the neutron energy spectrum seen by the sample. A large solid angle is subtended by a sample close to the target encompassing a wide range of neutron energies, lowering the average neutron energy of the irradiation seen by the sample. Taking these effects into consideration, the actual neutron energy spectrum is difficult to predict, depending not only on the facility, but the irradiation configuration. There have been two notable calculations of neutron energy spectra from different types of sources. Ricci⁽²¹⁾ performed calculations for a typical target assembly of a small neutron generator with a 150-keV accelerating potential. Calculations were also

Figure III-1. Neutron Energy vs. Angle of Emission for Deuteron Energies of 100-500 keV.

made in 1957 for a 600-keV accelerator then in operation at the Los Alamos Scientific Laboratory by Seagrave et al.⁽²²⁾, for several angles. Their results for 0° and 90° are shown in Fig. III-2.

The sharply defined peak at 90° underscores the deuteron energy loss in the target prior to the DT reaction. It is seen from Fig. III-1, the neutron energy near 90° is independent of deuteron energy, unlike the 0° case.

There is a small anisotropy in the neutron distribution from the DT reaction. The yield is somewhat forward peaked, so that the flux at 180° is only 86% of the flux at 0° .

B. Neutron Generators

The DT reaction came into wide use with the introduction of a relatively inexpensive neutron generator originally marketed by Texas Nuclear Corporation. The Nuclear Engineering Program at the University of Illinois has two such neutron generators. Although these are now older units, the principles of operation are the same as newer sources.

The principal components of a neutron generator are an ion source, an accelerating potential, a tritium target, and an adequate high vacuum system. These parts will be described briefly as an explanation of how a neutron generator works. Prud'homme⁽²⁴⁾ has provided a detailed description of these components of a Texas Nuclear neutron generator.

Three types of ion sources are employed on neutron generators: the radiofrequency (RF) ion source, the Penning ion source, and the duoplasmatron ion source. Each has its own advantages and

Figure III-2. DT-Neutron Energy Spectrum at 0 and 90 Degrees for a Deuteron Energy of 600 keV.

disadvantages⁽¹⁷⁾ and a source is chosen based upon its compatibility with other accelerator components. An RF source is most likely to be used on a 150-keV accelerator because it is most efficient at producing deuterons; the atomic-to-molecular deuterium ratio is about 9 for an RF source. (Both machines at the University of Illinois have RF sources.) When D_2 -ions are accelerated through 150 kV, each deuteron receives only 75 keV, which is below the maximum cross section for the DT reaction.

An RF source is a glass bottle encircled by two metal rings connected to a 30-100 MHz oscillator. The radiofrequency radiation is absorbed by the D_2 causing dissociation and ionization. A characteristic bright pink plasma is thus produced in the bottle. A positive extraction voltage of 5-10 kV is applied to the closed end of the bottle to push the deuterons out into the accelerating section, and a -30 kV lens voltage is applied to attract and focus the beam.

A Penning ion source (PIG, after the Penning ion vacuum gauge) uses two cold cathodes and a high, DC potential anode to induce ionization.⁽¹⁷⁾ The source chamber is in a magnetic field so electrons accelerated from the cathodes to the anode travel long, circular routes, creating an intense plasma. A PIG source is capable of higher beam currents than an RF source, and the PIG source has a higher efficiency for D_2 usage. The atomic-to-molecular ratio of the PIG source is only 0.4, however, and they are most likely to be used on 300-400 kV accelerators.

The duoplasmatron (DP) source is capable of higher beam currents than the PIG source. For example, the DP source on the RTNS-I supplies

25-30 mA of D^+ . A hot filament supplies electrons, which spiraling through a magnetic field create a dense plasma. The beam is extracted through a series of precision anodes. The DP source is more complicated in construction than RF or PIG sources, and its proper operation requires careful attention to exit canal alignment. Usually DP sources appear only on very elaborate accelerators with high beam currents.

There are four types of high-voltage (HV) power supplies currently being used on neutron generators: the Cockcroft-Walton HV supply, the Insulating Core Transformer (ICT), the Van de Graaff electrostatic belt generator, and the electrostatic rotor machines built by Tunizi-Sames in France.⁽¹⁶⁾ Discussion of the ICT will be deferred until the next section because it is a central feature of the RTNS-I. Van de Graaff generators are usually used for potentials considerably larger than 150-400 kV, and electrostatic rotor machines are generally not found in the United States, so these two HV supplies will not be described here.

Most neutron generators have Cockcroft-Walton supplies, including those at the University of Illinois. A 150-kV Cockcroft-Walton supply features a transformer to step up the line voltage to 75 kV AC, which is then input to a rectifying, voltage-multiplier circuit producing the 150 kV DC accelerating potential. The accelerating tube, about 50 cm long, separates the ion-source components from the bulk of the neutron generator at electrical ground.

The ions coast down a drift tube after being accelerated until they strike the tritium target. A Cu disk is used as the target with

tritium adsorbed onto a thin Ti or Zr layer. Copper is needed to conduct heat from the 150 W of beam power (1 mA at 150 kV) to the water coolant on the backside of the target. Because a hydride is not formed with every metal atom, a loading of about 1:1 of tritium to Ti or Zr is typical. The 1-in diameter targets used at the University of Illinois are loaded with 5 Ci of tritium. Loss of tritium is due primarily to off-gassing rather than DT reactions, so nearly all of the tritium is pumped away by the vacuum system.

Like any accelerator, a neutron generator must have a good vacuum, less than 10^{-6} torr, for successful operation. Nonionized deuterium and off-gassed tritium must be removed during operation or the vacuum will quickly deteriorate. Diffusion pumps were used in early neutron generators, but because oil vapors can coat surfaces inside the accelerator, and there is no adequate way of reducing the tritium hazard, diffusion pumps have been replaced by getter-ion pumps. Ion pumps are very efficient at hydrogen pumping, and, tritium atoms are buried permanently in the Ti cathode of the pump, as are the noble gases. One of the University of Illinois neutron generators has a large, 200 l/s ion pump, whereas the other has a Ti-sublimation pump and a small ion pump to remove inert gases.⁽²⁵⁾ The sublimation pump deposits very thin layers of Ti on the walls of its chamber, allowing the formation of hydrides with the deuterium and tritium in the system.

A neutron generator, similar to those described above, may produce up to 10^{10} n/s, and a maximum flux of about 10^9 n/cm²-s. Advances in accelerator technology have increased these yields by more than a factor of ten. In addition, there has been a

dramatic improvement in sealed-tube 14-MeV neutron sources; Fanger et al.⁽²⁶⁾ are testing a sealed-tube source with a cylindrical acceleration structure capable of producing 5×10^{12} n/s, with a maximum flux of 5×10^{10} n/cm²-s. This system is very expensive, however, and the tubes have only 40-80 h lifetimes.

C. The Rotating Target Neutron Source, RTNS-I

The RTNS-I was installed at LLNL in 1966 for physics and radiochemistry research. It is generally referred to as the ICT because the high voltage is supplied by an Insulating Core Transformer purchased from High Voltage Engineering Corporation. Both RF and duoplasmatron ion sources are available; the RF source allows nanosecond pulsing but has a much smaller steady-state output than the DP source. Ionized deuterium atoms and molecules are segregated magnetically near the source so that only one component, usually D^+ , is accelerated.

Because of the very high power requirements (10 kW), an ICT HV supply was chosen for the RTNS-I. The ICT generates 300-500 kV by incorporating a strong magnetic flux through a central iron core which is segmented by thin plastic sheets of great dielectric strength. Each segment is electrically insulated from the next and a rectifying circuit at every segment produces about 25 kV DC.^(27,28) The primary windings producing the magnetic flux are on the outside (ground) of the supply, and have a frequency of about 1000 Hz. The number of segments determines the voltage. Several atmospheres of SF_6 insulate the core components from the ICT tank enclosure. The ICT is able to produce a very steady current and a HV with only 3% ripple.

In 1971, the RTNS-I was modified to produce higher yields by upgrading the ICT HV supply, and making several additional alterations.⁽²⁹⁾ Table III-1 is a list of the output specifications of the ICT and the neutron source. It should be noted, as of this writing, the 1971 HV supply is out of service and the original supply is being used. The facility is located in two rooms adjacent to Building 212, which houses the control room. The ICT, ion-source components, accelerator, and a number of vacuum pumps and focusing magnets are in the machine room, pictured in Fig. III-3. Neutrons are produced in the target pit, a large room with ample space for experimental equipment, containing more pumps and focusing magnets, and the rotating target. The beam line follows a diagonal path toward the floor of the target pit so the target is below ground level.

A sketch of the target assembly is shown in Fig. III-4. Note the location of the bearing and seal. The target and its in-vacuum support structure (but not the water catch cage or coolant plumbing) rotate on this seal designed by Rex Booth.⁽³⁰⁾ The end of the assembly, including the water catch cage, moves up and down about the pivot point allowing the beam to strike another band of the target. In 1980, a new "sandwich" target was tested featuring an intricate network of coolant channels etched inside a two-layer target. No catch cage was required so one could see the target turning and experimental apparatus could be moved a millimeter closer to the beam spot.

Targets have been continually enlarged and improved.^(29,31)

Presently they are constructed of a 0.15%-Zr alloy of Cu, shaped into a section of a sphere. Tritium is adsorbed onto a 7.5-cm band of Ti at

Table III-1. RTNS-I Source Parameters

Deuteron energy	400 KeV
HV supply current	60 mA [*]
Target current	25 mA [*]
Beam spot diameter	0.6 cm
Target diameter	23 cm
Target rotation	1100 rpm
Source strength	6×10^{12} n/s [*]
Maximum flux	1.7×10^{12} n/cm ² -s [*]
Target lifetime	100 h

* In 1980-81, the RTNS-I operated at about 70% of these values because the ICT HV supply was damaged.

Figure III-3. RTNS-I Machine Room.

Figure III-4. Rotating Tritium Target Assembly at the RTNS-I.

the Isotopes Division of the Oak Ridge National Laboratory. About 1.7 tritium atoms are adsorbed per Ti atom, resulting in a total of 2000 Ci per target. When all the available bands on the target have dropped below 70% of their initial output, the target is replaced. Target lifetime is about 100 h.

The output characteristics of the RTNS-I have been studied in great detail. As already mentioned, deuterium ions may lose much of their energy in the target before initiating a DT reaction. The most probable deuteron energy for a DT reaction at the RTNS-I has been determined by time-of-flight neutron spectra to be about 200 keV, when the accelerating potential is 400 kV.⁽³²⁾ The RTNS-I has a neutron energy spectrum at 0° similar to that in Fig. III-2, but with a maximum energy of 15.6 MeV.

Of more immediate concern in activation analysis is the spatial distribution of the neutron flux. In 1972, Goldberg et al.⁽³³⁾ confirmed flux measurements agreed with values predicted by computations. Neutron flux varies with distance along the beam axis according to

$$\phi = \frac{Y}{4\pi a^2} \ln [1 + (a/Z)^2] \quad (\text{III-1})$$

where, Y = total neutron yield (n/s),

a = the radius of the beam spot, and

Z = the distance from the target.

The flux at any point a distance r away from the axis is

$$\phi = \frac{Y}{4\pi a^2} \ln \left\{ \frac{\sqrt{(a^2 + Z^2 - r^2)^2 + 4Z^2 r^2} + a^2 + Z^2 - r^2}{2Z^2} \right\} \quad (\text{III-2})$$

Figures III-5 and III-6 are plots of the flux versus Z and r, respectively, for $Y = 2.7 \times 10^{12} \text{ n/s}$ and $a = 0.3 \text{ cm}$, the beam conditions of most of the irradiations in the present work (after similar plots by Van Konynenburg⁽³⁴⁾). Step flux gradients may contribute to large errors in FNAA. As seen in Fig. III-5, small variations in the distance between the beam spot and the samples have a tremendous influence upon the neutron flux through the sample.

Figure III-5. Neutron Flux at Points along the Beam Axis vs. Distance from the Target.

Figure III-6. Neutron Flux at Points off the Beam Axis 0.6 and 1.1 cm from the Target.

IV. ACTIVATION ANALYSIS

In the previous chapters, characteristics of fast-neutron reactions and sources have been presented. Activation analysis using 14-MeV neutrons differs from thermal-neutron activation analysis primarily in the reactions that are possible and the irradiation conditions inherent in fast-neutron sources. Procedures developed for FNAA have taken these differences into account.

To place multielement FNAA in the context of the entire field of activation analysis, background information on the status of activation analysis and the methods employed is described below. After a discussion of the limitations and errors in activation analysis, the procedures for high-flux, multielement FNAA developed in the present work are presented. Finally, results of the analyses of standard reference materials are given as a demonstration of the successful application of these multielement FNAA procedures.

A. Current Status of Neutron Activation Analysis

Instrumental neutron activation analysis (INAA) has emerged in the last 15 years as one of the most accurate methods for multielement quantitative analysis. A summary of the development and applications of INAA with thermal neutrons is given in the following section. Fast-neutron activation analysis (FNAA), also used extensively in this period for single-element determinations, will be discussed in the succeeding section.

1. Thermal-Neutron Activation Analysis

Although NAA is nearly 50 years old, two technological developments were required before the method could achieve its present status. The introduction of nuclear reactors as common research tools allowed the production of sufficient activity to perform multielement analyses. Until the development of high-resolution, semiconductor detectors, however, it was necessary to chemically separate the activities of interest before counting. Utilizing Ge(Li) detectors, multichannel analyzers, and computer analysis, thermal-neutron activation analysis (TNAA) became an instrumental technique. As the term instrumental implies, chemical treatment of samples was no longer required. In the broadest sense, one needed only to prepare and package samples, irradiate them, and collect and analyze gamma-ray spectra to determine concentrations of more than 50 elements in the sample. Several excellent books describe the procedures necessary to minimize sources of error while obtaining the most sensitive and accurate results.^(16,35,36)

The profound impact the Ge(Li) detector would make in activation analysis was forecast by Heath⁽³⁷⁾ at the 1968 Conference on Modern Trends in Activation Analysis. In the following year, TNAA procedures were developed for applications ranging from air pollution studies by Gordon et al.⁽³⁸⁾ and later by Hopke et al.⁽³⁹⁾, to analysis of biological materials by Nadkarni and Ehmann.⁽⁴⁰⁾ The works cited above are typical of the period as many laboratories acquired the capacity for determinations of up to 30 elements with two or three irradiations, and two to four counts per sample. The particulars of the

irradiation-counting protocol varied according to the application; the literature should be consulted for details. In the last decade, much larger, and therefore, more efficient Ge(Li) detectors allowed further improvements in accuracy and sensitivity. There has been a tremendous increase in the number of TNAA applications in environmental and energy-related research, such as emissions from coal-fired power plants.^(41,42) In addition, numerous applications of TNAA in chemistry, biology, medicine, and geology, etc. have been reported. The Proceedings of the 1976 Conference on Modern Trends in Activation Analysis⁽⁴³⁾, for example, contains papers concerning trace elements in drinking water, atmospheric aerosol, sewage sludge, human hair, and many other samples.

Standard reference materials (SRM) have been very important in establishing the accuracy and reliability of TNAA (and other analytical techniques as well). The United States National Bureau of Standards (NBS) has introduced the materials so laboratories are able to test their analysis procedures on well characterized samples. A SRM is carefully collected, homogenized, and analyzed by several techniques before NBS certifies the concentrations of a set of elements in the material. Many additional elemental concentrations have been reported in the literature for these materials, such as the round robin analyses of NBS fly ash (SRM 1633) and coal (SRM 1632) by Ondov et al.⁽⁴⁴⁾ and, more recently, of newer coal SRM's 1632a and 1635 by Germani et al.⁽⁴⁵⁾ Biological SRM's, such as orchard and tomato leaves, bovine liver, spinach, and pine needles are also available. Gladney⁽⁴⁶⁾ recently compiled a thorough review of all NBS

materials, in which he not only compiled mean values and uncertainties for elemental concentrations, but also lists every reported value from nearly 400 references. The United States Geological Survey and the International Atomic Energy Agency have also introduced SRM's and, although these materials are not certified, the compositions of all these materials have been the object of many studies. Analysis of an SRM may reveal errors in technique providing valuable feedback for the analyst and ultimately building confidence in new methods.

Although TNAA proved successful in a multitude of applications, it does have its limitations. Many other nuclear analytical methods have been developed emphasizing determinations of elements inaccessible by thermal-neutron activation. Epithermal⁽⁴⁷⁾ and fast neutrons, charged particles⁽⁴⁸⁾, and photons⁽⁴⁹⁾ have all been used to create activation products suitable for elemental analysis. X-ray emission spectra, whether excited by charged particles⁽⁵⁰⁾ or x-rays⁽⁵¹⁾, may be used for quantitative analysis, that can be competitive with TNAA in some applications. Spectra of prompt gamma rays emitted in radiative neutron-capture reactions have also been used for multielement analysis.⁽⁵²⁾ While none of these methods can usually duplicate the sensitivity or economy of TNAA for nearly as many elements, they often provide valuable complementary information. It is with this goal, the present work using high-flux, fast-neutron activation was initiated.

2. Fast-Neutron Activation Analysis

As sources of 14-MeV neutrons became available in the late

Fifties, radiochemists attempted the determination of elements impossible to analyze by TMAA, particularly O and Si. A very thorough book, Activation Analysis with Neutron Generators by Margolwalla and Przybylowicz⁽¹⁷⁾, serves as an excellent introduction and guide to the literature in this area. Because of the limited neutron fluxes from neutron generators, from 10^8 to 10^{10} n/cm²-s, analysis procedures were optimized for single-element determinations. The techniques required for O and Si determinations will be described here as they are representative of the status of FNAA at the start of the present work.

a. Oxygen. Much of the early work in FNAA was development of methods to determine O concentrations in various materials, particularly in steel. It is impossible to detect O by TMAA because ¹⁶O has a 99.8% isotopic abundance and ¹⁷O, its activation product, is stable. Because chemical measurements are very difficult, O content was often taken to be the difference from 100% of the sum of all the other major elements, a rather unreliable method subject to the cumulative errors of the other determinations.

The nuclear properties of ¹⁶O are favorable for analysis by 14-MeV neutron activation. The reaction, ¹⁶O(n,p)¹⁶N, can be used as long as the 7.2-s ¹⁶N activity can be counted quickly. Pneumatic sample-transfer systems were required to retrieve activated samples. Hoste et al.⁽⁵³⁾ developed a fast, reliable, and non-destructive method for O determinations in metals through automation of irradiations, sample transfer, and gamma-ray counting. Improvements in O analysis by FNAA have continued through the 1970's with development

of stronger neutron sources. Increasing sensitivities and more accurate treatment of problems due to neutron attenuation, interferences, and pulse pile-up effects in gamma-ray spectrometers have been presented by Beurton and Pillon.⁽⁵⁴⁾ FNAA determinations of O have also been made on geological samples and meteorites.⁽⁵⁵⁾ Finally, there has recently been considerable interest in the O content of various coals and coal derivatives for coal-combustion pollution studies.⁽⁵⁶⁾ The FNAA facility at the University of Illinois has been used by Raimondi⁽⁵⁷⁾ for O analysis of coal and fly ash.

All of the work cited above featured the same fundamental techniques of O analysis. Differences in the procedures are due to problems arising from the particular sample matrix in question or improvements in the analysis. FNAA is a very successful tool for O determinations because it is quick and inexpensive. It must be re-emphasized, however, this is a single-element technique requiring optimization for production and detection of ^{16}O at the expense of all other elements.

b. Silicon and other elements. FNAA can be used for Si determinations via the $^{28}\text{Si}(n,p)^{28}\text{Al}$ reaction. The procedure is similar to an O determination except longer irradiations and counts are needed for the 2-min ^{28}Al activity. Si analysis is impossible by TNAA so FNAA is widely used for geological and cosmological samples.⁽⁵⁸⁾ Although most applications are single-element determinations, Morgan et al.⁽⁵⁵⁾ increased the irradiation length, incorporated a Ge(Li) detector, and determined Mg, Al, and Fe as well.

Nargolwalla and Przybylowicz report FNAA determinations on 57 elements. Most of these applications are for very specific samples. A recent example was the determination of Ba in Barytes ores, in which the element of interest was the major constituent.⁽⁵⁹⁾

Positron-annihilation radiation has been used for the simultaneous determinations of N, F, and P in fossile bones, along with Ca using ^{44}K .⁽⁶⁰⁾ This four-element determination was possible because each element was a major constituent in a Ca matrix that caused no interference.

The flux limitations of neutron generators are chiefly responsible for the fact that FNAA has been used almost entirely for single, major element determinations. In the past three years, however, Segebade⁽⁶¹⁾ and Faanhof et al.⁽⁶²⁾ have performed multielement analysis with these low-flux fast-neutron sources and were able to determine 8-10 elements from among the major constituents. The maximum fast-neutron flux available from the RTNS-I is nearly 100 times the maximum flux produced by a typical neutron generator. It is the realization of this potential for trace multielement FNAA using the RTNS-I that is the primary goal of the present work.

B. Quantitative Analysis Methods

Two basic methods are employed in routine applications of NAA: absolute activation analysis and the comparison method. Descriptions of both methods will follow, with an emphasis on the latter because all of the quantitative analysis in this work employed the comparison method.

Absolute activation analysis is accomplished by direct solution of Eq. II-7, rewritten here with the elemental mass replaced by the product of its concentration, c_u , and the total sample mass, m_u , as follows:

$$A_u = b \epsilon \phi_u \sigma \frac{c_u m_u f L_o}{M} (1 - e^{-\lambda t_i}) e^{-\lambda t_u} \quad (\text{IV-1})$$

The subscript, u , is used to denote the unknown sample. In the case of absolute NAA, the concentration of an element can be calculated when all of the other parameters in Eq. IV-1 are known exactly, and the gamma-ray activity, A_u , is measured.

There are several conditions to be satisfied for successful use of the absolute method. The detector used for gamma-ray spectroscopy must be well calibrated so the absolute efficiencies are known for all gamma-ray energies and all counting geometries. In addition, there must be an accurate flux determination and heavy reliance upon tabulated nuclear data for gamma-ray branching ratios and cross sections. Alternatively, empirical data for induced gamma-ray activities following irradiation of all the elements of interest must be gathered prior to absolute NAA of unknown samples. The latter approach has been used by the Environmental Sciences Division at LLNL.^(63,64) In general, the absolute method is not widely used because of difficulties in meeting the above conditions.

The comparison method can eliminate the dependence upon several of the parameters difficult to ascertain. Standards of well known composition are irradiated and counted under exactly the same conditions

as the samples. Standards may be prepared for a specific application or may be SRM's. The activity from the standard in a particular photopeak, A_k , is given by Eq. IV-1 also, with the subscript, k, denoting "known":

$$A_k = b \epsilon \phi_k \sigma \frac{c_k m_k f L_o}{M} (1 - e^{-\lambda t_i}) e^{-\lambda t_k} \quad (\text{IV-2})$$

Dividing Eq. IV-1 by Eq. IV-2 and solving for the unknown concentration yields

$$c_u = c_k \frac{A_u}{A_k} \frac{m_k}{m_u} \frac{\phi_k}{\phi_u} \frac{e^{-\lambda t_k}}{e^{-\lambda t_u}} \quad (\text{IV-3})$$

Thus, the unknown concentration can be measured without knowledge of ϵ , b , σ , and t_i (and usually ϕ as well, in the case of TNAA). The comparison method is preferred at most laboratories; all of the NAA literature cited in Section A makes use of standards in the analysis of samples. Flux monitor foils are used when there may be flux variations, allowing determination of the ratio of the fluxes. The absolute value of the flux is not needed to solve Eq. IV-3. Because deviations from exactly the same irradiation and counting conditions for samples and standards may introduce many subtle errors, a detailed discussion of these conditions will be postponed until Section D.

C. Limitations of Activation Analysis

The ability to determine the concentration of an element by NAA, regardless of neutron energy, may be limited by three factors:

unfavorable nuclear properties of the activation product, interferences from other elements, and the sensitivity of NAA for the determination of the element. These factors will be explored in this section, particularly as they apply to FNAA.

Any one of a number of nuclear properties may make analysis of an element difficult or impossible:

1. Small cross sections for the production of useful activation products make some determinations difficult.
2. Reaction products from the most abundant target isotopes may be stable. TNAA is more susceptible to this problem because neutron capture is the only possible reaction. (In fact, exactly this situation exists for O and Si and led to the early applications of FNAA.) Stable products result from fast-neutron reactions also, and limit the options for analysis.
3. Many activation products decay directly to the ground state of the daughter nuclide without emitting any gamma rays, or the decay has very low gamma-ray intensities. (This is the case for TNAA of P for example, and is the reason FNAA is sometimes used for P determinations.)
4. Extremely short- or long-lived activation products are difficult to measure.

Because 2 or 3 activation products are usually available, FNAA is less affected by the above conditions.

The flexibility afforded FNAA by multiple reactions also leads to many serious interferences. TNAA is less likely to have limitations from interferences, but they can occur in some cases because most reactors have a fast-neutron flux component. A primary interference occurs when a particular radionuclide used for the analysis of an element is also produced from a second element in a sample or standard. For example, ^{28}Al used for a P determination may be produced from Si

as well. Many other possibilities for interference exist in FNAA; the most complete list has been compiled by Krivan and Krivan.⁽⁶⁵⁾

There also are many types of secondary interference.^(17,35) Charged particles produced in fast-neutron reactions can themselves induce a reaction in low-Z elements, for example, the $^{12}\text{C}(p,\gamma)^{13}\text{N}$ reaction has interfered with N determinations. A primary activity may also be enhanced by the decay of a short-lived parent activation product of another element, as in the growth of ^{47}Sc used for Ti determinations through the decay of ^{47}Ca .⁽⁶⁶⁾ Two activation products may decay into the same daughter nucleus and therefore have the same gamma rays, such as ^{44}K and ^{44}Sc . Finally, instrumental interferences may occur when a detector cannot resolve gamma-ray peaks with nearly the same energy.

While it is clear secondary interferences sometimes cause problems, of much greater concern are the primary interferences. Using the subscripts p and i to denote primary and interference, respectively, Eq. IV-1 must have an additional term for the photopeak activity in the sample from a second element:

$$A_u = b\epsilon\phi_u\sigma_p \frac{c_{up}m_u^fL_o}{M_p} (1-e^{-\lambda t_i}) e^{-\lambda t_u} \\ + b\epsilon\phi_u\sigma_i \frac{c_{ui}m_u^fL_o}{M_i} (1-e^{-\lambda t_i}) e^{-\lambda t_u} \quad (\text{IV-4})$$

A similar expression is true for A_k , the activity in the standard, with the subscript, k, everywhere replacing u above. As in the development of Eq. IV-3, dividing Eq. IV-4 by its analogous expression

for A_k and solving for the concentration of the primary element yields

$$c_{up} = c_{kp} \left\{ 1 + I_{ip} \frac{c_{ik}}{c_{kp}} \right\} \frac{A_{uk} \phi_k e^{-\lambda t_k}}{A_{ku} \phi_u e^{-\lambda t_u}} - I_{ip} c_{ui} \quad (IV-5)$$

where I_{ip} is the ratio of primary activity to interfering activity produced in an irradiation of equal masses of both:

$$I_{ip} = \frac{\sigma_i f_i M_p}{\sigma_p f_p M_i} \quad (IV-6)$$

By comparing Eq. IV-5 to Eq. IV-3, several points are immediately obvious:

1. The concentration of the interfering element in the sample, c_{ui} , must be determined (using a different activation product) in order to calculate c_{up} .
2. Because reaction cross sections appear explicitly in Eq. IV-5, there are often large uncertainties in I_{ip} , as seen in Table IV-4.
3. The expression in brackets in Eq. IV-5 can be much larger than unity depending on I_{ip} and the concentrations. This case occurs when the majority of the standard activity is from the interfering element.
4. Since c_{up} is a difference of numbers which may have large uncertainties, it is possible to have extremely large errors in c_{up} , and the interference will simply make the determination impossible. It is, therefore, very unlikely a determination could be made for the case of two sources of interference.

In spite of the possible problems outlined above, Eq. IV-5 is a relatively simple correction for an interference and can successfully be used in all but extreme cases.

Interferences should be avoided whenever possible. Frequently in FNAA, an alternative activation product can be used for a determination. It is sometimes possible to omit an interfering element from standards, simplifying Eq. IV-5. In many cases, however, it is simply impossible to omit the interfering elements without forfeiting an elemental determination. Finally, Eq. IV-5 does not apply to the case of an FNAA determination with an interference from thermal-neutron absorption. A correction for this case can easily be obtained in the same manner but is not presented because the thermal-neutron flux in the present work was shown to be negligible.⁽⁶⁷⁾

The third limitation mentioned at the start of this section is the sensitivity of NAA for a particular element. The sensitivity is not only a function of the nuclear properties of the element, but depends on the specifics of the analysis and the sample matrix. An elemental concentration cannot be determined if gamma-ray activity from the activation product is too low with respect to the background. A detection limit can be obtained, however, if the gamma ray is observed in the standard spectrum. Many criteria are used to set detection limits⁽³⁵⁾ but the condition most often used is

$$A_u < 2\sigma_B \quad (IV-7)$$

where, A_u is the maximum activity in the sample spectrum that would not be observed, and σ_B is the standard deviation of the background over that region in the sample spectrum containing the peak in the standard spectrum. A_u is then inserted into Eq. IV-3, now an inequality, and c_u is the detection limit. The background standard

deviation is the square root of the sum of the counts in the channels of the region of interest.

The sensitivity, expressed as a mass, is the minimum amount of an element that can be detected by a particular method. Many examples of sensitivities for TNAA and FNAA have been reported in the literature, but one should interpret these values with caution because assumptions are always made in the calculations. Different sample types may have radically different sensitivities for the same element under identical analysis procedures.

D. Errors in Activation Analysis

It has already been stated changes in irradiation or counting conditions of the samples and standards can introduce errors in the comparison method of NAA. Sources of errors arising from irradiation and counting conditions will be presented in this section, along with miscellaneous sources of error, and a discussion of flux monitoring. Much of what follows is applicable to the absolute method of NAA as well as the comparison method. Errors particularly likely to occur in FNAA will be emphasized.

1. Irradiation-Related Errors

Variations in neutron flux need not lead to errors in activation analysis if flux monitors can be used to accurately represent the neutron fluence through the sample. There are circumstances where this is not the case, however. For example, it follows from the discussion in Chapter II, that variable neutron attenuation in samples and standards may lead to different average fluxes, whereas, flux-monitor

foils placed between the source and the samples would indicate identical fluxes. This particular error, having been a problem in low-flux FNAA of large samples⁽¹⁷⁾, can be corrected using the expression for the average flux (Eq. II-16) rather than the flux calculated directly from the foil activity.

Neutron attenuation is related to another subtle source of error in FNAA. As mentioned previously, many fast-neutron cross sections vary appreciably between 13 and 15 MeV. Inelastic scattering may degrade the neutron energy in the sample, without necessarily lowering the neutron energy below 13 MeV. Since the cross sections vary, however, the induced activity may vary between samples and standards if the average number of elastic collisions varies. This source of error should only affect those elements with excitation functions varying sharply with energy, and would be difficult to quantify because of the large uncertainties in cross-section data.

A major source of error is the existence of spatial flux variations over the irradiation location. While flux gradients may exist in nuclear reactors, they are rarely as pronounced as those in the vicinity of a 14-MeV neutron source (see Figs. III-5 and III-6). The existence of flux gradients is not intrinsically a source of error, but imposes the condition that samples and standards must be homogeneous. If an element is unevenly distributed through the sample or standard, the activity produced from that element will depend upon its location in the non-uniform flux. Thus, inhomogeneous standards or samples can cause large errors in FNAA, for which there is no correction. Finally, in a non-uniform neutron flux, the irradiation position must be

accurately reproducible.

2. Flux Monitoring

Variations in neutron source strength due to beam and target conditions necessitate accurate neutron flux monitors. Monitors currently in use fall into three major categories: neutron detectors, associated-particle detectors, and post-irradiation counting of activation foils. A review article by Iddings⁽⁶⁸⁾ presents many details on these monitors and also mentions a fourth (and the least reliable) method, ^{16}N monitoring of target cooling water.

Proportional counters filled with BF_3 or ^3He , and organic and inorganic scintillators are neutron detectors used for flux monitoring. If the output of a neutron detector is sent to a multichannel analyzer in the multiscale mode, the time dependence of the neutron source is also monitored.⁽⁵⁵⁾ Use of a neutron detector as an absolute flux monitor requires at least one measurement of the flux value (generally by foil activation) as a function of the monitor detector output.

Associated-particle detectors exploit the alpha particles emitted during the DT reaction to monitor the neutron source. Either surface barrier detectors or plastic scintillators may be used, but both must be built into the neutron generator (under vacuum) to count alpha particles. An advantage of a surface barrier detector is the ability to detect ^3He and protons, thus simultaneously monitoring DT and DD neutrons as the tritium is depleted.⁽⁶⁹⁾ Neither neutron detectors nor associated-particle detectors will yield any information regarding

neutron attenuation in the sample or spatial variations in the flux due to changing beam conditions.

Thin metal foils irradiated in intimate contact with samples provide the best monitor of integrated neutron flux received by the sample. By placing foils on both sides of the sample, the effects of flux gradients may be properly treated by taking the average activity of the two foils. Foils provide no information about the time dependence of the source strength, however, but associated-particle or neutron detectors may be used along with foils if the need arises. Several metals such as Al, Co, Cu, and Nb have been used; the final choice depends primarily upon irradiation length and counting convenience. For single element determinations, a flux-monitor reaction with a fast-neutron threshold energy similar to the reaction of interest is desirable. The so-called "Texas Convention" of 14-MeV neutron flux monitoring⁽⁷⁰⁾ used the $^{63}\text{Cu}(n,2n)^{62}\text{Cu}$ reaction because it has a threshold energy of 12 MeV, most similar to the 10-MeV threshold for the production of ^{16}N from oxygen.

3. Sources of Error in Gamma-Ray Spectroscopy

While irradiation-related errors are much more prevalent in FNAA than in TNAA, errors resulting from counting techniques can occur in any activation analysis application.

Perhaps the most serious source of error using the comparison method is counting geometry. Samples and standards must be counted in exactly the same positions relative to the detector. Good geometry is particularly important within a few centimeters of a Ge(Li) detector

because the geometrical efficiency factor⁽⁷¹⁾ will vary significantly over a few millimeters, and because corrections may be difficult as the detector volume is hidden inside the vacuum enclosure.

Errors may arise from the spectroscopy system itself. Electronic components, such as the amplifier and the analog-to-digital converter (ADC), must be properly adjusted to yield the best possible resolution and peak shape. The system should be stable to avoid drifting of the amplifier gain during a counting period. A description of gamma-ray spectroscopy procedures will be presented in Section E.

A source of error related to the spectroscopy system is the correction required for dead time and pulse pile-up. The time taken by the ADC to process a signal is generally several microseconds⁽⁷²⁾, which is longer than the charge-collection time of a Ge(Li) detector (about 100 ns) and, therefore, the major source of the system dead time. Any signal from the detector during the period the ADC is busy is lost. Signals contributing to a gamma-ray photopeak may also be lost via pulse pile-up. Peak pile-up occurs when two pulses are so close together that the system components treat the pulses as one. Most multichannel analyzers count live time as well as real time in order to correct for counts lost during system dead time. Errors may arise in using MCA live-time corrections in cases of short counts of rapidly decaying activities.⁽⁷³⁾ The addition of a fixed-frequency pulser to the preamplifier signal resulting in a pulser peak in the spectrum provides a more accurate live-time correction and a pile-up correction as well. The correction is made with the assumption that the ratio of true counts to measured photopeak area is equal to the ratio of the known number of

pulses to the pulser-peak area. A long-lived gamma-ray source may be used for the purpose, providing the source does not interfere with the gamma rays of interest.

Finally, errors in NAA can result from gamma-ray attenuation in samples and standards. Gamma-ray attenuation increases with sample size and, therefore, has been a problem in FNAA in the past. An average gamma-ray intensity, I_{AV} , from a sample of thickness, d , can be calculated in a manner analogous to the neutron attenuation case in Chapter II

$$I_{AV} = \frac{I_0}{\mu d} (1 - \exp [-\mu d]) \quad (IV-9)$$

where, I_0 = gamma-ray intensity with no attenuation, and

μ = the linear absorption coefficient.

The linear absorption coefficient of a sample is given in terms of tabulated⁽⁷⁴⁾ mass attenuation coefficients of its n major constituents

$$\mu = \rho \sum_{i=1}^n c_i (\mu/\rho)_i \quad (IV-10)$$

where, ρ = sample density, and

$(\mu/\rho)_i$ = mass attenuation coefficient of element i (cm^2/g).

Absorption coefficients are energy dependent so corrections must be calculated for each gamma ray. If the attenuation differs between the samples and standards, Eq. IV-9 can be used to calculate the true intensity from the measured intensity, which is I_{AV} . In general, gamma-ray attenuation is rarely a problem in NAA for photon energies greater than 100 keV.

4. Additional Sources of Error

Results of any analysis are meaningless if a sample is not representative of the material to be analyzed. Thus, samples for NAA must be carefully collected and handled so as to avoid the addition of contaminants or the loss of volatile elements. Directions are given for the use of standard reference materials to insure homogeneous samples. Moisture must be removed by desiccating, baking, or freeze-drying to the correct dry weight.

Samples are generally counted in the polyethylene vials or bags in which they have been irradiated. Packaging materials must be cleaned by leaching in acid and thoroughly rinsed with de-mineralized water, and must not be contaminated by touching or with unclean instruments. Radioisotopes may still be produced from trace elements in polyethylene and blank sample packages should be irradiated to determine the package contribution to the measured activity.

E. Procedures for Multielement FNAA

In order to develop and test FNAA procedures, three NBS standard reference materials were chosen as samples: fly ash (SRM 1633), orchard leaves (SRM 1571), and bovine liver (SRM 1577). Elemental concentrations of these materials are well known, several certified by NBS, and many more widely reported in the literature and most recently compiled by Gladney.⁽⁴⁶⁾ Procedures presented in this section evolved from many irradiations of these materials, first for qualitative information regarding the radioisotopes produced in 14-MeV neutron irradiations, followed by several quantitative analyses.

1. Sample and Standard Preparation

Standard reference materials must be used according to instructions given by the NBS Certificate of Analysis for the material. Although the materials are carefully collected and homogenized, a minimum sample size of 250 mg is recommended for any analysis to insure a homogeneous sample. Biological samples must be dried in air in a 85° oven for at least four hours before weighing and packaging. Samples varied from 250 mg to about 500 mg.

Standards were prepared by thoroughly mixing many dry chemical compounds, either oxides, salts, or metal powders. In the case of standards used to analyze the biological samples, powdered, ultra-pure graphite was added to dilute the mixture.⁽⁷⁵⁾ Concentrations of the major elements in the standards were approximately equal to the concentrations of the matrix elements in the samples so gamma-ray spectra from the samples and standards had the same major peaks. The constituents of the standards used for the most recent analysis of fly ash following short irradiations are listed in Table IV-1. Trace elements, Sc, Ni, Cu, Zn, Ga, Rb, Zr, Mo, Sb, Cs, and Pb, were mixed separately with graphite powder and then small amounts of the trace element mixture were added to the standards before final mixing. This was the only practical method to include trace amounts of material and insure homogeneity.

2. Irradiations

For most of the irradiations, the RTNS-I source strength was $2.6-2.9 \times 10^{12}$ n/s. A pneumatic sample-transfer (rabbit) system was

Table IV-1. Constituents of the Standards Used to Analyze
NBS Fly Ash Following Short Irradiations.

<u>Element</u>	<u>Compound used in standard</u>	<u>Concentration (µg/g unless % indicated)</u>
Mg	MgO	2.03 %
Al	Al ₂ O ₃	13.69 %
Si	SiO ₂	17.73 %
K	KCl	2.25 %
Cl	KCl	2.44 %
Ca	CaCO ₃	6.15 %
Sc	Sc ₂ O ₃	1810
Fe	Fe ₂ O ₃	6.04 %
Ni	Ni	912
Cu	Cu	1420
Zn	ZnO	1640
Ga	Ga ₂ O ₃	246
Rb	RbCl	788
Sr	SrCl ₂ ·6H ₂ O	4840
Zr	ZrO ₂	5340
Mo	(NH) ₆ Mo ₇ O ₂₄ ·4H ₂ O	520
Sb	Sb ₂ O ₄	395
Cs	CsCl	1270
Ba	BaCO ₃	6840
Pb	Pb(NO ₃) ₂	1720

essential to most efficiently irradiate many samples and to retrieve them quickly to count short-lived activities. The existing rabbit system at the RTNS-I, refurbished in 1980, is described in the Appendix, which also includes instructions for its use and a source listing of one of the PASCAL programs for automatic cyclic activation.

The design of the rabbits (see Fig. A-3 in the Appendix) evolved from several experiments. Specifications for rabbit performance were based primarily on safety considerations for the decay-heat studies described in Chapter VI. The snap-in cap was introduced to insure that the cap holding the sample in place is not removed in transit (the rabbit velocity exceeds 70 m/s). Samples are sealed in polyethylene capsules to guard against spilling radioactive material even after hundreds of cycles through the system.

Polyethylene was chosen because it contains no elements with Z greater than 6, and therefore should have produced no interfering gamma rays upon 14-MeV neutron irradiation. Unfortunately, small amounts of O, Al, and Si were present in the polyethylene even after thorough cleaning so it was sometimes necessary to subtract blank values of ^{27}Mg and ^{28}Al . It is not unusual to find some residual O in polyethylene⁽⁷⁶⁾, but the large amounts of O found in other plastics will produce copious quantities of ^{16}N upon 14-MeV neutron irradiation, interfering with any other short-lived activities. Low-density polyethylene proved to be much more resilient than high-density polyethylene, that often cracked in transit.

The irradiation geometry using the rabbit system is shown in Fig. A-5. A minimum distance of about 0.6 cm separates the sample

capsules from the neutron source. The extreme flux gradients over the sample volume necessitated a measurement of the average flux. Sample capsules filled with Fe_2O_3 were irradiated and by counting the ^{56}Mn activity produced, the average flux through the sample volume was determined to be $2.0 \times 10^{11} \text{ n/cm}^2\text{-s}$ at a source strength of $2.7 \times 10^{12} \text{ n/s}$.

A flux monitor foil was placed between the sample capsule and the cap for all irradiations. For all but cyclic FNAA, Al foils were used and ^{24}Na was counted to ascertain the relative fluxes for the samples and standards. Titanium foils were used for the cyclic activation producing $^{46}\text{Sc}^{\text{III}}$ with a 19-s half life and an isomeric transition of 142 keV. The short half life left little residual activity at the start of a new cycle. Elemental Ti was used, however, and extraneous gamma-rays from ^{47}Sc and ^{48}Sc were also produced.

3. Gamma-Ray Spectroscopy

All gamma-ray spectra were collected on one of several Ge(Li) detectors, with efficiencies in the range of 12-17% (relative to a 7.5 cm X 7.5 cm NaI(Tl) detector) and resolutions ranging from 1.9 to 2.5 keV at 1332 keV. The spectroscopy systems consisted of a detector, a preamplifier, an amplifier, a linear stretcher, an ADC, and an MCA. Several different analyzers were used for various experiments including a Nuclear Data ND 6603 and ND 6620, a Canberra 80, and a Data General NOVA 1200 minicomputer. Dead-time and pile-up corrections were usually made using the 392-keV gamma-ray from a ^{113}Sn source placed near the detector. A pulser was used in some of the earlier experiments but

unstable temperatures in the counting room caused considerable drifting of pulser peaks. Some Ge(Li) spectra were collected on the detectors located in the Nuclear Chemistry Division counting facilities.⁽⁷¹⁾

Five gamma-ray spectra from Ge(Li) detectors are shown in Section F. Several striking features of these spectra are worth noting here in order to better interpret the data. The most obvious feature of these spectra is the resolution. The ability to resolve closely spaced gamma rays (such as the 843.8- and 846.8-keV peaks from ^{27}Mg and ^{56}Mn , respectively) allows analysis codes to accurately calculate these peak areas.

Other features of Ge(Li) spectra are direct results of the mechanisms of photon interactions with the detector volume. Most gamma rays in the range of interest, 100-2300 keV, undergo Compton scattering in the detector. As the photon energy increases past 1022 keV, however, there is an increasing probability of pair production. The background in a Ge(Li) spectrum is due largely to the Compton continuum below the major photopeaks, caused by the escape from the detector of the scattered photon. When a photon interacts with a detector to create an electron-positron pair, it is possible one or both of the 511-keV gamma rays from positron annihilation will escape the detector. If such discrete amounts of energy escape, so-called escape peaks result in the spectrum. For example, the ^{28}Al decay is accompanied by a 1779-keV gamma ray which often interacts with the detector via pair production, resulting in a single-escape peak at 1268 keV and a double-escape peak at 757 keV. All major gamma rays greater than about 1300 keV will cause escape peaks in the spectrum, and frequently an escape peak will be

observed from a gamma ray that is beyond the highest energy being collected in the MCA. Escape peaks are often useful in NAA, but sometimes lead to instrumental interferences.

Many computer codes have been developed to search for peaks in Ge(Li) spectra and calculate peak areas and uncertainties. GAMANAL, a code developed at LLNL⁽⁷¹⁾, was used to analyze most gamma-ray spectra in the present work. The many complicated algorithms that determine the background, set energy-calibration and peak-shape parameters, search for peaks, resolve multiplets, and fit Gaussian shapes to peaks for area determinations, are described fully in Reference 71. As many as 15 peaks can be used for energy calibration and the calculation of peak-shape parameters. For the purpose of FNAA, only two peaks were used because a linear calibration was adequate.

Data collected on the ND 6603 analyzer were analyzed by a peak search routine developed by Nuclear Data for the ND 6620 minicomputer. This code is necessarily less complex than GAMANAL; a description appears in ND users's manuals but is proprietary information. Peaks are located by the correlation method. A general description of the use of correlation techniques for finding peaks is given by Black⁽⁷⁷⁾, although the method employed by ND is somewhat different.

F. Results and Discussion

1. Radioisotopes Observed in Standard Reference Materials

Over 80 activation products from 37 elements have been observed in the three NBS standard reference materials following 14-MeV neutron irradiations of various lengths. The reactions observed and their cross

sections are listed in Tables IV-2 through IV-4, categorized according to half life of the radioisotope produced. Major gamma rays from these activation products are also listed. If a radioisotope appears more than once in these tables the half-life and decay information are not repeated; the open spaces in the table serve to flag these potential interferences.

The interferences are listed in Table IV-5 with the ratios of interfering activity to primary activity (I_{ip} from Eq. IV-6) for each case. Although I_{ip} may be very small, the relative concentrations of the elements may dictate an interference correction. Frequently, the interfering element is not present in the sample in significant quantities, but may be an important source of the primary activity in the standard. If the primary element and the interfering element are reversed, the new value of I_{ip} is the reciprocal of the value listed in Table IV-5.

Gamma-ray spectra collected in the most recent analysis are shown in Figs. IV-1 to IV-5. A spectrum collected shortly after a fast-neutron irradiation of fly ash is dominated by ^{27}Mg , ^{28}Al , and ^{29}Al , as shown in Fig. IV-1. In general, spectra following short irradiations contain activation products from the major constituents. Several escape peaks are visible in Fig. IV-1, denoted by (SE) and (DE), as is the 392-keV peak from ^{113}Sn used for dead-time and pile-up corrections. Figure IV-2 is a spectrum from fly ash taken two days after a 45-min irradiation, and is dominated by ^{24}Na . In addition to activity from the major constituents, Mg, Al, and Ca, peaks from Ti and Ba are observed. The same sample counted one week after the

Table IV-2. Radioisotopes with half lives less than 1 hour observed in 14-MeV neutron activation.

Element sought	Reaction and product	14.9-MeV neutron ^a cross section (mb)	Product ^{b,c} half life	Gamma-ray ^{b,c} energies (keV)
O	$^{16}\text{O}(n,p)^{16}\text{N}$	34 ± 4	7.13 S	6129, 7116
F	$^{19}\text{F}(n,p)^{19}\text{O}$	18 ± 5	29.6 S	197, 1357
Na	$^{23}\text{Na}(n,\alpha)^{20}\text{F}$	150 ± 20	11.0 S	1634
	$^{23}\text{Na}(n,p)^{23}\text{Ne}$	43 ± 5	37.6 S	440
Mg	$^{26}\text{Mg}(n,\alpha)^{23}\text{Ne}$	72 ± 10		
	$^{25}\text{Mg}(n,p)^{25}\text{Na}$	40 ± 4	60.0 S	975, 380, 585
Al	$^{27}\text{Al}(n,p)^{27}\text{Mg}$	75 ± 7	9.46 M	844, 1014, 171
Si	$^{30}\text{Si}(n,\alpha)^{27}\text{Mg}$	110 ± 65		
	$^{29}\text{Si}(n,p)^{29}\text{Al}$	147 ± 18	6.6 M	1273
	$^{28}\text{Si}(n,p)^{28}\text{Al}$	210 ± 20	2.24 M	1779
P	$^{31}\text{P}(n,\alpha)^{28}\text{Al}$	115 ± 12		
Cl	$^{35}\text{Cl}(n,2n)^{34}\text{Cl}^m$	12	32.0 M	146(IT), 2128
	$^{37}\text{Cl}(n,p)^{37}\text{S}$	41 ± 4	5.0 M	3103, 2081(DE)
K	$^{39}\text{K}(n,2n)^{38}\text{K}$	5.1 ± 0.5	7.61 M	2167
	$^{41}\text{K}(n,\alpha)^{38}\text{Cl}$	31 ± 5	37.2 M	2167, 1642
Ca	$^{44}\text{Ca}(n,p)^{44}\text{K}$	36 ± 7	22.1 M	1157
Cr	$^{52}\text{Cr}(n,p)^{52}\text{V}$	96 ± 10	3.76 M	1434
Mn	$^{55}\text{Mn}(n,\alpha)^{52}\text{V}$	32 ± 5		
Fe	$^{54}\text{Fe}(n,2n)^{53}\text{Fe}$	22 ± 2	8.51 M	378
Zn	$^{64}\text{Zn}(n,2n)^{63}\text{Zn}$	204 ± 16	38.1 M	669, 962
Rb	$^{85}\text{Rb}(n,2n)^{84}\text{Rb}^m$	670 ± 200	20.1 M	248, 465, 216
Sr	$^{84}\text{Sr}(n,p)^{84}\text{Rb}^m$	47 ± 7^d		
	$^{88}\text{Sr}(n,p)^{88}\text{Rb}$	17 ± 2	17.7 M	898, 1836
Zr	$^{90}\text{Zr}(n,2n)^{89}\text{Zr}^m$	130	4.18 M	588(IT)
	$^{91}\text{Zr}(n,p)^{91}\text{Y}^m$	20	49.7 M	556(IT)
Ba	$^{138}\text{Ba}(n,2n)^{137}\text{Ba}^m$	1250 ± 100	2.55 M	662(IT)

a. Borman et al., Ref. 7

b. Chart of the Nuclides, Ref. 78

c. Table of Isotopes, Ref. 79

d. Ref. 13

IT: Isomeric transition

DE: Double escape peak

Table IV-3. Radioisotopes with half-lives between than 1 hour and 3 days, observed in 14-MeV neutron activation.

Element sought	Reaction and product	14.9-MeV neutron ^a cross section (mb)	Product ^{b,c} half-life	Gamma-ray ^{b,c} energies (keV)
Al	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	111 ± 4	15.0 H	1369, 1732(10)
Mg	$^{24}\text{Mg}(n,p)^{24}\text{Na}$	180 ± 18		
P	$^{31}\text{P}(n,p)^{31}\text{Si}$	83 ± 5	2.62 H	1266
K	$^{41}\text{K}(n,p)^{41}\text{Ar}$	48 ± 10	1.83 H	1294
Ca	$^{44}\text{Ca}(n,\alpha)^{41}\text{Ar}$	35 ± 8		
	$^{43}\text{Ca}(n,p)^{43}\text{K}$	500	22.3 H	618, 373
	$^{42}\text{Ca}(n,p)^{42}\text{K}$	180 ± 10	12.4 H	1525
Sc	$^{45}\text{Sc}(n,\alpha)^{42}\text{K}$	54 ± 6		
	$^{45}\text{Sc}(n,2n)^{44}\text{Sc}^m$	155 ± 15	2.44 D	271(IT)
Ti	$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	120 ± 20	3.42 D	159(IT)
	$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	61 ± 6	43.7 H	984, 1038, 1312
Fe	$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	103 ± 6	2.58 H	847, 1811
Ni	$^{58}\text{Ni}(n,2n)^{57}\text{Ni}$	35 ± 3	36.1 H	1378
Cu	$^{65}\text{Cu}(n,2n)^{64}\text{Cu}$	975 ± 50	12.7 H	1346
	$^{65}\text{Cu}(n,p)^{65}\text{Cu}$	23 ± 3	2.52 H	1482
Zn	$^{64}\text{Zn}(n,p)^{64}\text{Cu}$	170 ± 15		
	$^{68}\text{Zn}(n,\alpha)^{65}\text{Ni}$	10 ± 1		
	$^{70}\text{Zn}(n,2n)^{69}\text{Zn}^m$	744 ± 100	13.8 H	439(IT)
Ga	$^{69}\text{Ga}(n,p)^{69}\text{Zn}^m$	24 ± 6		
	$^{69}\text{Ga}(n,2n)^{68}\text{Ga}$	1070 ± 90	68.1 M	1077
Rb	$^{85}\text{Rb}(n,p)^{85}\text{Kr}^m$	4.5 ± 0.5	4.48 H	151
Sr	$^{88}\text{Sr}(n,\alpha)^{85}\text{Kr}^m$	70d		
	$^{86}\text{Sr}(n,2n)^{85}\text{Sr}^m$	276 ± 30	67.7 M	232(IT), 151
	$^{88}\text{Sr}(n,2n)^{87}\text{Sr}^m$	354 ± 30	2.81 H	388(IT)
Zr	$^{90}\text{Zr}(n,2n)^{89}\text{Zr}$	790 ± 50	78.5 H	909
	$^{90}\text{Zr}(n,p)^{90}\text{Y}^m$	46 ± 4	3.19 H	479(IT)
Mo	$^{100}\text{Mo}(n,2n)^{99}\text{Mo}$	1390 ± 60	66.0 H	141
Sb	$^{123}\text{Sb}(n,2n)^{122}\text{Sb}$	1300	2.68 D	564
Ba	$^{136}\text{Ba}(n,2n)^{135}\text{Ba}^m$	700 ± 80	28.7 H	268(IT)
	$^{134}\text{Ba}(n,2n)^{133}\text{Ba}^m$	1600 ^e	38.9 H	276(IT)
Pb	$^{204}\text{Pb}(n,2n)^{203}\text{Pb}$	1840 ± 130	52.1 H	279

a. Ref. 7

b. Ref. 78

c. Ref. 79

d. Ref. 13

e. Eq. II-2

IT: Isomeric transition

DE: Double escape peak

Table IV-4. Radioisotopes with half lives greater than 3 days observed in 14-MeV neutron activation.

Element sought	Reaction and product	14.9-MeV neutron ^a cross section (mb)	Product ^{b,c} half life	Gamma-ray ^{b,c} energies (keV)
Na	$^{23}\text{Na}(n,2n)^{22}\text{Na}$	54 ± 5	2.60 Y	1274
Ca	$^{48}\text{Ca}(n,2n)^{47}\text{Ca}$	1070 ± 360	4.54 D	1297, 489, 808
Ti	$^{50}\text{Ti}(n,\alpha)^{47}\text{Ca}$	10 ± 2		
	$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	270 ± 25	83.8 D	889, 1131
Cr	$^{52}\text{Cr}(n,2n)^{51}\text{Cr}$	358 ± 25	27.7 D	320
Mn	$^{55}\text{Mn}(n,2n)^{54}\text{Mn}$	850 ± 60	312 D	835
Fe	$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	315 ± 25		
	$^{54}\text{Fe}(n,\alpha)^{51}\text{Cr}$	96 ± 10		
Co	$^{59}\text{Co}(n,p)^{59}\text{Fe}$	82 ± 8	44.6 D	1099, 1292
	$^{59}\text{Co}(n,2n)^{58}\text{Co}$	735 ± 55	70.8 D	811
Ni	$^{58}\text{Ni}(n,p)^{58}\text{Co}$	330 ± 30		
	$^{58}\text{Ni}(n,n')^p)^{57}\text{Co}$	520 ± 15^d	271 D	122
Zn	$^{66}\text{Zn}(n,2n)^{65}\text{Zn}$	740 ± 60	244 D	1116
As	$^{75}\text{As}(n,2n)^{74}\text{As}$	1085 ± 75	17.8 D	596, 635
Se	$^{76}\text{Se}(n,2n)^{75}\text{Se}$	944 ± 65	119 D	265, 136
Rb	$^{85}\text{Rb}(n,2n)^{84}\text{Rb}$	1430	32.9 D	882
	$^{87}\text{Rb}(n,2n)^{86}\text{Rb}$	750	18.7 D	1077
Sr	$^{84}\text{Sr}(n,p)^{84}\text{Rb}$	96 ± 8		
	$^{86}\text{Sr}(n,p)^{86}\text{Rb}$	45 ± 4		
	$^{86}\text{Sr}(n,2n)^{85}\text{Sr}$	1060 ± 105	64.8 D	514
Y	$^{89}\text{Y}(n,2n)^{88}\text{Y}$	1015 ± 70	107 D	898, 1836
Zr	$^{96}\text{Zr}(n,2n)^{95}\text{Zr}$	1529 ± 141	64.0 D	757, 724
Nb	$^{93}\text{Nb}(n,2n)^{92}\text{Nb}$	450 ± 30	10.1 D	935
Mo	$^{92}\text{Mo}(n,p)^{92}\text{Nb}$	60 ± 10		
Sb	$^{121}\text{Sb}(n,2n)^{120}\text{Sb}$	620	5.76 D	1172, 197, 1023
I	$^{127}\text{I}(n,2n)^{126}\text{I}$	1650 ± 140	13.0 D	389, 666
Cs	$^{133}\text{Cs}(n,2n)^{132}\text{Cs}$	1600 ± 110	6.47 D	668
Ba	$^{132}\text{Ba}(n,p)^{132}\text{Cs}$	200 ^e		
	$^{132}\text{Ba}(n,2n)^{131}\text{Ba}$	1600	11.7 D	496, 124, 216
	$^{134}\text{Ba}(n,2n)^{133}\text{Ba}$	940 ± 80	10.7 Y	356
Ce	$^{140}\text{Ce}(n,2n)^{139}\text{Ce}$	1760 ± 110	137 D	166
	$^{142}\text{Ce}(n,2n)^{141}\text{Ce}$	1850 ± 160	32.5 D	145
Tl	$^{203}\text{Tl}(n,2n)^{202}\text{Tl}$	1850 ± 160	12.2 D	439

a. Ref. 7

b. Ref. 78

c. Ref. 79

d. Ref. 17

e. Eq. II-4

Table IV-5. Interferences in Multielement FNAA and the ratio of interfering activity to primary activity from equal masses of the two elements.

<u>Radionuclide</u>	<u>Primary element</u>	<u>Interfering element</u>	<u>I_{ip}</u>	<u>Uncertainty (%)</u>
²³ Ne	Na	Mg	0.174	18
²⁴ Na	Mg	Al	0.704	11
²⁷ Mg	Al	Si	0.044	60
²⁸ Al	P	Si	1.86	14
⁴¹ Ar	K	Ca	0.222	31
⁴² K	Ca	Sc	41.3	12
⁴⁷ Ca	Ca	Ti	0.217	39
⁵¹ Cr	Cr	Fe	0.017	13
⁵² V	Cr	Mn	0.376	19
⁵⁴ Mn	Mn	Fe	0.021	11
⁵⁸ Co	Co	Ni	0.308	12
⁶⁴ Cu	Cu	Zn	0.268	10
⁶⁵ Ni	Cu	Zn	0.258	16
⁶⁹ Zn ^m	Zn	Ga	2.88	18
⁸⁴ Rb ^{m+g}	Rb	Sr	5x10 ⁻⁴	33
⁸⁵ Kr ^m	Rb	Sr	16.6	44
⁹² Nb ^m	Nb	Mo	0.019	18
¹³² Cs	Cs	Ba	1x10 ⁻⁴	50

Figure IV-1. Gamma-Ray Spectrum from NBS Fly Ash Following a Short 14-MeV Neutron Irradiation.

Figure IV-2. Gamma-Ray Spectrum from NBS Fly Ash Following an Intermediate 14-MeV Neutron Irradiation.

Figure IV-3b. Gamma-Ray Spectrum from NBS Fly Ash 7 Days after an Intermediate 14-MeV Neutron Irradiation.

Figure IV-4. Gamma-Ray Spectrum from NBS Orchard Leaves Following an Intermediate 14-MeV Neutron Irradiation.

Figure IV-5. Gamma-Ray Spectrum from NBS Bovine Liver Following an Intermediate 14-MeV Neutron Irradiation.

irradiation had substantial activity from many trace elements, as seen in Fig. IV-3. Several activation products with long half lives that were observed in previous experiments after a 4-h irradiation can be seen in Fig. IV-3.

Spectra from orchard leaves and bovine liver are shown in Figs. IV-4 and IV-5, respectively. These spectra were collected the day of the irradiation and are dominated by the activation products from Mg, Cl, K, Ca, and Fe, although peaks from Zn, Rb, and Sr are also visible. The 558-keV peak in Fig. IV-5 is a prompt gamma ray from the neutron capture reaction in ^{113}Cd in the Cu-Cd x-ray absorber placed over the detector.

2. Preliminary Results

A preliminary analysis of the three SRM's proved the feasibility of multielement FNAA while demonstrating the need for more reliable and less expensive procedures. Reference 67 is a report of these experiments so the concentrations determined will not be repeated here. It is useful to summarize the procedures, however, in order to chronicle the development of the final procedures described in Section E.

The analysis was conducted in two parts. First, three irradiations (10-60 s, 25-60 min, and 4-5 h) of sample-standard pairs were made without the use of the rabbit system. Samples and standards were packaged in polyethylene bags and folded together during the irradiation to lessen the effects of flux gradients. Trace elements were introduced to the standards as liquids, and major constituents were added as dry powders after the solute had evaporated from the

polyethylene bags. The rabbit system was available for the second part of the experiment, in which the short irradiations were repeated. Samples were again sealed in polyethylene bags, but because the rabbits were too small for sample-standard pairs, each was irradiated separately with an Al flux monitor.

The concentrations of the following elements were determined in at least one the samples: Na, Mg, Al, Si, P, K, Sc, Ti, Fe, Co, Ni, Zn, As, Se, rb, Sr, Y, Zr, Mo, Sb, Ba, Ce, Tl, and Pb (20 in fly ash, 12 in orchard leaves, and 10 in bovine liver). Nearly half of the concentrations differed by more the 25% from generally accepted values.

The major source of error in the first part of this experiment was the combination of poor irradiation geometry and inhomogeneous standards. Procedures for preparing standards similar to those outlined in Section E were developed for the second part of the experiment with much more accurate results. The need for a reproducible irradiation geometry and more sturdy sample containers led to the rabbit design described in the Appendix.

Two additional changes were suggested by the preliminary work. First, the lack of induced activity in the biological samples coupled with the observation of activation products with half lives less than a minute, pointed towards cyclic activation as a method to enhance the activities from short-lived radionuclides. Second, the prohibitive expense of long irradiations dictated the need to maximize the number of elements determined while minimizing the necessary beam time.

In addition to demonstrating the feasibility of multielement FNAA, the preliminary experiments showed that several potentially

disasterous problems associated with 14-MeV neutron activation would not pose major problems in the present work:

1. While interferences abound, only two elemental determinations, Cr and Mn in fly ash, were totally obscured by interfering activity.
2. The thermal-neutron flux was measured to be about 1×10^7 n/cm²-s by a 45-min irradiation of Co and counting the ⁶⁰Co. Therefore, interference from neutron-capture reactions was shown to be negligible.
3. The size and geometry of samples were such that the neutron flux was not significantly attenuated by interactions within the sample. Using the removal cross sections from Margolwalla and Przybylowicz in Eq. II-15, the maximum attenuation in samples and standards is less than 1%.
4. Gamma-ray attenuation was slightly greater, as much as 7% for 100-keV photons, but the attenuation was nearly identical for samples and standards so no correction was needed.

3. Analysis of Standard Reference Materials

Following the procedures described in Section E, multielement FNAA of fly ash, orchard leaves, and bovine liver was performed using cyclic activation, and short (1-5 min) and intermediate (45-90 min) irradiations. The cyclic FNAA included 15 cycles with a 1-5 s irradiation, a 30-s delay for the decay of ¹⁶N, and a 30-s count. To cut costs, long irradiations of the previous work were not repeated. The concentrations determined by FNAA are given in Tables IV-6 through IV-8, along with values from the compilation by Gladney⁽⁴⁶⁾ for comparison. Activation products used for each determination are listed also, and many notes concerning interferences and irradiations are included as well. Values for Co, As, Se, Y, Ce, and Tl, obtained from the long irradiations in the preliminary work, are presented in order to

Table IV-6. Summary of elemental concentrations in NRS fly ash determined by FNNA (ug/g unless % indicated).

Element sought	Isotope used	Present Work	Compilation by Gladney ⁴⁶	Comments
Na	²⁰ F, ²² Na	3330 ± 170	3200 ± 340	Cyclic or long irradiation
Mg (%)	²⁴ Na	2.1 ± 0.5	1.6 ± 0.3	Large Al interference
Al (%)	²⁷ Mg	12.2 ± 0.5	12.5 ± 1.0	Small Si interference
Si (%)	^{28,29} Al	22.4 ± 1.6	20.2 ± 2.5	
K (%)	³⁸ Cl	1.8 ± 0.3	1.70 ± 0.09	1642-keV peak used to avoid intf. from ³⁸ K
Ca (%)	⁴⁷ Ca	4.40 ± 0.18	4.6 ± 0.4	Interference from ⁵⁰ Ti
Sc	⁴⁴ Sc ^m	29 ± 3	26 ± 2	
Ti	^{46,48} Sc	7600 ± 200	7200 ± 720	⁴⁷ Sc daughter of ⁴⁷ Ca
Cr	⁵¹ Cr	< 150	129 ± 11	Very large Fe intf.
Fe (%)	⁵⁶ Mn	6.40 ± 0.15	6.2 ± 0.4	⁵¹ Cr, ⁵⁴ Mn also
Co	⁵⁹ Fe	45 ± 16	40 ± 3	Long irradiation
Ni	⁵⁷ Co	106 ± 12	98 ± 11	(n,d+n'p+pn') reaction
Zn	⁶⁹ Zn	< 300	213 ± 13	
As	⁷⁴ As	68 ± 12	61 ± 3	Long irradiation
Se	⁷⁵ Se	35 ± 13	9.6 ± 0.8	Long irradiation
Rb	⁸⁴ Rb	102 ± 5	114 ± 10	Small Sr interference
Sr	⁸⁷ Sr ^m	1310 ± 60	1380 ± 190	
Y	⁸⁸ Y	150 ± 15	62 ± 10	Long irradiation
Zr	⁸⁹ Zr	380 ± 20	300 ± 75	
Mo	⁹⁹ Mo	22.3 ± 1.6	29 ± 6	
Sb	^{122,120} Sb	8.3 ± 1.8	6.9 ± 0.4	
Cs	¹³² Cs	10 ± 1	8.6 ± 1.8	
Ba	¹³⁵ Ba ^m	2550 ± 110	2700 ± 170	¹³³ Ba ^m , ¹³⁷ Ba also
Ce	^{139,141} Ce	136 ± 8	152 ± 11	Long irradiation
Tl	²⁰² Tl	18 ± 6	4.1 ± 0.9	Long irradiation
Pb	²⁰³ Pb	100 ± 25	72 ± 5	Very weak peak

Table IV-7. Summary of elemental concentrations in NBS orchard leaves (SRM 1571) determined by FNNA ($\mu\text{g/g}$ unless % indicated).

Element sought	Isotope used	Present Work	Compilation by Gladney ⁴⁶	Comments
F	^{19}F	< 60	4.3	Cyclic FNAA
Na	^{23}Na	< 70	100 ± 40	Cyclic FNAA
Mg	^{24}Mg	6100 ± 1100	6070 ± 580	Cyclic FNAA
Al	^{27}Al	380 ± 100	310 ± 100	Blank subtracted
P (%)	^{31}P	0.20 ± 0.02	0.2 ± 0.02	Si interf. from blank
Cl	^{35}Cl	950 ± 70	720 ± 50	
K (%)	^{39}K	1.46 ± 0.20	1.46 ± 0.06	Cyclic FNAA
Ca (%)	^{40}Ca	1.97 ± 0.15	2.02 ± 0.09	$^{42}, ^{43}\text{K}$ also
Ti	^{48}Ti	30 ± 4	20 ± 20	
Cr	^{52}Cr	< 2	2.5 ± 0.3	Large Fe interference
Fe	^{56}Fe	282 ± 21	280 ± 50	^{51}Cr , ^{54}Mn also
Co	^{59}Co	< 8	0.2 ± 0.1	Long irradiation
Ni	^{58}Ni	< 5	1.7 ± 1.0	
Zn	^{66}Zn	< 40	25 ± 3	
As	^{75}As	10.6 ± 0.5	11.5 ± 2.1	Long irradiation
Rb	^{85}Rb	10 ± 1	11.6 ± 1.2	$^{84}\text{Rb}^m$ also
Sr	$^{87}\text{Sr}^m$	39 ± 2	36 ± 4	
Y	^{89}Y	< 1.1	---	Long irradiation
Zr	^{89}Zr	3 ± 1	2.0 ± 1.8	
Mo	^{99}Mo	< 0.8	1.4 ± 1.6	
Sb	$^{120}, ^{122}\text{Sb}$	2.7 ± 0.4	3.0 ± 0.4	
Cs	^{132}Cs	< 0.3	0.03 ± 0.01	
Ba	$^{135}\text{Ba}^m$	80 ± 22	45 ± 8	
Ce	$^{139}, ^{141}\text{Ce}$	< 2	1.0 ± 0.1	Long irradiation
Pb	^{203}Pb	42 ± 9	46 ± 4	

Table IV-8. Summary of elemental concentrations in NBS bovine liver (SRM 1577) determined by FNNA (ug/g unless % indicated).

<u>Element sought</u>	<u>Isotope used</u>	<u>Present Work</u>	<u>Compilation by Gladney⁴⁶</u>	<u>Comments</u>
Na	²³ Ne	2000 $\pm$ 150	2440 $\pm$ 230	Cyclic FNAA
Mg	²⁴ Na	700 $\pm$ 20	587 $\pm$ 20	Negligible Al intf.
Al	²⁷ Mg	< 80	30 $\pm$ 20	Blank > signal
P (%)	²⁸ Al	1.10 $\pm$ 0.15	1.05 $\pm$ 0.16	Si intf. from blank
Cl	³⁴ Cl ^m	3500 $\pm$ 200	2680 $\pm$ 160	
K (%)	³⁸ Cl	1.18 $\pm$ 0.10	0.94 $\pm$ 0.10	
Ca (%)	⁴⁷ Ca	< 0.1	0.12 $\pm$ 0.02	
Sc	⁴⁴ Sc	< 0.8	0.001	
Ti	⁴⁸ Sc	< 4	2	
Cr	⁵¹ Cr	< 0.3	0.7 $\pm$ 0.7	Fe interference
Fe	⁵⁶ Mn	334 $\pm$ 10	263 $\pm$ 13	
Co	⁵⁹ Fe	0.31 $\pm$ 0.12	0.24 $\pm$ 0.06	Long irradiation
Cu	⁶⁵ Ni	270 $\pm$ 90	189 $\pm$ 21	Zn interference
Zn	⁶⁷ Cu	200 $\pm$ 40	132 $\pm$ 10	^{63,69} Zn also
Ga	⁶⁸ Ga	< 20	--	
As	⁷⁴ As	< 1.3	0.05 $\pm$ 0.01	Long irradiation
Rb	⁸⁴ Rb	15 $\pm$ 2	19.5 $\pm$ 3.6	Negligible Sr intf.
Sr	⁸⁷ Sr ^m	2.0 $\pm$ 0.8	0.2	
Zr	⁸⁹ Zr	< 0.5	--	
Mo	⁹⁹ Mo	3.7 $\pm$ 0.4	3.1 $\pm$ 0.5	
Sb	^{120,122} Sb	< 0.5	--	
Cs	¹³² Cs	< 0.2	0.2	
Ba	¹³⁵ Ba ^m	< 20	--	
Pb	²⁰³ Pb	< 11	0.35 $\pm$ 0.06	

list the cumulative results of the analyses. (As can be seen in Fig. IV-3, however, had these elements been included in the standards, they may have been determined in fly ash irradiated only 45 min.)

Concentrations of 24 elements, representing 50% of the sample mass, were determined in fly ash. The agreement with the compilation is good, the most notable discrepancies are Se, Y, and Tl because of standard inhomogeneities in the preliminary work. Interference from Al nearly obscured ^{24}Na activity from Mg, and Fe interference made Cr and Mn determinations impossible, but in general, interferences were not a major problem. Cyclic activation provided only a Na determination for fly ash and even that could have been obtained from the intermediate data if Na was present in the standards irradiated 45 min. Thus, the cyclic activation appears to be impractical, given the required beam time, for samples with inorganic matrices such as fly ash. The procedure recommended for fly ash is a 1-2 min irradiation and counting within 10 min, and a 45-min irradiation with two or three counts in the following week. Spectra obtained in this way would be similar to Figs. IV-1 to IV-3.

The analysis of the biological samples also represent an improvement in sensitivity and accuracy over the previous work. Still, many elements are present in concentrations below the detection limits of multielement FNAA. A total of 15 elements in orchard leaves and 12 in bovine liver were determined. Cyclic activation was more useful for these samples, but the rabbit system and even the rabbits themselves introduced substantial ^{27}Mg and ^{28}Al activities in blank capsules. (The rabbit system is unquestionably contaminated with dirt

and rust. The polyethylene capsules were probably contaminated by machining during fabrication, and, although they were leached in HNO_3 , they still contained some Al and Si.) Subtraction of the blank caused problems with Al and P determinations.

The agreement with the compilation is not as close as in the case of fly ash. This is probably due to the practice of diluting the standards with graphite. The final mixture may have separated from the lower-density graphite, or the entire mixture may simply have settled into a smaller volume. In either case, an uneven distribution of the standard material would account for the discrepancies.

G. Conclusions and Recommendations

Analysis of the biological SRM's points to the need for further improvements in the procedures. The cyclic FNAA will require further development to optimize the irradiation-counting sequences. Rabbits and capsules should be modified to reduce the contamination. The procedure of mixing standard components with graphite should be studied and probably modified. Finally a rabbit able to deliver larger samples, 1-2 g, would no doubt increase the sensitivity. Presently, the analysis of orchard leaves or bovine liver requires about twice the beam time as is necessary for fly ash, for half as many elemental determinations.

High-flux, multielement FNAA is a reliable and accurate method of analysis. It is clearly more practical for inorganic samples such as fly ash, soils, and rocks. Further applications of this technique will depend on economic factors and the availability of intense sources of 14-MeV neutrons. Presently, the RTNS-I is the only fast-neutron source

available for high-flux FNAA. Much cheaper sealed-tube sources are being developed that nearly equal the RTNS-I source strength. If such sources are able to produce fluxes greater than 10^{11} n/cm²-s at a lower cost, the procedures developed here may become practical in the near future. Otherwise, a period of 10-20 years may pass before there are fast-neutron sources strong enough to make multielement FNAA economically practical.

V. DENSITY DIAGNOSTICS OF INERTIAL-CONFINEMENT-FUSION TARGETS

In the last three years, neutron activation techniques have been used to infer the density of the deuterium-tritium fuel at the time of thermonuclear burn for the Shiva laser-fusion facility at LLNL. A description of the method currently used for this diagnostic is presented in the following section. Several 14-MeV neutron irradiations were performed at the RTNS-I in the summer of 1979 to test the suitability of various materials potentially useful for more advanced nuclear diagnostics. These irradiations, described below, confirmed that if Br could be introduced as a seed gas in the DT fuel, it could yield a direct density measurement. The 14.1-MeV neutron cross section for the $^{79}\text{Br}(n,2n)^{78}\text{Br}$ reaction was measured at the RTNS-I using the activation method, and is described in the final section.

A. Density Measurements by 14-MeV Neutron Activation

The 20-beam Shiva laser was brought on line in 1978 with the capacity to deliver 30 TW in 100-ps pulses or 15 kJ in 1-ns pulses to illuminate and compress targets approximately 150 μm in diameter. Shiva is a neodymium-glass laser with a 1.06 μm wavelength.⁽⁸⁰⁾ Targets used in laser-fusion experiments are of two basic types: exploding-pusher targets or ablative targets.⁽⁸¹⁾ The exploding-pusher targets are very thin-walled (1-3 μm) glass microspheres loaded with 10-50 atmospheres of DT fuel, and have attained very high fuel temperatures, up to 10 keV, and neutron yields of up to 3×10^{10} n. Ablative targets have 5- μm thick glass walls coated with a 15- μm layer of fluorocarbon plastic ablator. Such targets have lower

neutron yields and temperatures, but are capable of being compressed to much higher densities. A typical ablative target is sketched in Fig. V-1.

The burn efficiency of inertial-confinement fusion (ICF) is proportional to the areal density, that is, the density-radius product, ρR , of the fuel. The confinement time of a sphere of a dense plasma is proportional to its radius divided by the speed of sound in the plasma.⁽⁸²⁾ Since the burn rate is proportional to the density, it is the areal density that characterizes the quality of the confinement and the thermonuclear burn, in a manner analogous to Lawson's criterion for magnetic fusion. Areal densities greater than 0.3 g/cm^2 , the range of the 3.6-MeV alpha particles produced in the DT reaction, will be required to provide additional heat to the plasma from fusion reactions to ultimately produce more energy than was expended in the laser illumination. A thorough introduction to laser-fusion physics is given by Breuckner and Jorna.⁽⁸³⁾ The Shiva laser experiments fall about two orders of magnitude in density below that required for breakeven, so the emphasis at LLNL has been to achieve the highest densities possible from ablative targets.

Density diagnostics, using charged particles and x-rays, which were very successful for exploding-pusher targets, are hampered for ablative targets by the lower temperatures and the increased densities of the pusher material. A new diagnostic technique, using activation products from the thermonuclear neutrons, was therefore developed by E. M. Campbell.⁽⁸⁴⁾ The glass pusher, containing silicon, is activated by 14-MeV neutrons through the $^{28}\text{Si}(n,p)^{28}\text{Al}$ reaction. By

Laser conditions

20 beam illumination

$$E_L = 4 \text{ kJ}$$

$$\tau = 200 \text{ psec}$$

20% absorbed

Performance

$$N_Y \sim 1 \times 10^8$$

$$\rho \Delta R_{\text{SiO}_2} \sim 8 \text{ mg/cm}^2$$

Initial inner
diameter $\sim 140 \mu\text{m}$

Figure V-1. Ablative Inertial-Confinement-Fusion Target Used at the Shiva Laser Facility.

collecting target debris, and rapidly retrieving and counting the ^{28}Al , the areal density of the pusher is measured and the fuel density can be inferred.

Assuming a spherically symmetric implosion with no pusher-fuel mixing, and a burn time that is short compared to the time taken by the hydrodynamic processes, the number of ^{28}Al atoms produced, N^* , is related to the neutron yield, Y , and the pusher areal density, $\rho\Delta R$, by the following expression:

$$N^* = \sigma(L_o/A_w) f Y (\rho\Delta R) \quad (\text{V-1})$$

where, A_w = the average atomic weight of the pusher, and

f = the fractional abundance of ^{28}Si in the pusher.

Because N^* may be as low as a few hundred atoms, a low-background, high-efficiency β - γ coincidence detector is needed. A 4π NE 102 fluor, 5 cm X 15 cm, is used as a beta-detector, and the fluor is placed inside the well of a 25 cm X 25 cm NaI(Tl) detector serving as a gamma-ray detector.⁽⁸⁴⁾ Target debris is collected on a 20- μm thick Ti foil, positioned near the target as shown in Fig. V-2. The foil is retrieved immediately after the shot and wrapped around the finger of the beta fluor, which is reassembled and inserted into the NaI detector as shown in Fig. V-3. The number of counts recorded, N_c , is related to N^* by

$$N_c = N^* \epsilon_c \epsilon_d (1 - e^{-\lambda t_c}) e^{-\lambda t} \quad (\text{V-2})$$

where, ϵ_c = the fraction of ^{28}Al atoms collected, and

Figure V-2. Apparatus for Collection of ICF Target Debris.

Figure V-3. Coincidence Detector Used for 14-MeV Neutron Activation Diagnostic of ICF Target Pusher Areal Density.

ϵ_d = the efficiency of the coincidence detector.

The collection fraction is determined in a different shot using the same collection geometry and an identical target that has been irradiated with thermal neutrons, producing ^{24}Na from Na present in the silicate pusher.⁽⁸⁵⁾ Debris is collected in the usual manner, and the fraction of ^{24}Na collected is assumed to be the collection efficiency for ^{28}Al as well.

Fuel densities of ablative targets have been determined using this neutron activation diagnostic. As an example, for a yield of 6.7×10^8 neutrons, the pusher areal densities of such targets were measured to be about 0.006 g/cm^2 , and a fuel density of 1-2 g/cm^3 , up to 10 times liquid-hydrogen density, was inferred.⁽⁸⁶⁾ This particular shot, represents a 50-fold volume compression, and a fuel areal density of up to 0.0001 g/cm^2 , a factor of about 300 below breakeven. There is no simple relationship between the pusher areal density and the fuel density during thermonuclear burn. Auerbach et al.⁽⁸⁷⁾ have presented a detailed discussion of how physical measurements, such as the pusher areal density, are interpreted along with complex computer simulations to ascertain the fuel density. Finally, it should be noted that densities measured by neutron activation for simple exploding-pusher targets are in good agreement with the other density diagnostics⁽⁸¹⁾, giving confidence that the method is reliable.

B. Direct Measurements of the Fuel Areal Density

Because of the complex relationship between the pusher areal density and the fuel density, it is highly desirable to make a direct measurement of the fuel areal density. Such a measurement could be achieved by 14.1-MeV neutron activation if a tracer gas could be introduced into the fuel and if sufficient amounts of an activation product could be created. The relationship between the areal density of the fuel and the number of atoms activated from the tracer is⁽⁸⁸⁾

$$N^* = \frac{3}{4} \frac{\sigma}{M_{tr}} L_o f_{tr} \left\{ \frac{\rho_{tr}}{\rho_{DT}} \right\}_o Y (\rho R)_{DT} \quad (V-3)$$

where, $\left\{ \frac{\rho_{tr}}{\rho_{DT}} \right\}_o$ = the initial density ratio in the tracer-fuel mixture,

M_{tr} = the atomic weight of the tracer gas, and

f_{tr} = the isotopic abundance of the tracer nuclei.

The requirements for the ideal tracer gas are:

1. The gas must yield an activation product upon 14-MeV neutron bombardment, with a half life long enough to allow time to collect the debris but short enough to conveniently count a substantial fraction of the decays (a half life between 1 and 10 min).
2. The gas must mix well with the fuel and not react chemically with the pusher materials.
3. There must be a method to measure the collection fraction for the gas using a long-lived (half life greater than 10 h) activation product, preferably from a thermal-neutron irradiation.
4. The tracer gas must have minimal interference with the thermonuclear burn, that is, it should have Z as low as possible.

5. The decay of the activation product must be simple in order to use a high efficiency detection scheme.

Four reactions that meet some of these requirements are given in Table V-1; none of the reactions meet all of the requirements. The major disadvantages of Ar and N are the low cross sections and the need for indirect collection-fraction measurements. The problem with Br lies in target fabrication difficulties. Bromine gas is extremely reactive, causing design difficulties for target fabrication apparatus, and raising the possibility that Br will form compounds with the pusher materials rather than remain in the gas phase mixed with the fuel.

Fast-neutron irradiations of various amounts of N, Ar, and Br, mixed with target material, were performed at the RTNS-I in order to determine if the activation products from the potential tracers could be observed in a silicate matrix. Argon-filled targets with 0.1 atm of Ar are produced in the routine target fabrication procedure, so these targets were irradiated in an attempt to observe ^{37}S or ^{40}Cl . Neither of these activation products could be observed in existing targets, however, in gamma-ray spectra collected with a Ge(Li) detector. Fast-neutron reactions with Si produced abundant quantities of ^{28}Al , ^{29}Al , and ^{27}Mg , and ^{23}Ne was produced from the Na in the glass.

In order to test the ability to observe ^{13}N in target debris, several samples of target material with varying amounts of N added (in the form of NH_4OH) were irradiated. The annihilation radiation thus produced was compared to the magnitude of the 511-keV peak from irradiated target material with no N added. Nitrogen

Table V-1. Trace Candidates for Use in a Direct Measurement of Fuel Areal Density of ICF Targets

<u>Gas</u>	<u>Reaction and product</u>	<u>14.1-MeV neutron cross section (mb)^a</u>	<u>Product half life^b</u>	<u>Decay^b</u>
N	$^{14}\text{N}(n,2n)^{13}\text{N}$	6.1 ± 0.4	9.96 M	β^+ , no gamma
Ar	$^{40}\text{Ar}(n,p)^{40}\text{Cl}$	15.7 ± 2.0^c	1.35 M	β^+ , γ : 1461, 2840
Ar	$^{40}\text{Ar}(n,\alpha)^{37}\text{S}$	13.0 ± 1.5	5.05 M	β^- , γ : 3103
Br	$^{79}\text{Br}(n,2n)^{78}\text{Br}$	862 ± 50	6.45 M	β^+ , γ : 614

a) Bormann et al., Ref. 7

b) Chart of the Nuclides, Ref. 78

c) 14.4-MeV neutron cross section

concentrations of greater than 1% by weight were required to observe an enhanced 511-keV peak. In typical ICF targets, the presence of N_2 at 1% of the target weight would correspond to a partial pressure of 4 atm of N_2 , a factor of 40 greater than the present partial pressure of Ar , and probably enough of a contaminant to affect target performance.

Finally, a sequence of irradiations of target material containing various amounts of Br (introduced as a solution of $NaBr$) was performed. Because the cross section for production of ^{78}Br is much greater than those for the N and Ar reactions, enhanced annihilation radiation was observed in the samples with only 0.2% Br by weight. A target with such a Br content would have 0.1 atm of Br_2 , which is an acceptable amount. The results of these irradiations gave additional support for an attempt to use Br as a tracer gas in Shiva experiments.

Since the above irradiations, progress has been made in the use of Br. A batch of microspheres containing Br has been produced by the target fabrication group.⁽⁸⁹⁾ Chemical analysis of these targets showed that nearly 90% of the Br diffused through the pusher and was in the gas phase inside the target, and only 10% had remained in the glass (the target fabrication group was fearful that Br would react strongly with the pusher material). A thermal-neutron irradiation of such a target was performed and a collection-fraction measurement showed it is possible to collect more than 50% of the Br target debris.⁽⁹⁰⁾ As of this writing, Br-doped targets have not been used for an areal density measurement.

C. Measurement of the $^{79}\text{Br}(n,2n)^{78}\text{Br}$ Cross Section

Anticipating the use of Br for laser-fusion diagnostics, it was necessary to ascertain the value of the cross section for the production of ^{78}Br from 14.1-MeV neutron reactions. A quick survey of the literature indicated a range of reported cross sections from 870 to 1340 mb for 14.1-MeV neutrons. Since most of the measurements were made before the introduction of Ge(Li) detectors, it was decided to measure the cross section using high-resolution gamma-ray spectroscopy at the RTNS-I.

Procedures set forth by Rao and Fink⁽⁹⁸⁾ for the activation method using the mixed-powder technique were followed for this measurement. Thoroughly homogenized mixtures of NaBr, Fe_2O_3 , and Al_2O_3 were made in order to use both Fe and Al as flux monitors. Because these compounds were mixed together, the Br and the flux monitors necessarily experience the same flux, regardless of attenuation or flux gradients.

Irradiation geometry was very important, however, because the excitation function for the $^{79}\text{Br}(n,2n)^{78}\text{Br}$ reaction shows an increase of about 25% over the range of 14-15 MeV. The irradiation position used for activation analysis and decay-heat studies is along the beam line so the neutrons are emitted at 0° with an average neutron energy of 14.9 MeV. To assure a neutron energy of 14.1 MeV, the samples must be irradiated at 90° . The RTNS-I can be equipped with a conical tritium target, allowing the beam to strike the wall of the cone at a sharp angle, giving direct access to the space adjacent to the beam spot but perpendicular to the beam direction. Unfortunately, the

irradiation end of the rabbit system was too large to fit into this space, and as a result, the samples were irradiated at about 45° with an average neutron energy of about 14.6 ± 0.2 MeV.

The relationship between the unknown cross section, σ_u , and the cross section of the monitor reaction, σ_m , is

$$\sigma_u = \sigma_m \frac{\epsilon_m b_m \lambda_u c_f m M}{\epsilon_u b_u \lambda_m c_f m M} \times \frac{(1-e^{-\lambda_m t_i}) e^{-\lambda_m t} (1-e^{-\lambda_m t_c})}{(1-e^{-\lambda_u t_i}) e^{-\lambda_u t} (1-e^{-\lambda_u t_c})} \quad (V-4)$$

where, C is the the total number of counts in the gamma-ray peak, and the remaining variables are the same as for Eq. II-7. The nuclear properties of Br, Fe, and Al needed to calculate the cross section are summarized in Table V-2. Three samples were irradiated for 5 min at an RTNS-I source strength of 2×10^{12} n/s. (The value of the flux was not measured for this experiment because only the relative efficiencies of the Ge(Li) detector were known.) Samples were counted for 10 min after a decay period of 10 min; there was ample activity in the major peaks so the uncertainties of the peak areas were 1% or less. Ratios of ^{56}Mn to ^{27}Mg confirmed that the samples were homogeneous, correct values for nuclear properties were employed, and the 843.6-keV and 846.6-keV peaks were adequately resolved and analyzed.

Using Fe as the flux monitor, the cross section was determined to be 1020 ± 90 mb whereas a value of 990 ± 80 was obtained using Al as the monitor. There was no evidence to suggest one of the monitors was superior to the other, so the average value of

Table V-2. Nuclear properties of Br, Fe, and Al used in the determination of the $^{79}\text{Br}(n,2n)^{78}\text{Br}$ reaction cross section.

	^{79}Br	^{56}Fe	^{27}Al
activation product	^{78}Br	^{56}Mn	^{27}Mg
half life ^a	6.45 min	2.580 H	9.45 min
isotopic abundance ^a	0.5069	0.918	1.00
gamma rays ^b (keV)	613.6	846.8	843.8, 1014.4
intensity ^b (uncertainty)	0.136 (5)	0.9887 (4)	0.73 (1), 0.292 (9)
cross section		112±6	78±7

a. Chart of the Nuclides, Ref. 78

b. Table of Isotopes (7th Ed), Ref. 79

the $^{79}\text{Br}(n,2n)^{78}\text{Br}$ reaction cross section at 14.6 MeV is 1000 ± 60 mb. Bormann et al.⁽⁷⁾ recommend a value of 932 ± 60 for 14.5-MeV neutrons, so there is considerable overlap in the uncertainties in these two values. The increase of the excitation function with energy can account for the 14.6-MeV cross section being slightly greater than the 14.5-MeV cross section.

A measurement of the cross section at 14.1 MeV is still needed, however. Perhaps the activation method using Ge(Li) spectroscopy is inappropriate for counting ^{78}Br because of the 4% uncertainty in the intensity of the 614-keV gamma ray. It may be advantageous to employ a high-efficiency detector for annihilation radiation, as 92% of the decays of ^{78}Br are by β^+ emission. In either case, a precision sample holder should be designed for use with the conical target at the RTNS-I so only 14.1 MeV neutrons are incident upon the samples.

D. Conclusions

The technique of using the 14.1-MeV neutrons from the thermonuclear burn to diagnose ICF targets is still in its infancy. The Shiva laser was the first ICF facility to yield enough neutrons to produce detectable amounts of activation products. The next generation laser, NOVA, scheduled for completion in 1985, is expected to yield 10^{14} - 10^{16} neutrons per shot, enough to require a shielded target chamber. There will be much more flexibility in planning neutron-activation diagnostics for NOVA than currently exists for Shiva. In addition to advanced fuel-density diagnostics, it may be possible to coat the silicate pusher with a layer of a metal, and activate a tracer

in the fuel, ^{28}Al in the glass, and an additional activation product from the metal. A Ge(Li) detector would probably be needed to analyze these more complex spectra. Such an arrangement would allow simultaneous measurements of the areal densities of all three target components, and thus provide a great deal of information on the implosion process.⁽⁹⁰⁾

Advanced neutron-activation diagnostics will have to be thoroughly tested by fast-neutron irradiations at the RTNS-I before being implemented at NOVA, since the cost per shot will probably exceed \$250,000. As Icf experiments approach breakeven later this decade, whether driven by lasers or charged particles, fast-neutron irradiations and high-resolution gamma-ray spectroscopy will be required to test the feasibility of neutron-activation diagnostics and search for competing reactions from charged particles.

VI. FISSION DECAY-HEAT STUDIES

Detailed knowledge of decay properties of fission fragments is required for many nuclear engineering applications, such as emergency cooling system designs and radioactive waste management. While the short-lived fission products have been studied by traditional chemical separations, there are still great uncertainties in the decay characteristics of many short-lived products with half lives less than 1 min. For example, in the latest compilation of the ENDF/B fission product file of 800 radioisotopes, over 500 required theoretical estimates of their decay energies.⁽⁹²⁾ For many short-lived products, half lives are not accurately known and there may be many unknown isomers.

For several years, the Nuclear Chemistry Division at LLNL has been studying decay schemes of short-lived fission products. Methods for rapid chemical separations in either a batch mode or in continuous operation have been developed to study radioisotopes with half lives between 1 and 100 s.⁽⁹³⁾ In the batch mode, uranyl solutions were irradiated in the Livermore Pool-Type Reactor, rapidly retrieved, subjected to an automated sequence of chemical separations (depending on the element being studied), and finally counted with a system of Ge(Li) detectors. This method has been used for studies of As, Br, Sb, and Te. A continuous system features a gas jet to carry fission products from a thin foil of ^{235}U through a series of traps for chemical separations.⁽⁹⁴⁾ Selenium isotopes with half lives as short as 1 s have been studied in this manner.

As an alternative to chemical separations of fission products, certain radioisotopes of interest in decay-heat studies can be produced by fast-neutron irradiation of stable isotopes. The rabbit system at the RTNS-I was refurbished primarily for the purpose of developing this alternative. Chapter VI presents an example of 14-MeV production of one decay-heat isotope, ^{148}Pr , for subsequent gamma-ray spectroscopy.

A. Fast-Neutron Irradiation of ^{148}Nd to Produce ^{148}Pr

A number of radioisotopes that contribute significantly to reactor decay heat can be produced via (n,p) reactions upon 14-MeV neutron irradiation of stable isotopes. By irradiating mass-separated isotopes, the activity produced is primarily from 2 or 3 radioisotopes from the principle fast-neutron reactions. For example,

^{82}As , ^{100}Nb , or ^{148}Pr , can be produced through fast-neutron irradiations of ^{82}Se , ^{100}Mo , or ^{148}Nd . In the case of short-lived isomers of ^{82}As (13.7 s and 19 s) and ^{100}Nb (1.5 s and 3.1 s), the sample must contain no oxygen or the Compton edge from ^{16}N will obscure many low-intensity, low-energy gamma rays.

Irradiation of ^{148}Nd was chosen to test the ability to study ^{148}Pr using the improved rabbit system at the RTNS-I, because there is considerable controversy regarding the decay of ^{148}Pr , and because the longer half life allows the decay of ^{16}N before counting. In particular, it was desirable to observe gamma rays of 3 to 5 MeV that may occur in the decay of ^{148}Pr . Since the Q-value of the decay of ^{148}Pr is estimated to be 4.8 MeV⁽⁹⁵⁾, these

high-energy gamma-rays are possible, but they have not been previously observed. A significant fraction of the decay heat may be contained in these high-energy gamma-rays, even though they have lower intensities relative to the low-energy gamma rays. From gamma-ray spectra collected on a single Ge(Li) detector, the energies and intensities of the gamma rays from ^{148}Pr have been determined.

1. Background

There is considerable disagreement between the two most recent papers, both published in 1979, concerning the decay of ^{148}Pr . Taylor and Singh⁽⁹⁶⁾ used 14-MeV neutron irradiations of ^{148}Nd to produce ^{148}Pr and ^{145}Ce . A decay scheme was proposed based on γ - γ coincidence measurements using a Ge(Li) detector gated by a 7.5 cm X 7.5 cm Na(I) detector. The half life of ^{148}Pr was determined to be 2.28 ± 0.05 min based on the decay of the 302-keV gamma-ray activity, in a separate experiment using the $^{150}\text{Nd}(d,\alpha)$ reaction.

At the same time, Ikeda et al.⁽⁹⁵⁾ reported the results of their work on ^{148}Pr chemically separated from mixed fission fragments by rapid paper electrophoresis. Two Ge(Li) detectors were used in these γ - γ coincidence measurements. In addition, γ - γ anisotropy measurements, and β -ray singles and β - γ coincidence measurements were made. The major difference between the results of Ikeda et al. and those of Taylor and Singh is that the former report the existence of two isomers of ^{148}Pr . The half lives were determined using the 302-keV and the 451-keV peaks to be 2.27 ± 0.04 and 2.0 ± 0.1 min, respectively; no isomeric transition was observed. The decay scheme proposed by Ikeda

et al., presented in Fig. VI-1, shows the energy levels, transitions, and gamma-ray intensities of both isomers.

Several additional discrepancies exist between these two works. Taylor and Singh report levels at 1727, 2381, 2431, and 2579 keV, not present in Fig. VI-1, and do not propose levels at 723, 1512, and 1683 keV. The gamma-ray intensities also differ greatly for several peaks because the relative intensities are normalized to the 302-keV activity, which is dominant in the decay of both isomers. Therefore, the intensities reported by Taylor and Singh are generally much lower than those of Ikeda et al. as the 302-keV activity is split in the later work between both isomers.

2. Irradiations of ^{148}Nd at the RTNS-I

In order to resolve the above discrepancies, and to confirm or disprove the existence of two isomers, a series of irradiations of ^{148}Nd was performed at the RTNS-I. Twenty, 100-ng samples of Nd_2O_3 containing 98% ^{148}Nd (obtained from the Oak Ridge Isotopes Division) were sealed in polyethylene capsules 0.1-cm thick as described in the Appendix. Each capsule was irradiated for 70-90 s at a 14-MeV neutron flux of about 2×10^{11} n/cm²-s. Two, 2-min counts were then taken following a 40-s delay for the decay of ^{16}N . The 8192-channel gamma-ray spectra were obtained with an ORTEC coaxial Ge(Li) detector with an efficiency of about 13% and resolution of 2.0 keV at 1332 keV; a Canberra model 80 MCA was used to collect and store data on magnetic tape. The gain was set to detect gamma rays in the energy range from 0 to 5 MeV.

$^{148}\text{Pr } 2.27 \pm 0.04 \text{ m}$
(1⁺)
 $Q_\beta = 4.8 \pm 0.2 \text{ MeV}$

$^{148}\text{Pr } 2.0 \pm 0.1 \text{ m}$
(4⁺)
 $Q_\beta = 4.8 \pm 0.2 \text{ MeV}$

Figure VI-1. Decay Schemes of ^{148}Pr Isomers Proposed by Ikeda et al. (Several of the gamma-ray intensities are slightly different from the final version in Ref. 95.)

Each sample was automatically cycled through an irradiation-counting sequence 10 times, after which the cumulative data was recorded and a new sample was irradiated. A series of 10 cycles required about an hour of beam time. Samples were changed frequently because of the growth of 1.7-h ^{149}Nd produced from the ^{150}Nd present in the samples. A total of 50 samples were irradiated, so that each capsule was used 2 or 3 times over the course of the 5-day experiment. Two blank samples were also irradiated under the identical conditions to determine the gamma rays originating from the trace constituents in the polyethylene and the contaminants introduced by the rabbit system.

The singles spectra were added together and the resulting spectra were analyzed for gamma-ray peak energies and areas using the LLNL code GAMANAL.⁽⁷¹⁾ Detector efficiency coefficients for use with GAMANAL were determined earlier by Lin *et al.*⁽⁹⁷⁾ The linearity coefficients needed for GAMANAL were determined from a calibration spectrum of ^{56}Co , $^{110}\text{Ag}^{\text{m}}$, ^{134}Cs , and ^{133}Ba .

B. Results and Discussion

The sum of the second 2-min count spectra (the second-slice) is plotted in Fig. VI-2. In addition to ^{148}Pr and ^{145}Ce , the following radioisotopes were produced from the other isotopes of Nd in the samples: ^{141}Nd , ^{147}Nd , ^{149}Nd , $^{144}\text{Pr}^{\text{m}}$, ^{145}Pr , and ^{146}Pr . Several more isotopes were observed from contaminants from the rabbit system: ^{20}F , ^{23}Ne , ^{24}Na , ^{25}Na , ^{27}Mg , ^{28}Al , ^{29}Al , ^{37}S , ^{38}K , and ^{56}Mn .

Figure VI-2a.

Figure VI-2b.

Figure VI-2d.

Figure VI-2e.

Figure VI-2f.

None of the blank activity resulted in serious interferences, but the ^{28}Al activity was rather strong and resulted in a substantial Compton edge from the 1779-keV peak. The second slice was plotted to better show the high-energy gamma rays, which were more apparent when the ^{16}N had completely decayed. There are many unidentified peaks in Fig. VI-2, some of which may be from the decay of ^{148}Pr but have not been previously observed. Two gamma rays, the 2086-keV peak reported by Ikeda et al. for ^{148}Pr , and the 1118-keV peak reported by Pfeiffer et al. ⁽⁸⁶⁾ for ^{145}Ce , are absent from all of the spectra. All the other gamma rays previously reported ^(83,84,86) are identified.

The energies and intensities obtained in the present work for all the gamma rays observed by either Ikeda et al. or Taylor and Singh are listed in Table VI-1. The uncertainties in the energies listed in Table VI-1 are less than 0.2 keV. In general, the intensities are in agreement with those of Taylor and Singh. The singles data reported here provide no direct method of discriminating between gamma rays from two isomers. Therefore, it is not surprising the intensities are similar to Taylor and Singh. There is indirect evidence, however, even from the singles data, that there are indeed two isomers. Assuming the decay scheme for the 2.0-min ^{148}Pr is correct, and neglecting the 451-keV activity from the 2.3-min isomer, the fraction of the 302-keV activity from the 2.0-min isomer is 37% of the total 302-keV activity. Since many of the intensities reported in the present work are 30-50% lower than those reported by Ikeda et al., it follows that if the 302-keV activity were divided as indicated between the two isomers, the

Table VI-1. Energies and relative intensities of gamma rays in the decay of ^{148}Pr produced in high-flux, 14-MeV neutron irradiations of ^{148}Nd .

<u>Energy (keV)</u>	<u>Relative intensity (%)</u>	<u>Notes</u>	<u>Energy (keV)</u>	<u>Relative intensity (%)</u>	<u>Notes</u>
246.2	1.3 $\pm$ 0.1	a,c	1248.5	4.6 $\pm$ 0.2	
301.8	100.	c	1327.3	0.36 $\pm$ 0.07	b
450.7	18.9 $\pm$ 0.6	c	1343.3	0.55 $\pm$ 0.06	
490.0	4.3 $\pm$ 0.2		1357.5	4.7 $\pm$ 0.2	
499.5	1.5 $\pm$ 0.1	b	1381.5	2.0 $\pm$ 0.1	
522.7	2.5 $\pm$ 0.2	c	1427.0	0.50 $\pm$ 0.06	
615.2	2.6 $\pm$ 0.1		1521.2	1.0 $\pm$ 0.1	
636.4	1.1 $\pm$ 0.1		1556.6	1.7 $\pm$ 0.1	
660.3	1.8 $\pm$ 0.1		1579.7	0.69 $\pm$ 0.07	
697.6	11.2 $\pm$ 0.4	c	1682.8	0.31 $\pm$ 0.03	b
721.5	2.8 $\pm$ 0.1		1726.7	0.55 $\pm$ 0.11	b
825.2	1.9 $\pm$ 0.1		1771.9	0.71 $\pm$ 0.07	
869.4	5.6 $\pm$ 0.3		1907.9	0.91 $\pm$ 0.08	
902.9	1.3 $\pm$ 0.1		1931.6	0.51 $\pm$ 0.05	
935.7	0.77 $\pm$ 0.05	c	2079.7	1.6 $\pm$ 0.1	
947.1	2.2 $\pm$ 0.1		2104.9	0.05 $\pm$ 0.03	a
999.1	0.30 $\pm$ 0.05		2130.2	1.6 $\pm$ 0.1	
1023.0	4.5 $\pm$ 0.2		2143.5	0.39 $\pm$ 0.06	
1080.6	1.1 $\pm$ 0.2	b	2223.7	0.43 $\pm$ 0.04	a
1106.4	1.1 $\pm$ 0.2	c	2542.5	0.14 $\pm$ 0.03	a
1132.3	1.9 $\pm$ 0.1	b	2629.5	0.89 $\pm$ 0.06	
1157.5	0.93 $\pm$ 0.06		2735.9	0.33 $\pm$ 0.03	
1170.9	0.82 $\pm$ 0.08		2982.8	0.31 $\pm$ 0.03	a
1209.9	4.4 $\pm$ 0.4	d			

a) Gamma-ray not reported by Taylor and Singh.

b) Gamma-ray not reported by Ikeda et al.

c) From 2.0-min isomer according to Ikeda et al.

d) Interference correction made due to ^{145}Ce .

relative intensities calculated for gamma rays from the 2.3-min isomer would agree more closely with those reported by Ikeda et al.

To expand on the points above, the 302-keV activity was divided into 2.3-min and 2.0-min components, and a second set of relative intensities was calculated based on the two-isomer decay scheme of Ikeda et al. Both sets of intensities are listed in Table V-2, along with the intensities reported by Taylor and Singh and Ikeda et al., for comparison. Gamma rays with relative intensities greater than 1% are included in Table V-2, with the exceptions of 721 and 1210 keV, which suffer interference from ^{145}Ce . The agreement between the one-isomer intensities from this work, and those of Taylor and Singh can easily be seen, although there may be a small but systematic discrepancy. Intensities from the present work are slightly greater than those reported by Taylor and Singh at low energies, and the reverse is true at high energies. Relative intensities calculated based on the two-isomer assumption agree fairly well with those of Ikeda et al., for two thirds of the gamma rays listed. Intensities marked with an asterisk are primarily due to the 2.0-min isomer, and these intensities were calculated with respect to the 451-keV peak.

As additional evidence, the relative intensities of the 451-, 1106-, and 1557-keV gamma rays, major peaks in the decay of the 2.0-min isomer not present in the decay of the other isomer, decrease from the first-slice spectrum to the second by about 20-30%. A decrease indicates that these gamma-ray activities are decaying at a faster rate than the 302-keV peak. While on first inspection, the intensities given in Table VI-1 support the decay scheme of Taylor and Singh, they are

Table VI-2. Comparisons of Relative Gamma-Ray Intensities of ^{148}Pr with Those Reported by Taylor and Singh, and Ikeda et al.

Energy (keV)	Present work (single isomer)	Taylor and Singh	Present work (two isomers)	Ikeda et al.
302	100.0	100.0	100.0	100.0
450	18.9 $\pm$ 0.6	17.1 $\pm$ 0.9	100.0	100.0*
490	4.3 $\pm$ 0.2	3.2 $\pm$ 0.3	6.8 $\pm$ 1.5	0.6 $\pm$ 0.1
499	1.5 $\pm$ 0.1	1.6 $\pm$ 0.4	2.4 $\pm$ 0.5	
523	2.5 $\pm$ 0.2	1.6 $\pm$ 0.4	14 $\pm$ 3 *	1.8 $\pm$ 0.3*
615	2.6 $\pm$ 0.1	2.0 $\pm$ 0.2	4.1 $\pm$ 1.0	3.7 $\pm$ 0.3
636	1.1 $\pm$ 0.1	1.0 $\pm$ 0.2	1.7 $\pm$ 0.2	2.4 $\pm$ 0.2
660	1.8 $\pm$ 0.1	1.2 $\pm$ 0.3	2.8 $\pm$ 0.6	2.8 $\pm$ 0.2
697	11.2 $\pm$ 0.4	11.0 $\pm$ 0.6	59 $\pm$ 15 *	80 $\pm$ 7 *
825	1.9 $\pm$ 0.1	2.2 $\pm$ 0.2	3.0 $\pm$ 0.6	2.1 $\pm$ 0.2
869	5.6 $\pm$ 0.3	6.2 $\pm$ 0.6	8.9 $\pm$ 0.9	6.1 $\pm$ 0.4
903	1.3 $\pm$ 0.1	1.2 $\pm$ 0.3	2.1 $\pm$ 0.4	2.3 $\pm$ 0.2
947	2.2 $\pm$ 0.1	2.3 $\pm$ 0.2	3.5 $\pm$ 0.7	2.3 $\pm$ 0.2
1023	4.5 $\pm$ 0.2	6.2 $\pm$ 0.6	7.2 $\pm$ 1.5	7.8 $\pm$ 0.6
1080	1.1 $\pm$ 0.2	1.3 $\pm$ 0.4	1.7 $\pm$ 0.11	
1106	1.1 $\pm$ 0.2	1.4 $\pm$ 0.4	5.7 $\pm$ 1.2 *	8.2 $\pm$ 0.7*
1132	1.9 $\pm$ 0.1	2.2 $\pm$ 0.2	3.0 $\pm$ 0.6	
1157	0.93 $\pm$ 0.06	1.6 $\pm$ 0.4	1.5 $\pm$ 0.3	1.9 $\pm$ 0.2
1170	0.82 $\pm$ 0.08	1.6 $\pm$ 0.4	1.3 $\pm$ 0.3	1.0 $\pm$ 0.1
1248	4.6 $\pm$ 0.2	6.3 $\pm$ 0.6	7.3 $\pm$ 1.5	5.3 $\pm$ 0.4
1357	4.7 $\pm$ 0.2	7.5 $\pm$ 0.8	7.5 $\pm$ 1.5	9.0 $\pm$ 0.6
1381	2.0 $\pm$ 0.1	2.9 $\pm$ 0.3	3.2 $\pm$ 0.7	3.7 $\pm$ 0.3
1521	1.0 $\pm$ 0.1	1.0 $\pm$ 0.2	1.6 $\pm$ 0.3	1.2 $\pm$ 0.1
1556	1.7 $\pm$ 0.1	2.3 $\pm$ 0.2	9.0 $\pm$ 1.8 *	9.8 $\pm$ 0.8*
2079	1.6 $\pm$ 0.1	1.9 $\pm$ 0.2	2.5 $\pm$ 0.5	

also consistent with the two-isomer decay scheme, as seen in Table V-2. Coincidence data will be required to ultimately determine the number of isomers.

Many additional gamma rays, with energies greater than 3 MeV, were observed in the singles data. Several of these gamma rays suggest that three new energy levels may be proposed for ^{148}Nd at 4180, 4115, and 3282 keV. The energies and intensities of gamma rays that may originate from these levels are listed in Table VI-3. Since many of these peaks are weak, with poorly defined centroids, and there were no energy calibration peaks above 3103 keV, the energy uncertainties for these peaks and levels are at least 1 keV. Therefore, proposed gamma-ray transitions between these levels and the levels given by Ikeda et al. were included in Table VI-3 if the gamma-ray energy was within 2 keV of the apparent transition energy. Intensities were calculated using the second-slice data because several of the peaks were obscured by the background in the first-slice spectrum. Note that the three levels in the two-isomer scheme not proposed by Taylor and Singh, 723, 1512, and 1683 keV, are levels into which several of the gamma rays in Table V-3 decay. Although coincidence data is required to confirm these energy levels, the number of gamma rays that fit into the decay scheme from these levels is strong evidence for their existence.

It is clear from the singles data from ^{148}Pr that high-flux, 14-MeV neutron activation provides an excellent opportunity to study the decay of certain important decay-heat isotopes. The high flux available from the RTNS-I allowed much greater sensitivity than had been possible

Table VI-3. High-energy gamma rays in the decay of ^{148}Pr that support the existence of three new energy levels in ^{148}Nd .

<u>Energy level (keV)</u>	<u>Transition energy (keV)</u>	<u>Final state of transition</u>	<u>Relative intensity (%)</u>
4180	4180	G.S.	0.05 ± 0.01
	3879	302	0.04 ± 0.01
	3425	753	0.03 ± 0.01
	3159	1023	0.03 ± 0.01
	2933	1249	0.04 ± 0.01
	2670	1512	very weak
	2500	1683	0.04 ± 0.02
4115	4115	G.S.	0.07 ± 0.01
	3812	302	0.03 ± 0.01
	3198	917	0.07 ± 0.02
	3092	1023	0.05 ± 0.02
	2604	1512	0.03 ± 0.02
	2434	1683	0.10 ± 0.02
	2042	2074	0.10 ± 0.02
3282	3282	G.S.	0.11 ± 0.01
	2982	302	0.26 ± 0.02
	2558	724	0.03 ± 0.02
	2257	1023	0.05 ± 0.03

previously, resulting in the observation of many heretofore unobserved gamma rays. Although coincidence data is needed to fix the final decay scheme of ^{148}Pr , many of these gamma rays fit into the decay scheme of Ikeda et al., when three levels are added. The complete decay scheme of ^{148}Pr will be established from the coincidence-data analysis currently in progress. The improved polyethylene rabbits with very low oxygen concentrations will enable the study of even shorter-lived radioisotopes. Samples of ^{82}Se and ^{100}Mo , containing no oxygen, will enable future studies of ^{82}As and ^{100}Nb .

VII. SUMMARY

The fast-neutron irradiations described in Chapters IV, V, and VI, demonstrate the usefulness of high-flux, 14-MeV neutron activation for a wide variety of applications. Improvements in the rabbit system made it possible to exploit the unique nature of the RTNS-I for the three applications presented in this report.

The multielement fast-neutron activation analysis of standard reference materials is by far the most thorough analysis of any samples to date using 14-MeV neutron activation. Procedures developed for high-flux FNAA resulted in an accurate and reliable method of elemental analysis. Four elements, Si, P, Tl, and Pb, difficult to determine by thermal-neutron activation analysis, have been determined by FNAA as part of these multielement analysis procedures. The limiting factor upon the applicability of FNAA at the RTNS-I is the cost per sample. Because samples can only be irradiated one at a time, FNAA cannot compete with TNAA in analysis cost. Further improvements in the sample irradiation system at the RTNS-I will not substantially reduce the cost of FNAA. Therefore, future use of the FNAA procedures developed here depend on availability of still more intense fast-neutron sources, such as the existing RTNS-II or anticipated experimental fusion devices. The development of such sources could result in multielement FNAA becoming a practical analytical tool in 10 to 20 years.

Fast-neutron irradiations of inertial-confinement-fusion target materials mixed with potential seed gases for direct fuel-density diagnostics have demonstrated that Br can be a suitable tracer. The

large cross section for the production of ^{78}Br allows enhanced annihilation radiation to be observed from small amounts of Br in a silicate matrix. The future for 14-MeV neutron activation diagnostics is very bright. NOVA, the next generation laser at LLNL, will cause targets to yield several orders of magnitude more neutrons than Shiva can presently produce. At that time, the accuracy of the diagnostic method will be limited by the cross-section uncertainties rather than the lack of induced activity. It will then be important that the cross sections be meticulously measured at 14.1 MeV.

The use of the high-flux RTNS-I facility for producing decay-heat isotopes for the purpose of decay-scheme studies has also been successful. Many low-intensity, high-energy gamma rays were observed because of the increased sensitivity of the RTNS-I irradiations over previous work. Activity for contaminants in the rabbits did not adversely affect the ability to observe very weak peaks. Thus, the only requirement for studying shorter-lived radioisotopes, such as ^{100}Nb and ^{82}As , is that the samples themselves contain no oxygen.

Improvement of the rabbit system at the RTNS-I has allowed high-flux, fast-neutron activation to become a valuable tool for these applications. Although there is some question as to whether FNAA will prove to be practical, there are many applications in advanced ICF diagnostics and fission decay-heat studies of current interest. It is hoped that the facilities and the procedures developed in the present work will be further utilized in the future.

REFERENCES

1. Anthony Foderaro, The Elements of Neutron Interaction Theory, The MIT Press, Cambridge, Massachusetts 1971.
2. S. Pearlstein, "Analysis of Neutron Emission Spectra from 14-MeV Neutron Reactions", Nucl. Sci. Eng. 68, 55-60 (1978).
3. B. Strohmaier, M. Uhl, and W.K. Matthes, "Application of Nuclear Reaction Models for Neutron Nuclear Data Evaluation: Statistical and Optical Model Calculations for $^{134-138}\text{Ba}$ ", Nucl. Sci. Eng. 65, 368-384 (1978).
4. M. Blann, "A Priori Pre-Equilibrium Decay Models", Nucl. Phys. A213, 570-588 (1973).
5. L.F. Hansen, S.M. Grimes, R.J. Howerton, and J.D. Anderson, "Calculations of the Neutron Emission Spectra from Materials Bombarded with 14-MeV Neutrons", Nucl. Sci. Eng. 61, 201-211 (1976).
6. S.M. Grimes, R.C. Haight, K.R. Alvar, H.H. Barschall, R.R. Borchers, "Charged-Particle Emission in Reactions of 15-MeV Neutrons with Isotopes of Chromium, Iron, Nickel, and Copper", Phys. Rev. C 19, 2127-2137 (1979).
7. M. Bormann, H. Neuert, and W. Scobel, "Tables and Graphs of Cross-Sections for (n,p), (n, γ), and (n,2n) Reactions in the Neutron Energy Region 1-37 MeV", from Handbook on Nuclear Activation Cross-Sections, Technical Reports Series No. 156, 87-272 (IAEA, Vienna, 1974).
8. Wen-deh Lu, N. Ranakumar, and R.W. Fink, "Activation Cross Sections for (n,2n) Reactions at 14.4 MeV in the Region Z=40-60: Precision Measurements and Systematics", Phys. Rev. C 1, 350-357 (1970).
9. S.M. Qaim, "Activation Cross Sections, Isomeric Cross-Section Ratios, and Systematics of (n,2n) Reactions at 14-15 MeV", Nucl. Phys. A185, 614-624 (1972).
10. S.M. Grimes, R.C. Haight, and J.D. Anderson, "Measurement of Sub-Coulomb Barrier Charged Particles Emitted from Aluminum and Titanium Bombarded by 15-MeV Neutrons", Nucl. Sci. Eng. 62, 187-194 (1977).
11. S.M. Qaim, "A Survey of Fast-Neutron Induced Reaction Cross-Section Data", Proc. Conf. on Nuclear Cross Sections and Technology, Vol. I, Washington, D.C., March 3-7, 1975.

12. Wen-deh Lu and R.W. Fink, "Applicability of the Constant-Nuclear-Temperature Approximation in Statistical-Model Calculations of Neutron Cross Sections at 14 MeV for Medium-Z Nuclei", *Phys. Rev. C* 4, 1173-1181 (1971).
13. N.I. Molla and S.M. Qaim, "A Systematic Study of (n,p) Reactions at 14 MeV", *Nucl. Phys.* A283, 269-288 (1977).
14. V.N. Levkovskii, "Empirical Behavior of the (n,p) Cross Section for 14-15 MeV Neutrons", *Soviet Physics: JETP* 18, 213-217 (1964).
15. W.W. Givens, W.R. Mills, and R.L. Caldwell, "Cyclic Activation Analysis", *Nucl. Instr. Meth.* 80, 95-103 (1970).
16. J. Hoste, J. Op de Beeck, R. Gijbels, F. Adams, P. Van den Winkle, and D. DeSoete, Instrumental and Radiochemical Activation Analysis, (Chemical Rubber Company Press, Cleveland, 1971).
17. S.S. Nargolwalla and E.P. Przybylowicz, Activation Analysis with Neutron Generators, (John Wiley & Sons, New York, 1973).
18. S.J. Bame, Jr. and J.E. Perry, Jr., " $T(d,n)He^4$ Reaction", *Phys. Rev.* 107, 1616-1620 (1957).
19. J. Benevinste and J. Zenger, "Information on the Neutrons Produced in the $H^3(d,n)He^4$ Reaction", Lawrence Radiation Laboratory, UCRL-4266, (1954).
20. J.L. Fowler and J.E. Brolley, "Monoenergetic Neutron Techniques in the 10- to 30-MeV Range", *Rev. Mod. Phys.* 28, 103-134 (1956).
21. E. Ricci, "Output Spectrum from 14-MeV Neutron Generators", *J. Inorg. Nucl. Chem.* 27, 41-52 (1964).
22. J.D. Seagrave, E.R. Grayes, S.J. Hipwood, and S.J. McDole, " $D(d,n)He^3$ and $T(d,n)He^4$ Neutron Sources", Los Alamos Informal Report, LAMS-2162 (1957).
23. J.E. Brolley, Jr. and J.L. Fowler, "Monoenergetic Neutron Sources: Reactions with Light Nuclei", in Fast Neutron Physics, Vol. 1, J. B. Marion and J. L. Fowler, editors, (Interscience, New York, 1960).
24. J.T. Prud'homme, Texas Corporation Neutron Generators, (Texas Nuclear Corporation, Austin, Texas, 1962).
25. R. Steinberg and D.L. Alger, "A Solution to Tritium Pump

- Contamination for Small Accelerators", NASA Technical Memorandum TMX-52578, Lewis Research Center, Cleveland, Ohio (1969).
26. H.-U. Fanger, R. Pepelnik, and W. Michaelis, "Fast-Neutron Activation Analysis with Short-Lived Nuclides", J. Radioanal. Chem. 61, 147-163 (1981).
 27. P. Kleinheins, "Low Energy Accelerators", Kerntechnik 11, 684-686 (1969).
 28. Technical Staffs of High Voltage Engineering Corp., Burlington, Mass. and Applied Radiation Corp., Walnut Creek., Cal., "Three High-Power Accelerators for Radiation Processing", Nucleonics 18(8), 54-57 (1960).
 29. R. Booth, J.C. Davis, C.L. Hanson, J.L. Held, C.M. Logan, J.E. Osher, R.A. Nickerson, B.A. Pohl, and B.J. Schumacher, "Rotating Target Neutron Generators", Nucl. Instr. Meth. 145, 25-39 (1977).
 30. R. Booth, "High-Speed Rotating Vacuum Seal", Nucl. Instr. Meth. 59, 131-135 (1968).
 31. R. Booth and H.H. Barschall, "Tritium Target for Intense Neutron Source", Nucl. Instr. Meth. 99, 1-4 (1972).
 32. C. Wong, J.D. Anderson, P. Brown, L.F. Hansen, J.L. Kammerdiener, C. Logan, and B. Pohl, "Livermore Pulsed Sphere Program: Program Summary Through July 1971", Lawrence Livermore Laboratory, UCRL-51144 (1971).
 33. E. Goldberg, R. Griffith, and C. Logan, "Peak Flux Measurements of the LLL High-Intensity 14-MeV Neutron Source", Lawrence Livermore Laboratory, UCRL-51317 (1972).
 34. R. Van Konynenburg, "Neutron Dosimetry (14 MeV) for Foil Samples Irradiated with the Lawrence Livermore Laboratory Rotating Target Neutron Source", Lawrence Livermore Laboratory, UCRL 51393 Rev. 1 (1972).
 35. Paul Kruger, Principles of Activation Analysis, (Wiley-Interscience, New York, 1972).
 36. D. DeSoete, R Gijbels, and J. Hoste, Neutron Activation Analysis, (John Wiley & Sons, New York, 1974).
 37. R.L. Heath, "Gamma-Ray Spectrometry and Automated Data Analysis Systems for Activation Analysis", Modern Trends in Activation Analysis, Vol. II, James R. DeVoe, Editor, (National Bureau of Standards Special Publication 312, June 1969).

38. W.H. Zoller and G.E. Gordon, "Instrumental Neutron Activation Analysis of Atmospheric Pollutants Utilizing Ge(Li) Gamma-Ray Detectors", *Anal. Chem.* 42, 257-265 (1970).
39. P.K. Hopke, E.S. Gladney, G.E. Gordon, W.H. Zoller, and A.G. Jones, "The Use of Multivariate Analysis to Identify Sources of Selected Elements in the Boston Urban Aerosol", *Atmos. Environ.* 10, 1015-1025 (1976).
40. R.A. Nadkarni and W.D. Ehmann, "Determination of Trace Elements in Biological Standard Kale by Neutron Activation Analysis", *J. Radioanal. Chem.* 3, 175-185 (1969).
41. D.H. Klein, A.W. Andren, J.A. Carter, J.F. Emery, C. Feldman, W. Fulkerson, W.S. Lyon, J.C. Ogle, Y. Talmi, R.I. Van Hook, and N. Bolton, "Pathways of Thirty-seven Trace Elements Through Coal-Fired Power Plant", *Environ. Sci. Technol.* 9, 973-979 (1975).
42. J.M. Ondov, R.C. Ragaini, and A.H. Biermann, "Elemental Emissions from a Coal-Fired Power Plant. Comparison of a Venturi Wet Scrubber System with a Cold-Side Electrostatic Precipitator", *Environ. Sci. Technol.* 13, 598-607 (1979).
43. 1976 International Conference: Modern Trends in Activation Analysis, Munich, 13-17 September 1976, from *J. Radioanal. Chem.* 37(1977).
44. J.M. Ondov, W.H. Zoller, K. Olmez, N.K. Araz, G.E. Gordon, L.A. Ranticelli, K.H. Able, R.H. Filby, K.R. Shah, and R.C. Ragaini, "Elemental Concentrations in the National Bureau of Standards' Environmental Coal and Fly Ash Standard Reference Materials", *Anal. Chem.* 47, 1102-1109 (1975).
45. M.S. Germani, Inci Gokmen, A.C. Sigleo, G.S. Kowalczyk, Ilhan Olmez, A.M. Small, D.L. Anderson, M.P. Failey, M.C. Gulovali, C.E. Choquette, E.A. Lepel, G.E. Gordon, and W.H. Zoller, "Concentrations of Elements in the National Bureau of Standards' Bituminous and Subbituminous Coal Standard Reference Materials", *Anal. Chem.* 52, 240-245 (1980).
46. Ernest Gladney, "Compilation of Elemental Concentration Data for NBS Biological and Environmental Standard Reference Materials", Los Alamos Scientific Laboratory Informal Report, LA-8438-MS (1980).
47. E.S. Gladney, D.B. Curtis, D.R. Perrin, J.W. Owens, and W.E. Goode, "Nuclear Techniques for the Chemical Analysis of Environmental Materials", Los Alamos Scientific Laboratory Informal Report, LA-8192-MS, January 1980.

48. J.R. McGinley and E.A. Schweikert, "Multielement Charged Particle Activation Analysis with X-Ray Counting", Anal. Chem. 48, 429-435 (1976).
49. A. Chattopadhyay and R.E. Jervis, "Multielement Determination of Market-Garden Soils by Instrumental Photon Activation Analysis", Anal. Chem. 46, 1630-1639 (1974).
50. N.F. Mangelson, M.W. Hill, K.K. Nielson, D.J. Eatough, J.J. Christensen, R.M. Izatt, and D.O. Richards, "Proton Induced X-Ray Emission Analysis of Pima Indian Autopsy Tissues", Anal. Chem. 51, 1187-1194 (1979).
51. R.E. Giauque, R.B. Garrett, and L.Y. Goda, "Determination of Forty Elements in Geochemical Samples and Coal Fly Ash by X-Ray Fluorescence Spectrometry", Anal. Chem. 49, 1012-1017 (1977).
52. M.P. Failey, D.L. Anderson, W.H. Zoller, G.E. Gordon, and R.M. Lindstrom, "Neutron-Capture Prompt Gamma-Ray Activation Analysis for Multielement Determination in Complex Samples", Anal. Chem. 51, 2209-2221 (1979).
53. J. Hoste, D. DeSoete, and A. Speecke, "The Determination of Oxygen in Metals by 14-MeV Neutron Activation Analysis", European Atomic Energy Community report, EUR 3565e, (Brussels, 1967).
54. G. Beurton and R. Pillon, "Experimental Improvements of the Sensitivity of Neutron Activation Analysis of Oxygen in Metals", J. Radioanal. Chem. 40, 189-201 (1977).
55. J.W. Morgan and W.D. Ehmann, "14 MeV Neutron Activation Analysis of Rocks and Meteorites", Proc. of the NATO Advanced Study Institute - Activation Analysis in Geochemistry and Cosmochemistry, Kjeller, Norway, Sept. 7-12, 1970 (Universitetsforlaget, Oslo, 1971).
56. L.L. Chyi, W.D. James, W.D. Ehmann, G.H. Sun, and C.E. Hamrin, Jr., "14 MeV Activation Analysis of Oxygen and Nitrogen in Coal", Proc. Fourth Conf. on the Scientific and Industrial Applications of Small Accelerators, North Texas State University, October 1976.
57. J.R. Vogt and W.D. Ehmann, "The Nondestructive Determination of Silicon and Oxygen in Meteorites by Fast-Neutron Activation Analysis", Proc. Texas A&M Conference on Modern Trends in Activation Analysis, (McGraw-Hill, New York, 1966).
58. P.J. Raimondi, "The Use of Fast Neutron Activation Analysis

for the Determination of the Oxygen Content of Coal and Flyash", M.S. Thesis, University of Illinois (1981).

59. K. Randle and E. Hartman, "The Determination of Barium in Barytes Ores by 14 MeV Neutron Activation Analysis", Radiochem. Radioanal. Letters 32(5-6), 239-246 (1978).
60. E. Eisenbarth and P. Hille, "A Nondestructive Method for Age Determination of Fossile Bone", J. radioanal. Chem. 40, 203-211 (1977).
61. CH. Segebade, "Simultane Instrumentalle Analyse Einger Haupt-und Nebenelemente In Calciumcarbid Durch Aktivierungsanalyse Mit 14 MeV-Neutronen", J. Radioanal. Chem. 43, 243-251 (1978).
62. A. Faanhof, H.A. Das, and J. Zonderhuis, "Possibilities of the Elemental Analysis of Dry Biological Material by Fast Neutron Activation Analysis", J. Radioanal. Chem. 56, 173-184 (1980).
63. R.C. Ragaini, R.E. Heft, and D. Garvis, "Neutron Activation Analysis at the Livermore Pool-Type Reactor for the Environmental Research Program", Lawrence Livermore Laboratory Report UCRL-52092, 1976.
64. R.E. Heft, "A Consistant Set of Nuclear Parameters Values for Absolute INAA", Proc. of Topical Meeting on Atomic and Nuclear Methods in Fossile Energy Research, Mayaguez, Puerto Rico, December 1-4, 1981.
65. V. Krivan and K. Krivan, "Tabulation of Calculated Data on Primary Interferences in 14-MeV Neutron Activation Analysis", J. Radioanal. Chem. 29, 145-173 (1976).
66. R.E. Williams, P.K. Hopke, and R.A. Meyer, "Procedures for Multielement Analysis Using High-Flux Fast-Neutron Activation", Proc. 1981 Conf. on Modern Trends in Activation Analysis, Toronto, June 15-19, 1981, in press.
67. R.E. Williams, P.K. Hopke, and R.A. Meyer, "Trace Multielement Analysis Using High-Flux Fast-Neutron Activation", J. Radioanal. Chem. 63, 187-199 (1981).
68. F.A. Iddings, "A Study of Flux Monitoring for Instrumental Neutron Activation Analysis", Anal. Chim. Acta 31, 206-212 (1964).
69. N.E. Hertel and B.W. Wehring, "Absolute Monitoring of DD and DT Neutron Fluences Using the Associated-Particle Technique", Nucl. Instr. Meth. 172, 501-506 (1980).

70. R.L. Heath, Proc. Texas A&M Conf. on Modern Trends in Activation Analysis, Addendum, (McGraw-Hill, New York, 1966).
71. R. Gunnink and J.B. Niday, "Computerized Quantitative Analysis by Gamma-Ray Spectroscopy, Lawrence Livermore Laboratory Report UCRL-51061, Vol. I (1972).
72. Glenn F. Knoll, Radiation Detection and Measurement, (John Wiley & Sons, New York, 1979).
73. B.A. Roscoe and A.K. Furr, "Time Dependent Deadtime and Pile-up Corrections for Gamma Ray Spectroscopy", Nucl. instr. Meth. 140, 401-404 (1977).
74. E. Storm and H.I. Isreal, "Photon Cross Sections from 0.001 to 100 MeV for Elements 1 through 100", Los Alamos Scientific Laboratory Report LA-3753 (1967).
75. N. Stalnaker, M. Kahn, and B. Kenna, "Fast Neutron Activation: Sample Preparation Utilizing Graphite as a Diluent", J. Radioanal. Chem. 7, 81-87 (1971).
76. M. Pollermann, K. Zietlow, and K. Nagorny, "Activation Analysis Studies with Fast Neutrons of the Oxygen Contents of Various Polyethylene Plastics", Kerntechnik 15, 473-475 (1973).
77. W.W. Black, "Application of Correlation Techniques to Isolate Structure in Experimental Data", Nucl. Instr. Meth. 71, 317-327 (1969).
78. F.W. Walker, G.J. Kirouac, and F.M. Rourke, Chart of the Nuclides, Twelfth Edition, (Knolls Atomic Power Laboratory, Schenectady, N.Y., General Electric Company 1977).
79. C.M. Lederer and V.S. Shirley, Table of Isotopes, Seventh Edition (John Wiley & Sons, New York 1978).
80. Erlan S. Bliss, "Alignment of the Twenty-Beam Shiva Laser", from Energy and Technology Review, UCRL-52000-78-12, 1-10 (1978)
81. Stephen M. Lane and E. Michael Campbell, "Density Diagnosis of Fusion Targets by Neutron Activation", from Energy Technology and Review, UCRL-52000-80-1, 10-18 (1980).
82. J. Nuckolls, L. Wood, A. Thiesson, and G. Zimmerman, "Laser Compression of Matter to Super-High Densities: Thermonuclear (CTR) Applications", Nature 239, 139-142 (1972).

83. K.A. Brueckner and S. Jorna, "Laser Driven Fusion", Rev. Mod. Phys. 46, 325-367 (1974).
84. E.M. Campbell, W.M. Ploeger, P.H. Lee, and S.M. Lane, "Exploding-Pusher-Tamper Areal Density Measurement by Neutron Activation", Appl. Phys. Lett. 36(12), 965-967 (1980).
85. E.M. Campbell, H.G. Hicks, W.C. Mead, L.M. Coleman, C.W. Hatcher, J.H. Dellis, M.J. Boyle, J.T. Larson, and S.M. Lane, "Use of a Radioactive Tracer to Determine the Fraction of Fusion Target Debris Collected", J. Appl. Phys. 51(12), 6065-6067 (1980).
86. S.M. Lane, E.M. Campbell, and C. Bennett, "Measurement of DT Neutron-Induced Activity in Glass Microshell Laser Fusion Targets", Appl. Phys. Lett. 37(7), 600-601 (1980).
87. J.M. Auerbach, W.C. Mead, E.M. Campbell, D.L. Mathews, D.S. Bailey, C.W. Hatcher, L.N. Koppel, S.M. Lane, P.H.Y. Lee, K.R. Manes, G. McClellen, D.W. Phillion, R.H. Price, V.C. Rupert, V.W. Slivinski, and C.D. Swift, "Compression of Polymer-Coated Laser-Fusion Targets to Ten Times Liquid DT Density", Phys. Rev. Lett. 44, 1672-1675 (1980).
88. E.M. Campbell, S.M. Lane, Y.L. Pan, J.T. Larson, R.J. Wahl, and R.H. Price, "Determination of the Fuel Density-Radius Product of Inertial Confinement Fusion Targets by Neutron Activation", J. Appl. Phys. 51(12), 6062-6064 (1980).
89. R.L. Morrison, "Addition of Bromine as a Diagnostic Gas to Inertial Confinement Fusion Target Microspheres", Proc. 27th National American Vacuum Society Symposium, Detroit, Michigan, October 14-17, 1980.
90. Stephen M. Lane, Lawrence Livermore National Laboratory, private communication.
91. P. Venugopala Rao and R.W. Fink, "Neutron Reaction Cross Sections at 14.4 MeV", Phys. Rev. 154, 1023-1028 (1967).
92. Richard A. Meyer, Lawrence Livermore National Laboratory, private communication.
93. R.A. Meyer, "Rapid Nuclear Chemistry for Determining the Properties of Short-Lived Fission Products", from Energy and Technology Review, UCRL-52000-78-9, 19-29 (1978).
94. R.A. Meyer, "Studies of Short-Lived Fission Products", from Energy and Technology Review, UCRL-52000-80-6, 8-16 (1980).

95. Y. Ikeda, H. Yamamoto, K. Kawade, T. Takeuchi, T. Katoh, and T. Nagahara, "Decay of the ^{148}Pr Isomers to Levels of ^{148}Nd ", J. Phys. Soc. Jpn. 47, 1039-1046 (1979).
96. H.W. Taylor and B. Singh, "The Decay of 2.8 Min ^{145}Ce and 2.3 Min ^{148}Pr ", J. Phys. G: Nucl. Phys. 5, 1445-1460 (1979).
97. J. Lin, E.A. Henry, and R.A. Meyer, "Detection Efficiency of Ge(Li) and HPGe Detectors for Gamma Rays up to 10 MeV, Lawrence Livermore National Laboratory, UCRL-84536 (1980).
98. B. Pfeiffer, F. Schussler, J. Blachot, S.J. Feenstra, J. van Klinken, H. Lawin, E. Monnard, G. Sadler, H. Wollnik, K.D. Wunsch, and the JOSEF-, LOHENGRIN-, and OTIS-collaboration, "Nuclear Spectroscopy of Neutron-Rich A=145 Nuclei", Z. Physik A 287, 191-210 (1978).

APPENDIX

A PNEUMATIC SAMPLE-TRANSFER SYSTEM FOR USE WITH
THE LAWRENCE LIVERMORE NATIONAL LABORATORY
ROTATING TARGET NEUTRON SOURCE (RTNS-1)

PREFACE

The Appendix is a report, written for internal distribution at the Lawrence Livermore National Laboratory (UCRL-19169), documenting the modifications in the rabbit system at the RTNS-I. A description of the system and instructions for its use are presented.

UCRL-19161

A PNEUMATIC SAMPLE-TRANSFER SYSTEM FOR USE WITH
THE LAWRENCE LIVERMORE NATIONAL LABORATORY
ROTATING TARGET NEUTRON SOURCE (RTNS-1)

Robert E. Williams
Nuclear Chemistry Division

July 1981

This is an informal report intended primarily for internal or limited external distribution. The opinions and conclusions stated are those of the author and may or may not be those of the Laboratory.

Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under Contract W-7405-Eng-48.

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial products, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government thereof, and shall not be used for advertising or product endorsement purposes.

INTRODUCTION

A pneumatic sample-transfer system is needed to be able to rapidly retrieve samples irradiated with 14-MeV neutrons at the Rotating Target Neutron Source (RTNS-I). The rabbit system, already in place for many years, has been refurbished with modern system components controlled by an LSI-11 minicomputer. Samples can now be counted three seconds after an irradiation.

There are many uses for this expanded 14-MeV neutron activation capability. Several fission products difficult to isolate from mixed fission fragments can be produced instead through (n,p) or (n, α) reactions with stable isotopes. Mass-separated samples of Nd, Mo, and Se, for example, can be irradiated to produce Pr, Nb, and As radionuclides sufficient for decay scheme studies. The system may also be used for multielement fast-neutron activation analysis because the neutron flux is greater than 2×10^{11} n/cm²-sec. Single element analyses of Si and O are also possible. Finally, measurements of fast-neutron cross sections producing short-lived activation products can be performed with this system.

A description of the rabbit system and instructions for its use are presented in this report. Users should contact Gene Henry in the Nuclear Chemistry Division; he is responsible for the use of the facility.

FAST-NEUTRON IRRADIATIONS

A sample-transfer system is absolutely essential if one is to irradiate many samples with 14-MeV neutrons at the RTNS-I. Not only is it very inconvenient to shut down the accelerator and go into the target pit to retrieve samples, constant repetition of such a practice poses an extreme radiation hazard. In addition, the rabbit system is able to retrieve samples in just three seconds, making possible measurements of short-lived radioisotopes. For these reasons, the rabbit system was installed at the RTNS-I in the late 1960's and has twice been modified.

The system consists of the rabbit loader, an irradiation station, and the rabbit dropout. The sample loader, pictured in Fig. 1, also has a rabbit catcher and plunger to position a sample before a Ge(Li) detector. From this point, the rabbit may easily be returned to the RTNS-I for a second irradiation. Thus, samples may be cycled back and forth for irradiation and counting as often as needed. The irradiation station, Fig. 2, is colinear with the deuteron beam, and can be positioned about 0.5 mm from the rotating target. Rabbits are held in place by air pressure, and in the event of a rabbit system malfunction, the rabbit will fall away from the neutron source. Rabbits are removed from the system at the sample dropout. They travel between the sample loader and the irradiation station through 65 m of polyethylene tubing. Figure 3 is a schematic of the entire system.

Originally, small incandescent lamps and photosensors were used to detect the arrival of a rabbit, signaling to shut off the air supply. Because these controls grew very unreliable, an LSI-11 was incorporated and software developed to control the system. Infra-red photosensors were installed also,

Figure 1. Rabbit loader and plunger. A rabbit is placed in the loader (circular section) with the sample facing to the left, and then sent to the RTNS-I. If it is to be counted, it is returned to the plunger at the end of the line and positioned in front of the detector. An infra-red sensor is shown on the far left.

Figure 2. The irradiation station adjacent to the RTNS-I target assembly.

Figure 3. Schematic of the rabbit system showing the location of major system components, valves (lettered), and sensors (S1-5).

because they are not adversely affected by dust as were the visible-light sensors, and because the infra-red sensors easily clamp around the outside of the polyethylene tube. The above modifications, completed in November 1980, have resulted in a very reliable system.

Rabbits remaining from the original system also were the source of problems. Lids, which snapped over the end of the rabbit to hold the samples in place, frequently were removed in transit, getting stuck in the tubing and sometimes spilling the samples. The design of these rabbits was complex and fabrication costs were high. A new rabbit was designed, therefore, with a snap-in cap (yet to fail in 2500 cycles) and a more simple design. Sample capsules, somewhat smaller than those in use with earlier rabbits, are about 1.4 cm in diameter and 0.1 to 0.5 cm thick. The largest capsule, with a sample volume of, 0.7 cm^3 , is adequate for most purposes. A rabbit, cap, and sample capsule are shown in Fig. 4.

Low-density polyethylene was used for the rabbits because it contains (in principle) no elements with proton numbers greater than 6, and because the low-density polyethylene is much less brittle than high-density polyethylene, which tended to break apart in the system. For analysis of short-lived radioisotopes, it is crucial that there be little oxygen in the polyethylene. Upon 14-MeV neutron irradiation, ^{16}N is produced from ^{16}O . The short half life, 7.2 s, and very high-energy gamma ray, 6.1 MeV, of ^{16}N can cause great problems in measurements of short-lived activities. Blank rabbits are not entirely clean, however, as ^{28}Al and ^{27}Mg activity, from Si and Al, are easily detected after short irradiations. Most of the contamination is on the outside of the rabbits, but there is activity from blank sample capsules indicating trace quantities of Al and Si on or in the polyethylene capsules.

Figure 4. Rabbit body, sample capsule, and snap-in cap. The scale pictured is calibrated in cm.

Polyethylene rods were purchased from Cadillac Plastics and the rabbits were machined by Beimer's Machine Works in Pleasanton (PO # 1503101, Job Order 639233); contact Joe Bohac (Nuclear Chemistry Division) for details.

The irradiation geometry is depicted in Fig. 5. About 0.5 cm separates the sample from the actual neutron source, the deuteron beam spot on the rotating target. Although the maximum fast-neutron flux from the RTNS-I is about 2.0×10^{12} n/cm²-s, the flux decreases rapidly with the distance (1,2). It was therefore necessary to measure the flux through the sample volume. A capsule filled with Fe₂O₃ was irradiated producing ⁵⁶Mn. Using a value of 103 ± 6 mb for the ⁵⁶Fe(n, p) ⁵⁶Mn cross section at 14.9 MeV, the average flux was determined to be 2.0×10^{11} n/cm²-s at a RTNS-I source strength of 2.7×10^{12} n/s. The average neutron energy in the forward direction (along the beam line) is 14.9 MeV. Since many fast-neutron cross sections vary significantly between 13 and 15 MeV (3), care must be taken when employing nominally 14-MeV neutron cross-section data.

SYSTEM DESCRIPTION

A rabbit is placed in the loader, with the sample facing forward, and sent to the RTNS-I target by activating Valve A while the default position of Valve B is the exhaust. It is held in place during irradiation by low pressure flow through Valve E. To return the rabbit, Valve B is actuated with Valves A and E in the exhaust position. A switch (activated by Valve G) allows the removal of the rabbit from the system at the dropout or placement of the sample before a detector in the plunger, where it can be cycled through another irradiation. Valves H and J operate the dropout and the plunger, respectively.

Figure 5. Details of the 14-MeV neutron irradiation geometry utilizing the RTNS-I rabbit system.

Five sensors are used by computer programs operating the system to detect the presence of a rabbit and actuate valves accordingly. Sensors 1 through 4 are infra-red photosensors that clamp around the outside of the rabbit tube, and can detect a rabbit through the polyethylene walls. Two sensors are at the target end; a backup exists to avoid replacing a sensor in a high radiation field. Sensor 5 is a microswitch activated only when a rabbit has been placed before the detector.

An LSI-11 has been programmed in PASCAL to operate the rabbit system in either a user-controlled mode or an automated cyclic-activation mode. Documentation for the LSI-11 and its interface to the system valves is available from the Nuclear Chemistry Division Electronics Engineering Shop. The computer controls all the valves, interrogates the sensors, sets irradiation and counting times, and gates the detector when the count should begin according to whether or not the irradiation sequence was correctly executed. Any gamma-ray spectrometer may be used; the only interface between the LSI-11 and the spectrometer is the +5 volt gate.

System software is described in Appendix C. Instructions in the following section will enable a user to start the LSI-11 and operate the rabbit system without consulting this appendix. Familiarity with the RT-11 operating system is required to make any changes in the software. Digital Equipment Corp. provides many manuals available at the LLNL Small Computer User's Library. Most useful among these are the Introduction to RT-11 and the RT-11 User's Guide. The PASCAL implementation is described in the QMSI PASCAL-1 Manual, Version 1.1 for RT-11.

INSTRUCTIONS

Prior to operation of the Rabbit System, the accelerator operators should be notified. The system should be thoroughly tested before it is moved into position at the RTNS-I target. A small distance, about 0.5 mm, should separate the rabbit irradiation end from the target so vibrations will not cause the end cap to touch the target. To operate the system, apply the air pressure and electric power, and start the LSI-11.

A. Apply Air Pressure

Air pressure in the rabbit delivery line must NEVER exceed 35 psig as measured by the gauge atop the source tank under the stairs in the RTNS-I target pit. The large regulator on the supply line to the tank is used to adjust the pressure. Before each experiment, the trap at the source end of the supply line should be cleared of rust accumulated from the Building 212 supply. The operators should be notified about any operations in the target pit.

In addition to the rabbit line, three other valves are air-pressure actuated. The solenoid valve at the plunger should receive about 10 psig. Both the rabbit line switch (on the west wall) and the dropout ball-valve need about 40 psig. The regulator for the switch is located outside the trailer where there is a valve to supply all the valves in T940.

B. Electric Power

Power to T940 is turned on at the circuit breaker inside the door to the left. All of the rabbit system valves draw power from a relay box on the west

wall of T940. The computer terminal, sensor multiplexor, valve controller, floppy-disk drive, and teletype are all plugged into the LSI-11 rack.

C. LSI-11 Rabbit System Controller

After turning on the terminal and the disk drive, place a "PASCAL System Disk" in Drive 0 and a "PASCAL Work Disk" in Drive 1. Start the LSI-11 with the power button located on its back panel. The terminal should respond with "\$". To load the operating system into the LSI-11, i.e., to Bootstrap the system, type "DX" followed by return. The user should have some experience with RT-11 to better understand the system. (If the LSI-11 does not respond with a "\$" initially, type 173000G. If there is still no "\$", turn off the LSI-11, open its container and check to see that all the boards are firmly connected to the processor back plane and that all cables to peripherals are connected. If it fails again seek professional help.)

D. Software

Each work disk should contain the Rabbit System software; for a list type DIR. Programs that run the system are in the following files:

1. BYHAND.SAV - All actions are user-initiated. Sensors are ignored.
BYHAND is used to check the air pressure needed to send rabbits, and may be useful in the event a rabbit is stuck.
2. MANUAL.SAV - The sensors are used to shut off the air when a rabbit reaches its destination. Send and return commands are required from the user. MANUAL provides a total system check-out and is the best program for long irradiations.

3. CYCLE.SAV - The rabbit is automatically cycled through a preset number of irradiation/counting sequences. A file, FIL.TXT, is created to store irradiation and counting times. A failure of any kind in a cycle will interrupt the series and ask the user whether or not to continue.
4. CYCLE6.SAV - A count-start pulse replaced the gate in CYCLE for use with the NOVA analyzer. Otherwise, CYCLE6 is the same as CYCLE.
5. CYCLES.SAV - No file is created by CYCLES, which enables restarting another series of cycles without ending the program.
6. ACYCLE.SAV - As in CYCLE, irradiation and counting times are set, but the user initiates each cycle on command. ACYCLE also creates FIL.TXT.
7. ZCYCLE.SAV - ZCYCLE is initialized to give a start pulse also, but is otherwise identical to ACYCLE.

To run one of the above programs, *****.SAV, type RUN ***** . All the programs are user-interactive; necessary actions are prompted by queries. At several points in each program, the user may abort the procedure. In the event of a disaster, type "CTRL-C" twice to terminate any program. Read the section about On-Line Debugging to be able to shut off any valves left activated following CTRL-C. Or, of course, turn off all the valves at the relay box power switch.

(Several other files are related to the rabbit system:

1. *****.PAS - Over twenty PASCAL source listings of the main programs and external procedures needed to change the programs.

2. *****.OBJ -

Compiled versions of the above ready to be linked together if there are NO changes in the global variables.

None of the above is required to execute the four rabbit controller programs so they need not appear on the user's PASCAL Work Disk.)

E. Sending Rabbits

Cycle the system without a rabbit first, testing all the components thoroughly. Snap a cap into a rabbit and place it in the sample loader. Be certain the cover of the loader is secure before sending a rabbit. Run the program of your choice; all programs prompt the user for instructions.

CONCLUSION

Incorporation of an LSI-11 and infra-red photo sensors have greatly increased the reliability of the RTNS-I rabbit system. The instructions describe the normal operations of the system; consult the appendix for trouble shooting hints. Before attempting any irradiations, consult the Operational Safety Procedure 212.11 for T940 and the Hazards Control technician for Building 212.

ACKNOWLEDGEMENTS

The following people have contributed much to improving the rabbit system: Howard Spracklen, Dave Tinoko, and Ken DeWitt, for their help with the LSI-11 and controls; Dennis Chakedis and Gary Power for making modification to the rabbit lines; Richard Jesse for help with the software needed to utilize the sensor multiplexor; Dick Peterson for help in rabbit design. The author appreciates their help.

The author particularly wishes to thank Dick Meyer and Gene Henry for their support and assistance in this effort.

REFERENCES

1. Richard A. Van Konynenburg, "Neutron Dosimetry (14 MeV) for Foil Samples Irradiated with the Lawrence Livermore National Laboratory Rotating Target Neutron Source", UCRL-51393 Rev. 1 (1974).
2. E. Goldberg, R. Griffith, and C. Logan, " Peak Flux Measurements of the LLNL High-Intensity 14-MeV Neutron Source", UCRL-51317 (1972).
3. M. Bormann, H. Neuert, and W. Scobel, "Tables and Graphs of Cross-Sections for (n,p), (n, α), and (n,2n) Reactions in the Neutron Energy Region 1-37 MeV", Handbook on Nuclear Activation Cross-Sections, IAEA Technical Reports Series No. 156, Vienna, 1974.
4. Peter Grogono, Programming in PASCAL (Revised Edition), Addison-Wesly Publishing Company, Inc., Reading, Massachusetts, 1980.

APPENDIX A: ON-LINE DEBUGGING TECHNIQUE (ODT)

ODT is a powerful tool allowing a user to step through the execution of a program, monitoring machine language by displaying memory contents. Consult the Introduction to RT-11 for a complete description of ODT. For the purpose of manipulating the rabbit system by changing the contents of a memory location, one needs to know only a few commands.

At any time, the contents of a memory location may be displayed. Simply type "BREAK" and the LSI-11 interrupts whatever it is doing and responds with "@". The user responds with the location to be displayed followed by "/", which prints the contents of the location and opens it so the user may enter new contents. Whether or not new contents are entered, "RETURN" closes the location and the LSI-11 prompts again with "@". A sequence of locations can be examined using "LINE FEED", which closes a location and opens the next.

The major use of ODT with the Rabbit System will be manual control of a valve. All of the commands for actuating or or turning off valves are entered into the location 166660. Table 1 lists each valve, its function, and the value of location 166660 needed to turn the valve on or off. To accomplish this type "BREAK", then 166660/, the command from the table, and "RETURN". To return from ODT type "P" and "RETURN" (if the LSI-11 responds to this with "file?", type "CTRL-C").

TABLE 1. VALVE CONTROL COMMANDS

<u>Valve</u>	<u>Controller No.</u>	<u>Function</u>	<u>ON/OFF</u>	<u>Command</u>
A, D	0	Send rabbit	ON OFF	0 100000
B	1	Return rabbit	ON OFF	10 100010
E, F	2	Irradiation	ON OFF	20 100020
H	4	Dropout	OPEN CLOSE	40 100040
J	5	Plunger	DOWN UP	50 100050
G	6	Rabbit-line switch	DROPOUT DETECTOR	60 100060
	7	5-v detector gate	OPEN CLOSED	70 100070

APPENDIX B: TROUBLE-SHOOTING

Table 2 is a summary of several problems likely to occur upon extensive use of the rabbit system. Possible causes are listed for a number of symptoms and remedies are suggested. Read the section about sensitivity adjustments to the sensors before making such adjustments.

TABLE 2. TROUBLE-SHOOTING

TROUBLE	CAUSE	REMEDY
Sensor fails to see rabbit	Rabbit getting stuck in tube	1) Make sure that no sharp turns have been introduced by moving equipment.
		2) Check air pressure to the switch and see if it is operating properly.
		3) Check for build up of crud inside tube.
	Sensor Malfunctions	
	1) Sensitivity needs adjustment	1) Each sensor card should have its red light on with no rabbit present and off only when a rabbit breaks the beam. See the section on adjusting the sensitivity and attempt to correct the problem before trying the following remedies.
	2) Defective sensor	1) Test sensor in another circuit board. If it fails replace the sensor with another in the original board and re-adjust the sensitivity.
	3) Defective circuit boards	1) Replace green circuit board and adjust its sensitivity.
		2) Return first green board changing the larger L-shaped board the sensor is connected to. Leave no blanks between the cables at either end of the series of boards!

TABLE 2. TROUBLE-SHOOTING (Continued)

TROUBLE	CAUSE	REMEDY
Sensor sees rabbit when there is none	Sensor is too sensitive	Adjust sensitivity.
	Sensor incorrectly mounted on tube	Check for obstructions, alignment.
Plunger fails to catch rabbit	Bad timing	1) Experiment with different settings of the plunger delay time set in the programs and
	Bounces out of plunger	2) Adjust the flow through the two exhaust lines at the plunger so that more flow goes through the rabbit line.
LSI-11 "freezes"	Only God knows	1) Try "CTRL-C" a few times. 2) "Break" and Type P then "Return". 3) "Break" and re-bootstrap with 173000G which should return a \$. 4) Completely shut down computer, terminal and disk drive and start again.

Adjusting Sensor Sensitivity

Each green circuit board in the sensor multiplexor contains a small variable resistance used for sensitivity adjustments. Looking at the board from the back of the multiplexor box, a CLOCKWISE turn of the screw DECREASES the sensitivity (do this if the red light is always off). The sensitivity adjustment is itself very sensitive; make small changes (less than a quarter turn) and turn off and on again the multiplexor power. If the sensor light is on but the sensor fails to see a rabbit, increase the sensitivity slightly and try again several times before a further adjustment.

If many adjustments are needed, the program TESTA.SAV interrogates the sensors and returns "TRUE" if the sensor sees the rabbit (the light will go out also). Be sure to put the sensor on a length of 1-1/8" ID polyethylene tubing so conditions while running TESTA match the rabbit system. TESTA is less convenient for adjusting sensors 1 and 2 in the RTNS-I target pit. (Sensor 1 is closest to the target. These sensors are redundant; the user may choose either sensor for use in any program, in case one fails, and the pit is "too hot" to enter for maintenance.)

Each sensor consists of two circuit boards and the sensor itself. If you believe a sensor is bad, plug it into a board that you know is working properly. It is best to turn the multiplexor off while making any changes. Make sure ALL 26 pins are plugged into the connector. If this sensor network now fails, the sensor itself is bad, otherwise one of the circuit boards is bad. There are many spare boards in the multiplexor box. Return the sensor to the board that fails and try 2 or 3 green boards, making many sensitivity adjustments. If the sensor still fails, swap L-shaped boards (leaving no blanks in the chain) keeping the original green board, and repeat sensitivity adjustments. It is easy to misalign the 26-pin connector, so double check.

APPENDIX C: SOFTWARE

Detailed knowledge of the programs controlling the rabbit system is not needed to operate the system. This section is included in the event that changes are needed in the software. An understanding of the RT-11 operating system, especially editing and file manipulation, and the OMSI (Oregon Minicomputer Software Inc.) implementation of PASCAL is required to make any changes.

Except for BYHAND, all of the programs are broken into many external procedures, a list of global variables, and a main program. Procedures can be individually edited and compiled with the global variables without altering the rest of the program. The final program is obtained by linking all of the object files created by compilation of the main program and the procedures. For instructions regarding the PASCAL compiler consult the OMSI PASCAL manual.

Because PASCAL operates via a once-through compiler, all constants, variables, procedures, and functions must be defined before being used in the program. When a procedure is being individually compiled, all other procedures to be called must be declared as external with an OMSI PASCAL "EXTERNAL" statement (see the source listings of CYCLE following this appendix). Table 3 gives the object-file names and correct order for the system programs. File names are usually the first few letters of the procedure name, which, like all identifiers in PASCAL, can be of any length. An excellent reference, serving as a text book, is Programming in PASCAL, by Peter Grogono, which is available from the Small Computer User's Library.

TABLE 3. RABBIT SYSTEM PROGRAMS AND THEIR
CONSTITUENT PROCEDURES AND FUNCTIONS.

MANUAL	CYCLE/ACYCLE ⁺ (CYCLE6/ZCYCLE ⁺)*	CYCLES
MAIN1	MAIN2 ⁺	MAIN4
GLOB1	GLOBAL	GLOBAL
SHOWHO	SHOWH2	SHOWH2
INIT	INIT2	INIT2
	INITI2	INITI2
INITIA	IRRADI*	IRRAD6
SEE	SEE2	SEE2
SEND	SEND2	SEND2
BRINGI	BRING2	BRING2
RETURN	RETUR2	RETUR2
RETRIE	RETRI2	RETRI2
	KEEPTR	
	SECOND	SECOND
	START	START

⁺MAIN3 for ACYCLE and ZCYCLE

*Detector "start-pulse" rather than gate in IRRAD6.

Source listings of all necessary program pieces are kept on floppy disks in T940, and are identified as filename.PAS. Object files of each are identified as filename .OBJ, while executable programs have the identifier .SAV. If a program change does not introduce new global variables, a newly created object file can be linked with the previously existing object files. An example of the linking procedure, for CYCLE, is as follows:

```
R LINK (return)
```

```
*CYCLE=MAIN2,SHOWH2,INIT2,INITI2,IRRADI,/C (return)
```

```
*SEE2,SEND2,BRING2,RETUR2,RETRI2,/C (return)
```

```
*KEEPT2,SECOND,START,PASCAL (return)
```

```
*CTRL-C
```

The file, CYCLE.SAV, is thus created and can be executed with the command RUN CYCLE. The prompt from the linker is "*", while: "/C" tells the linker the command is continued on the next line. PASCAL.OBJ the last object file in any link operation, is the OMSI PASCAL support library.

Finally, the source listings of the components of CYCLE are presented as an example of the system software. Comments for the purpose of documentation are enclosed in special brackets (* like this *).

```

C)   FILE : GLOBAL.PAS   *)

PROGRAM CYCLE (INPUT,OUTPUT,INFILE);

(*  FILE OF ALL CONSTANTS AND VARIABLES DEFINED OVER ALL OF THE PROGRAM  *)
(*  "CYCLE".  MOST ARE SELF-EXPLANATORY.  TIMES ARE LISTED IN HOURS FOR  *)
(*  USE WITH THE OMST PASCAL FLOATING-POINT LINE CLOCK.  *)

(*  THE "ORIGIN" COMMAND ALLOWS THE PROGRAM TO CHANGE THE VALUE OF A  *)
(*  PARTICULAR MEMORY LOCATION.  ANY NUMBER FOLLOWED BY "B" IS AN  *)
(*  OCTAL NUMBER.  *)

(*  THE CONSTANTS ARE AS FOLLOWS :  *)
(*  INTERROGATE   = NUMBER OF INTERROGATIONS IN 5 SECONDS  *)
(*                  (DEFAULT AIR SHUT-OFF TIME)  *)
(*  PLUNGDELAY     = TIME (H) BETWEEN DETECTION OF RABBIT  *)
(*                  BY SENSOR 3 AND PLUNGER DOWN  *)
(*  DROPDELAY     = TIME (H) BETWEEN DETECTION OF RABBIT  *)
(*                  BY SENSOR 4 AND OPEN DROP-OUT  *)
(*  SEEPIT        = SENSOR NUMBER IN TARGET PIT  *)
(*  CNTDELAY       = TIME (H) BETWEEN IRRAD AND COUNT  *)

CONST      (*  RABBIT SYSTEM DEFAULT PARAMETERS  *)

            INTERROGATE   = 5682;          PLUNGDELAY   = 5.505E-5;
            DROPDELAY     = 1.111E-3;      SEEPIT       = 1;

            (*  IRRADIATION CYCLE DEFAULT PARAMETERS  *)

            NUMCYCLES     = 10;            IRRLENGTH    = 1.3888E-3;
            CNTDELAY      = 8.333E-4;      CNTLENGTH    = 9.333E-3;
            WAIT          = 5.555E-4;      MAXLEN       = 70;

            (*  VALVE CONTROL CONSTANTS FOR DRV-11  *)

            SENDIT        = 0B;            SENDOFF      = 100000B;
            RETURNON      = 10B;           RETURNOFF    = 100010B;
            IRRADON       = 20B;           IRRADOFF     = 100020B;
            DROPIT        = 40B;           CLOSEIT      = 100040B;
            PLUNGDOWN     = 50B;           PLUNGUP      = 100050B;
            DROP-OUT      = 60B;           DETECTOR     = 100060B;
            GATEUPEN      = 70B;           GATTCLOSE    = 100070B;

TYPE      COL      = 1..MAXLEN;
          LINE      = ARRAY [COL] OF CHAR;
          TABLE    = FILE OF LINE;

VAR       COUNT,SEECYCLES,SENSOR,PITSSENSOR,PLUNGSENSOR,
          CYCLENUM, IHOUR,IMIN,ISEC,TENTHS,CYCLES      : INTEGER;

          PLUNGTIME,T1,T2,DROPTIME,IRRADLENGTH,
          WAITNEXT,WAITING,IRRADON,IRRADLEN,IRRADEND,
          COUNTBEGIN,COUNTEND,COUNTDELAY,COUNTLENGTH  : REAL;

          INFILE      : TABLE;

          REPLY        : CHAR;

          CONTINUE,SENDOK,RETURNOK,ALLOK,AOF,ATCOUNT  : BOOLEAN;

          RABBITID     : ARRAY [ 1..30 ] OF CHAR;

          VALVE0 ORIGIN 166660B,
          VALVE1 ORIGIN 166660B,
          VALVE2 ORIGIN 166660B,
          VALVE4 ORIGIN 166660B,
          VALVE5 ORIGIN 166660B,
          VALVE6 ORIGIN 166660B,
          GATE ORIGIN 166660B   : INTEGER;

```

```
(*E+  EXTERNAL TO CYCLE  *)
```

```
PROCEDURE SHOWHOW;
```

```
BEGIN
```

```
(*  SHOWHOW  *)
```

```

WRITELN('  PROGRAMS CYCLE AND ACYCLE PRESET THE IRRADIATION AND COUNTING')
WRITELN('PARAMETERS SO THAT THE LSI-11 CYCLES THE RABBIT SYSTEM.  ACYCLE');
WRITELN('REQUIRES A START SIGNAL FROM THE USER FOR EVERY CYCLE WHEREAS CYCLE'
WRITELN('WILL AUTOMATICALLY STEP THROUGH A PRESET NUMBER OF CYCLES.  BOTH');
WRITELN('CREAT A FILE, FIL.TXT, RECORDING FOR EACH CYCLE THE IRRADIATION');
WRITELN('AND COUNTING TIMES, AND A MESSAGE, "OK" OR "NO DATA", INDICATING');
WRITELN('WHETHER OR NOT THE CYCLE WAS COMPLETED SUCCESSFULLY.  THIS FILE MAY'
WRITELN('BE INSPECTED AFTER PROGRAM EXECUTION ON THE TERMINAL WITH THE ');
WRITELN('COMMAND TYPE FIL.TXT OR IT CAN BE PRINTED ON THE TELETYPE WITH THE')
WRITELN('COMMAND PRINT FIL.TXT.  THIS FILE IS DESTROYED WITH THE NEXT');
WRITELN('EXECUTION OF EITHER PROGRAM !');
WRITELN;
WRITELN('  DURING THE OPERATION OF CYCLE, FAILURE OF ANY OF THE SENSORS');
WRITELN('TO SEE THE RABBIT WILL CAUSE NO DATA TO BE TAKEN (VIA THE GATE)');
WRITELN('AND WILL INTERRUPT THE PROGRAM.  THE USER MAY THEN DECIDE TO STOP');
WRITELN('THE RUN OR TO CONTINUE.  IF THE PROGRAM IS ABORTED AT THIS POINT,');
WRITELN('FIL.TXT WILL CONTAIN RECORDS OF THE IRRADIATIONS COMPLETED.  ');
WRITELN;
WRITELN('  MORE INFORMATION ON THE RABBIT SYSTEM SOFTWARE IS CONTAINED');
WRITELN('IN FILE RABBIT.TXT. ');
WRITELN;
WRITE('WHEN READY TO CONTINUE, TYPE "RET":');
READLN;READ(REPLY);

```

```
END;
```

```
(*  SHOWHOW  *)
```

```
(*  FILE : INIT.PAS  *)
```

```
(*E+  EXTERNAL PROCEDURE *)
```

```
PROCEDURE INIT;
```

```

(*  A VERY SHORT PROCEDURE TO INITIALIZE GLOBAL VARIABLES THAT ARE  *)
(*  RABBIT-SYSTEM OR IRRADIATION PARAMETERS TO DEFAULT VALUES LISTED *)
(*  AS CONSTANTS.  EXISTS MAINLY TO FACILITATE CHANGES IN DEFAULT  *)
(*  VALUES IF THEY ARE EVER NEEDED.  *)

```

```
BEGIN
```

```
(*  INIT  *)
```

```

PLUNGTIME := PLUNGDELAY;
DROPTIME := DROPDELAY;
PITSENSOR := SEEPIT;
SEECYCLES := INTERROGATE;
CYCLES := NUMCYCLES;
IRRADLENGTH := IRKLENGTH;
COUNTDELAY := CNTDELAY;
COUNTLENGTH := CNTLENGTH;
WAITANEXT := WAIT
END;
```

```
(*  INIT  *)
```

```

FILE : INITIA.PAS      *)

(*!!*) EXTERNAL PROCEDURE *)

PROCEDURE INITIALIZE;

(* PRESENTS RABBIT-SYSTEM PARAMETERS TO THE USER AND ALLOWS CHANGES *)
(* FOR THIS EXECUTION OF "CYCLE". THE USER MAY SPECIFY: 1) THE TIME *)
(* DELAYS BETWEEN THE "RABBIT-HAS-RETURNED" SIGNAL AND ACTIVATION OF *)
(* THE PLUNGER AND THE OPENING OF THE DROPOUT, 2) THE DEFAULT AIR- *)
(* PRESSURE SHUT-OFF TIME IN CASE A RABBIT IS NOT SEEN, AND *)
(* 3) WHICH SENSOR TO BE USED AT THE TARGET END. *)

CONST   SECS      = 3600;
        CYCLETIME = 8.8E-4;      (* TIME (S) NEEDED FOR ONE INTERROGATION *)
                                      (* OF A SENSOR *)

VAR     CHOICE    : CHAR;
        D,P,T    : REAL;
        S        : INTEGER;
        LIST,CHANGES : BOOLEAN;

BEGIN                                     (* INITIALIZE *)

    T := SEECYCLES * CYCLETIME;
    P := PLUNGTIME * SECS;
    D := DROPTIME * SECS;
    S := PITSENSOR;

    WRITELN;WRITELN;

    REPEAT
        WRITELN('THE ADJUSTABLE PARAMETERS, THEIR VALUES, AND ONE-LETTER CODES'
                  ' ARE:');
        WRITELN;
        WRITELN('PLUNGER DELAY TIME (P) = ',P:7:3, ' SEC');
        WRITELN('DROPOUT DELAY TIME (D) = ',D:5:1, ' SEC');
        WRITELN('DEFAULT SHUT-OFF (T) = ',T:5:1, ' SEC');
        WRITELN('PIT SENSOR 1 OR 2 (S) = ',S:3);

        LIST := FALSE;
        CHANGES := TRUE;

        REPEAT
            WRITELN;
            WRITELN('TYPE ONE-LETTER CODE (P,D,T,S) TO CHANGE A PARAMETER. ');
            WRITE('!!' TO LIST, 'X' TO EXIT, 'N' TO EXIT WITHOUT CHANGES: ');
            READLN;READ(CHOICE);
            CASE CHOICE OF
                'D' : BEGIN
                    WRITELN;
                    WRITE('DROPOUT DELAY TIME (D) = ',D:5:1, ' SEC');
                    READLN;READ(D);
                END;
                'S' : BEGIN
                    WRITELN;
                    WRITE('PIT SENSOR IS NOW (S) = ',S:3, ' SEC');
                    READLN;READ(S);
                END;
            ELSE
                WRITELN;
            END;
        UNTIL CHOICE = 'X' OR CHOICE = 'N';

    WRITELN;

```

```

'T' : BEGIN
    WRITELN;
    WRITE('DEFAULT AIR SHUT-OFF TIME (T) = ',T;5:1,' : ');
    READLN;READ(T);
END;

'P' : BEGIN
    WRITELN;
    WRITELN('***** CAUTION *****');
    WRITELN('CHANGING P EFFECTS RABBIT SYSTEM PERFORMANCE!');
    WRITELN('BEST VALUE IS :',P;LUNGTIME*SECS;7:3,' SEC');
    WRITE('PLUNGER DELAY TIME (P) = ',P;7:3,' : ');
    READLN;READ(P);
END;

'L' : BEGIN
    CHANGES := FALSE;
    LIST := TRUE;
END;

'N' : EXIT;

'X' : CHANGES := FALSE;

ELSE CHANGES := TRUE
END
UNTIL CHANGES = FALSE;

(* CASES *)

UNTIL LIST = FALSE;
IF CHOICE = 'X' THEN
    BEGIN
        PLUNGTIME := P/SECS;
        DROPTIME := D/SECS;
        PITSENSOR := S;
        SEECYCLES := TRUNC ( T/CYCLETIME );
    END;
    (* UPDATE *)
END;
(* INITIALIZE *)

```

```

(*      FILE : IRRADI.PAS      *)

(*4E4 EXTERNAL TO CYCLE *)

PROCEDURE IRRADINIT;

(*  ALLOWS THE USER TO SET IRRADIATION/COUNTING PARAMETERS FOR A SERIES *)
(*  OF CYCLIC ACTIVATIONS: 1) NUMBER OF CYCLES, 2) IRRADIATION LENGTH, *)
(*  3) DELAY BEFORE COUNTING, 4) COUNT LENGTH, AND 5) WAIT PERIOD   *)
(*  BEFORE THE NEXT CYCLE.      *)

CONST  SECS      = 3600;
       CYCLETIME = 8.8E-4;

VAR     CHOICE      : CHAR;
       CHOOSE,LIST  : BOOLEAN;
       I,D,W,C      : REAL;
       M            : INTEGER;

BEGIN                                     (*  IRRADINIT  *)

    (*  INITIALIZE TO DEFAULT VALUES  *)

    M := CYCLES;
    I := IRRADLENGTH * SECS;
    D := COUNTDELAY * SECS;
    C := COUNTLENGTH * SECS;
    W := WAIT4NEXT * SECS;

    (*  LIST CURRENT IRRADIATION PARAMETERS  *)

    REPEAT                               (*  UNTIL LIST = FALSE  *)

        WRITELN;WRITELN;
        WRITELN('THE IRRADIATION-CYCLE PARAMETERS, THEIR VALUES, AND THEIR ONE');
        WRITELN('LETTER ACCESS CODES ARE:');
        WRITELN;
        WRITELN('NUMBER OF CYCLES           (M) = ',M:4);
        WRITELN('IRRADIATION LENGTH         (I) = ',I:5:1,' SEC');
        WRITELN('DELAY BEFORE COUNT          (D) = ',D:5:1,' SEC');
        WRITELN('COUNT LENGTH (REAL TIME) (C) = ',C:5:1,' SEC');
        WRITELN('WAIT BETWEEN CYCLES        (W) = ',W:5:1,' SEC');
        WRITELN;WRITELN;

        LIST := FALSE;
        CHOOSE := TRUE;

        REPEAT                           (*  MAKE CHANGES  *)

            WRITELN;WRITELN;
            WRITELN('TYPE ONE-LETTER CODE (M,I,D,C, OR W) TO CHANGE A PARAMETER. ');
            WRITELN('L TO LIST PRESENT VALUES. X TO EXIT, OR N '
                    'TO EXIT WITHOUT CHANGES. ');

            WRITE('ENTER OPTION: ');
            READLN;CHOOSE := TRUE;
            WRITELN;WRITELN;

            CASE CHOICE OF

                'M' : BEGIN

```

```

        WRITELN('NUMBER OF CYCLES (N) = ',M:4,' : ');
        READLN;READ(M);
        WRITELN(' M:4,' CYCLES')
    END;

'I' : BEGIN
    WRITELN('IRRADIATION LENGTH (I) = ',I:5:1,' : ');
    READLN;READ(I);
    WRITELN(' I:5:1,' SEC')
    END;

'D' : BEGIN
    WRITELN('DELAY BEFORE COUNT (D) = ',D:5:1,' : ');
    READLN;READ(D);
    WRITELN(' D:5:1,' SEC')
    END;

'C' : BEGIN
    WRITELN('COUNT LENGTH (C) = ',C:5:1,' : ');
    READLN;READ(C);
    WRITELN(' C:5:1,' SEC')
    END;

'W' : BEGIN
    WRITELN('WAIT BETWEEN CYCLES (W) = ',W:5:1,' : ');
    READLN;READ(W);
    WRITELN(' W:5:1,' SEC')
    END;

'L' : BEGIN
    CHOOSE := FALSE;
    LIST := TRUE;
    END;

'N' : EXIT;

'X' : CHOOSE := FALSE;

ELSE CHOOSE := TRUE

END
(* CASES *)
UNTIL CHOOSE = FALSE

UNTIL LIST = FALSE;

IF CHOICE = 'X' THEN
    BEGIN
        CYCLES := M;
        IRRADIATION := I/SECS;
        DELTA := D/SECS;
        COUNTLENGTH := C/SECS;
        WAITNEXT := W/SECS;
        END;
    (* UPDATED *)

    (* IRRADIANT *)

```

```

(*) FILE : SEE.PAS      *)

(**E+ EXTERNAL FUNCTION *)
FUNCTION SEE ( SENSOR : INTEGER ) : BOOLEAN;

(* "FUNCTION SEE" INTERROGATES A SENSOR, SPECIFIED WHEN SEE IS CALLED, *)
(* UNTIL IT SEES A RABBIT OR EXCEEDS A PRESET NUMBER OF INTERROGATIONS. *)
(* "SEE" HAS A BOOLEAN VALUE; IT RETURNS 'TRUE' OR 'FALSE' TO THE *)
(* POINT FROM WHICH IT WAS CALLED. *)

(* THE DEFAULT AIR-PRESSURE TIME LIMIT IS SET BY SPECIFYING THE NUMBER *)
(* OF INTERROGATIONS BEFORE SEE RETURNS 'FALSE'. A FIVE SECOND DEFAULT *)
(* REQUIRES 56,820 INTERROGATIONS. SINCE THIS IS GREATER THAN THE *)
(* ALLOWED MAXIMUM, THE INTERROGATE COMMAND IS NESTED FURTHER IN A *)
(* DECAYE LOOP. THE ACTUAL TIME ELAPSED IN ONE INTERROGATION IS ABOUT *)
(* 88 MICROSECONDS. *)

(* A DRV-11 BOARD PROVIDES THE INTERFACE BETWEEN THE LSI-11 AND THE *)
(* SENSOR MULTIPLEXOR. DOCUMENTATION CONCERNING THE USE OF THIS *)
(* SYSTEM IN NURE, INCLUDING PROGRAMS IN MACRO-11, IS ON A DISK AT *)
(* T-940 IN THE FILE, FILE.MAC, AND FURTHER INFORMATION IS INCLUDED *)
(* IN UCR1-81773, 'ARITHMETIC CIRCUIT ELIMINATES ADDRESS JUMPERS AND *)
(* DIP SWITCHES' BY RICHARD JESSE. CONSULT THE LSI-11, PDP-11/03 *)
(* USER'S MANUAL FOR A DESCRIPTION OF THE DRV-11. *)

(* SEE FIRST DISABLES THE INTERRUPT. THEN, (256 - SENSOR NUMBER) IS *)
(* LOADED IN THE DRV-11 OUTPUT, SEEOUT, AND THE ZERO-ENABLE IS OFF. *)
(* A CARRY FROM THE 8-BIT ADDRESS OF SEEOUT SPECIFIES WHICH SENSOR IS *)
(* BEING INTERROGATED. THE DRV-11 INPUT IS EQUAL TO 'SIGNAL' ( 'B' *)
(* IMPLIES OCTAL ) IF A RABBIT BREAKS THE IR-BEAM TO THE SENSOR. *)

CONST INRUPT = 100B;
      ZEROEN = 20000B;
      SIGNAL = 100000B;

VAR SEECR ORIGIN 167710B;
    SEEOUT ORIGIN 167712B;
    SEEIN ORIGIN 167714B : INTEGER;
    SAW : BOOLEAN;

    DECAYE, INDEX : INTEGER;

BEGIN
    (* FUNCTION SEE *)
    SEE := FALSE;
    SEECR := SEECR AND NOT INRUPT;
    SEEOUT := ((SEEOUT AND NOT 377B) + ( 256 - SENSOR )) AND NOT ZEROEN;
    FOR DECAYE := 1 TO 10 DO
    BEGIN
        FOR INDEX := 1 TO SEECYCLES DO
            IF ( SEEIN AND SIGNAL ) = 0
            THEN SEE := FALSE
            ELSE BEGIN SEE := TRUE; SAW := TRUE; EXIT END; (* SAW RABBIT *)
        IF SAW = TRUE THEN EXIT END;
    END;

    (* FUNCTION SEE *)

```



```

(*      FILE : SEND.PAS      *)

(*!E+ EXTERNAL PROCEDURE *)

FUNCTION SEE(SENSOR : INTEGER) : BOOLEAN; EXTERNAL;

PROCEDURE SEND;

(* 'SEND' OPENS VALVE A TO SEND THE RABBIT TO THE ICT AND THEN CALLS *)
(* 'SEE'. IF 'SEE' RETURNS 'TRUE', VALVE A IS CLOSED, VALVE E IS *)
(* OPENED TO HOLD THE RABBIT IN PLACE, AND THE MESSAGE, 'RABBIT *)
(* AT PIT' IS DISPLAYED. IF 'SEE' RETURNS 'FALSE' AFTER THE DEFAULT *)
(* TIME HAS ELAPSED, VALVE A IS CLOSED AND A WARNING GIVEN. *)

VAR      ATPIT : BOOLEAN;

BEGIN                                     (* SEND *)

  SENSOR := PITSSENSOR;
  VALVES := FLURIGUP;
  VALVE0 := SENDIT;
  ATPIT := SEE (SENSOR);
  IRRADBEGIN := TIME;
  VALVE0 := SENDOFF;

  IF ATPIT THEN                          (* SAW RABBIT AT PIT *)
    BEGIN                                (* VALVE2 HOLDS RABBIT UP TO TARGET *)
      VALVE2 := IRRADON;
      SENDOK := TRUE;
      WRITELN;WRITELN('RABBIT AT TARGET.')
    END

  ELSE
    BEGIN                                (* BAD NEWS *)
      SENDOK := FALSE;
      WRITELN;
      WRITELN('#####');
      WRITELN('***');
      WRITELN('!!!      RABBIT NOT AT IRRADIATION STATION !      ***');
      WRITELN('***');
      WRITELN('#####');
      WRITELN;

    END;                                (* BAD NEWS *)

END;                                    (* SEND *)

```

```

(*      FILE : BRING2.PAS      *)

(**$+ EXTERNAL PROCEDURE *)

FUNCTION SEE(SENSOR : INTEGER) : BOOLEAN; EXTERNAL;

PROCEDURE BRINGITBACK ( SENSOR : INTEGER );

(* "BRINGITBACK" CLOSES VALVE E, OPENS VALVE B TO BRING THE RABBIT *)
(* BACK TO T-940, AND CALLS "SEE". THE PLUNGER IS ACTIVATED AFTER A *)
(* DELAY OF "PLUNGTIME" FOLLOWING THE CLOSING OF VALVE B.      *)

VAR      ATLAB      : BOOLEAN;

BEGIN
                                (* BRINGITBACK *)
    VALVE2 := IRRADOFF;
    VALVE1 := RETURNOK;
    ATLAB := SEE (SENSOR);

    IF NOT ATLAB
    THEN
                                (* BAD NEWS *)
        BEGIN
            RETURNOK := FALSE;
            WRITELN;
            WRITELN('*****');
            WRITELN('***                               ***');
            WRITELN('***          RABBIT NOT AT TRAILER !          ***');
            WRITELN('***                               ***');
            WRITELN('*****');
            WRITELN;

            END
                                (* BAD NEWS *)

        ELSE RETURNOK := TRUE;

        VALVE1 := RETURNOFF;
        T1 := TIME;
        REPEAT
            T2 := TIME
        UNTIL T2 - T1 > PLUNGTIME;
        VALVES := PLUNGDOWN
    END;
                                (* BRINGITBACK *)

(*      FILE : RETURN.PAS      *)

(**$+ PROCEDURE EXTERNAL *)
PROCEDURE SEND; EXTERNAL;
PROCEDURE BRINGITBACK ( SENSOR : INTEGER ); EXTERNAL;

PROCEDURE RETURN;

(* "RETURN" CALLS "SEND", MARKS THE IRRADIATION TIME, AND CALLS *)
(* "BRINGITBACK". IT IS USED TO CYCLE THE RABBIT BACK TO THE *)
(* DETECTOR.      *)

BEGIN
                                (* RETURN *)
    SEND;
    IF SENDOK
    THEN
        REPEAT
                                (* LOOP UNTIL PRESET IRRADIATION *)
                                (* TIME IS UP *)
            IRRADEND := TIME
        UNTIL ( IRRADEND - IRRADBEGIN ) > IRRADLENGTH

        ELSE IRRADEND := TIME;
        SENSOR := 3;
        BRINGITBACK (SENSOR);
        AOK := SENDOK AND RETURNOK;

    END;
                                (* RETURN *)

```

```

(*)      FILE : RETRIE.PAS      *)

(**E+  EXTERNAL TO CYCLE *)
PROCEDURE BRINGITBACK ( SENSOR : INTEGER ); EXTERNAL;
FUNCTION SEE (SENSOR : INTEGER ) : BOOLEAN;  EXTERNAL;

PROCEDURE RETRIEVE;

(* 'RETRIEVE' BRINGS A RABBIT TO THE DROPOUT TO REMOVE IT FROM THE *)
(* SYSTEM.  IT SENDS THE RABBIT, OPENS THE SWITCH TO THE DROPOUT, *)
(* CALLS 'BRINGITBACK', AND DROPS THE RABBIT INTO A LEAD PIG.    *)

CONST   PAUSE1  = 1E-4;
        PAUSE2  = 1E-3;

VAR     T3,T4,T5,T6,T7          : REAL;
        ATPIT   : BOOLEAN;

BEGIN
  SENSOR := 4;
  VALVE0 := SENDIT;
  ATPIT := SEE (PITSSENSOR);
  T3 := TIME;
  VALVE0 := SENDOFF;
  VALVE6 := DROPOUT;
  REPEAT
    T4 := TIME
  UNTIL T4 - T3 > PAUSE1;
  BRINGITBACK (SENSOR);
  T5 := TIME;
  REPEAT
    T6 := TIME
  UNTIL T6 - T5 > DROPTIME;
  VALVE4 := DROFIT;
  VALVE6 := DETECTOR;
  REPEAT
    T7 := TIME
  UNTIL T7 - T6 > PAUSE2;
  VALVE4 := CLOSING;
  VALVE5 := FLURGUP;

END;

```

```

(*)      RETRIEVE      *)

```

```

(*)      FILE : SECOND.PAS      *)

(**E+  PROCEDURE EXTERNAL TO CYCLE *)

PROCEDURE SECONDS ( VAR  T : REAL );

(* FOR EACH TIME ORIGINALLY RECORDED IN DECIMAL HOURS, "SECONDS" *)
(* RETURNS HOURS, MINUTES, SECONDS, AND TENTHS OF SECONDS.    *)

VAR     RMIN,RSEC          : REAL;

BEGIN
  IHOUR := TRUNC(T);
  RMIN := (T - IHOUR ) * 60;
  IMIN := TRUNC(RMIN);
  RSEC := ( RMIN - IMIN ) * 60;
  ISEC := TRUNC(RSEC);
  TENTHS := ROUND ((RSEC-ISEC) * 10 );

  IF TENTHS = 10 THEN BEGIN ISEC := ISEC + 1; TENTHS := 0;END;
  IF ISEC = 60 THEN BEGIN IMIN := IMIN + 1; ISEC := 00;END;
  IF IMIN = 60 THEN BEGIN IHOUR:=IHOUR+1; IMIN := 00; END;

END;

```

```

(*)      SECONDS      *)

```

```

(*)      FILE : KEEPTR.PAS      *)

(=*E+  PROCEDURE EXTERNAL TO CYCLE  *)
PROCEDURE SECONDS ( VAR T : REAL );  EXTERNAL;

PROCEDURE KEEPTRACK;

( *  SETS UP 'FIL.TXT' TO STORE TIMES FOR EACH CYCLE AND WRITES THE  *)
( *  TIMES AFTER EACH CYCLE.      *)

VAR      CYCLENUM,I,J      : INTEGER;
          T      : ARRAY [1..4] OF REAL;
          ANS     : CHAR;

BEGIN

                                ( *  KEEPTRACK  *)

IF COUNT = 1 THEN BEGIN      ( *  INITIALIZE FILE  *)
  WRITELN(INFILE,' '18,'RECORD OF IRRADIATIONS');
  WRITELN(INFILE);WRITELN(INFILE);
  WRITELN(INFILE,'CYCLE      IRRAD START      IRRAD END      COUNT START
                                COUNT END      MESSAGE');
  WRITELN(INFILE);
END;                          ( *  INITIALIZATION  *)

  CYCLENUM := COUNT;
  TC1J := IRRADBEGIN;
  TC2J := IRRADEND;
  TC3J := COUNTBEGIN;
  TC4J := COUNTEND;
  WRITE(INFILE,CYCLENUM : 3);
  FOR J:= 1 TO 4 DO
    BEGIN
      SECONDS (TCJJ);          ( *  WRITE THE TIME  *)
      WRITE(INFILE,' '5,1HOUR;2,' '5,1MIN;2,' '5,1SEC;2,' '5,TENTHS;1);
    END;                      ( *  WRITING TIMES  *)

  IF ALLOC THEN WRITELN(INFILE,' OK')
  ELSE WRITELN(INFILE,' NO DATA');

END;                          ( *  KEEPTRACK  *)

```

```

(*) FILE : START.PAS      *)
(************************************************************************)
(*IF+ EXTERNAL TO CYCLE *)
PROCEDURE SHOWHOW; EXTERNAL;
PROCEDURE INIT; EXTERNAL;
PROCEDURE INITIALIZE; EXTERNAL;
PROCEDURE IRRADINIT; EXTERNAL;

PROCEDURE START;

(* "START" IS HALF OF THE MAIN PROGRAM, CALLING INITIALIZATION *)
(* PROCEDURES AND GIVING INSTRUCTIONS. *)

VAR      PRELIMINARIES      : BOOLEAN;

BEGIN                                     (* START *)

    WRITE('DO YOU NEED INSTRUCTIONS? Y OR N(RETURN) :');
    READLN;READ(REPLY);
    IF REPLY = 'Y' THEN SHOWHOW;

    WRITELN;WRITE('NEED TO INITIALIZE PARAMETERS? Y OR N(RET) :');
    READLN;
    READ(REPLY);
    INIT;
    IF REPLY = 'Y' THEN INITIALIZE;

    IRRADINIT;
    PRELIMINARIES := FALSE;

    WHILE PRELIMINARIES = FALSE DO
        BEGIN                                (* LOOP TO GET READY *)

            WRITELN;WRITELN;
            WRITELN('WHEN READY TO START THE IRRADIATION CYCLE TYPE "RET".');
            WRITELN('OTHERWISE, TYPE "H" FOR HELP, "N" TO ABORT THE RUN,');
            WRITELN('"I" TO INITIALIZE THE RABBIT SYSTEM PARAMETERS, OR');
            WRITELN('"C" TO INITIALIZE IRRADIATION-CYCLE PARAMETERS. ');
            WRITELN;WRITE('ENTER OPTION: ');
            READLN;READ(REPLY);
            CASE REPLY OF

                ' ' : PRELIMINARIES := TRUE;
                'H' : SHOWHOW;
                'I' : INITIALIZE;
                'C' : IRRADINIT;
                'N' : WRITE('TO ABORT THE RUN, TYPE "CTRL-C"');
            ELSE PRELIMINARIES := FALSE
            END;
        END;
    END;
END;                                     (* CASES *)
                                     (* PRELIMINARIES LOOP *)

END;                                     (* START *)

```

```
(* FILE : MAIN.FAS *)
```

```
PROCEDURE START; EXTERNAL;
PROCEDURE SHOWHOW; EXTERNAL;
PROCEDURE INIT; EXTERNAL;
PROCEDURE INITIALIZE; EXTERNAL;
PROCEDURE IRRADINIT; EXTERNAL;
FUNCTION SEE ( SENSOR : INTEGER ) : BOOLEAN; EXTERNAL;
PROCEDURE SEND; EXTERNAL;
PROCEDURE BRINGITBACK ( SENSOR : INTEGER ); EXTERNAL;
PROCEDURE RETURN; EXTERNAL;
PROCEDURE SECONDS ( VAR T : REAL ); EXTERNAL;
PROCEDURE KEEPTRACK; EXTERNAL;
PROCEDURE RETRIEVE; EXTERNAL;
```

```
(* AFTER "MAIN" CALLS "START", IT BEGINS THE CYCLE LOOP, COUNTING *)
(* THE CYCLES. IT CALLS "RETURN" AND IF EVERYTHING WAS OK, DISPLAYS *)
(* THE RETURN TIME, AND STARTS THE DETECTOR. ANY ERRORS REQUIRE THE *)
(* USER TO RESTART THE SERIES OR TO ABORT. "MAIN" TIMES THE COUNT *)
(* AND THE WAIT BETWEEN CYCLES, AND THEN CALLS "KEEPTRACK". *)
```

```
BEGIN (* CYCLE; MAIN PROGRAM *)
  COUNT := 0;
  START; (* CALLS INITIALIZATION ROUTINES *)
  CONTINUE := TRUE;
  REWRITE(INFILE, 'OK:FIL', 'TXT');
  WHILE CONTINUE DO (* CYCLE RABBITS *)

    BEGIN

      RETURN; (* SENDS AND RETURNS RABBIT ONCE
      COUNT := COUNT + 1;
      PLUNGSENSOR := 5;
      SENSOR := PLUNGSENSOR;
      T2 := ( T2 - IRRADEND ) * 3600; (* RETURN TIME IN SECONDS *)
      ATCOUNT := SEE (SENSOR); (* RABBIT IN PLUNGER AT DETECTOR?
      IF (RETURNOK AND ATCOUNT) THEN
        WRITELN('RABBIT AT DETECTOR, TIME = ', T2/3:1, ' SECS');

      ALLOK := AOK AND ATCOUNT;
      IF ALLOK
      THEN
        BEGIN (* EVERYTHING IS COOL, *)
          BEGIN (* START COUNTING AFTER *)
            REPEAT (* PRESET DELAY TIME *)
              COUNTBEGIN := TIME
            UNTIL (COUNTBEGIN - IRRADEND) > COUNTDELAY;
            GATE := GATEOPEN;
            WRITELN('CYCLE = ', COUNT :3, ' ALL OK. ');
          END (* START COUNTING *)
        ELSE
          BEGIN (* THERE ARE PROBLEMS *)
            WRITELN; WRITELN;
            WRITE('TO RESTART, TYPE "RET", TO ABORT TYPE "N": ');
            READLN; READ( REPLY );
            IF REPLY = 'N' THEN EXIT; (* EXIT CYCLE LOOP *)
          END;
        END;
      END;
```

```

REPEAT
    COUNTEND := TIME
    UNTIL ( COUNTEND - COUNTBEGIN ) > COUNTLENGTH;
    GATE := GATECLOSE;                (* STOP COUNTING *)

    KEEPTIME;                          (* STORE TIME INFORMATION *)

    REPEAT
        WAITING := TIME
        UNTIL ( WAITING - COUNTEND ) > WAITANEXT;    (* AGAIN *)

        IF COUNT = CYCLES THEN EXIT;    (* ALL DONE! *)

    END;                                (* CYCLE LOOP *)

    VALVES := PLUNGUP;
    WRITE('DO YOU NEED TO RETRIEVE RABBIT? Y OR N(RET): ');
    READLN;READ(REPLY);
    IF REPLY = 'Y' THEN RETRIEVE;
    WRITELN (' COUNT =',COUNT);
    CLOSE(INFILE)

END.                                    (* MAIN *)

```

VITA

Robert Eugene Williams was born October 19, 1948 in Albany, New York. He attended elementary and high school in the Ravena-Coeymans-Selkirk Central Schools. Mr. Williams received a B.S. degree in Physics from the State University of New York at Albany in 1971, and began his graduate training at Albany State also, receiving a M.S. degree in Physics in 1974. He then entered the Nuclear Engineering Program at the University of Illinois, earning a M.S. degree in 1976 before undertaking his Ph. D. research. With a fellowship from the Associated Western Universities, Inc., Mr. Williams completed his research at the Lawrence Livermore National Laboratory from 1979 to 1981, where he also was employed in the Laser Fusion Program temporarily in 1979. He may currently be found at the Center for Nuclear Studies at Memphis State University, Memphis, Tennessee.

ACKNOWLEDGEMENTS

I wish to express my sincere appreciation to my thesis advisor, Professor Philip K. Hopke, for introducing me to the world of activation analysis, assisting through those 16-h days at the RTNS, but mostly for sticking with this project and exploring every avenue for my support. I also wish to thank Professor Bernard W. Wehring for exhibiting infinite patience with me in the laboratory when most people would have changed the locks. I am indebted to Dr. Richard A. Meyer for cutting through vast layers of red tape to get me to LLNL and into the fast-chemistry group, and for the many lessons in viticulture, certainly not wasted, even if I was. I also thank Dr. Eugene A. Henry for his frequent assistance and tutelage in complex nuclear spectroscopy experiments.

This thesis could not have been completed without a fellowship granted by the Associated Western Universities, Inc., for which I am indebted to Dr. Donald Walker. I must also credit Y-Division at LLNL for temporary employment in 1979 enabling me to avoid bankruptcy, for which I thank Dr. E. Michael Campbell and Dr. Stephen M. Lane.

A multitude of secretarial and technical people have been of great assistance to me both at the University of Illinois and at LLNL, and I thank them all for a thousand favors rendered. I especially thank Joyce Plis for coordinating publications with the Technical Information Department at LLNL, and Barb Cummins for much-needed last minute typing.

Life as a graduate student would be intolerable but for the camaraderie of other students, for which I am eternally grateful. I am referring to the gang of four at Albany State, the great group in the Nuclear Radiation Laboratory (for which I thank Dr. Marvin Wyman for assembling at this point on the globe), and the guys behind the fences at "Teller Tech". I also wish to thank Earl Minor for sharing my experiences at LLNL, and Brad Roscoe for much assistance through the years and particularly in the final days.

I wish to express my deepest appreciation to my roommate, Dan Alpert, for helping me cope with the day to day frustrations of getting a thesis topic off the ground, and for PROVIDING the dirty socks.

Finally, I want to express my love and appreciation for my mother and father, for their love and many sacrifices on my behalf. I deeply regret that I did not finish in time for my father to see this long-anticipated document.