

DETERMINATION OF OXYGEN
BY NEUTRON ACTIVATION ANALYSIS

by

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B.S., National University of Mexico, 1971

A MASTER'S THESIS

submitted in partial fulfillment of the
requirements for the degree

MASTER OF SCIENCE

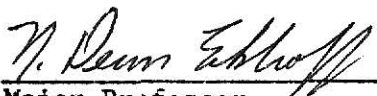
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NOMENCLATURE

$\dot{N}$	Time rate of change of the number of product nuclei
N	Number of product nuclei
N_0	Number of parent nuclei
σ	Microscopic neutron absorption cross sections ($\text{cm}^2/\text{nucleus}$)
Φ	Neutron flux (neutrons/ cm^2 sec)
λ	Decay constant (sec^{-1})
A_0	Activity at end of irradiation (desintegrations/sec)
A	Activity after an interval of time has been elapsed
t_e	Elapsed time after irradiation
μ	Subscript denoting "unknown" sample
k	Subscript denoting "known" sample
f_u	Subscript for fluence monitor of unknown
f_k	Subscript for fluence monitor of known
R	Subscript for "reference" sample
A^*	Activity per unit mass (desintegrations/sec gm)
k_1	Rate constant of the reaction C-0
k_2	Rate constant of the reaction C+0
m	Mass (grams)
M	Mass (grams)
h	Planck's constant
c	Velocity of light
w	Stretching frequency (min^{-1})
k	Boltzmann's constant
T	Temperature ($^{\circ}\text{C}$)
t_d	Decay time (sec)
t_i	Irradiation time (sec)
t_c	Counting time (sec)

l	Minimum left channel
r	Minimum right channel
C_i	Contents of channel i
t_o	Irradiation time
X_c	Centroid of peak
MR	Mole ratio
R	Rate of production (reactions/sec)
C	Total number of counts accumulated during a counting interval
ϵ	Overall-all counting efficiency
B	Total number of counts from background accumulated during a counting interval
f	Fraction of an element in a given sample
th	Subscript denoting "thermal" neutron flux
F	Subscript denoting fluence monitor
σ	Standard deviation
MW	Molecular weight of a given sample
t_1	Decay time (sec)
t_2	Counting time
N_T	Number of product nuclei
P	Net peak area under photopeak of interest
t_D	Irradiation time (120 sec)
ψ	Time wave-function
ϕ_o	Neutron flux at rotary specimen rack (1.8×10^{12} n/sec ² sec)
M	Slope of a straight line
B	Y intercept of a straight line

1. Introduction

Neutron activation analysis (NAA) has been widely used in the determination of the composition of a variety of materials. A large body of reference literature exists with large additions being made annually. Of interest in this work is the literature dealing with activation analysis used to determine ^{18}O in samples of distilled water, sugars, organic materials, as well as inorganic materials (2,3,5,6).

Oxygen-18 is used as a stable isotope tracer to follow the mechanics of various enzymatic reactions involving several sugars (1), to study the hydrolysis of a considerable number of organic esters of inorganic oxyacids (5), and to study the hydrolysis of amides (6). So, the importance of this stable isotope has created the need for rapid and nondestructive analytical techniques.

1.1 Other Methods for Determining ^{18}O

The importance of oxygen-18 as a tracer for oxygen in various fields of research created the need for suitable analytical techniques. The common method of determining ^{18}O content is by mass spectrometry (4,8), but radioactivation techniques have been tried as well (2,3,26,28,29). Although mass spectrometry is a very sensitive high precision method the destruction of samples is sometimes of great concern. It frequently happens that other methods are preferred even though they are less accurate and sensitive than mass spectrometry. Density measurement techniques (27) have been used to determine ^{18}O . However, the difficulties of sample preparation and the need for relatively large samples limit the use of this procedure in most tracer experiments. Also density measurement techniques are destructive. A brief description of some methods is given below.

1.1.1 Mass Spectrometry

Analysis for ^{18}O has been performed mainly by one of two methods which involve combined mass spectrometric techniques: the classical combustion train-isotope ratio technique (8) and gas-chromatographic quadrupole mass spectrometry (4). Basically, a mass spectrometer is a quantitative and qualitative analyzer for most components in all types of samples - gas, liquid, or solid, but with some volatility limitations. A complete analysis is obtained from submicrogram samples in a few seconds or minutes. The range is from ppm or ppb to 100 percent purity; accuracy is generally $\pm 1\%$; the specificity is good. The instruments are moderately complex and costly. Particular disadvantages are the need to isolate the oxygen of sample as a simple gas, e.g., CO or CO_2 for water samples, prior to mass analysis, requires complex vacuum apparatus as well as complicated gas collecting devices resulting in a relatively long procedure and the complete destruction of the sample analyzed.

1.1.2 Activation Analysis

Activation analysis is a competitive alternative to mass spectrometry or density measurements. The activation methods are non-destructive, by nature, as well as rapid, sensitive and versatile.

In general, analysis of ^{18}O is carried out by measuring the activity induced after bombardment by neutrons or other sub-atomic particles. Some radioactivation methods used to determine ^{18}O are:

a) γ detecting from the reaction $^{18}\text{O}(\alpha, n\gamma) ^{21}\text{Ne}$ (28).

α - Particles accelerated in a cyclotron to an energy of 4.6 MeV are used. The ^{18}O concentration is determined from the γ -ray spectrometry ($E_\gamma = 350$ keV) emitted by the excited ^{21}Ne nucleus. The absolute sensitivity

of the method is about 10^{-9} g for ^{18}O . Of great concern is the backing material as well as the kind of sample to be analyzed because of possible γ -ray interference of competitive γ -energies.

b) β and γ detecting from the reaction $^{18}\text{O}(p,n)^{18}\text{F}$ (2,30).

The ^{18}F nuclei formed in this reaction are β^+ emitters and have a decay period of 110 min. A measure of the 0.511 -MeV annihilation radiation from ^{18}F yields a measure of the ^{18}O (2) (detection by an assembly of NaI(Tl) detectors), or as a result of many hours of tracing the decay of the activity of ^{18}F with a beta counter (Geiger-Muller counter) (30) or an autoradiography where beta emission is measured by the blackening of photographic emulsions (30). To avoid possible interferences from the reactions $^{16}\text{O}(p,\alpha)^{13}\text{N}$ and $^{13}\text{C}(p,n)^{13}\text{N}$ ($T_{1/2} = 10$ min) a storage of two hours is required. Also the β^+ is interfered with by other radioisotopes that yield a 511 keV annihilation peak. However, the method is comparable in accuracy to mass spectrometry and it can be used to analyze water samples as small as 1.5 μl .

c) Counting of delayed neutrons of Nitrogen-17 (26).

Neutron counting of 4.14-second Nitrogen-17 is used for the analysis of ^{18}O in solutions containing known amounts lithium-6. ^{17}N is produced by the successive nuclear reactions $^6\text{Li}(n,\alpha)t$; $^{18}\text{O}(t,\alpha)^{17}\text{N}$ upon irradiation with thermal neutrons. The total delayed neutron emission of a sample irradiated to saturation at a thermal neutron flux of 10^{13} n/cm² sec is about 4×10^5 neutrons per mg of ^6Li per atom percent ^{18}O . The ^{17}N formation is proportional to the atom fraction of ^{18}O and is independent of the sample size. This method requires access to large thermal neutron fluxes; also, the dilution of ^6Li and ^{18}O sample is required. Of great concern are: the possible interference of reactions produced with fast neutrons, the need for a rapid transfer system, and the use of an assembly of B^{10}F_3 neutron counters

and associated electronics. The method is excellent in that it shows high specificity and can measure ^{18}O in very small samples.

d) Thermal neutron activation analysis.

The purpose of this work was to develop a NAA technique, using thermal neutrons, to analyze samples having ^{18}O as a constituent. The NAA method is limited because the activation cross section for the reaction $^{18}\text{O}(n,\gamma)^{19}\text{O}$ and the abundance of ^{18}O are both low; 0.21 mb, and 0.204%, respectively. Of great concern also is the relatively short half life of ^{19}O , 26.9 seconds, which requires a fast transfer system. This work was mainly oriented toward the solution of such problems, with emphasis of the use of a simple manual transport method.

In order to understand better the steps performed in this work, a basic formulation on NAA is given. More detailed information can be found, for example, in references (9,10).

2 Basic Standard Comparator Technique in Neutron Activation Analysis

NAA is a procedure in which a sample is exposed to a flux of neutrons and by neutron absorption the sample becomes radioactive. For such nuclear reactions the activity of the product nuclide is directly proportional to the number of atoms of the parent nuclide; this activity can be used to calculate the amount of the parent which was originally in the sample being studied.

The rate of change of the product nuclei, $\dot{N}$, is described by

$$\dot{N} = N_0 \sigma \Phi - \lambda N, \quad (2.1)$$

where

$\dot{N}$ = rate of change of the number of product nuclei N ,

N_0 = number of parent nuclei,

σ = microscopic cross section for the reaction under consideration,

Φ = irradiation neutron flux,

λ = decay constant of the product nuclei.

The solution to equation 2.1 is

$$A_o = N_o \sigma \Phi (1 - e^{-\lambda t_o}) , \quad (2.2)$$

where

A_o = activity of the product at end of irradiation,

t_o = irradiation time,

Φ = irradiation flux.

The activity after an elapsed time t_e from the end of the irradiation would then be A , where

$$A = A_o e^{-\lambda t_e} . \quad (2.3)$$

The standard comparator method of radioactivation analysis is based on simultaneous analysis of a standard sample of known composition (the "known" sample) and a sample of unknown composition (the "unknown" sample) for the desired element in which both samples are treated identically. Under such conditions there are several common parameters such as the irradiation times, the half-lives and the reaction cross sections; then, for the unknown and known samples,

$$\frac{A_{ou}}{A_{ok}} = \frac{N_u \sigma_u \Phi_u (1 - e^{-\lambda t_{ou}})}{N_k \sigma_k \Phi_k (1 - e^{-\lambda t_{ok}})} , \quad (2.4)$$

where

N = number of parent nuclei (N_o),

t_o = irradiation time.

Then, for equal irradiation times and cross section, the number of target atoms of the unknown sample is given by

$$N_u = N_k \frac{\phi_k}{\phi_u} \frac{A_{ou}}{A_{ok}}, \quad (2.5)$$

where all the activities have been normalized to a unit mass. The subscripts u and k denote values for the unknown and known samples, respectively. In this method, however, equal conditions of sample and comparator must be carefully maintained. Both must be prepared so that they both experience identical irradiation under the same flux conditions. To achieve these conditions it is necessary to use a suitable fluence monitor. Each known and unknown sample must be irradiated with a fluence monitor which serves to normalize all the samples to identical irradiation conditions. Thus, from equation 2.2, the neutron flux can be calculated as follows

$$\phi = \frac{A_{of}}{N_{of}\sigma_f} (1 - e^{-\lambda_f t}) . \quad (2.6)$$

where the parameters, were defined below Eq. 2.2, are for data for the fluence monitor used.

Therefore, Equations 2.5 and 2.6, can be used to find the number of target nuclide, N_u , of the unknown sample as follows:

$$N_u = N_k \frac{A_{ofk}}{A_{ofu}} \frac{A_{ou}}{A_{ok}}, \quad (2.7)$$

where A_o is the activity per unit mass and the subscript f refers to the fluence monitors for the unknown (u) and known (k) samples, respectively.

The most common procedure in NAA is to measure the gamma radiation associated with the activity A. Since most samples contain several types of parent nuclei, the high resolution Germanium (Lithium-drifted) detector

has been widely used because it can differentiate between 20 to 100's electronvolts for different gamma-ray energies.

Gamma-ray spectrometry is discussed by many authors (9,17,18,19,20) and will not be covered in this work. Among the different modes of gamma-ray interaction with matter, the photoelectric and the Compton processes are of special interest. The total area under the photopeak is directly proportional to the weight of the element from which radiation is produced; thus, the photoelectric process is used to identify and quantify a given radionuclide. The presence of the Compton scattering continuum limits the measurement of the photopeak. It is considered as an undesirable background in the measurement of photopeaks, especially for photopeaks of low energy.

Basically equation 2.7 was used throughout this work. However, gamma radiation detection introduces a new parameter to be taken in consideration. Efficiency and geometric considerations, which are functions of the gamma-ray energy, are most important. However, in the comparator method they must cancel and equation 2.7 is ultimately adequate for calculations. Appendix A gives a more detailed formulation of the development of equation 2.7.

3. Description of Experimental Apparatus

The Kansas State University TRIGA Mark II nuclear reactor was the source of thermal neutrons for this work. Thermal neutron flux levels at several locations are shown in Table 3.1. Slight variations in these flux values would be possible, but order of magnitude differences would be unlikely.

The irradiation position throughout this work was the rotary specimen rack (RSR) of the TRIGA reactor. The RSR is a ring of sample holders which can be rotated around the core. Access to the RSR is provided by an aluminum tube which extends from the top of the reactor to the RSR.

Table 3.1

Thermal Neutron Flux Values for the TRIGA Mark II Nuclear Reactor
Operating at 250 kW (7).

Irradiation Position	RSR	Fuel-element Ring D	Fuel-element Ring F	Reactor Pool Outside Reflector
Neutron Flux (n/cm ² sec)	1.8×10^{12}	4.9×10^{12}	4.3×10^{12}	6.8×10^{11}

To satisfy the requirements of a fixed position for irradiation, an RSR irradiation vial was modified. This modification consisted of filling the vial with paraffin wax, then five holes were drilled in the wax, as shown in Fig. 3.1. The center hole, 15/32 inch diameter (1.19 cm) and 1.25 inches (3.175 cm) depth, was large enough to hold the polyethylene vial where samples are to be handled. The other four holes, 0.125 inches (0.3175 cm) diameter and 0.5 inch (1.27 cm) depth, were drilled around the center hole and separated from each other by approximately 90°. These holes held the iron wires, which were used as fluence monitors.

A Canberra coaxially drifted Ge(Li) semiconductor detector model 7227 (37.5 mm dia., and 22.5 mm length) was used for gamma-ray detection. This detector was located within the reactor bay. A Canberra, model 970, FET-cooled preamplifier coupled directly to the Ge(Li), was used to couple the detector through a long cable to the main amplifier, an ORTEC model 451. The main amplifier was coupled directly to a Northern Scientific analog-to-digital converter, model NS-636, and its associated multichannel analyzer equipment. Data were then transferred and stored on magnetic tape, through the Northern Scientific, model NS-408. Often a Nova 1220 minicomputer, from Data General Corporation, was used to control the electronic system.

To reduce the background interferences when counting, a lead wall was placed between the detector and the reactor. This shielding, inverted-L

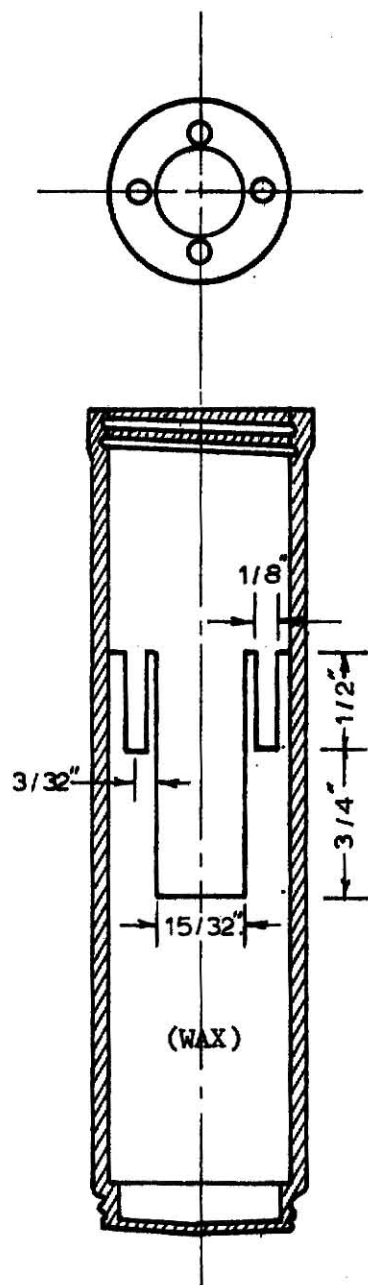


Fig. 3.1 Modified RSR irradiation vial.

in shape, was 8 inches (20.32 cm) thick and 30 inches (76.2 cm) high. The relative position in the reactor bay of this assembly was at the southwest corner at approximately 10 feet (3m) from the concrete wall shield of the reactor (see Fig. 3.2).

A plexiglass device was also constructed to provide accurate positioning of the samples above the Ge(Li) detector so that consistent counting geometry could be maintained throughout the experiment. Figure 3.3 is an illustration of the irradiation vials used in this work (7/16 inch (1.11 cm) outside diameter and one inch long). Figure 3.4 is a diagram of the plexiglass holding device used to maintain constant counting geometry with respect to the Ge(Li) detector.

An IBM 370/158 digital computer was used in processing the data for this work.

4. Experimental

4.1 Preliminary Investigations

Several preliminary experiments were performed with the purpose of gaining information that could be used in the final analyses.

The relatively short-lived radioisotope product ^{19}O and the low thermal activation cross section of the parent ^{18}O were of paramount importance because of possible interferences from other activated products. Data about nuclides of interest are included in Figure 4.1. Short irradiation times (usually about 2 minutes at 250 kW) for complex samples were preferred to reduce interferences with the 197 Kev gamma-ray from ^{19}O . Also, several tests were developed for a simple and quick manual method for transporting samples after irradiation.

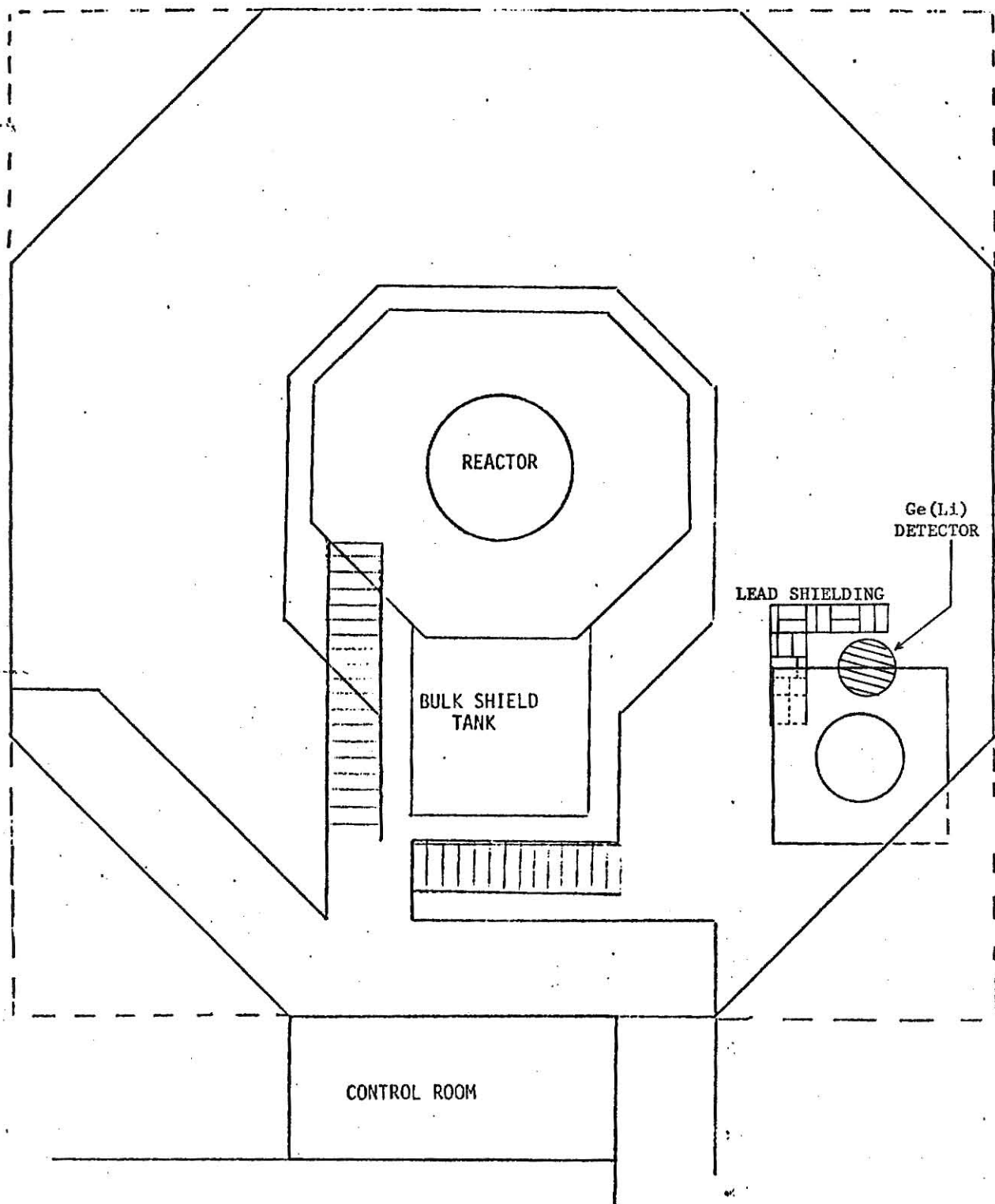


Figure 3.2 Plan View of Reactor Bay with Location of
Detector Shown.

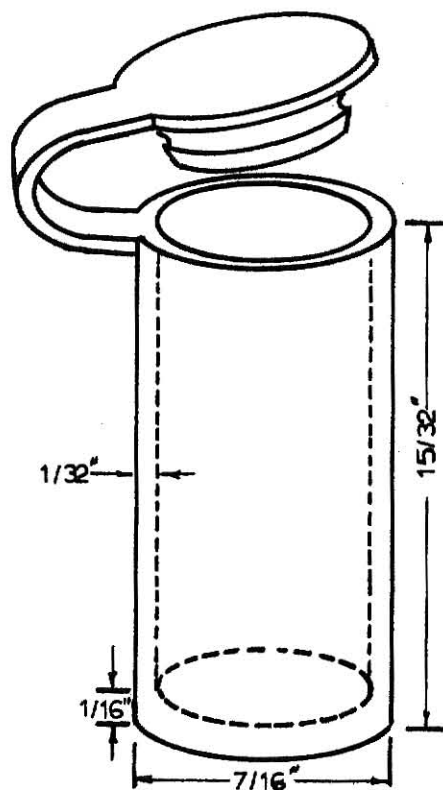


Fig. 3.3 Polyethylene irradiation container
(Olympic Plastics, L.A., Calif.).

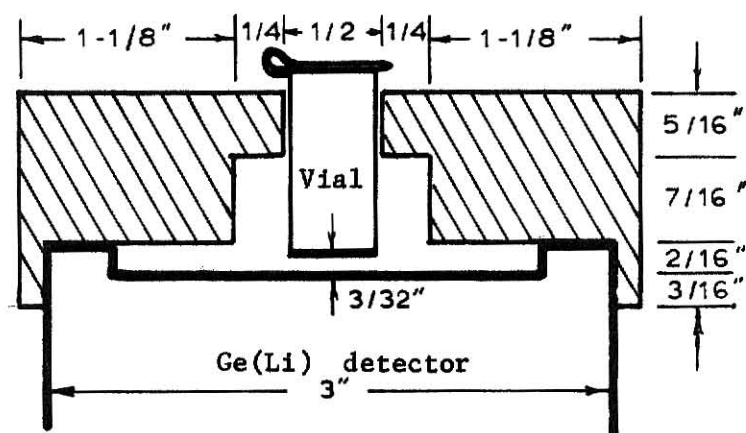
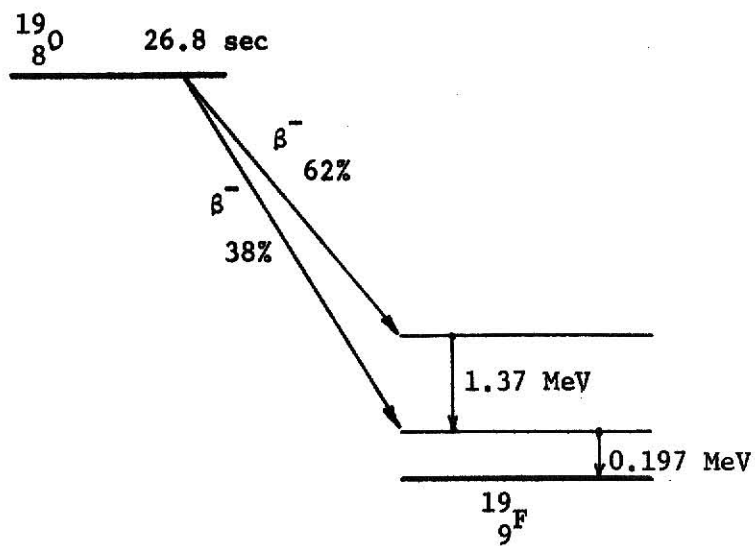


Fig. 3.4 Plexiglass device used to fix the position of the sample vials on the detector for counting.

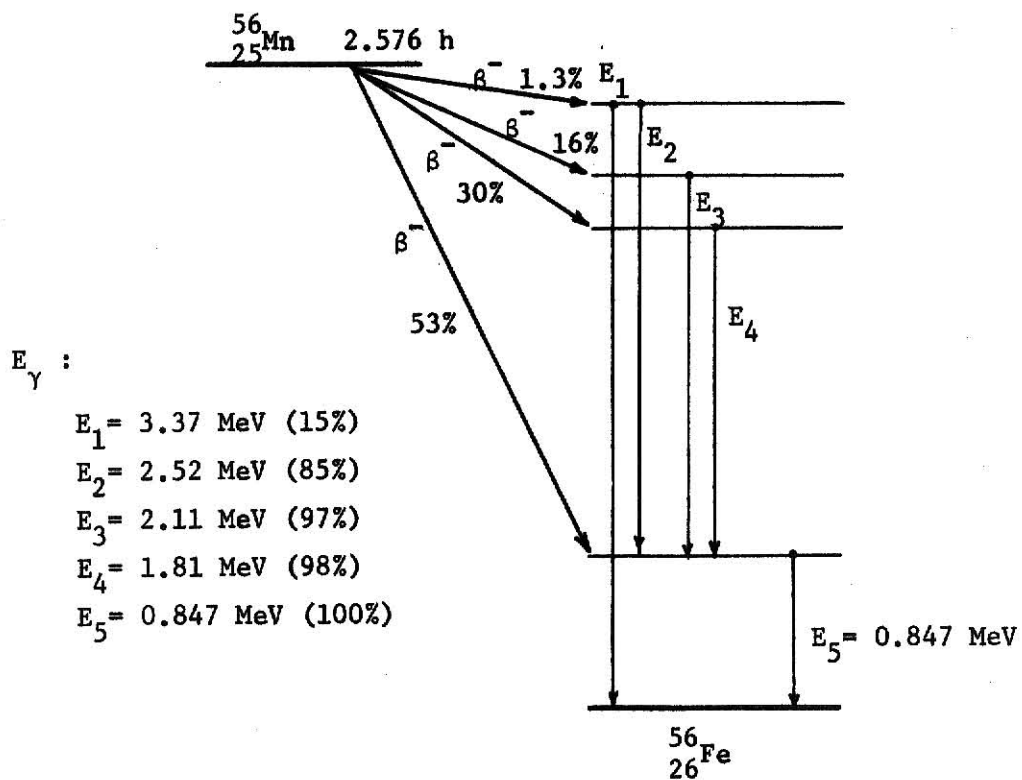
Previous work was reported to have irradiated sugar (samples)-water (references) pairs by manually transporting the samples. This procedure was repeated in order to record the transporting and cooling times before counting. The recorded times for six sets of samples varied from 30 seconds (the best time) to 3 minutes. The average was approximately 2 minutes. For each recorded time the following steps were performed:

- a) after completing the irradiation, the irradiation vial was taken from the RSR, b) both unknown and reference samples were removed from the irradiation vial while the samples were being manually transported from the top of the reactor to the NAA laboratory (for the case with the Ge(Li) detector in its normal laboratory location), c) at the same time, the iron wire fluence monitor for the unknown sample was removed, d) the sample was placed on the Ge(Li) detector, e) a spectrum was accumulated for 30 seconds, f) after accumulating the spectrum for the first sample, the sample was removed, g) the reference sample was placed on the Ge(Li) detector, and h) the spectrum for the reference was accumulated for 30 seconds.

Although these steps (except for the extreme hurry involved in this work) are the normal steps to follow in the comparator technique in NAA, it is easy to understand their limitations when a short-lived radioisotope, as ^{19}O , is to be analyzed. Therefore, this procedure was considered as impractical because of the slowness of the procedure. It just required too many factors for good control. Further the times recorded justified the decision that a faster method was needed and one in which fewer number of steps were required would be desirable.



(a)



(b)

Figure 4.1 Simplified decay schemes for (a) ^{19}O and (b) ^{56}Mn (16).

Table 4.1. Results of experiments performed to find the best manual method for transporting samples from the irradiation source to the counting system.

Exp. #	Method of Transporting	Material	Average Decay Time (sec)	Mole Ratio (Average) Sample/H ₂ O (Expected 6:1)	% Error $\frac{(6-\text{Found}) \times 100}{6}$	Number of Samples	
						Irr.	Count.
1	"Running"	Glucose	98.7	--	--	3	3
		Fructose	104.0	--	--	3	3
		H ₂ O	41.5	--	--	8	8
2	"Basket"	Gluc.	60	4.35 $\pm$ 0.30	25.6	5	1
		Fruc.	40	6.69 $\pm$ 0.15	-11.6	5	4
		H ₂ O	96.6			10	5
3	at Reactor Bay	Gluc.	15.5	8.75 $\pm$ 0.17	-45.8	5	4
		Fruc.	19.3	5.55 $\pm$ 0.11	7.5	5	3
		H ₂ O	57.9			10	7

Several methods were suggested, and the following were considered and/or tested (see Table 4.1):

a) An automatic system for transferring samples from the irradiation source to the radiation detector system. Although this method would be best - the most efficient and quickest - it might involve several technical problems in design and construction due to building design, and safety regulations, for example. This method was considered to be beyond the scope of this work.

b) Another method tested was the use of a small basket for transporting the irradiation vial from the top of the reactor down to an exit door in the path way to the NAA laboratory, the normal site of the counting system. The basket was attached to a cord tied at both ends, at the top of the reactor and near the door to the NAA laboratory. The basket was allowed to fall by gravity. However, due to the speed gained when the basket slid down the cord, problems of handling the irradiation vials were typical. Most of the samples were lost from the impact and the iron wires were confused for the two samples. Therefore, this was also an impractical procedure, and it was eliminated.

c) After the failure or rejection of the methods described in a) and b), it was decided that only improvement for more rapid transportation could be obtained by moving the counting-analyzer system, or the counting system only, to the reactor bay. This was the simplest method available because typical cooling times of 15 seconds (about one half of an ^{19}O half life) were obtained. Although the number of steps required in the comparator technique were not reduced, there was enough "gain" in time to allow for performing the experiment more efficiently.

The sacrifices made with this method were the high background during reactor operation and electronic noise caused by sources in the line voltages (due to pumps of the reactor cooling system, air compressors, etc.). Also, at least two people were required to perform the experiment; one at the irradiation source and the other at the counting system. However, this was the most feasible and simplest method, and was used successfully in this work.

In Table 4.1 the most important results and data of experiments described in this section are summarized.

4.2 Measurements for Record

All the irradiations were performed in the RSR (positions 1 or 2) of the KSU-TRIGA reactor. Maximum power levels were desired in this work so that irradiation times could be small enough to induce radioactivation in the ^{18}O while minimizing interferences from longer-lived radionuclides in the sample.

4.2.1 Analyzed Samples

Most of the samples for analysis in this work consisted of matrices of distilled water, α -D-Fructose, and α -D-Glucose. However, several wheat samples, alcohol-water mixtures, and other oxides were analyzed also.

The sugar samples were dried before irradiation for 24 hours or more at 50 °C. The water, wheat, alcohol, and oxide samples were not treated in any way.

All samples were contained in the small polyethylene vials. All vials were carefully cleaned before filling with samples. They were washed with detergent (laboratory grade) and were chemically treated,

consisting of immersing in nitric acid (1%) for 5 minutes or longer. Rinse was performed with distilled water. After cleaning, plastic gloves were used to handle the vials and to avoid any possible contamination.

Table 4.2 shows the approximate weights of typical samples.

Table 4.2 Typical Sample Weights.

Sample	Weight (grams)	Comments
α -D-Glucose	0.8	Purchased: Mallinckrodt Inc., (St. Louis, MO)
α -D-Fructose	0.8	Purchased: Fisher Scientific Co., (Fair Lawn, NJ)
Distilled Water	1.2	
Alcohol-H ₂ O	0.9	50% water-50% ethanol
Wheat	0.7-1.0	
Oxides	0.2	

All weighing of samples and polyethylene vials was performed on a Mettler balance with an accuracy of approximately ± 0.0001 gm.

Often distilled water was used as the reference standard in this comparator NAA work. The natural concentration of ^{18}O in H_2O was assumed to be 0.204% (15).

4.2.2 The Neutron Fluence Monitor

Iron wire was chosen as a fluence monitor for this work principally for two reasons. First, it was easy to manipulate, and second it did not require a container.

^{56}Mn was formed in sufficient quantity to observe good counting statistics in the 0.86 Mev photopeak. The formation of the ^{56}Mn was due principally to the (n,p) reaction with ^{56}Fe .

Each iron wire was approximately 1/64 inch (0.1588 cm) in diameter and of analytical reagent grade. Two ways were used to place the iron wires for monitoring the neutron flux near the samples. In one, the iron wires were wrapped through the hinge of the cover of the polyethylene irradiation vial. In this case, each iron wire was approximately 1.5 inches (3.81 cm) long and 0.067 grams in weight. In the second case, the iron wires were placed in quadruplicate, each one approximately 3/8 inches (0.9525 cm) long and 0.009 grams in weight, at 90° positions around the sample. In Fig. 3.1 the positions of the iron wires in the irradiation vial for this later case are shown. The reasons for placing the iron wires were a) easy and fast removal in the first case, and b) better flux monitoring (fixed geometry) in the second case.

4.2.3 Irradiation Conditions

The two different forms of using iron wires described in last reaction corresponded to two different irradiation conditions, as well. In both cases the typical irradiation time was two minutes and the reactor power level was about 225 kw (1.8×10^{12} n/cm² sec; see Table 3.1) the maximum attainable. Before all the irradiations the reactor was brought to full power level. The irradiation vial containing the sample(s) and the iron wire(s) was then inserted into the RSR. At the end of the irradiation time the capsule was retrieved by a cord previously tied to it. After retrieving the capsule, it was inserted into the counting system.

Both operations were accomplished as quickly as possible. Two stop watches were used; one to record the irradiation time, the other to record the elapsed time between the end of an irradiation and the beginning of counting the sample to accumulate the gamma-ray spectrum. The reactor was not scrammed between individual irradiations of samples.

The difference between the two irradiation schemes consisted of:

a) In the first the unknown (sugars) and the standard reference (H_2O) samples were irradiated together to maintain consistent irradiation conditions. Since the radionuclide of interest, ^{19}O , was short-lived (29 seconds), it was only feasible to irradiate one sample-reference pair at one time. There were 20 glucose samples and 20 fructose samples along with their respective reference H_2O samples. There were approximately 15 and 50 seconds decay times for the unknown and the reference samples, respectively. These times corresponded approximately to a measurement of time between the end of irradiation and the beginning of counting.

b) In the second irradiation conditions, a fixed geometry was the typical consideration. In this case only one sample, unknown or reference, was irradiated along with four iron wires (flux monitors). It was assumed that the iron wires could normalize the data to achieve conditions needed for the comparator technique (see Theory, Section 2). Typical decay times before counting were approximately 15 seconds. The iron wires were allowed to decay for approximately an hour between the end of irradiation of samples and the beginning of counting.

4.2.4 Counting Procedures

A Ge(Li) semiconductor detector was used for the counting of all samples. Additional details on the equipment can be found in the "Description of Apparatus," Section 3. Data for a particular sample spectrum were, in general, accumulated in 1024 channels configuration of the multichannel analyzer. The analyzer dead time was never greater than 10% for any of the measurements. The samples irradiated by pairs of unknown-reference were counted for 32 "live" seconds (i.e., the analyzer automatically corrects for analysis time). For any other case, including data for iron wires, a counting time of 60 "live" seconds, was the standard time.

Because the gamma-ray energy of interest was only 197 keV, amplification gains in the spectroscopy amplifier were set so that 197 keV corresponded approximately to channel 200. However, such amplification gains were somewhat variable due to the use of the long coaxial cable between the Ge(Li) counting and the analyzer.

A Nova computer code was developed to be used for automatic control of the analyzer and the magnetic tape units. With the Nova, the following consecutive steps were performed: a) start the accumulating in PHA-add mode for the previously selected time, b) deposit data on magnetic tape, c) increment tag word for recording the next spectrum, and d) erase the analyzer memory. The starting of the cycle was performed externally by a push-button control at the Ge(Li) system.

Because of the importance of counting geometry in the comparator technique of NAA, the special sample holder of plexiglass was constructed. (See Fig. 3.4). The device was placed directly on the top of the Ge(Li) detector. The principal characteristic is the center hole, 15/32 inch (1.1906 cm) diameter, which provided accurate positioning of samples, so that unknown and reference samples were approximately of equal cylindrical volume sources of irradiation. Similar sample containers were also used to count iron wires at same position. The detector was partially surrounded by a lead shield to reduce background interference.

A normal procedure in NAA before counting is to transfer samples to previously weighed, nonirradiated polyethylene vials. In this work, it was not possible to perform this step because of the short half-life of ^{19}O , and interference background from external contamination for the irradiated vials was possible.

5. Analysis of Results

This work for determining the ^{18}O was developed primarily to provide a easy method of measuring the concentration of ^{18}O in sugars, in connection with tracer studies of the mechanism of reactions in the sugar series (1). Before other biological samples of unknown ^{18}O content, as wheat for example, could be processed, the precision and accuracy of the thermal NAA method was investigated by calculating the molecular ratio of sugar ($\alpha\text{-D-Glucose}$ and $\alpha\text{-D-Fructose}$) to distilled water (standard reference) of ^{18}O levels.

5.1 Experimental Results Without Fixed Geometry Conditions

Within the conditions of the comparator technique in NAA, two sets of twenty pairs, sugar-water samples, were irradiated and analyzed (see Table 5.1). More information on samples, irradiation, and counting procedures can be found in "Measurements for Records" section of this work.

The area under the desired photopeak was calculated by the Gregory's formula of numerical integration which is based on a finite difference technique (13). More information is found in Appendix B.

In Table 5.1 the analysis performed for the two sets of sugar samples is shown. The mole ratio of oxygen in sugar was determined on the basis of equation 2.7 by use of the following expression:

$$\text{Mole Ratio} = \frac{\frac{A^* (\text{sugar})}{A^* (\text{Fe sugar})}}{\frac{A^* (\text{water})}{A^* (\text{Fe Water})}} \times \frac{\text{Mol. weight (sugar)}}{\text{Mol. weight (H}_2\text{O)}}, \quad (5.1)$$

ILLEGIBLE DOCUMENT

**THE FOLLOWING
DOCUMENT(S) IS OF
POOR LEGIBILITY IN
THE ORIGINAL**

**THIS IS THE BEST
COPY AVAILABLE**

TABLE 5.1 RATIO OF ACTIVITY OF SUGAR TO DISTILLED WATER (5:1-MOLE BASIS)
EACH SAMPLE HAS ITS OWN REFERENCE. NO FIXED-GEOMETRY DURING
COUNTING OR IRRADIATING.

SAMPLE	EXPECTED MOLE RATIO	OBSERVED MOLE RATIO	KINETIC MULTIPLIER	CORRECTED MOLE RATIO	% error ^b
GLUCOSE S.# A1	6:1	6.036 (0.215) ^a	1.063	6.416 (0.229)	-6.937
GLUCOSE S.# B1	6:1	4.433 (0.153)	1.063	4.712 (0.163)	21.467
GLUCOSE S.# C1	6:1	8.771 (0.304)	1.063	9.271 (0.323)	-54.509
GLUCOSE S.# D1	6:1	6.286 (0.201)	1.063	6.661 (0.214)	-11.358
GLUCOSE S.# E1	6:1	4.425 (0.150)	1.063	4.708 (0.159)	21.530
GLUCOSE S.# F1	6:1	7.819 (0.261)	1.063	8.312 (0.277)	-39.532
GLUCOSE S.# G1	6:1	6.511 (0.211)	1.063	6.921 (0.224)	-15.357
GLUCOSE S.# H1	6:1	5.679 (0.178)	1.063	6.037 (0.189)	-6.610
GLUCOSE S.# I1	6:1	6.510 (0.203)	1.063	6.920 (0.221)	-15.130
GLUCOSE S.# J1	6:1	6.515 (0.229)	1.063	6.925 (0.244)	-15.431
GLUCOSE S.# K1	6:1	5.452 (0.179)	1.063	5.795 (0.190)	-3.408
GLUCOSE S.# L1	6:1	6.368 (0.202)	1.063	6.769 (0.215)	-12.012
GLUCOSE S.# M1	6:1	5.414 (0.175)	1.063	5.755 (0.186)	-4.081
GLUCOSE S.# N1	6:1	6.958 (0.239)	1.063	7.356 (0.276)	-23.273
GLUCOSE S.# O1	6:1	6.215 (0.195)	1.063	6.611 (0.207)	-10.176
GLUCOSE S.# P1	6:1	5.962 (0.176)	1.063	6.359 (0.200)	-5.779
GLUCOSE S.# Q1	6:1	6.268 (0.192)	1.063	6.663 (0.205)	-11.068
GLUCOSE S.# R1	6:1	5.619 (0.175)	1.063	5.973 (0.188)	-3.451
GLUCOSE S.# S1	6:1	4.551 (0.147)	1.063	4.837 (0.156)	14.556
GLUCOSE S.# T1	6:1	5.996 (0.190)	1.063	6.374 (0.202)	-5.735
AVERAGE =		6.063 (0.046)		6.471 (0.049)	-7.854
FRUCTOSE, S# 1	6:1	5.507 (0.203)	1.063	5.854 (0.216)	-2.426
FRUCTOSE, S# 3	6:1	5.016 (0.149)	1.063	5.332 (0.158)	11.137
FRUCTOSE, S# 5	6:1	4.637 (0.143)	1.063	4.929 (0.157)	17.547
FRUCTOSE, S# 7	6:1	6.664 (0.229)	1.063	7.084 (0.243)	-19.070
FRUCTOSE, S# 9	6:1	5.565 (0.200)	1.063	5.915 (0.212)	-3.408
FRUCTOSE, S# 11	6:1	5.024 (0.177)	1.062	5.345 (0.188)	10.999
FRUCTOSE, S# 13	6:1	6.635 (0.230)	1.063	7.053 (0.245)	-17.545
FRUCTOSE, S# 15	6:1	7.690 (0.273)	1.063	8.175 (0.291)	-36.242
FRUCTOSE, S# 17	6:1	7.978 (0.286)	1.063	8.481 (0.304)	-41.345
FRUCTOSE, S# 19	6:1	5.942 (0.131)	1.063	6.264 (0.133)	-4.393
FRUCTOSE, S# 21	6:1	4.340 (0.129)	1.063	4.614 (0.137)	23.103
FRUCTOSE, S# 23	6:1	6.179 (0.213)	1.063	6.598 (0.227)	-9.473
FRUCTOSE, S# 25	6:1	5.928 (0.210)	1.063	6.301 (0.223)	-5.023
FRUCTOSE, S# 29	6:1	5.649 (0.196)	1.063	6.005 (0.208)	-6.077
FRUCTOSE, S# 31	6:1	2.016 (0.073)	1.063	2.143 (0.073)	64.289
FRUCTOSE, S# 33	6:1	5.409 (0.187)	1.063	5.750 (0.199)	-4.173
FRUCTOSE, S# 35	6:1	5.254 (0.202)	1.063	5.641 (0.216)	-5.151
FRUCTOSE, S# 37	6:1	6.331 (0.215)	1.063	6.750 (0.228)	-12.163
FRUCTOSE, S# 39	6:1	5.944 (0.197)	1.063	6.324 (0.209)	-5.394
AVERAGE =		5.672 (0.046)		6.029 (0.049)	-5.488

^a The values in parentheses are standard deviations.

^b The error percentage is calculated by dividing the difference of the expected mole ratio and the corrected mole ratio by the expected mole ratio.

where the molecular weights are 180.16 and 18.015 for sugar and H_2O , respectively. The expected mole ratio, i.e., the theoretical value, is 6:1 calculated by the ratio of oxygen in the sugar (the chemical formula is $C_6H_{12}O_6$) to the oxygen in H_2O . We assumed a natural occurrence of ^{18}O in both cases. The ratio of sugar to water for ^{18}O levels was expected to be smaller than the desired value (16). Then the kinetic multiplier of 1.063 was used to correct the experimental ratio. This kinetic effect was assumed to appear due to the preference of ^{16}O and ^{17}O over ^{18}O when sugars are being produced. The value 1.063 was calculated at 25 °C using the following equation (1,5,22)

$$\frac{k_1}{k_2} = (m_2/m_1)^{1/2} [1 + G(u)\Delta u] , \quad (5.2)$$

where

$\frac{k_1}{k_2}$ = maximum isotope effect to be expected in the chemical kinetic for the hypothetical process, $C-O \rightarrow C+O$,

m = reduced mass of the fragment,

$u = hcw/kT$,

Δu = difference in the value of u for the two isotopic species,

$G(u)$ = function for which numerical values are available from tables (22),

h = Planck's constant,

c = velocity of light,

w = average value for the stretching frequency of the bonds of the process,

k = Boltzmann's constant,

T = temperature.

The accuracy, or % of error, in Table 5.1 was calculated as follows:

$$\% \text{ Error} = \frac{\text{Desired Value} - \text{Found Value}}{\text{Desired Value}} \times 100 . \quad (5.3)$$

The precision or standard deviation of each measurement was calculated via the comparator technique where common parameters in numerator and denominator are canceled (assuming as constants the neutron flux and N , as was explained in Section 2). The main contributor is the error in the measurement of the area under photopeak of interest. Detailed formulation can be found in Appendix C.

5.2 Discussion of the Source of Errors

It is well recognized that any physical magnitude can never be measured exactly, i.e., with no error. Progressively more elaborate experimental or theoretical efforts result only in reducing the possible error of the determination. In reporting the result of any measurement it is therefore obligatory to specify also the probability that the result is in error by some specific amount, since a gamble on relative correctness is always involved in all physical determinations. The discussion of source of errors, presented here, describes several types of error, including gross mistakes, to determine the quality or reliability of data, particularly data of the type encountered in the method presented in this work.

Several experiments were performed to discover the effect of the following points as sources of errors:

- a. Inadequate manipulation of samples
- b. Incorrect weighing of samples and recording of time
- c. Improper method of area calculation
- d. Irregularities in position during irradiation and/or counting
- e. Significant variations in the reactor power level during sample irradiation
- f. Instability of the electronic counting system

Manipulation of Samples

Special care was taken during preparation of samples, as explained in Section 4.2, "Analyzed Samples." However, most of the careful handling was to avoid contamination of the samples. There were, however, some other variations, sometimes out of control of the experimenter which might have introduced errors in the measurements. Loss of material was observed in some experiments due to the splitting of the sample vials (from impact when the samples were dropped from the reactor top to be inserted into the counting system). Considerable water was observed in RSR, so an increase of radioactive mass was also possible.

Because most of the samples were prepared one day before the irradiation and stored at room temperature, some evaporation (principally in the water samples and very critically in the alcohol mixtures) could cause a decrease of sample mass. In one water sample up to a 2% decrease of mass from the time of preparation to next day 6 hours after irradiation was recorded; a sample of water-alcohol mixture showed a 9.3% decrease during a similar elapsed time. No change was observed in any of the sugar samples.

A final contributor of error could have been a possible oxygen exchange with air during the preparation of the samples. This exchange might have changed measurements of the oxygen fraction.

The set of samples presented in Table 5.1 was then analyzed using a fixed water reference sample for all groups of sugars and the other sets of distilled water. Four water references, two from each group of sugars, were selected to be the standard reference. They correspond to the glucose samples, H1 and Q1, and to the fructose samples 9, and 23. Also a sample of glucose (labeled C1) and one of fructose (labeled 9) were used as standard reference samples. Their respective group samples were compared. In

Table 5.2 a summary of the results of these analyses is presented. A considerable improvement with respect to results in Table 5.1 was observed in the percentage error as well as the distribution of the percentage error. These results confirmed that errors due to manipulation of sample were of some importance.

Error Due to Incorrect Weighing of Samples and Recording of Time

In the standard comparator technique it is normal to consider the area under the photopeak as the main contributor to the uncertainty of the measurements of the activity of an activated sample. It is also considered that the parameters related to time measurements can be very well determined. The standard deviation of each individual mole ratio in Table 5.1 was calculated considering the error in the area determination for each unknown sample and it's reference sample as well as their respective fluence monitors.

However, as was pointed out in the introduction of this work, the irradiation time and the decay time (or transfer time) were of concern, i.e., the difference in half-lives between ^{19}O (29.3 sec) and ^{56}Mn (2.56 hr) might result in the ^{56}Mn not being sensitive enough to time variations. The possible variations in the uncertainty of the determination of an activity, per unit mass, as a result of time variations were calculated. Fluctuations as large as 10% of the decay and irradiation times measurements and 5% in counting time were considered in the calculation of the standard deviation associated with the activity per unit mass. The general expression is given:

$$R^* = K \frac{\text{Area}}{\text{Mass}} [e^{-\lambda t_d} (1 - e^{-\lambda t_i}) (1 - e^{-\lambda t_c})]^{-1}, \quad (5.1)$$

where K is a constant containing the over-all counting efficiency, the isotopic abundance, the mass number, etc.,

TABLE 5.2 Average Mole Ratios (gm- mole basis) for Experiment 4. Each sample was normalized for the neutron fluence.

MATERIAL	ACTUAL MOLE RATIO	M E T H O D 1 ^a		M E T H O D 2		M E T H O D 3		No. of Meas. ≥ 10% error ^c		
		FOUND	% ERROR ^b	FOUND	% ERROR	FOUND	% ERROR	1	2	3
Group 1 ^d										
Glucose	6	6.18 (0.06) ^g	-2.9	6.47 (0.05)	-7.9	6.38 (0.06)	-6.4	13	13	13
Fructose	6	5.87 (0.06)	2.1	6.03 (0.05)	-0.5	5.94 (0.06)	1.0	12	10	10
Group 2										
Glucose	6	6.10 (0.06)	-1.7	6.31 (0.05)	-5.1	5.88 (0.05)	2.0	6	10	5
Water (G) ^e	1	1.01 (0.01)	-0.5	0.98 (0.01)	1.3	0.93 (0.01)	6.7	7	9	8
Fructose ^f	6	5.66 (0.06)	5.7	6.40 (0.05)	-6.7	5.72 (0.05)	4.6	8	10	7
Water (F) ^f	1	1.00 (0.01)	0.01	1.09 (0.01)	-9.9	0.99 (0.01)	0.7	10	10	8
Group 3										
Glucose	6	6.07 (0.05)	-1.2	7.11 (0.05)	-18.5	6.07 (0.05)	-1.1	6	14	8
Water (G)	1	0.99 (0.01)	0.1	1.12 (0.01)	-11.9	0.97 (0.01)	3.5	7	12	8
Fructose	6	5.63 (0.05)	6.2	7.22 (0.06)	-20.3	5.91 (0.05)	1.5	9	19	4
Water (F)	1	0.99 (0.01)	0.5	1.24 (0.01)	-23.9	1.03 (0.01)	-2.5	10	16	8
Group 4										
Glucose	6	6.87 (0.07)	-14.5	6.10 (0.05)	-1.7	6.32 (0.06)	-5.4	14	10	9
Water (G)	1	1.13 (0.01)	-13.1	0.96 (0.01)	4.4	1.01 (0.01)	-0.8	11	9	5
Fructose	6	6.37 (0.07)	-6.2	6.19 (0.05)	-3.3	6.16 (0.06)	-2.6	8	7	6
Water (F)	1	1.13 (0.01)	-13.3	1.07 (0.01)	-6.7	1.07 (0.01)	-7.1	13	9	9
Group 5										
Glucose	6	6.25 (0.06)	-4.2	6.22 (0.05)	-3.6	6.02 (0.05)	-0.3	9	10	6
Water (G)	1	1.03 (0.01)	-2.9	0.97 (0.01)	2.6	0.96 (0.01)	4.1	8	10	8
Fructose	6	5.78 (0.06)	3.4	6.31 (0.05)	-5.2	5.86 (0.06)	2.3	7	8	5
Water (F)	1	1.03 (0.01)	-2.6	1.09 (0.01)	-8.8	1.02 (0.01)	-1.8	8	10	8
Group 6										
Glucose	1	0.91 (0.01)	8.7	0.91 (0.01)	8.9	0.91 (0.01)	8.7	9	11	7
Fructose	1	1.07 (0.01)	-7.4	1.05 (0.01)	-5.0	1.08 (0.01)	-7.6	8	8	9

^a Areas under photopeaks calculated by: Method 1 = Graphical Method (GM) with fixed-minima for background subtraction
 Method 2 = Gregory's equation with no background subtraction.
 Method 3 = GM with variable-minima for background subtraction.

^b The error percentage is calculated by dividing the difference of the actual mole ratio and the calculated mole ratio by the actual mole ratio.

^c The number of individual measurements out of the total number of samples (20 glucoses and 19 fructoses) of which the percentage error is $\geq 10\%$.

^d The groups are formed from the different standard references used:

Group 1 = each sample has its own water reference.

Group 2 = reference is the water sample H2 (glucose group).

Group 3 = reference is the water sample Q2 (glucose group).

Group 4 = reference water sample 10 (fructose group).

Group 5 = reference water sample 24 (fructose group).

Group 6 = for glucoses the glucose sample C1; for fructoses the fructose sample 9.

^e The water samples with (G) were used as references for the glucose samples.

^f The water samples with (F) were used as references for the fructose samples.

^g The values in parentheses are standard deviations.

t_d is the decay time,

t_i is the irradiation time,

t_c is the counting time.

The results of these calculations, see Appendix C, have shown net differences in the standard deviation up to three times larger than the same measurement with the standard deviation for the area alone. The main contributor is the 10% variation in the decay time. If a 10% variation in mass occurs, the difference could be as large as 5 times, especially for photopeaks which are not well-defined (low counting statistics). For the iron wires there were differences as large as 10 times, when similar calculations were performed. For this work, it must be noted that variations as large as mentioned above were never observed except for the mass as was explained.

Method of Area Calculation

As was earlier mentioned, the area under the photopeak of the gamma-ray spectrum associated with the activity of the sample being analyzed is directly proportional to the weight of the desired element in sample. Therefore, the correct determination of the area is a very important step in the analysis, so much so that special works have been reported by several researchers (9,11,12,21). Most of these methods were developed for gamma-ray spectra obtained with a NaI(Tl) detector system which exhibits a principal characteristic of a large number of channels to define any photopeak, i.e., poor energy resolution compared to Ge(Li). For a high resolution Ge(Li) system, a few channels are required to define similar photopeaks. Methods in the first case include simple graphical stripping to a more sophisticated "smoothing" of data using the facilities of a

large digital computer. Although subjectivity is always present in the analysis, these methods have proved to be satisfactory, especially for the gamma-ray spectra that show well-resolved photopeaks. All methods can use spectra from Ge(Li) systems; however it was felt that sophisticated methods (24,25) should not necessarily give a considerable improvement when compared to the simpler graphical method because of the few channels that define the photopeak. The graphical method is introduced in Appendix B of this work.

Analysis of the same samples as presented in Table 5.1 was performed using Gregory's equation, see Appendix B, for the area calculation. For this method it was assumed that for well-resolved full-energy peaks, in which the Compton continuum is reasonably flat, the area under the top six channels around the maximum defines a fixed fraction of the total area of photopeak and is constant for spectra of similar samples (unknown and reference samples). This last assumption could not be completely fulfilled, in particular for the reference water samples, where fluctuations in mass and irradiation and counting times might produce considerable variations in the number of counts in the desired photopeak. Because it is necessary to determine how much of a given photopeak is actually due to photoelectric events of gamma-rays and how much is due to the Compton effect, the subtraction of the later portion of the area under the peak from the total area is the usual practice (9,11,12,21). In Gregory's method that subtraction was not performed. Then, two different approaches were used, explained below and in Appendix B.

In these two methods as well as in Gregory's method, the response of the pulse-height analyzer is represented graphically as rectangles whose areas are proportional to the number of counts in the channels. In the two

new methods, for "background" subtraction, a baseline extrapolated between the valleys adjacent to the photopeak was estimated. Among several possible ways to estimate the baseline, a straight line between the minima is the simplest, though not necessarily the best. However, that is the one that was used for both methods. The difference between both methods is the way of finding the minimum on each side of the peak. In one method the minima are the fourth channel before and after the channel corresponding the maximum number of counts of the peak. In the other method both minima are chosen as follows: for a peak with a maximum in channel "n", for the left minimum, the average value of the content from channels n-10 to channel n-2 is calculated. The minimum is chosen as " C_l " for which

$$C_l \leq \sum_{i=n-10}^{n-2} C_i + \left(\sum_{i=n-10}^{n-2} C_i \right)^{1/2} . \quad (5.2a)$$

For the right side, the average value is calculated from channel n+2 to channel n+10. The minimum is chosen as " C_r " for which

$$C_r \leq \sum_{j=n+2}^{n+10} C_j + \left(\sum_{j=n+2}^{n+10} C_j \right)^{1/2} . \quad (5.2b)$$

More details are given in Appendix B. The effective area under the photopeak is then calculated as the sum of the counts of channel between minima (inclusive). The estimated background is subtracted by averaging three adjacent channels on both minima (two before the left minimum and the left minimum; two after the right minimum and minimum). The baseline (background) may be calculated, subjectively, with a considerable variation. When the photopeak is clearly defined, the fractional error in the photopeak area may be quite small because the background is small compared to the photopeak. The error becomes more significant as the mixture of radio-

nuclides increases and the lower energy peaks represent increasingly smaller fractions of the total counts in the photopeak.

The data in Table 5.1 were analyzed using these two methods. Results are summarized in Table 5.2. Thus it is possible to compare the results from the three methods.

Table 5.2 is a summary of average results of the calculated molecular ratios and the associate percentage error. Columns 8, 9, and 10 represent the spread of the percentage error of individual measurements which are equal to or greater than 10%. The first two lines are the average values of Table 5.1 by the different area-methods.

From Table 5.2 it can be seen that a particular set of samples is better defined by either selecting one method of area calculation and/or a specific standard reference, see for example lines one and three where Glucose samples favor using the method of fixed minimum for area calculation and a fixed standard reference. The situation can be reversed for other methods of area calculation or other references, see lines eleven and fourteen where Gregory's method gives the better results.

When samples and references are identical either method of area calculation can be used, see lines nineteen and twenty.

The main difference between those methods is in the distribution of percentage error of individual samples. From this point of view Gregory's method proved to be unsuccessful while the variable minimum - finding method was the most adequate. Posterior studies determined that Gregory's method seemed to be the least appropriate method in this work, see successive Tables 5.3 to 5.6. Under low statistics (poorly defined peaks), Gregory's equation overestimates the evaluation of peaks, probably because the method does not subtract for the background and some points of the background are included in the true area.

Geometry During Irradiation and Counting

The results given in Tables 5.1 and 5.2 were obtained while trying to keep the transfer time as small as possible and trying to treat the unknown and reference sample in similar fashion. The position of the samples and fluence monitors were uncertain resulting in a great possibility of errors. Iron wires were wrapped around the hinge of the polyethylene vial instead of wrapped around the center of such a vial.

It was felt that the fluence monitor did not normalize the data to the neutron flux to which the sample was exposed. Hence a considerable error in the analysis of data was introduced.

Although the geometric position for counting samples is important in the comparator technique, it was not as critical as the position for irradiation. However it was decided to keep the sample vial fixed with respect to the reactor core during irradiation and the Ge(Li) detector during counting, for subsequent experiments (see the description of irradiation and counting conditions for fix-geometry in Section 3, "Description of Apparatus").

To study the effect that these iron wires could have on the analysis, a simple analysis was performed, and the results are given in Table 5.3. Thus, the same set of data was analyzed considering the neutron flux as constant. Also, the same analysis was performed assuming no normalization for neutron flux. In Table 5.3, notice there is a considerable improvement in the fluctuation of the percentage error for individual samples (see columns eight to ten). There was no noticeable change in the average percentage errors, however, the values tend to be more in accord with the expected lower concentration of ^{18}O in sugar than in water (positive percentage error predominates the results given in Table 5.3 compared to the observed negative tendency of the results given in Table 5.1).

TABLE 5.3 Average Mole Ratios (gm- mole basis) for Experiment 4. Samples are not normalized for the neutron fluence.

MATERIAL	ACTUAL MOLE RATIO	M E T H O D 1		M E T H O D 2		M E T H O D 3		No. of Meas. > 10% error		
		FOUND	% ERROR	FOUND	% ERROR	FOUND	% ERROR	1	2	3
Group 1										
Glucose	6	6.01 (0.05)	-0.2	6.23 (0.04)	-3.8	6.18 (0.05)	-2.9	8	5	7
Fructose	6	5.93 (0.05)	1.3	6.09 (0.04)	-1.6	6.04 (0.05)	-0.7	10	8	7
Group 2										
Glucose	6	5.79 (0.05)	3.5	5.98 (0.04)	1.7	5.51 (0.04)	8.1	6	4	6
Water(G)	1	0.97 (0.01)	3.0	0.96 (0.01)	4.2	0.89 (0.01)	10.3	4	7	10
Fructose	6	5.98 (0.05)	0.4	6.42 (0.04)	-7.0	5.73 (0.04)	4.4	3	6	3
Water(F)	1	1.02 (0.01)	-1.8	1.07 (0.01)	-6.8	0.96 (0.01)	4.3	5	9	7
Group 3										
Glucose	6	5.72 (0.04)	4.7	6.69 (0.04)	-11.6	5.95 (0.04)	0.9	7	12	4
Water(G)	1	0.96 (0.01)	4.3	1.09 (0.01)	-9.4	0.97 (0.01)	2.7	5	11	4
Fructose	6	5.90 (0.05)	1.6	7.29 (0.05)	-21.5	6.19 (0.05)	-3.1	3	18	3
Water(F)	1	1.01 (0.01)	-0.6	1.21 (0.01)	-21.2	1.03 (0.01)	-3.3	4	15	4
Group 4										
Glucose	6	5.94 (0.05)	1.0	5.86 (0.04)	2.4	6.03 (0.05)	-0.3	4	4	5
Water(G)	1	0.99 (0.01)	0.4	0.95 (0.01)	4.7	0.99 (0.01)	1.5	4	6	4
Fructose	6	6.13 (0.05)	-2.2	6.38 (0.04)	-6.3	6.26 (0.05)	-4.3	4	5	3
Water(F)	1	1.05 (0.01)	-4.7	1.06 (0.01)	-6.4	1.05 (0.01)	-4.7	6	9	3
Group 5										
Glucose	6	5.29 (0.04)	11.8	5.66 (0.04)	5.6	5.60 (0.04)	6.6	11	6	4
Water(G)	1	0.89 (0.01)	11.2	0.92 (0.01)	7.8	0.92 (0.01)	8.3	11	7	7
Fructose	6	5.46 (0.05)	9.0	6.17 (0.04)	-2.8	5.83 (0.05)	2.9	8	5	3
Water(F)	1	0.93 (0.01)	7.3	1.03 (0.01)	-2.7	0.97 (0.01)	2.9	9	8	6
Group 6										
Glucose	1	0.87 (0.01)	13.3	0.89 (0.01)	10.3	0.92 (0.01)	8.2	13	9	6
Fructose	1	0.95 (0.01)	5.2	0.97 (0.01)	3.3	0.95 (0.01)	4.6	4	6	3

For explanations see footnotes in Table 5.2.

TABLE 5.4 Average Mole Ratios (gm- mole basis) for Experiment 5.

Part A: Each sample normalized for the neutron fluence.

MATERIAL	ACTUAL MOLE RATIO	METHOD 1		METHOD 2		METHOD 3		No. of Meas. ≥ 10% error		
		FOUND	% ERROR	FOUND	% ERROR	FOUND	% ERROR	1	2	3
Group 1 ^d										
Fructose	6	7.0 (0.70)	-16.6	7.88 (0.43)	-31.3	7.00 (0.60)	-16.5	3	3	3
Glucose	6	9.86 (0.84)	-64.3 _h	9.67 (0.47)	-61.2	11.86 (0.93)	-97.6	4	3	4
CaCO ₃	3	86.97(13.65)	*****	698.70(59.70)	*****	371.80(52.90)	*****	3	3	3
Water ₃ (L2)	1	0.73 (0.11)	26.9	0.87 (0.08)	13.2	0.68 (0.08)	31.6	1	1	1
CaHPO ₄	4	14.6 (2.00)	*****	152.44(11.93)	*****	9.26 (1.22)	*****	1	1	1
Group 2										
Fructose	6	9.58 (0.94)	-59.7	9.08 (0.49)	-51.3	10.22 (0.84)	-70.3	3	3	3
Glucose	6	13.5 (1.10)	*****	11.10 (0.50)	-85.7	17.34 (1.32)	*****	5	4	5
CaCO ₃	3	120.46(18.13)	*****	804.74(67.37)	*****	543.53(74.8)	*****	3	3	3
Water(K2)	1	1.37 (0.19)	-36.9	1.15 (0.10)	-15.2	1.46 (0.18)	-46.2	1	1	1
CaHPO ₄	4	19.94 (2.62)	*****	175.58(13.31)	*****	13.52 (1.70)	*****	1	1	1
Group 3										
Fructose	1	0.74 (0.10)	26.1	0.83 (0.06)	17.2	0.85 (1.00)	15.0	1	2	2
Glucose	1	0.98 (0.11)	1.6	1.14 (0.07)	-13.5	1.32 (0.15)	-32.4	4	3	2

Part B: Samples not normalized for neutron fluence.

Group 1										
Fructose	6	5.86 (0.23)	2.3	8.83 (0.19)	-47.2	6.22 (0.23)	-3.6	2	3	2
Glucose	6	8.09 (0.22)	-34.9	10.06 (0.16)	-67.6	8.82 (0.23)	-46.9	5	5	5
CaCO ₃	3	13.13 (0.57)	*****	98.02 (1.56)	*****	29.73 (0.93)	*****	3	3	3
Water (L2)	1	1.08 (0.05)	-7.9	1.16 (0.04)	-15.6	1.06 (0.05)	-6.1	0	1	0
CaHPO ₄	4	15.95 (1.11)	*****	138.29 (3.56)	*****	9.09 (0.77)	*****	1	1	1
Group 2										
Fructose	6	5.44 (0.21)	9.4	7.64 (0.16)	-27.3	5.86 (0.22)	2.4	3	2	2
Glucose	6	7.80 (0.50)	-25.1	8.70 (0.14)	-45.0	8.31 (0.21)	-38.5	4	5	3
CaCO ₃	3	12.18 (0.53)	*****	84.80 (1.3)	*****	28.02 (0.88)	*****	3	3	3
Water (K2)	1	0.93 (0.05)	7.3	0.87 (0.03)	13.5	0.94 (0.05)	5.8	0	1	0
CaHPO ₄	4	14.79 (1.03)	*****	119.64 (2.99)	*****	8.57 (0.72)	*****	1	1	1
Group 3										
Fructose	1	0.60 (0.03)	39.8	0.79 (0.02)	20.4	0.67 (0.04)	33.4	2	1	2
Glucose	1	0.93 (0.03)	7.1	1.06 (0.02)	-6.0	1.33 (0.04)	-32.6	2	1	3

For explanations see footnotes in Table 5.2.

^dThe standard references used are:

Group 1 = sample of distilled water K2.

Group 2 = water sample L2.

Group 3 = glucose G2 for glucoses; fructose B2 for fructoses.

There were 3 fructoses, 6 glucoses, 3 CaCO₃, 2 water samples, and 1 CaHPO₄ in each group.^hMore than hundred percent.

TABLE 5.5 Average Mole Ratios (gm- mole basis) for Experiment 5a.

Part A: Each sample normalized for the neutron fluence.

MATERIAL	ACTUAL MOLE RATIO	METHOD 1		METHOD 2		METHOD 3		No. of Meas. ≥ 10% error		
		FOUND	% ERROR	FOUND	% ERROR	FOUND	% ERROR	1	2	3
Group 1 ^d										
Fructose	6	7.45 (0.36)	-24.2	9.87 (0.29)	-64.5	6.60 (0.30)	-10.1	3	4	3
Glucose	6	5.60 (0.30)	7.3	8.86 (0.24)	-47.6	6.90 (0.30)	-14.2	2	5	3
CaCO ₃	3	102.14 (6.12)	*****	122.23 (4.31)	*****	18.05 (1.34)	*****	3	3	3
CaHPO ₄	4	30.40 (1.47)	*****	23.86 (0.68)	*****	34.87 (1.81)	*****	3	3	3
MgO	1	89.94 (4.00)	*****	61.01 (1.46)	*****	45.76 (2.56)	*****	3	3	3
Water(L2)	1	0.94 (0.08)	5.8	0.87 (0.05)	12.7	0.87 (0.07)	13.4	1	1	1
Group 2										
Fructose	6	7.91 (0.38)	-31.8	11.31 (0.33)	-88.5	7.63 (0.36)	-27.1	3	4	3
Glucose	6	5.90 (0.29)	1.6	10.15 (0.28)	-69.1	7.91 (0.34)	-31.9	2	5	3
CaCO ₃	3	108.39 (6.37)	*****	140.00 (4.90)	*****	20.84 (1.54)	*****	3	3	3
CaHPO ₄	4	32.26 (1.52)	*****	27.30 (0.80)	*****	40.37 (2.05)	*****	3	3	3
MgO	1	95.45 (4.14)	*****	69.88 (1.68)	*****	52.82 (2.89)	*****	3	3	3
Water(K2)	1	1.06 (0.09)	-6.1	1.15 (0.06)	-14.5	1.15 (0.09)	-15.4	1	1	1
Group 3										
Fructose	1	1.34 (0.09)	-34.1	1.55 (0.06)	-55.0	1.50 (0.10)	-48.3	3	3	3
Glucose	1	0.82 (0.05)	17.7	0.96 (0.03)	3.7	0.64 (0.04)	36.2	2	2	4

Part B: Samples not normalized for neutron fluence.

Group 1										
Fructose	6	10.63 (0.53)	-77.6	13.99 (0.39)	*****	9.67 (0.46)	-61.1	3	4	4
Glucose	6	5.72 (0.27)	4.7	9.10 (0.23)	-51.7	7.43 (0.31)	-23.9	3	5	4
CaCO ₃	3	250.70 (14.30)	*****	148.98 (5.31)	*****	35.40 (2.30)	*****	3	3	3
CaHPO ₄	4	71.10 (3.20)	*****	67.80 (1.80)	*****	103.70 (5.10)	*****	3	3	3
MgO	1	217.40 (8.80)	*****	164.95 (3.55)	*****	127.50 (6.70)	*****	3	3	3
Water(L2)	1	0.98 (0.08)	2.3	0.91 (0.05)	9.1	0.96 (0.07)	3.6	0	0	0
Group 2										
Fructose	6	10.91 (0.53)	-81.8	15.38 (0.44)	*****	10.02 (0.49)	-67.1	3	4	4
Glucose	6	5.86 (0.27)	2.4	10.01 (0.26)	-66.9	7.71 (0.32)	-28.5	3	5	3
CaCO ₃	3	256.60 (14.50)	*****	163.90 (5.90)	*****	36.70 (2.40)	*****	3	3	3
CaHPO ₄	4	72.80 (3.20)	*****	74.53 (1.99)	*****	107.50 (5.20)	*****	3	3	3
MgO	1	222.56 (8.82)	*****	181.54 (3.97)	*****	132.20 (6.80)	*****	3	3	3
Water(K2)	1	1.02 (0.08)	-2.4	1.10 (0.06)	-9.9	1.04 (0.08)	-3.7	0	0	0
Group 3										
Fructose	1	2.20 (0.16)	*****	2.25 (0.17)	*****	2.32 (0.16)	*****	3	3	3
Glucose	1	0.89 (0.06)	10.8	0.93 (0.03)	6.9	0.64 (0.03)	36.5	2	2	4

For explanations see footnotes in Table 5.2.

^dThe standard references are:

Group 1 = water sample K2.

Group 2 = water sample L2.

Group 3 = fructose B2 for fructoses; glucose J2 for glucoses.

Each group has 4 fructoses, 5 glucoses, 3 CaCO₃, 3 CaHPO₄, 3 MgO, and 2 water samples.

TABLE 5.6 Average Mole Ratios (gm- mole basis) for Experiment 6.

Part A: Each sample normalized for neutron fluence.

MATERIAL	ACTUAL MOLE RATIO	M E T H O D 1		M E T H O D 2		M E T H O D 3		No. of Meas. ≥ 10% error		
		FOUND	% ERROR	FOUND	% ERROR	FOUND	% ERROR	1	2	3
Group 1 ^d										
Water	1	0.89 (0.02)	11.1	0.96 (0.02)	3.9	0.91 (0.02)	9.1	1	0	1
Fructose	6	7.67 (0.15)	-27.8	7.44 (0.12)	-23.9	6.66 (0.12)	-10.9	4	4	2
Glucose	6	8.35 (0.17)	-39.2	7.70 (0.12)	-28.4	7.28 (0.13)	-21.1	4	4	4
Group 2										
Water	1	0.99 (0.02)	0.8	1.09 (0.02)	-9.4	0.95 (0.02)	5.4	1	2	1
Fructose	6	8.36 (0.16)	-39.3	8.26 (0.12)	-37.7	6.87 (0.12)	-14.4	4	4	4
Glucose	6	9.10 (0.17)	-51.7	8.56 (0.13)	-42.6	7.49 (0.13)	-24.9	4	4	4
Group 3										
Water	1	0.98 (0.02)	2.2	1.07 (0.02)	-7.1	0.94 (0.02)	5.8	1	1	1
Fructose	6	8.26 (0.16)	-37.7	8.12 (0.12)	-35.3	6.85 (0.12)	-14.1	4	4	2
Glucose	6	8.99 (0.17)	-49.9	8.41 (0.13)	-40.1	7.47 (0.13)	-25.5	4	4	4
Group 4										
Water	1	1.25 (0.02)	-24.8	0.98 (0.02)	1.8	1.28 (0.02)	-28.1	4	0	4
Fructose	6	10.10 (0.20)	-68.1	7.57 (0.12)	-26.2	8.79 (0.15)	-46.4	4	4	4
Glucose	6	10.98 (0.21)	-83.0	7.84 (0.12)	-30.7	9.59 (0.17)	-59.9	4	4	4
Group 5										
Water	1	0.95 (0.02)	5.3	0.91 (0.01)	8.9	0.99 (0.02)	1.5	1	2	1
Fructose	1	1.04 (0.02)	-4.0	1.07 (0.02)	-6.7	1.05 (0.02)	-5.1	0	2	0
Glucose	1	1.03 (0.02)	-2.5	0.89 (0.02)	10.4	1.00 (0.02)	-0.2	1	1	0

Part B: Samples not normalized for the neutron fluence.

Group 1										
Water	1	0.99 (0.01)	1.5	0.97 (0.01)	2.7	0.98 (0.01)	1.9	1	0	1
Fructose	6	7.73 (0.09)	-28.8	7.16 (0.07)	-19.3	6.85 (0.08)	-14.2	4	4	3
Glucose	6	7.99 (0.09)	-33.3	7.16 (0.06)	-19.3	7.22 (0.08)	-20.4	4	4	4
Group 2										
Water	1	0.99 (0.01)	0.6	1.01 (0.01)	-1.2	0.93 (0.01)	6.8	1	0	1
Fructose	6	7.78 (0.09)	-29.7	7.39 (0.07)	-23.2	6.58 (0.07)	-9.7	4	4	3
Glucose	6	8.05 (0.09)	-34.2	7.39 (0.07)	-23.1	6.94 (0.07)	-15.6	4	4	4
Group 3										
Water	1	0.99 (0.01)	0.8	1.02 (0.01)	-2.2	0.96 (0.01)	3.9	1	0	1
Fructose	6	7.77 (0.09)	-24.4	7.45 (0.07)	-24.1	6.75 (0.08)	-12.5	4	4	3
Glucose	6	8.04 (0.09)	-33.9	7.45 (0.07)	-24.1	7.11 (0.08)	-18.5	4	4	4
Group 4										
Water	1	1.17 (0.01)	-17.3	1.02 (0.01)	-2.2	1.20 (0.01)	-20.3	4	0	4
Fructose	6	8.90 (0.12)	-48.4	7.45 (0.07)	-24.1	8.10 (0.10)	-39.4	4	4	4
Glucose	6	9.21 (0.12)	-53.5	7.44 (0.07)	-24.1	8.53 (0.09)	-42.2	4	4	4
Group 5										
Water	1	0.89 (0.01)	11.2	0.97 (0.01)	2.7	0.96 (0.01)	4.1	1	0	1
Fructose	1	1.07 (0.02)	-6.9	1.10 (0.01)	-10.3	1.07 (0.02)	-6.9	1	2	0
Glucose	1	1.08 (0.02)	-7.7	0.92 (0.01)	8.1	1.02 (0.02)	-2.4	1	2	0

^d The groups are formed from the different standard reference used:

Group 1: the reference is the water sample 3.

Group 2: the reference is the water sample 5.

Group 3: the reference is the water sample 15.

Group 4: the reference is the water sample 19.

Group 5: the water sample 1 for water samples; the fructose 22 for fructoses; the glucose 12 for glucoses.

With the requirements of fixed geometry for irradiation and counting, three experiments were performed. These results are presented in Tables 5.4 and 5.5 in the same way as in Tables 5.2 and 5.3.

Observe that the results for experiments labeled 5 and 5a are poor. For this reason it was decided that in addition to gross mistakes, like the mixing of iron wires, and reactivation of some iron wires, there possibly were also errors introduced by fluctuations of the reactor power level and/or instabilities of the counting system.

In experiments 5 and 5a samples were counting using the plexiglass device described in Section 3, "Description of Apparatus," but without the central perforation. So, considerable absorption of gamma-rays might be possible. This was corrected for experiment 6. The results are better than those of experiments 5 and 5a.

Also, in experiment 5 and 5a there were introduced three new materials to analyze: CaCO_3 , CaHPO_4 , and MgO . However, we have not been able to explain the great discrepancy in the molecular ratio calculations (expected values were 3,4,and 1, for CaCO_3 , CaHPO_4 , and MgO , respectively, so it was decided to eliminate them in following experiments.

Error Due to Variations in the Reactor Power Level During Irradiation

In order to obtain the solution, Eq. 2.2, to the well known differential equation Eq. 2.1, which is used to describe the rate of change of the product nuclei, it is usually assumed that the neutron flux, which is a function of the reactor power level, is constant. This assumption is reasonable for most nuclear reactor irradiations, since most reactor irradiations are performed during relatively steady-state operations and for relatively short times. Some fluctuations are observed within a steady-state operation, and because of the relatively very short half-life

of ^{19}O it was of interest to find how much the activation of one sample containing oxygen as a component was affected by variations of a considerable duration during the irradiation time. Also, we wanted to see what the effect would be because of the difference in half-lives of ^{19}O and the fluence monitor ^{56}Mn .

For this purpose a regular square wave behavior was assumed for the power level with an average value considered for the flux at full power in accordance with the characteristics of KSU TRIGA Mark II reactor, (see Table 3.1 for an approximate flux value). A period of 20 seconds and variations in pulse height, as large as 1% and 5%, from the assumed constant level, were used to calculate a theoretical activation of samples. The percentage of error involved when "molecular ratios" (in this case 1:1) are to be calculated was observed. Step functions, positive and negative, at the end and at the beginning of irradiation were also included in the study (see Appendix D).

Then, Eq. 2.1 becomes

$$\frac{dN(t)}{dt} + \lambda N(t) = \sigma N_0 \phi(t) , \quad (5.3)$$

The solution to Eq. 5.3 is

$$N(t) = e^{-\lambda t_0} \sigma N_0 \int_0^{t_0} dt' e^{\lambda t'} \phi(t') \quad (5.4)$$

where t_0 is the irradiation time.

By numerical integration, the integral term of Eq. 5.4 can be approximated for ϕ_1 constant during the time interval $\Delta t = t_i - t_{i-1}$:

$$N(t) = \frac{\sigma N_0}{\lambda} e^{-\lambda t_0} \sum_{i=1}^m \phi_1(t_i) [e^{-\lambda t_i} - e^{-\lambda t_{i-1}}] , \quad (5.5)$$

where m is the number of intervals $\Delta t = t_i - t_{i-1} = 20$ seconds, t_0 is 120 seconds.

Ideal unit masses of "pure" ^{18}O and ^{56}Fe were considered for the analysis. The following Table 5.7 is a listing of the data used in the calculations.

Table 5.7 Data used for Ideal Square Wave Power Level Variations.

ϕ_0	$= 1.8 \times 10^{12} \text{ n/cm}^2\text{-sec at 250 Mw (at the RSR)}$
λ_{ox}	$= 0.02586 \text{ sec}^{-1}$
λ_{Fe}	$= 7.46 \times 10^{-5} \text{ sec}^{-1}$
σ_{ox}	$= 0.2 \times 10^{-27} \text{ cm}^2$
σ_{Fe}	$= 2.7 \times 10^{-24} \text{ cm}^2$

The seven different wave types and their respective functions of the term $\sum_i \phi_i [e^{\lambda_i t} - e^{\lambda_i t - 1}]$ are given in Table 5.8. Within the seven different wave types, two modes were also considered. Mode "B" corresponds to the respective wave type ending at a positive variation after irradiation and mode "A" is for the negative ending.

The theoretical activities at the end of irradiation as well as the normalization of sample with the flux monitor are given in Table 5.9. There were two theoretical samples (a reference and unknown, each with a neutron fluence reference) each exposed to thirteen irradiation patterns, which yields 169 possible unknown/reference combinations. The results are given in Table 5.10.

Results for such "molecular ratios" were for cases where there was believed to be more possibility of error. The maximum percentage error observed was 2.59% which corresponds to the ratio of "samples" 6 to 12 ("wave" ratio type 6A and 6A for 6, and "wave" ratio type 6B and 6B for 12). Probably the last error can be assumed as the maximum to be obtained under

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TABLE 5.8 THEORETICAL NEUTRON FLUXES.

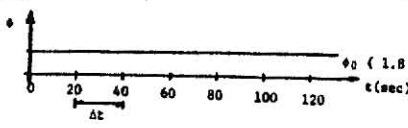
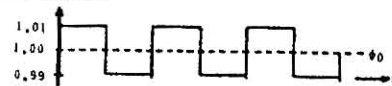
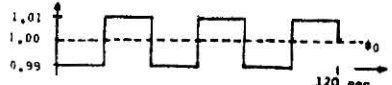
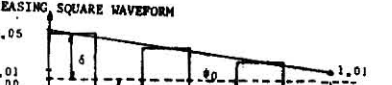
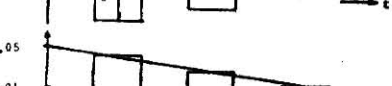
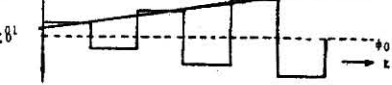
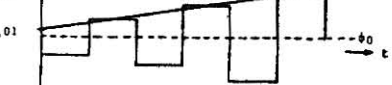
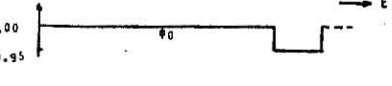
WAVEFORMS		WAVEFORM FUNCTION
1. CONSTANT		
		$\varphi(t) = \phi_0 (e^{\lambda_{120} t} - 1)$
2. SQUARE WAVEFORM		
case A		$\varphi(t) = \phi_0 (e^{\lambda_{120} t} - 1) [1.01 + 0.99e^{-\lambda_{120} 20} + 1.01e^{-\lambda_{120} 40} + 0.99e^{-\lambda_{120} 60} + 1.01e^{-\lambda_{120} 80} + 0.99e^{-\lambda_{120} 100}]$
case B		$\varphi(t) = \phi_0 (e^{\lambda_{120} t} - 1) [0.99 + 1.01e^{-\lambda_{120} 20} + 0.99e^{-\lambda_{120} 40} + 1.01e^{-\lambda_{120} 60} + 0.99e^{-\lambda_{120} 80} + 1.01e^{-\lambda_{120} 100}]$
3. SQUARE WAVEFORM		
case A		$\varphi(t) = \phi_0 (e^{\lambda_{120} t} - 1) [1.05 + 0.95e^{-\lambda_{120} 20} + 1.05e^{-\lambda_{120} 40} + 0.95e^{-\lambda_{120} 60} + 1.05e^{-\lambda_{120} 80} + 0.95e^{-\lambda_{120} 100}]$
case B		$\varphi(t) = \phi_0 (e^{\lambda_{120} t} - 1) [0.95 + 1.05e^{-\lambda_{120} 20} + 0.95e^{-\lambda_{120} 40} + 1.05e^{-\lambda_{120} 60} + 0.95e^{-\lambda_{120} 80} + 1.05e^{-\lambda_{120} 100}]$
4. DECREASING SQUARE WAVEFORM		
case A		$\varphi(t) = \phi_0 (e^{\lambda_{120} t} - 1) [1.047 + 0.953e^{-\lambda_{120} 20} + 1.033e^{-\lambda_{120} 40} + 0.967e^{-\lambda_{120} 60} + 1.02e^{-\lambda_{120} 80} + 0.98e^{-\lambda_{120} 100}]$
case B		$\varphi(t) = \phi_0 (e^{\lambda_{120} t} - 1) [0.960 + 1.040e^{-\lambda_{120} 20} + 0.973e^{-\lambda_{120} 40} + 1.027e^{-\lambda_{120} 60} + 0.987e^{-\lambda_{120} 80} + 1.013e^{-\lambda_{120} 100}]$
5. INCREASING SQUARE WAVEFORM		
case A		$\varphi(t) = \phi_0 (e^{\lambda_{120} t} - 1) [1.013 + 0.987e^{-\lambda_{120} 20} + 1.027e^{-\lambda_{120} 40} + 0.973e^{-\lambda_{120} 60} + 1.040e^{-\lambda_{120} 80} + 0.960e^{-\lambda_{120} 100}]$
case B		$\varphi(t) = \phi_0 (e^{\lambda_{120} t} - 1) [0.98 + 1.02e^{-\lambda_{120} 20} + 0.967e^{-\lambda_{120} 40} + 1.033e^{-\lambda_{120} 60} + 0.953e^{-\lambda_{120} 80} + 1.047e^{-\lambda_{120} 100}]$
6. STEP WAVEFORM		
case A		$\varphi(t) = \phi_0 [(e^{\lambda_{100} t} - 1) + 1.05(e^{\lambda_{120} t} - e^{\lambda_{100} t})]$
case B		$\varphi(t) = \phi_0 [(e^{\lambda_{100} t} - 1) + 0.95(e^{\lambda_{120} t} - e^{\lambda_{100} t})]$
7. STEP WAVEFORM		
case A		$\varphi(t) = \phi_0 [1.05(e^{\lambda_{120} t} - 1) + (e^{\lambda_{120} t} - e^{\lambda_{20} t})]$
case B		$\varphi(t) = \phi_0 [0.95(e^{\lambda_{120} t} - 1) + (e^{\lambda_{120} t} - e^{\lambda_{20} t})]$

TABLE 5.9 RESULTS BY ASSUMING WAVE FUNCTION BEHAVIOR FOR THE NEUTRON FLUX

PART A: THEORETICAL ACTIVITY AT END OF IRRADIATION

MODE	FLUX TYPE 1	TYPE 2	TYPE 3	TYPE 4	TYPE 5	TYPE 6	TYPE 7
OXYG. A	0.444918E 09	0.443792E 09	0.419240E 09	0.443909E 09	0.439173E 09	0.454324E 09	0.445626E 09
B		0.446043E 09	0.450546E 09	0.445927E 09	0.450663E 09	0.435513E 09	0.444209E 09
IRON A	0.625113E 13	0.625108E 13	0.625045E 13	0.627180E 13	0.623014E 13	0.630341E 13	0.630301E 13
B		0.625118E 13	0.625139E 13	0.621046E 13	0.627211E 13	0.619386E 13	0.619925E 13

PART B: ACTIVITY / GM FOR "OXYGEN" SAMPLES NORMALIZED FOR THE NEUTRON FLUENCE

RATIO MODE	SAMPLE NO.	RATIO TYPE	ACTIVITY	SAMPLE NO.	RATIO TYPE	ACTIVITY	SAMPLE NO.	RATIO TYPE	ACTIVITY
A:A	1	1:1	0.711740E-04	2	2:2	0.709666E-04	3	3:3	0.702767E-04
	5	5:5	0.704917E-04	6	6:6	0.720758E-04	7	7:7	0.707005E-04
B:B	8	2:2	0.713536E-04	9	3:3	0.720712E-04	10	6:4	0.715721E-04
	12	6:6	0.702570E-04	13	7:7	0.716553E-04	11	5:5	0.718518E-04
							4	4:4	0.707786E-04

TABLE 5.10 THEORETICAL MOLE RATIO FOR SAMPLE(I) TO SAMPLE (J).
EXPECTED VALUE IS 1:1

I : J	RATIO	% ERROR
1 : 2	1.002527	-0.253
1 : 3	1.012767	-1.277
1 : 4	1.005586	-0.559
1 : 5	1.009679	-0.968
1 : 6	0.987487	1.251
1 : 7	1.006697	-0.670
1 : 8	0.997485	0.252
1 : 9	0.987550	1.245
1 : 10	0.994438	0.556
1 : 11	0.990567	0.943
1 : 12	1.013052	-1.305
1 : 13	0.993282	0.672
2 : 1	0.997479	0.252
2 : 3	1.010214	-1.021
2 : 4	1.003051	-0.305
2 : 5	1.007133	-0.713
2 : 6	0.984998	1.500
2 : 7	1.004158	-0.416
2 : 8	0.994970	0.503
2 : 9	0.985061	1.494
2 : 10	0.991931	0.807
2 : 11	0.988070	1.193
2 : 12	1.010498	-1.050
2 : 13	0.990778	0.922
3 : 1	0.987394	1.261
3 : 2	0.989889	1.011
3 : 4	0.992909	0.709
3 : 5	0.996951	0.305
3 : 6	0.975039	2.496
3 : 7	0.994006	0.599
3 : 8	0.984910	1.509
3 : 9	0.975101	2.490
3 : 10	0.981902	1.810
3 : 11	0.978079	2.192
3 : 12	1.000280	-0.028
3 : 13	0.980761	1.924
4 : 1	0.994445	0.556
4 : 2	0.996958	0.304
4 : 3	1.007141	-0.714
4 : 5	1.004069	-0.407
4 : 6	0.982002	1.800
4 : 7	1.001104	-0.110
4 : 8	0.991943	0.806
4 : 9	0.982064	1.794

TABLE 5.10 (CONTINUED)

4	:	10	0.938914	1.109
4	:	11	0.985064	1.494
4	:	12	1.007424	-0.742
4	:	13	0.987764	1.224
5	:	1	0.990414	0.959
5	:	2	0.992917	0.708
5	:	3	1.003058	-0.306
5	:	4	0.975946	0.405
5	:	6	0.978021	2.198
5	:	7	0.997046	0.295
5	:	8	0.987923	1.208
5	:	9	0.978084	2.192
5	:	10	0.984905	1.509
5	:	11	0.981071	1.893
5	:	12	1.003341	-0.334
5	:	13	0.983761	1.624
6	:	1	1.012671	-1.267
6	:	2	1.015230	-1.523
6	:	3	1.025599	-2.560
6	:	4	1.018328	-1.833
6	:	5	1.022472	-2.247
6	:	7	1.019452	-1.945
6	:	8	1.010123	-1.012
6	:	9	1.000063	-0.006
6	:	10	1.007038	-0.704
6	:	11	1.003118	-0.312
6	:	12	1.025888	-2.589
6	:	13	1.005868	-0.587
7	:	1	0.993348	0.665
7	:	2	0.995858	0.414
7	:	3	1.006030	-0.603
7	:	4	0.998897	0.110
7	:	5	1.002962	-0.296
7	:	6	0.980918	1.908
7	:	8	0.990849	0.915
7	:	9	0.980981	1.902
7	:	10	0.987823	1.218
7	:	11	0.983977	1.502
7	:	12	1.006312	-0.631
7	:	13	0.986675	1.333
8	:	1	1.002522	-0.252
8	:	2	1.005055	-0.506

TABLE 5.10 (CONTINUED)

8	:	3	1.015321	-1.532
8	:	4	1.008121	-0.812
8	:	5	1.012224	-1.222
8	:	6	0.989978	1.002
8	:	7	1.909234	-0.923
8	:	9	0.990041	0.996
8	:	10	0.996946	0.305
8	:	11	0.993065	0.694
8	:	12	1.015606	-1.561
8	:	13	0.995787	0.421
9	:	1	1.012606	-1.261
9	:	2	1.015165	-1.517
9	:	3	1.025535	-2.553
9	:	4	1.018263	-1.826
9	:	5	1.022407	-2.241
9	:	6	0.999936	0.006
9	:	7	1.019387	-1.939
9	:	8	1.010059	-1.006
9	:	10	1.006974	-0.697
9	:	11	1.003054	-0.305
9	:	12	1.025823	-2.582
9	:	13	1.005804	-0.580
10	:	1	1.005592	-0.559
10	:	2	1.008134	-0.813
10	:	3	1.018432	-1.843
10	:	4	1.011209	-1.121
10	:	5	1.015326	-1.533
10	:	6	0.993010	0.699
10	:	7	1.012327	-1.233
10	:	8	1.003063	-0.306
10	:	9	0.993074	0.693
10	:	11	0.996107	0.389
10	:	12	1.018718	-1.872
10	:	13	0.998838	0.116
11	:	1	1.009522	-0.952
11	:	2	1.012074	-1.207
11	:	3	1.022411	-2.241
11	:	4	1.015162	-1.516
11	:	5	1.019294	-1.929
11	:	6	0.996891	0.311
11	:	7	1.016283	-1.628
11	:	8	1.006984	-0.698

TABLE 5.10 (CONTINUED)

11	:	9	0.996955	0.305
11	:	10	1.003908	-0.391
11	:	12	1.022699	-2.270
11	:	13	1.002741	-0.274
12	:	1	0.987116	1.288
12	:	2	0.989611	1.039
12	:	3	0.999719	0.028
12	:	4	0.992630	0.737
12	:	5	0.996670	0.333
12	:	6	0.974765	2.524
12	:	7	0.993726	0.627
12	:	8	0.984633	1.537
12	:	9	0.974827	2.517
12	:	10	0.981626	1.837
12	:	11	0.977804	2.220
12	:	13	0.980485	1.952
13	:	1	1.006763	-0.676
13	:	2	1.009307	-0.931
13	:	3	1.019616	-1.962
13	:	4	1.012386	-1.239
13	:	5	1.016507	-1.651
13	:	6	0.994166	0.583
13	:	7	1.013505	-1.350
13	:	8	1.004230	-0.423
13	:	9	0.994229	0.577
13	:	10	1.001163	-0.116
13	:	11	0.997266	0.273
13	:	12	1.019903	-1.990

irradiation conditions similar to those assumed here. However, it was felt that a reasonably steady-state behavior may be achieved. Therefore, we feel the introduction of large errors for non-stable flux conditions is not very likely.

Errors Due to Instability of the Counting System

Proper functioning of the associated electronic equipment in any experimental research is most important. The electronic capability of multichannel analyzers to assist in the determination of the amount of a radio-nuclide from a mixed gamma-ray spectrum is so important that most modern laboratories include multichannel analyzers as a standard part of their spectroscopy-detecting systems. The greatest concerns for these systems are the instrument deadtime, drift of the instrument between counts, and overall functioning of built-in devices.

In this work the linearity and drift of the Northern Scientific multichannel pulse-height analyzer was checked by using several gamma-ray reference sources, such as ^{60}Co , ^{22}Na , ^{137}Cs and ^{57}Co . The linearity (less than $\pm 1\%$ deviation) was satisfactory throughout all the experiments formed. The drift was negligible. It was possible to reproduce photopeak channel numbers within three channels of the same photopeak energies measured after almost six months had elapsed. This drift of the analyzer was checked using the following procedure. For a desired photopeak, the top three channels points were assumed to be represented by the quadratic function

$$X^2 + BX + CY + D = 0 , \quad (5.6)$$

where

B, C, and D are unknown constants,

X represents the channel number,

Y is the number of counts on each channel.

The centroid of peak was calculated by finding the maximum value for "Y". The centroid X_c is

$$X_c = -\frac{B}{2}. \quad (5.7)$$

It is sufficient to determine the parameter B to find X_c , so B was calculated by simple algebraic substitution of values of the three points mentioned earlier.

Although linearity and drift of the analyzer were always as good as was expected, there were observed extraneous signals, sporadic, which were distributed along all the spectrum being accumulated. The electronic noise was produced by a malfunctioning of the long coaxial cable which connected the Ge(Li) system in the reactor bay and the counting system in the laboratory room. The contribution of errors is easily observed in Tables 5.4, 5.5, and 5.6 for the experiments 5, 5a, and 6, respectively. This noise, however, was not observed during experiment 4; at least it was not as evident as in the other experiments.

A test was performed using the standard reference sources ^{60}Co , ^{137}Cs , and a mixed source, which includes up to 10 visible photopeaks. Several spectra were accumulated and the area ratio for each peak was calculated. The expected value was 1:1. Error percentages as large as 50% were observed for measurements of areas as well for peaks in the high energy regions and for peaks in the low energy regions.

A similar test was performed using a high precision pulse generator. The input signal was connected to the preamplifier of the spectroscopy system, to generate a nuclear-shaped pulse. Spectra were recorded using several pulse-height. Again the area ratios gave percentage errors similar to those obtained using the gamma-ray sources.

Finally we moved all the nuclear instrumentation to the reactor bay and used a short cable connection. With the entire electronic system operating stably, the error percentage was not larger than 3% when either the standard sources or the pulse generator were used. However, another problem appeared. Because variations in ambient temperature normally affect transistorized equipment, erratic behavior in the built-in clock was observed. Long periods of time with the electronic equipment in the reactor bay which is normally maintained at low temperature, at least low enough to effect the electronic equipment, caused this behavior. The typical experimental procedure is to run a relatively large number of samples which required a relatively long time to complete. The effect of temperature was of great concern, and it was decided that some erratic behavior in the electronics had to be accepted. The final method was then to use an external clock and the pulse generator to monitor the behavior of electronic system. When obvious mistakes were performed either due to malfunctioning of equipment or any other errors, the sample being suspected of having such errors was eliminated from further study. The final method is described in more detail in the section 6 "The Approved Method."

6. The Approved Method

After considering the possible source of errors discussed in section 5.9, a final experiment (labeled No. 7) was performed. In this manner, fixed geometry during irradiation and counting was used; the area calculations were performed by the graphical methods avoiding Gregory's equation because of the uncertainty of its results: Special care was taken to avoid or to discover (and eliminate) mistakes performed during manipulation of

the samples, and when obvious errors were observed, the samples were not included in the final results. The results were used to compare with the "good" results (see Tables 6.1 to 6.5). Because this experiment was performed with the electronic equipment in the reactor bay, careful observation was maintained when the spectra were being accumulated. Any undesirable spectrum was easily located and eliminated. To avoid any complication with malfunctioning of the built-in clock, an external clock was used to control the accumulation times. Also, a signal from a high-precision pulse generator (ORTEC Model 450), was used to monitor the accumulation times. The data were normalized for each particular accumulating times of each sample. The normalization was performed first, by calculating the area under the peak produced by the pulser and then, by dividing the area of the desired photopeak by this area. Therefore activity per unit mass per unit time was calculated for each sample and fluence monitor.

6.1 Results of Normalizing Data by the Area Produced from a Pulse Generator

Several samples consisting of six distilled water samples, three fructose samples, three glucose samples, three wheat samples, and three of a mixture (50:50 percent) water-alcohol (ethanol) samples were included in this experiment, labeled No. 7. The main characteristic of experiment No. 7 was the normalization of the photopeak area by the area produced by pulse generator. Results for the distilled water and sugar samples are given in Tables 6.1, 6.2, and 6.3. Results for the alcohol-water mixtures and wheat samples are discussed in sections 6.2 and 6.3, respectively.

In Table 6.1 the average mole ratios (gm-mole basis) for each group of water and sugar samples are given. In this table samples with obvious errors were eliminated, and the samples were compared to each one of the samples of water. Considering the percentage error for each case, water

TABLE 6.1 Average Mole Ratios (gm- mole basis) for Experiment 7. Each sample has been normalized for the neutron fluence and for the counting time. The normalization for the counting time was performed by dividing the area of the desired photopeak by the area of a peak formed from a signal from the Precision Pulse Generator (Ortec Model 419). Samples with obvious mistakes were not included in the record.

MATERIAL	ACTUAL MOLE RATIO	M E T H O D 1		M E T H O D 3		No. of Meas. ≥ 10% error	
		FOUND	% ERROR	FOUND	% ERROR	1	3
Group 1 ^d							
Water ^h	1	0.98 (0.04)	1.7	0.99 (0.03)	0.2	1	0
Fructose	6	6.27 (0.25)	-4.5	6.67 (0.25)	-11.2	0	2
Glucose	6	6.56 (0.33)	-9.4	6.34 (0.30)	-5.6	0	0
Group 2							
Water	1	1.10 (0.03)	-10.1	1.05 (0.03)	-5.4	1	0
Fructose	6	6.87 (0.21)	-14.5	6.97 (0.20)	-16.1	2	2
Glucose	6	7.19 (0.29)	-19.9	6.62 (0.25)	-10.2	2	1
Group 3							
Water	1	1.04 (0.03)	-4.3	0.96 (0.03)	-4.1	1	0
Fructose	6	6.57 (0.21)	-9.5	6.46 (0.19)	-7.7	2	1
Glucose	6	6.88 (0.28)	-14.7	6.13 (0.24)	-2.2	2	0
Group 4							
Water	1	0.87 (0.03)	13.2	0.94 (0.03)	6.3	3	0
Fructose	6	5.68 (0.22)	5.3	6.35 (0.23)	-5.8	1	1
Glucose	6	5.95 (0.29)	0.8	6.02 (0.28)	-0.4	0	0
Group 5							
Water	1	1.03 (0.04)	-3.1	1.06 (0.04)	-6.2	1	1
Fructose	6	6.51 (0.25)	-8.5	7.02 (0.26)	-16.9	2	2
Glucose	6	6.82 (0.34)	-13.6	6.66 (0.31)	-10.9	2	1
Over-all % ERROR			-6.1		-6.4		

For explanations see footnotes in Table 5.2.

^dThe groups are formed from the different standard reference used:
 Group 1 = the reference is the water sample 3.
 Group 2 = the reference is the water sample 27B.
 Group 3 = the reference is the water sample 33.
 Group 4 = the reference is the water sample 39.
 Group 5 = the reference is the water sample 3B.

^hTotal number of samples were: 5 distilled water, 3 fructoses, and 3 glucoses.

samples labeled 3 and 39 are the best references, and the water sample 27 B is the "worst" one. The over-all average percentage error established for this experiment is 6%. However, individual measurements can vary between a range of -20% to 13%. The values observed were frequently larger than the expected value. We were not able to explain that behavior.

In Table 6.2, the samples with obvious errors are included in the analysis. These groups of erroneous samples include one sample of distilled water labeled 27 (a decrease of 1.2% weight; re-activation of iron wire), and one sample of glucose (re-activation of iron monitor) labeled 1. The analysis gives poorer values, compared to Table 6.1, by the inclusion of these samples. The last group, for example, where the water sample labeled 27 is the reference, has percentage error as large as -56.8%. In Table 6.2, the over-all average percentage error estimated is -12%, and the range is from -57% to 17%. Also the results for the glucose samples are poorer compared to those in Table 6.1.

In Tables 6.1 and 6.2 the results with data normalized for counting time by the signal of the pulse generator are given. Only for comparison, results without normalization are shown in Table 6.3. A noticeable difference between the results given in Table 6.3 and Tables 6.1 and 6.2 is observed. The results in Table 6.3 are, in general, quite poor.

6.2 Oxygen Concentrations in Alcohol-Water Mixture

Four samples of a mixture of distilled water and ethanol (C_2H_5OH) were prepared on a volume basis of 50% for water. Typical mass for each sample was 1.2 grams. The expected mole ratio, sample to water, is calculated as follows

$$MR = \frac{2 \times 16}{46.07 + 18.01543} \quad (6.2.1)$$

TABLE 6.2 Average Mole Ratios (gm- mole basis) for Experiment 7. Each sample has been normalized for the neutron fluence and for the counting time. Record includes all of the samples analyzed in Table 6.1 plus the addition of samples with obvious mistakes.

MATERIAL	ACTUAL MOLE RATIO	M E T H O D 1		M E T H O D 3		No. of Meas. ≥ 10% error	
		FOUND	% ERROR	FOUND	% ERROR	1	3
Group 1 ^d							
Water	1	0.93 (0.03)	6.9	0.95 (0.03)	4.9	2	1
Fructose	6	6.27 (0.25)	-4.5	6.67 (0.25)	-11.2	0	2
Glucose	6	6.79 (0.28)	-13.3	6.63 (0.25)	-10.5	1	1
Group 2							
Water	1	1.04 (0.03)	-3.9	1.00 (0.03)	-0.2	2	1
Fructose	6	6.87 (0.21)	-14.5	6.97 (0.20)	-16.1	2	2
Glucose	6	7.45 (0.24)	-24.1	6.93 (0.21)	-15.4	3	2
Group 3							
Water	1	0.91 (0.02)	8.6	0.99 (0.03)	1.5	2	1
Fructose	6	6.46 (0.19)	-7.7	6.57 (0.21)	-9.5	2	1
Glucose	6	6.42 (0.20)	-7.0	7.12 (0.24)	-18.7	3	1
Group 4							
Water	1	0.89 (0.03)	10.6	0.83 (0.03)	17.4	4	1
Fructose	6	6.35 (0.23)	-5.8	5.68 (0.22)	5.3	1	1
Glucose	6	6.31 (0.24)	-5.1	6.16 (0.25)	-2.7	0	1
Group 5							
Water	1	1.01 (0.03)	-1.0	0.98 (0.03)	2.5	2	2
Fructose	6	7.02 (0.26)	-16.9	6.51 (0.25)	-8.5	2	2
Glucose	6	6.97 (0.27)	-16.2	7.06 (0.28)	-17.7	3	2
Group 6							
Water	1	1.37 (0.05)	-36.6	1.31 (0.04)	-31.2	5	5
Fructose	6	8.68 (0.36)	-44.6	8.76 (0.33)	-46.1	3	3
Glucose	6	9.41 (0.40)	-56.8	8.71 (0.34)	-45.1	3	3
Over-all % ERROR			-12.3			-11.7	

TABLE 6.3 Average Mole Ratios (gm- mole basis) for Experiment 7. Normalization for counting time was not performed. Record includes samples with obvious mistakes.

MATERIAL	ACTUAL MOLE RATIO	M E T H O D 1			M E T H O D 3			No. of Meas. > 10% error		
		FOUND	% ERROR		FOUND	% ERROR		1	3	
Group 1 ^d										
Water	1	1.18 (0.04)	-17.5		1.195 (0.033)	-19.5		4	3	
Fructose	6	6.78 (0.23)	-13.1		7.12 (0.22)	-18.4		2	2	
Glucose	6	7.62 (0.27)	-27.0		7.32 (0.24)	-22.0		3	2	
Group 2										
Water	1	1.03 (0.03)	-3.4		0.99 (0.02)	0.6		2	2	
Fructose	6	6.09 (0.14)	-1.5		6.09 (0.13)	-1.6		0	2	
Glucose	6	6.84 (0.17)	-14.0		6.27 (0.14)	-4.5		2	2	
Group 3										
Water	1	1.13 (0.03)	-13.1		1.03 (0.02)	-2.5		2	2	
Fructose	6	6.57 (0.15)	-9.5		6.25 (0.13)	-4.2		2	1	
Glucose	6	7.38 (0.18)	-23.0		6.43 (0.15)	-7.2		2	2	
Group 4										
Water	1	0.99 (0.03)	0.7		1.08 (0.03)	-8.2		3	1	
Fructose	6	5.89 (0.19)	1.9		6.54 (0.19)	-9.0		1	1	
Glucose	6	6.61 (0.22)	-10.2		6.73 (0.21)	-12.1		2	2	
Group 5										
Water	1	1.01 (0.03)	-1.2		1.11 (0.03)	-10.6		2	1	
Fructose	6	5.98 (0.19)	0.4		6.66 (0.20)	-11.0		0	2	
Glucose	6	6.72 (0.23)	-11.9		6.85 (0.22)	-14.2		2	2	
Group 6										
Water	1	0.76 (0.02)	24.5		0.72 (0.02)	27.7		5	5	
Fructose	6	4.71 (0.17)	21.4		4.71 (0.15)	21.5		3	3	
Glucose	6	5.295 (0.200)	11.8		4.84 (0.16)	19.3		1	3	

For explanations see footnotes in Table 5.2.

^dThe standard references are the same given in Table 6.2.

In this group of samples a noticeable decrease (9.3%) in mass was observed for one of the samples. Consequently it was eliminated from the analysis. For the other samples decreases in mass were apparent (about 1%). Therefore the average value for the mass were used in the analysis.

The analysis was the same as explained before (Section 6.1). In Table 6.4 these results for normalized data are given. One sample was eliminated, because its spectrum showed a "step" increase in the counts in the region of high energy, the normal position of the peak for pulser generator. In Table 6.5 the results of all three samples are given again without normalization. The percentage errors are, in general, large. This error percentage was calculated on the basis of a theoretical ratio 1:1, however these results show an apparent systematic error. Therefore, we assumed that the 1:1 ratio was probably in error. Therefore, we decided to calculate the results on the basis of 1.5:1. These results are given in Table 6.6. Notice the decrease in the percentage error in Table 6.6.

6.3 Concentration of Oxygen in Wheat

Three groups of 3 samples of standard wheat were irradiated in this work. We included wheat in our analysis of ^{18}O determination in order to include some samples with unknown concentration of oxygen to test the capabilities of the analysis method described in this work. For these samples the weight fraction of oxygen was calculated with water as the standard reference. Results are given in Table 6.7.

In Table 6.7 the average weight fraction of oxygen for each sample of the group is given. In each group, the fraction of oxygen of each sample was calculated by averaging all the individual measurements obtained by analysis with different water sample references. In the first two groups, two references were used; in the last group, six references were

TABLE 6.4 Average Mole Ratios (gm-mole basis) for three samples of ethanol-water mixture (50-50 volume). Each sample was normalized for the neutron fluence and for the counting time.

Part A: Record includes all three samples. The references are given in Table 6.2.

ACTUAL MOLE RATIO	M E T H O D 1		M E T H O D 3		No. of Meas. ≥ 10% error 1 3
	FOUND	% ERROR	FOUND	% ERROR	
1	1.13 (0.05)	-12.7	1.18 (0.05)	-17.9	3
1	1.24 (0.05)	-23.5	1.23 (0.05)	-23.1	3
1	1.18 (0.05)	-18.1	1.14 (0.04)	-14.2	3
1	1.02 (0.05)	-2.2	1.12 (0.05)	-12.1	3
1	1.17 (0.05)	-17.0	1.24 (0.06)	-23.9	3
1	1.56 (0.08)	-56.0	1.55 (0.07)	-54.9	3

Part B: Record for only two samples. One sample was eliminated because an erroneous spectrum in the region of the Pulse Peak was obtained. References are also reduced to the "better" water samples. See Table 6.1.

1	1.47 (0.08)	-47.3	1.56 (0.08)	-55.5	2
1	1.61 (0.07)	-61.4	1.62 (0.07)	-62.4	2
1	1.55 (0.07)	-54.5	1.51 (0.06)	-50.6	2
1	1.34 (0.07)	-33.6	1.48 (0.07)	-47.9	2
1	1.53 (0.07)	-53.1	1.64 (0.08)	-63.5	2

For explanations see footnotes in Table 5.2.

TABLE 6.5 Average Mole Ratios (gm-mole basis) for the three samples of ethanol-water mixture given in Table 6.4. No normalization for the counting time is included.

ACTUAL MOLE RATIO	M E T H O D 1		M E T H O D 3		No. of Meas. ≥ 10% error	
	FOUND	% ERROR	FOUND	% ERROR	1	3
1	1.63 (0.07)	-62.6	1.64 (0.06)	-63.7	3	3
1	1.46 (0.05)	-45.9	1.40 (0.04)	-40.1	3	3
1	1.57 (0.05)	-57.4	1.44 (0.04)	-43.7	3	3
1	1.41 (0.06)	-41.0	1.50 (0.06)	-50.4	3	3
1	1.43 (0.06)	-43.3	1.53 (0.06)	-53.2	3	3
1	1.13 (0.05)	-12.9	1.08 (0.04)	-8.3	3	3

For explanations see footnotes in Table 5.2.

TABLE 6.6 Average Mole Ratios (gm- mole basis) for the three samples of ethanol-water mixture given in Table 6.4. No normalizations for the counting time were performed. The actual mole ratio is assumed to be 1.5 : 1.

Part A: All three samples. The references are given in Table 6.2.						
ACTUAL MOLE RATIO	M E T H O D 1		M E T H O D 3		No. of Meas. ≥ 10% error	
	MOLE RATIO	% ERROR	MOLE RATIO	% ERROR	1	3
1.5	1.63 (0.07)	-8.4	1.64 (0.06)	-9.1	1	2
1.5	1.46 (0.05)	2.7	1.40 (0.04)	6.6	0	1
1.5	1.57 (0.05)	-4.9	1.44 (0.04)	4.2	0	0
1.5	1.41 (0.06)	6.0	1.50 (0.06)	0.3	0	0
1.5	1.43 (0.06)	4.5	1.53 (0.06)	-2.1	0	0
1.5	1.13 (0.05)	24.7	1.08 (0.04)	27.8	3	3

Part B: Record for two samples. The references are given in Table 6.1. One sample was not recorded due to the wrong spectrum accumulated.						
1.5	1.47 (0.08)	1.8	1.56 (0.08)	-3.7	0	0
1.5	1.61 (0.07)	-7.6	1.62 (0.07)	-8.3	1	0
1.5	1.55 (0.07)	-3.0	1.51 (0.06)	-0.4	0	0
1.5	1.34 (0.07)	10.9	1.48 (0.07)	1.4	1	0
1.5	1.53 (0.07)	-2.1	1.64 (0.08)	-9.0	0	1

For explanations see footnotes in Table 5.2.

TABLE 6.7 Oxygen weight fraction in Standard Wheat. Record is for nine samples, analyzed by triplicate in three experimental sets.

EXPERIMENT	CALCULATIONS BY		CALCULATIONS BY	
	METHOD	1	METHOD	3
5 ⁱ	0.636	(0.049)	0.673	(0.052)
5a ^j	0.487	(0.018)	0.522	(0.016)
7 ^k	0.525	(0.010)	0.487	(0.008)
AVERAGE	0.549	(0.018)	0.561	(0.018)

For explanations see footnotes in Table 5.2.

ⁱEach sample was measured by duplicate using values calculated for different standard references. References are given in Table 5.4.

^jEach measurement by duplicate using references given in Table 5.5.

^kEach sample was measured six times using the same references given in Table 6.4. Data are also normalized for the counting time.

used. Also the normalization by the pulser was performed in this last group. Similar results were obtained by using both graphical methods, the fixed and the variable minima methods.

If it is considered that typical wheat grain is approximately composed of 69.6% starch ($C_6H_{12}O_6$), 12.0% protein (C_5H_9OH), 2.1% fat ($(C_{17}H_{35}O_2)_3C_3H_5$), 2.5% cellulose ($C_6H_{12}O_6$), and 12% water (H_2O), where chemical formula are average values, then the results given in Table 6.7 are in very good agreement with these wheat compositions (23). However, we must point out that the analysis of wheat samples were only preliminary investigations.

7. Conclusions and Recommendations

The thermal neutron activation analysis can definitely be used to determine oxygen-18 concentrations in sugars, wheat, oxides, and water-alcohol mixtures. Although, other methods might be more accurate than the NAA method (about 6% accuracy for NAA compared to about 1-2% for mass spectroscopy), the main advantage of this method is in its simplicity and the non-destruction of the samples. We believe that improvements, such as an pneumatic transfer system and a more stable analyzer system, could improve the accuracy of the NAA method. The transfer method could be improved, by moving the counting system to the top of the reactor pool. However, this may not be practical due to the relatively large gamma-ray interference.

Future work delineating the sensitivity of ^{18}O concentration is also suggested. Even though the study was not directed to determine sensitivity, we believe that it is a few milligrams. However, a similar work reported sensitivities of few hundreds of micrograms (29).

8. Acknowledgements

The author would like to express his sincere gratitude to the professors of the Department of Nuclear Engineering at Kansas State University whose efforts to teach him surpassed his ability to reciprocate such endeavors. In particular, appreciation is extended to Dr. N. D. Eckhoff for his outstanding supervision and support throughout this work.

The author also wishes to acknowledge his financial support from the Department of Nuclear Engineering at Kansas State University, and the Consejo Nacional de Ciencia y Tecnologia in Mexico City, Mexico. Appreciation to the National Academy of Sciences and the International Atomic Energy Agency for their sponsorship is also extended. The assistance of Dr. J. K. Shultis, Mr. M. McEwan, Mr. S. Howe, and Mr. B. Lemmons is appreciated.

Finally, special thanks is given to God whose infinite love gave me Ursula, my wife.

Hector Ocampo Mansilla

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9. APPENDICES

APPENDIX A

Formulation of Mole Ratio

Consider an activated sample in which the radioactive isotope of interest is being produced at a rate R . Now we must relate R to a quantity that can be measured. The quantity we actually measure is the rate of desintegration of the radioactive nuclei at the time of measurement (usually some time after removal of sample from the neutron flux).

The buildup of radioactive nuclei during irradiation is described by (17,19)

$$\dot{N}(t) = R - \lambda N(t) , \quad (A.1)$$

where

N is the number of radioactive nuclei,

λ is the decay constant of the product,

R is the rate of production,

For the case of constant neutron flux throughout the sample, R is expressed by

$$R \text{ (reactions per sec)} = N_T \int_0^{\infty} \sigma_a(E) \phi(E) dE , \quad (A.2)$$

where

N_T is the total number of target atoms,

$\sigma_a(E)$ is the cross section for activation at the energy E ,

(σ is the cross section for the neutron process which causes the activations),

ϕ is the neutron flux.

In general, R may be a function of time. However, a very useful case is a constant, R , i.e., "burnup" of parent and daughter may be neglected. This also assumes that the neutron flux remains constant. Then, if we put a sample into the flux at time $t = 0$, the number of radioactive nuclei in the sample at time t is, i.e., the solution of Eq. A.1,

$$N(t) = \frac{R}{\lambda} (1 - e^{-\lambda t}) , \quad (A.3)$$

and the sample activity at time t is

$$A(t) \equiv \lambda N(t) = R(1 - e^{-\lambda t}) = A_0(1 - e^{-\lambda t}) . \quad (A.4)$$

The saturation activity A_0 is reached if the sample stays in the neutron flux for a very long time compared to the half-life where the activity of sample becomes equal to the production rate, R .

If the activity at the end of the irradiation is A_0 and a count is made between times t_1 and t_2 measured from the end of the exposure to the neutron flux, and using a detector with an over-all efficiency ϵ , the total observed number of counts, C , will be

$$C = \epsilon \int_{t_1}^{t_2} A_0 e^{-\lambda t'} dt' + B , \quad (A.5)$$

where B is the background count occurring in the interval t_1 and t_2 .

After integrating and rearranging Eq. A.5, we obtain

$$C = \epsilon \frac{A_0}{\lambda} (e^{-\lambda t_1} - e^{-\lambda t_2}) + B . \quad (A.6)$$

Combining Eqs. (A.4) and (A.6), we obtain for A_0 , the saturated activity,

$$A_0 = \lambda(C-B) [\epsilon e^{-\lambda t_1} (1 - e^{-\lambda t_0}) (1 - e^{-\lambda t_2})]^{-1} , \quad (A.7)$$

where t_0 is the irradiation time.

Since the samples we use in any experiment will generally have different masses, we must correct for the different masses as well as for the fraction of the isotope of interest. Thus, the saturation activity per unit mass A_0^* is given by

$$A_o^* \equiv \frac{A_o}{m} = \lambda(C-B) [mf \epsilon e^{-\lambda t_1} (1-e^{-\lambda t_o}) (1-e^{-\lambda t_2})]^{-1}, \quad (A.8)$$

where m is the sample mass, and f is the fraction of isotope of interest.

The confidence limits, or variance, of A_o^* is easily calculated knowing the variances on the sample count and the background count, as well as the variance in the other parameters of Eq. (A.8).

At thermal energies Eq. (A.2) can be expressed as

$$R = N_T \sigma_{ath} \phi_{th}, \quad (A.9)$$

where σ_{ath} is the thermal neutron activation cross section, and

ϕ_{th} is the average thermal neutron flux.

The comparator method of NAA is based on simultaneous analysis of a standard known sample for the desired elements in which the known and unknown samples are treated identically. Under such conditions the activities, for unknown and reference samples are proportional. Combining Eq. (A.9), for irradiation under thermal neutrons, and Eq. (A.4) we obtain

$$\frac{A_{ou}}{A_{ok}} = \frac{N_{Tu} \sigma_{au} \phi_u (1-e^{-\lambda t_{ou}})}{N_{Tk} \sigma_{ak} \phi_k (1-e^{-\lambda t_{ok}})}, \quad (A.10)$$

where the subscripts u and k denote unknown and known samples, respectively.

In Eq. (A.10) the subscript "th" for σ_a and ϕ were dropped for simplicity. A_o is calculated from Eq. (A.8). In Eq. (A.10) σ_{au} and σ_{ak} are equal for the same isotope of interest. The neutron flux ϕ is usually determined by an appropriate flux monitor. Combining Eqs. (A.4) and (A.8) and (A.9), we obtain the flux ϕ as

$$\phi = \frac{A_{oF} (1-e^{-\lambda_F t_{oF}})^{-1}}{N_{TF} \sigma_{aF}}, \quad (A.11)$$

where the subscript F is for the fluence monitor.

Let MR be the mole ratio of the fraction of isotope of interest in the unknown and the reference (known) sample, and defined as follows (gram-mole basis)

$$MR = \frac{f_u}{f_R}, \quad (A.12)$$

where f , the molecular fraction of isotope of interest is

$$f = \frac{\text{molecular weight of isotope}}{\text{total molecular weight in compound}}. \quad (A.13)$$

Combining Eqs. (A.8), (A.10), (A.11), and (A.12), we obtain MR as

$$MR = \frac{\lambda(C-B)_u [M_u \epsilon e^{-\lambda t_{1u}} (1 - e^{-\lambda t_{ou}}) (1 - e^{-\lambda t_{2u}})]^{-1} MW_u}{\lambda_F(C-B)_{Fu} [M_{Fu} \epsilon_F e^{-\lambda_F t_{1Fu}} (1 - e^{-\lambda_F t_{oFu}}) (1 - e^{-\lambda_F t_{2Fu}})]^{-1} MW_u} \cdot \frac{\lambda(C-B)_R [M_R \epsilon e^{-\lambda t_{1R}} (1 - e^{-\lambda t_{oR}}) (1 - e^{-\lambda t_{2R}})]^{-1} MW_R}{\lambda_F(C-B)_{FR} [M_{FR} \epsilon_F e^{-\lambda_F t_{1FR}} (1 - e^{-\lambda_F t_{oFR}}) (1 - e^{-\lambda_F t_{2FR}})]^{-1} MW_R}, \quad (A.14)$$

where

MW = molecular weight of sample,

F = subscript for fluence monitor,

u = subscript for unknown sample,

R = subscript for reference sample.

For equal irradiation and counting times, decay constants for fluence monitors and samples, and over-all efficiencies, Eq. (A.14) becomes

$$MR = \frac{(C-B)_u [M_u e^{-\lambda t_{1u}}]^{-1} MW_u}{(C-B)_{Fu} [M_{Fu} e^{-\lambda_F t_{1Fu}}]^{-1} MW_u} \cdot \frac{(C-B)_R [M_R e^{-\lambda t_{1R}}]^{-1} MW_R}{(C-B)_{FR} [M_{FR} e^{-\lambda_F t_{1FR}}]^{-1} MW_R}. \quad (A.15)$$

The variance of MR is calculated by the well-known equation (19,25):

$$\sigma^2[f(x_1, x_2, \dots, x_n)] = \left(\frac{\partial f}{\partial x_1}\right)^2 \sigma_{x_1}^2 + \left(\frac{\partial f}{\partial x_2}\right)^2 \sigma_{x_2}^2 + \dots + \left(\frac{\partial f}{\partial x_n}\right)^2 \sigma_{x_n}^2, \quad (\text{A.16})$$

where f is a function of n variables, each one with a variance σ^2 .

In general all the parameters, but (C-B), are considered to be well determined, then the variance of MR will be a function of the variance for each (C-B) term. Then, Eq. (A.15) can be represented by

$$\text{MR} = f(x, y, z, t) = k \frac{\frac{x}{y}}{\frac{z}{t}}, \quad (\text{A.17})$$

or

$$\text{MR} = k \frac{xz}{yt}, \quad (\text{A.18})$$

where k is a constant.

Then the variance of MR is, from Eq. (A.16)

$$\sigma^2(\text{MR}) = \left(\frac{kz}{yt}\right)^2 \sigma_x^2 + \left(\frac{kx}{yt}\right)^2 \sigma_z^2 + \left(-\frac{kxz}{y^2t}\right)^2 \sigma_y^2 + \left(-\frac{kxz}{yt^2}\right)^2 \sigma_t^2, \quad (\text{A.19})$$

or

$$\sigma^2(\text{MR}) = \left(\frac{kxz}{yt}\right)^2 \left[\frac{\sigma_x^2}{x^2} + \frac{\sigma_z^2}{z^2} + \frac{\sigma_y^2}{y^2} + \frac{\sigma_t^2}{t^2} \right], \quad (\text{A.20})$$

The standard deviation is then

$$\sigma(\text{MR}) = \text{MR} \left[\left(\frac{\sigma_P}{P_u}\right)^2 + \left(\frac{\sigma_P}{P_r}\right)^2 + \left(\frac{\sigma_P}{P_{Fu}}\right)^2 + \left(\frac{\sigma_P}{P_{FR}}\right)^2 \right]^{1/2}, \quad (\text{A.21})$$

where $P \equiv (\text{C-B}) =$ net peak area under the photopeak of interest for samples and fluence monitors.

In this work, we were interested in the isotope ^{18}O , in samples of sugar (glucoses and fructoses), water, wheat, and other oxides. Therefore, MR is known in most of the samples (except for wheat) and the desired value is in each case

a) For H_2O , $\text{MW} = 18.0153$,

$$f(\text{fraction of oxygen}) = \frac{16}{18.0153},$$

b) For sugar (glucose and fructose have same chemical formula $\text{C}_6\text{H}_{12}\text{O}_6$),

$$\text{MW} = 180.16$$

$$f(\text{fraction of oxygen}) = \frac{6 \times 16}{180.16};$$

MR for sugar to water the desired ratio is

$$\text{MR} = \frac{6 \times 16}{16} = \frac{6}{1},$$

c) For cases where sugars were used as a reference and as the unknown sample or where the water samples were the reference and unknown samples,

$$\text{MR} = \frac{1}{1}.$$

In the case of wheat samples, where the molecular composition is not well known, Eq. (A.15) is modified to find the fraction of oxygen:

$$f_{\text{oxygen}} = \frac{\frac{(C-B)_u [M_u e^{-\lambda t_{1u}}]^{-1}}{(C-B)_{Fu} [M_{Fu} e^{-\lambda_F t_{1Fu}}]^{-1}}}{\frac{(C-B)_R [M_R e^{-\lambda t_{1R}}]^{-1}}{(C-B)_{FR} [M_{FR} e^{-\lambda_F t_{1FR}}]^{-1}}} \cdot \frac{16}{18.01534}, \quad (\text{A.22})$$

where the reference sample was always H_2O . The standard deviation for Eq. (A.22) is

$$\sigma(f_{\text{oxygen}}) = f_{\text{oxygen}} \left[\left(\frac{\sigma_{P_U}}{P_U} \right)^2 + \left(\frac{\sigma_{P_R}}{P_R} \right)^2 + \left(\frac{\sigma_{P_{Fu}}}{P_{Fu}} \right)^2 + \left(\frac{\sigma_{P_{FR}}}{P_{FR}} \right)^2 \right]^{1/2}. \quad (\text{A.23})$$

APPENDIX B

Area Calculation Methods

B.1 Gregory's Equation Method

The Gregory's equation is a numerical approximation to the integration of a function, which graphically represents the area under the curve. This method is based on finite-difference techniques, which will be briefly introduced here. All operations and derivations are illustrative rather than rigorous. For a rigorous development we suggest readers see reference 13, or any other Advanced Mathematics text.

Suppose that we have a function $y = f(x)$ and if $f(x_i)$ and $f(x_j)$ are any two of its values, then the first divided difference of $f(x)$ is defined by the formula

$$f(x_i, x_j) \equiv \frac{f(x_i) - f(x_j)}{x_i - x_j} . \quad (B.1)$$

Similarly, if $f(x_i, x_j)$ and $f(x_j, x_k)$ are the two first divided differences of $f(x)$ having one argument, x_j , in common, then the second divided difference of $f(x)$ is defined by the formula

$$f(x_i, x_j, x_k) \equiv \frac{f(x_i, x_j) - f(x_j, x_k)}{x_i - x_k} . \quad (B.2)$$

Proceeding inductively, we can define a divided difference of any order. Usually the values of x in a set of data will be equally spaced, and the differences of the function will be based on sets of consecutive functional values. When this is the case, the denominators in the divided differences of any given order are all equal, and it is customary to omit them. This leads to a modified set of quantities known simply as the "differences" of the function. If the constant difference between successive values of x is h , so that the general value of x in the set is

$$x_k = x_0 + kh \quad k = \dots, -2, -1, 0, 1, 2, \dots \quad (\text{B.3})$$

and the corresponding functional value is

$$y_k = f(x_k) = f(x_0 + kh) = f_k \quad (\text{B.4})$$

then, Eqs. (B.1) and (B.2) can be defined by the formula

$$\Delta f_k = f_{k+1} - f_k \quad (\text{B.5a})$$

and

$$\Delta^2 f_k = \Delta(\Delta f_k) = \Delta f_{k+1} - \Delta f_k. \quad (\text{B.5b})$$

In general, for positive integral values of n ,

$$\Delta^n f_k = \Delta(\Delta^{n-1} f_k) = \Delta^{n-1} f_{k+1} - \Delta^{n-1} f_k \quad (\text{B.6})$$

The following table shows several orders of the difference operator Δ .

Table B.1. Difference table.

x	f	Δ	Δ^2	Δ^3	Δ^4	Δ^5
x_0	f_0	Δf_0	$\Delta^2 f_0$	$\Delta^3 f_0$	$\Delta^4 f_0$	$\Delta^5 f_0$
x_1	f_1	Δf_1	$\Delta^2 f_1$	$\Delta^3 f_1$	$\Delta^4 f_1$	
x_2	f_2	Δf_2	$\Delta^2 f_2$	$\Delta^3 f_2$	$\Delta^4 f_2$	
x_3	f_3	Δf_3	$\Delta^2 f_3$	$\Delta^3 f_3$		
x_4	f_4	Δf_4				
x_5	f_5					

Several properties can be associated with the difference operator Δ , as well as another notations, and associated functions. We are interested only in possible applications like interpolation, in which the method can be used to find the derivative of a tabular function. Among several formulas

for interpolation, for example, the Lagrange's formula, the forward Gregory-Newton formula, the forward Newton-Gauss formula, and the Laplace-Everett formula. The forward and backward Gregory-Newton formulae, for instance, can be used to give an operational derivation of the famous Euler-Maclaurin summation formula

$$\sum_{i=0}^n f_i = \frac{1}{h} \int_{x_0}^{x_n} f(x) dx + \frac{1}{2} (f_0 + f_n) + \sum_{i=1}^{\infty} \frac{B_{2i}}{(2i)!} h^{2i-1} (f_n^{(2i-1)} - f_0^{(2i-1)}) . \quad (\text{B.7})$$

where the B's are the Bernoulli numbers. The first few nonzero values are

$$\begin{aligned} B_0 &= 1 & B_1 &= -\frac{1}{2} & B_2 &= \frac{1}{6} & B_4 &= -\frac{1}{30} \\ B_6 &= \frac{1}{42} & B_8 &= -\frac{1}{30} & B_{10} &= \frac{5}{66} & B_{12} &= -\frac{691}{2730} \end{aligned}$$

As these values suggest, B_{2n} becomes infinite as $n \rightarrow \infty$.

If Eq. (B.7) is solved for the integral, we obtain

$$\int_{x_0}^{x_n} f(x) dx = h \sum_{i=0}^n f_i - \frac{h}{2} (f_0 + f_n) - \sum_{j=1}^{\infty} \frac{B_{2j}}{(2j)!} h^{2j} (f_n^{(2j-1)} - f_0^{(2j-1)}) , \quad (\text{B.8})$$

which is a fundamental formula of numerical integration. For functions defined only by a table of values it is usually convenient to have an integration formula, like Eq. (B.8), in which the "correction terms" are expressed as differences rather than as derivatives. To obtain such a formula from Eq. (B.8), we need only replace the derivatives $f'_0, f'''_0, \dots$ by means of appropriate interpolation formulas, as well as the derivatives f'_n, f'''_n , by means of

$$f'(x_0) = \frac{1}{h} (\Delta f_{-1} + \frac{1}{2} \Delta^2 f_{-2} + \frac{1}{3} \Delta^3 f_{-3} + \frac{1}{4} \Delta^4 f_{-4} + \dots) , \quad (\text{B.9a})$$

$$f'''(x_0) = \frac{1}{h^3} (\Delta^3 f_{-3} + \frac{3}{2} \Delta^4 f_{-4} + \dots) . \quad (\text{B.9b})$$

(For a complete development see reference 13, pages 116-130). This gives

$$\begin{aligned}
\int_{x_0}^{x_n} f(x)dx &= h\left(\frac{f_0}{2} + f_1 + \dots + f_{n-1} + \frac{f_n}{2}\right) - \frac{h^2}{12} \left[\frac{1}{h} (\Delta f_{n-1} + \frac{\Delta^2 f_{n-2}}{2} + \frac{\Delta^3 f_{n-3}}{3} + \dots) \right. \\
&\quad \left. - \frac{1}{h} (\Delta f_0 - \frac{\Delta^2 f_0}{2} + \frac{\Delta^3 f_0}{3} + \dots) + \frac{4h}{720} \left[\frac{1}{h^3} (\Delta^3 f_{n-3} + \frac{3}{2} \Delta^4 f_{n-4} + \dots) \right. \right. \\
&\quad \left. \left. - \frac{1}{h^3} (\Delta^3 f_0 - \frac{3}{2} \Delta^4 f_0 + \dots) \right] + \dots \right]
\end{aligned}$$

or

$$\begin{aligned}
\int_{x_0}^{x_n} f(x)dx &= h \left(\frac{1}{2} f_0 + f_1 + \dots + f_{n-1} + \frac{1}{2} f_n \right) - \frac{h}{2} (\Delta f_{n-1} - \Delta f_0) \\
&\quad - \frac{h}{24} (\Delta^2 f_{n-2} + \Delta^2 f_0) - \frac{19h}{720} (\Delta^3 f_{n-3} - \Delta^3 f_0) - \frac{3h}{160} (\Delta^4 f_{n-4} + \Delta^4 f_0) - \dots
\end{aligned}$$

(B.10)

Equation (B.10) is known as Gregory's Formula of numerical integration. When the correction terms are neglected Eqs. (B.8) and (B.10) reduce to the well-known trapezoidal rule of integration.

We used the Gregory's Formula, Eq. (B.10), as a method to evaluate the area of a photopeak (using $h=1$, equal spaced channels). The validity of this method may be justified by the following:

a) For the NAA comparator technique samples are treated identically, where masses, irradiation, decay and cooling times are similar; therefore, the expected peaks will be alike or proportional. For the absolute technique, the method may not be useful.

b) For well-resolved photopeaks, in which the background is reasonably flat, the area N , between the top six channels, as we choose, defines a fixed fraction of the total peak area S which includes the background (see Fig. B.1.1).

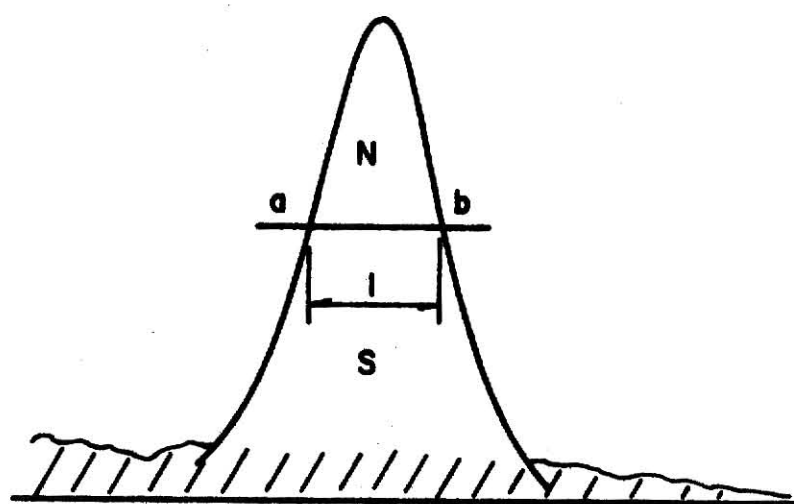


Fig. B.1.1. An ideal well-resolved peak.

This means that the ratio of the area N above a horizontally intersecting line of constant length " l " is proportional to the total area S and therefore varies directly with the gamma-ray intensity.

c) For any modern spectroscopy system of high resolution, as a Ge(Li) detector, the top section of a peak in a monoenergetic gamma-ray spectrum would be quite symmetrically distributed and can be similarly reproduced for similar peak-energy. Only a few channels are required to define any photopeak and the high ratio of the peak-area to total area that often distinguishes such a gamma-ray spectrum.

B.2 Graphical Methods

Another method of analysis of gamma-ray spectra is the graphical method. This method is the most popular for its simplicity which allows us to calculate areas quickly by hand calculators or by computers without requiring complicated programs. In this method the region of interest, or photopeak, is represented graphically as rectangles whose areas are proportional to the number of counts in the channels (see Fig. B.2.1). Summation of the rectangles from b_n through a_o to a_n gives the value that represents the total counts contained under the peak, shown graphically as the area T in Fig. B.2.1.

T is formed by the net area N , the area of interest, plus the background B ; therefore, the choice of a_n and b_n , the minima at both sides, are important and ultimately defines the adequate selection of background for finding the "true" area that defines the gamma-ray intensity associated with such a peak. We used the two methods which are described below.

Fixed-Minima Method

In the first method, we defined a fixed minima at both sides. The net area N is calculated as follows: For a maximum number of counts at channel n ,

the minima are C_l and C_r where $C_l = n-3$ and $C_r = n+3$. Seven fixed points define the total area T.

Thus, the net area N is

$$N = T - B, \quad (B.11)$$

where

$$T = \sum_{i=n-3}^{n+3} C_i, \quad (B.12)$$

C_i represents the counts contained into channel i.

For B, the background, the two adjacent channels at both minima are included in order to calculate an "average minima" value. Then a straight-line is used to connect both minima. Below that line is assumed to be the background, B. Therefore, B is calculated as (area under the trapezoid as can be seen in Fig. B.2.2).

$$B = \frac{7}{2} \left[\frac{1}{3} \sum_{j=n-5}^{n-3} C_j + \frac{1}{3} \sum_{k=n+3}^{n+5} C_k \right], \quad (B.13)$$

Variable-Minima Method

In the other graphical method used, the minima at both sides of the peak are found according to the characteristic number of channels that defines the peak, which may change from spectrum to spectrum. If n represents the channel of the maximum, the contents of the channels n - 10 to n - 2 are averaged, i.e.,

$$\bar{C} = \sum_{i=n-10}^{n-2} C_i \quad (B.14)$$

then the left minimum, l , is determined by scanning each channel from n-2 to n-8 until the following is satisfied,

$$C_l \leq \bar{C} + (\bar{C})^{1/2}. \quad (B.15)$$

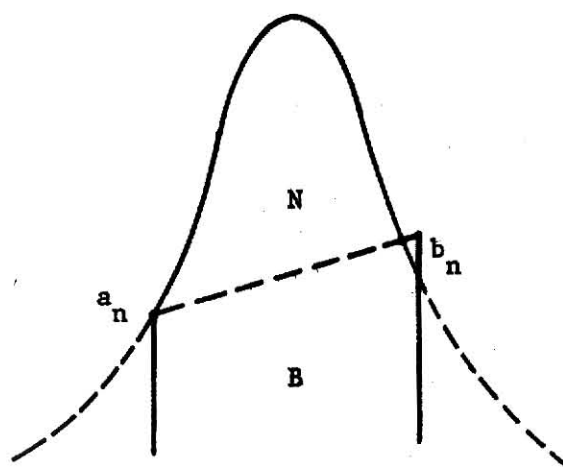
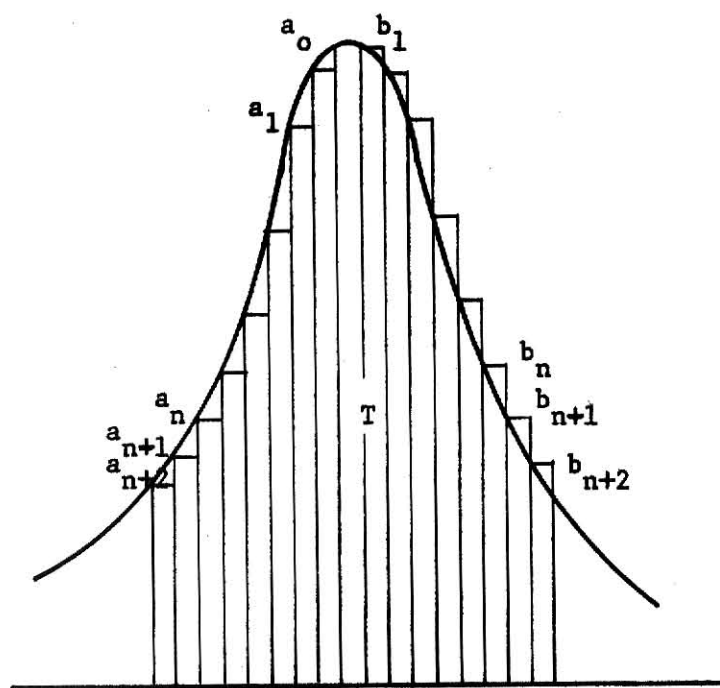


Fig. B.2.1 Graphical representation of a photopeak.

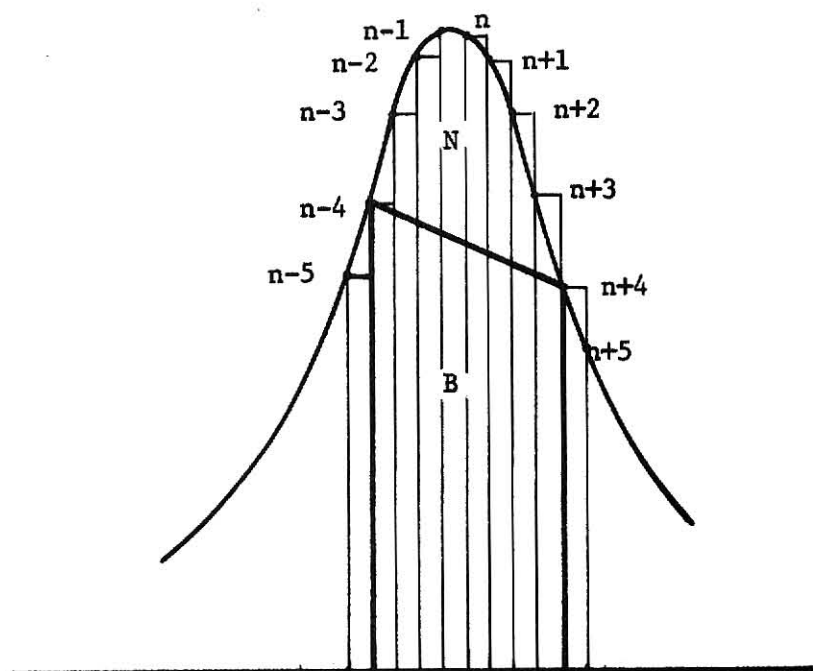


Fig. B.2.2 Histogram for a peak represented by Fixed-Minima Graphical Method.

In a similar way for the right minimum, r , the contents of channels $n + 2$ to $n + 8$ are averaged to find

$$\bar{C} = \sum_{i=n+2}^{n+8} C_i , \quad (B.16)$$

then r is found by scanning channels $n+2$ to $n+8$ until the condition

$$C_r \leq \bar{C} + (\bar{C})^{1/2} , \quad (B.17)$$

is satisfied.

The background is determined to be

$$B = \frac{7}{2} \left[\frac{1}{3} \sum_{i=\ell-2}^{\ell} C_i + \frac{1}{3} \sum_{i=r}^{r+2} C_j \right] , \quad (B.18)$$

and the total area is calculated as

$$T = \sum_{k=\ell}^r C_k . \quad (B.19)$$

The net area N is calculated by Eq. (B.11).

B.3 Standard Deviation of Area

We calculated the uncertainty in the determination of the net area under the peak, irrespective of the method used, to be found by

$$\text{STD. DV.} = (\text{AREA})^{1/2} . \quad (B.20)$$

Equation B.20 is true because the statistical distribution often associated with nuclear process is the Poisson's distribution, where the standard deviation of the mean is given as in Eq. (B.20). Methods, such as the graphical ones, can be given with more rigour for standard deviations. It is possible that with Eq. (B.20), the standard deviation is an over-estimated value, if compared with other values calculated by other methods.

APPENDIX C

Error Propagation Due to Uncertainty in Mass and Time Measurements

Let R^* be the activity per unit mass, defined by

$$R^* = K \frac{P}{M} [e^{-\lambda t_1} (1 - e^{-\lambda t_0}) (1 - e^{-\lambda t_2})]^{-1}, \quad (C.1)$$

where K is a constant which includes

$$K \equiv \frac{\lambda}{f\epsilon}, \quad (C.2)$$

λ = decay constant of the product,

f = molecular fraction of desired isotope,

ϵ = over-all counting efficiency,

P = net area under desired photopeak,

M = sample mass,

t_0 = irradiation time,

t_1 = decay time,

t_2 = counting time.

Assume that each parameter in Eq. (C.1) is measured with some error, and given by σ (standard deviation), i.e., $t_0 \pm \sigma_0$, $M \pm \sigma_M$, $P \pm \sigma_P$, $t_1 \pm \sigma_1$, and $t_2 \pm \sigma_2$.

The uncertainty in the exponential functions due to the errors in times can be calculated by (19,25)

$$\sigma^2(f) = \sum_{i=1}^n \left(\frac{\partial f}{\partial x_i} \right)^2 (\sigma_{x_i})^2, \quad (C.3)$$

where f is a function of the x_i ($i=1, \dots, n$) variables.

Thus, for

$$f_o \equiv f(t_o) = 1 - e^{-\lambda t_o} . \quad (C.4)$$

The S.D. σ_{f_o} is calculated to be

$$\sigma_{f_o} = \left\{ \left[\frac{\partial}{\partial t_o} (1 - e^{-\lambda t_o}) \right]^2 (\sigma_{t_o})^2 \right\}^{1/2} , \quad (C.5)$$

or

$$\sigma_{f_o} = \lambda e^{-\lambda t_o} \sigma_{t_o} . \quad (C.6a)$$

Similarly, for $f_1 \equiv f(t_1) = e^{-\lambda t_1}$ and $f_2 \equiv f(t_2) = 1 - e^{-\lambda t_2}$, the S.D. are

$$\sigma_{f_1} = \lambda e^{-\lambda t_1} \sigma_{t_1} , \quad (C.6b)$$

$$\sigma_{f_2} = \lambda e^{-\lambda t_2} \sigma_{t_2} , \quad (C.6c)$$

Equation (C.1) can be represented as

$$R^* = K \frac{x}{y} , \quad (C.7)$$

where

$$\begin{aligned} x &\equiv P e^{\lambda t_1} = P/f_1 \\ y &\equiv M (1 - e^{-\lambda t_o})(1 - e^{-\lambda t_2}) = M f_o f_2 . \end{aligned} \quad (C.8)$$

From Eq. (C.3), the standard deviation for x and y are calculated from the following:

$$\begin{aligned} \frac{\sigma_{R^*}}{R^*} &= \left[\left(\frac{\sigma_x}{x} \right)^2 + \left(\frac{\sigma_y}{y} \right)^2 \right]^{1/2} = \\ &= \left[\left(\frac{\sigma_P}{P} \right)^2 + \left(\frac{\sigma_M}{M} \right)^2 + \left(\frac{\sigma_{f_o}}{f_o} \right)^2 + \left(\frac{\sigma_{f_1}}{f_1} \right)^2 + \left(\frac{\sigma_{f_2}}{f_2} \right)^2 \right]^{1/2} , \end{aligned} \quad (C.9)$$

or explicitly,

$$\frac{\sigma_{R^*}}{R^*} = \left(\frac{\sigma_P}{P}\right)^2 + \left(\frac{\sigma_M}{M}\right)^2 + \left[\frac{\lambda e^{-\lambda t_o} \sigma_{t_o}}{(1-e^{-\lambda t_o})}\right]^2 + \left[\frac{\lambda e^{-\lambda t_1} \sigma_{t_1}}{e^{-\lambda t_1}}\right]^2 + \left[\frac{\lambda e^{-\lambda t_2} \sigma_{t_2}}{(1-e^{-\lambda t_2})}\right]^2 \}^{1/2} \quad (C.10)$$

It is general practice to assume that σ_M , σ_{t_o} , σ_{t_1} , and σ_{t_2} can be neglected and that the main contributor to the standard deviation of R^* is the uncertainty in the determination of the area under peak of interest. However, we are interested to find the difference in the standard deviation for R^* when calculated by considering the error in all the terms as compared to the standard deviation when error in only the area is considered. As an example the values given in Table C.1 are given for such calculations.

Table C.1 Typical example data.

Parameter	(Fluence Monitor)	
	Oxygen	Iron
λ	0.02586 sec ⁻¹	7.46 x 10 ⁻⁵ sec ⁻¹
t_o	120 sec	120 sec
t_1	15 sec	2 x 10 ³ sec
t_2	60 sec	60 sec
P	2000 counts	4000 counts
M	1 gram	0.03 grams

Let us assume the following standard deviation values:

$$\begin{aligned}\sigma_P &= \sqrt{P} \\ \sigma_M &= 10^{-4} M \\ \sigma_{t_o} &= 10\% t_o \\ \sigma_{t_1} &= 10\% t_1 \\ \sigma_{t_2} &= 5\% \text{ of } t_2\end{aligned}$$

Substituting into Eq. (C.10), we find:

a) for Oxygen

$$\frac{\sigma_{R^*_{ox}}}{R^*_{ox}} = \left\{ \left[\frac{(2000)^{1/2}}{2000} \right]^2 + \left[\frac{1 \times 10^{-4}}{1} \right]^2 + \left[\frac{(0.02586)(e^{-(0.02586)(120)})(12)}{(1 - e^{-(0.02586)(120)})} \right]^2 \right. \\ \left. + [(0.02586)(1.5)]^2 + \left[\frac{(0.02586)(e^{-(0.02586)(60)})(3)}{(1 - e^{-(0.02586)(60)})} \right]^2 \right\}^{1/2}$$

or

$$\frac{\sigma_{R^*_{ox}}}{R^*_{ox}} = \{(5 \times 10^{-4}) + (10^{-8}) + (2.13 \times 10^{-4}) + (1.51 \times 10^{-3}) \\ + (4.35 \times 10^{-4})\}^{1/2}$$

$$\frac{\sigma_{R^*_{ox}}}{R^*_{ox}} \approx 5.16 \times 10^{-2} \quad (\text{for area only: } 2.2 \times 10^{-2})$$

where the main contributor of error is the 10% in t_1 (decay time).

b) For Oxygen with $\sigma_{t_1} = 1\%$ of t_1

$$\frac{\sigma_{R^*_{ox}}}{R^*_{ox}} = \{(5 \times 10^{-4}) + (10^{-8}) + (2.13 \times 10^{-4}) + (1.51 \times 10^{-5}) \\ + (4.35 \times 10^{-4})\}^{1/2}$$

$$\frac{\sigma_{R^*_{ox}}}{R^*_{ox}} \approx 3.4 \times 10^{-2} \quad (\text{area only: } 2.2 \times 10^{-2})$$

where the σ_p , and σ_{t_0} and σ_{t_2} contributed in the approximately the same value.

c) For Oxygen, with $\sigma_{t_1} = 10\%$ of t_1 and $\sigma_M = 9\%$ of M

$$\frac{\sigma_{R^*_{ox}}}{R^*_{ox}} = \{(5 \times 10^{-4}) + (8.1 \times 10^{-3}) + (2.13 \times 10^{-4}) + (1.51 \times 10^{-3}) \\ + (4.35 \times 10^{-4})\}^{1/2}$$

$$\frac{\sigma_{R^*_{ox}}}{R^*_{ox}} \approx 10.4 \times 10^{-2} \quad (\text{area only: } 2.2 \times 10^{-2})$$

where the main contributors of error are the 9% of M (mass) and the 10% of t_1 (decay time).

d) For the fluence monitor for $\sigma_M = 10^{-4} M$ (We assumed that there were no changes in masses because the fluence monitors were solid material),

$$\begin{aligned} \frac{\sigma_{R^*_F}}{R^*_F} = & \left\{ \left[\frac{(4 \times 10^3)^{1/2}}{4 \times 10^3} \right]^2 + \left[\frac{0.03 \times 10^{-4}}{0.03} \right]^2 \right. \\ & + \left[\frac{(7.46 \times 10^{-5})(12)(e^{-(7.46 \times 10^{-5})(120)})}{(1 - e^{-(7.46 \times 10^{-5})(120)})} \right]^2 \\ & + \left[\frac{(7.46 \times 10^{-5})(2 \times 10^3)(10^{-1})}{(1 - e^{-(7.46 \times 10^{-5})(120)})} \right]^2 \\ & \left. + \left[\frac{(7.46 \times 10^{-5})(3)(e^{-(7.46 \times 10^{-5})(60)})}{(1 - e^{-(7.46 \times 10^{-5})(60)})} \right]^2 \right\}^{1/2} \end{aligned}$$

$$\frac{\sigma_{R^*_F}}{R^*_F} = \{(2.5 \times 10^{-4}) + (10^{-8}) + (9.9 \times 10^{-3}) + (2.23 \times 10^{-4}) + (2.5 \times 10^{-3})\}^{1/2}$$

$$\frac{\sigma_{R^*_F}}{R^*_F} \approx 1.13 \times 10^{-1} \quad (\text{area only: } 1.6 \times 10^{-2})$$

where the main contributors to error are the 10% of t_1 (decay time) and the 5% of the t_2 (counting time).

Let assume that the molecular ratio (MR) of the unknown and reference samples, normalized for neutron flux (see Appendix A) can be determined to have an error percentage of

$$\frac{\sigma_{MR}}{MR} \times 100 = \left[\left(\frac{\sigma_{R^*ox}}{R^*ox} \right)^2_{Unk} + \left(\frac{\sigma_{R^*ox}}{R^*ox} \right)^2_{Ref} + \left(\frac{\sigma_{R^*F}}{R^*F} \right)^2_{F Unk} + \left(\frac{\sigma_{R^*F}}{R^*F} \right)^2_{F Ref} \right]^{1/2} \times 100, \quad (C.11)$$

Let us assume the case of errors due to areas only, and values similar for each similar sample, i.e., for samples of oxygen and for fluence monitors. In this case the percentage error will be

$$\frac{\sigma_{MR}}{MR} \times 100 = [(2.2 \times 10^{-2})^2 + (2.2 \times 10^{-2})^2 + (1.6 \times 10^{-2})^2 + (1.6 \times 10^{-2})^2]^{1/2} \times (100)$$

$$\frac{\sigma_{MR}}{MR} \approx 4\% \quad (\text{actually values calculated were in the range of 3\% or less}).$$

Now, let us consider that case a applies for unknown sample, case b for the reference and case d for both fluence references. Then the percentage error is

$$\frac{\sigma_{MR}}{MR} \times 100 = [(5.16 \times 10^{-2})^2 + (10.4 \times 10^{-2})^2 + 2(1.13 \times 10^{-1})^2]^{1/2} \quad (100)$$

$$\frac{\sigma_{MR}}{MR} \times 100 \approx 17.1\% \quad (\text{four times larger than for the area alone})$$

Consider case b and c for the unknown and reference samples, respectively, and case d for fluence monitors.

$$\frac{\sigma_{MR}}{MR} \times 100 \approx 19.4\% \quad (\text{five times larger than for the area alone}).$$

APPENDIX D

Theoretical Mole Ratio and Activities by Assuming Wave-Function Behavior for the Neutron Flux

The rate of change of the product nuclei, $\dot{N}(t)$, is described by the differential equation (see Appendix A)

$$\dot{N}(t) = N_0 \sigma \phi - \lambda N(t) , \quad (D.1)$$

where

N_0 = number of atoms of parent nuclei,

σ = activation cross section,

ϕ = neutron flux,

λ = decay constant of the product.

To find a solution for Eq. (D.1), it is often considered that ϕ and N_0 are constant and independent of time. These assumptions are reasonable for most nuclear reactor irradiations since most of the nuclear irradiations are performed during relatively steady-state operations and relatively short times.

However, we were interested in finding the changes in activity if changes in flux were of considerable amplitude during a certain period of irradiation time, which was sufficiently long compared to the half live, e.g., for the case of activation of a short-lived isotope, such as ^{19}O . Therefore, a wave function neutron flux was considered (ideal wave-train of constant frequency) with the restriction of the average value being constant (and a fixed-value).

In Eq. (D.1) neutron flux becomes time dependent:

$$\dot{N}(t) + \lambda N(t) = \sigma N_0 \phi(t) . \quad (D.2)$$

The solution of Eq. (D.2) is (14,28)

$$N(t) = [e^{-\lambda \int_0^{t_D} dt''}] \sigma N_0 \int_0^{t_D} e^{\lambda \int_0^{t'} dt''} \phi(t') dt' , \quad (D.3)$$

or

$$N(t) = e^{-\lambda t_D} \sigma N_0 \int_0^{t_D} dt' e^{\lambda t'} \phi(t') , \quad (D.4)$$

where t_D is the irradiation time.

Assume that $\phi(t)$ is a constant, $\phi_i(t_i)$, over the interval $\Delta t = t_i - t_{i-1}$. Under such conditions, by numerical integration, Eq. (D.4) can be given as:

$$N(t) = \sigma N_0 e^{-\lambda t_D} \sum_{i=1}^M \phi_i(t_i) \int_{t_{i-1}}^{t_i} dt' e^{\lambda t'} , \quad (D.5)$$

or

$$N(t) = \sigma N_0 e^{-\lambda t_D} \sum_{i=1}^M \phi_i(t_i) [e^{\lambda t_i} - e^{\lambda t_{i-1}}] , \quad (D.6)$$

where M is the number of intervals of Δt .

Assuming samples of unit masses (1 gram) of pure ^{18}O with fluence monitors, also of unit masses, of pure ^{56}Fe , we want to calculate the activity per unit mass and normalized for neutron flux, for different wave-function neutron fluxes. Thereafter, the mole ratio of such samples will be calculated (see Appendix A for more information about mole ratio formulae).

In order to find a detailed description of Eq. (D.6), several cases of wave-functions will be discussed. Let $\psi(t)$ be

$$\psi(t) = \sum_{i=1}^M \phi_i(t_i) [e^{\lambda t_i} - e^{\lambda t_{i-1}}] . \quad (D.7)$$

Table D.1 gives the data used for calculations.

Table D.1 Data used in time-dependent neutron flux calculations.

ϕ_0 (average constant flux) = 1.8×10^{12} n/cm ² -sec		
$t_D = 120$ sec		
$\Delta t = 20$ sec		
$M = 6$ intervals		
Avogadro's number (A#) = 6.023×10^{23}		
Parameter	Oxygen	Iron
λ	0.02586 sec^{-1}	$7.46 \times 10^{-5} \text{ sec}^{-1}$
σ	$0.2 \times 10^{-27} \text{ cm}^2$	$2.7 \times 10^{-24} \text{ cm}^2$
Atomic weight (Ae)	17.99916	55.934934

Using data from Table D.1, we define the constant k as

$$k = \frac{N_o \sigma}{\lambda} e^{-\lambda t_D} = \frac{A\# \sigma}{Ae \lambda} e^{-\lambda t_D}, \quad (\text{D.8})$$

the constant k is calculated:

$$k(\text{oxygen}) = 1.162143 \times 10^{-5},$$

$$k(\text{iron}) = 3.8625 \times 10^2.$$

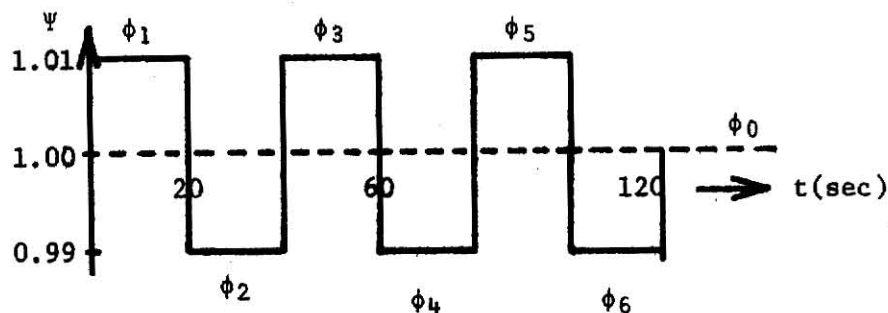
$\psi(t)$, which represents the flux behavior, is calculated for the following functions:

1. For constant flux ϕ_0 , Eq. D.7 becomes

$$\psi(t) = \phi_0 (e^{\lambda t_D} - 1). \quad (\text{D.9})$$

2. For a square wave of amplitude equal to $\pm 1\%$ of ϕ_0 .

Case a



Equation D.7 is

$$\begin{aligned}\psi(t) &= \sum_{i=1}^6 \phi_i [e^{\lambda t_i} - e^{\lambda t_{i-1}}] = \sum_{i=1}^6 \phi_i e^{\lambda t_{i-1}} [e^{\lambda(t_i - t_{i-1})} - 1] \\ &= \sum_{i=1}^6 \phi_i e^{\lambda t_{i-1}} [e^{\lambda \Delta t} - 1],\end{aligned}\quad (\text{D.10})$$

or

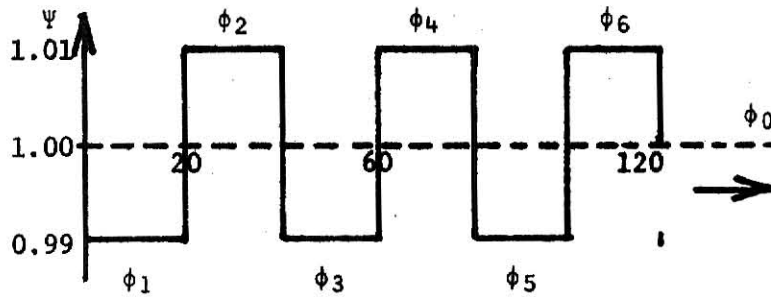
$$\begin{aligned}\psi(t) &= (e^{\lambda 20} - 1) \{ \phi_1 + \phi_2 e^{\lambda 20} + \phi_3 (e^{\lambda 20})^2 \\ &\quad + \phi_4 (e^{\lambda 20})^3 + \phi_5 (e^{\lambda 20})^4 + \phi_6 (e^{\lambda 20})^5 \},\end{aligned}\quad (\text{D.11})$$

where

$$\phi_1 = \phi_3 = \phi_5 = \phi_o \quad (1.01), \quad (\text{D.12a})$$

$$\phi_2 = \phi_4 = \phi_6 = \phi_o \quad (0.99). \quad (\text{D.12b})$$

Case b



For Eq. (D.11) with values ϕ_i :

$$\phi_1 = \phi_3 = \phi_5 = \phi_o \quad (0.99), \quad (\text{D.13a})$$

$$\phi_2 = \phi_4 = \phi_6 = \phi_o \quad (1.01). \quad (\text{D.13b})$$

3. For a square wave of amplitude equal to $\pm 5\%$ of ϕ_0 , Eq. (D.11) applies again and the ϕ_i are defined as

case a

$$\phi_1 = \phi_3 = \phi_5 = \phi_0 (1.05) , \quad (D.14a)$$

$$\phi_2 = \phi_4 = \phi_6 = \phi_0 (0.95) . \quad (D.14b)$$

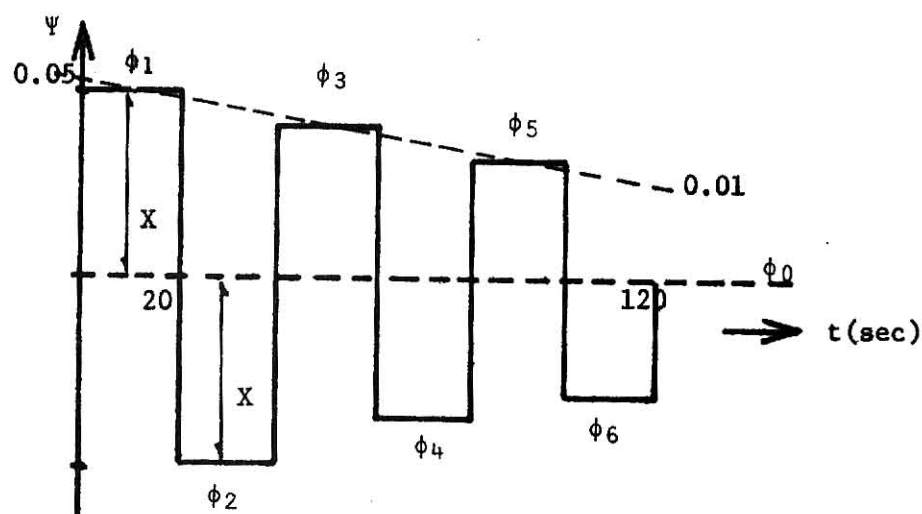
case b Equation (D.11) with ϕ_i defined as

$$\phi_1 = \phi_3 = \phi_5 = \phi_0 (0.95) , \quad (D.15a)$$

$$\phi_2 = \phi_4 = \phi_6 = \phi_0 (1.05) . \quad (D.15b)$$

4. For a decreasing damping wave. Equation (D.4) is to be described for each ϕ_i of each case.

Case a



Each ϕ_i is described by the equation of a straight line (14):

$$\phi_i = Mt_i + B, \quad (D.16)$$

where M = slope of line,

B = intercept (or initial amplitude) of line.

For this line, M and B are

$$M = \frac{(0.01 - 0.05) \phi_o}{t_D - 0} = -0.04 \frac{\phi_o}{t_D}, \quad (D.17a)$$

and

$$B = 0.05 \phi_o. \quad (D.17b)$$

For each ϕ_1, ϕ_3 and ϕ_5 an amplitude, x , above the constant level ϕ_o is calculated by assuming the midpoint for the respective period, was the midpoint a square wave. The negative cycles ϕ_2, ϕ_4, ϕ_6 have the same amplitude, below the level ϕ_o , as their respective positive part.

The midpoint of $P_1(t_i, \phi(t_i))$ and $P_2(t_{i+1}, \phi(t_{i+1}))$ is (14)

$$\text{Midpoint of } P_1 P_2 = \left(\frac{t_{i+1} + t_i}{2}, \frac{\phi(t_{i+1}) + \phi(t_i)}{2} \right), \quad (D.18)$$

where $\phi(t_i)$ and $\phi(t_{i+1})$ are calculated from Eq. D.16.

Therefore, x , the variable amplitude, is

$$x = \frac{[M(t_{i+1}) + B] + [M(t_i) + B]}{2}, \quad (D.19)$$

or

$$x = \frac{M}{2} (t_{i+1} + t_i) + B. \quad (D.20)$$

Each ϕ_i , i odd, is calculated as follows

$$\phi_i = \phi_o + x, \quad i=1,3,5, \quad (D.21a)$$

and each ϕ_i , i even, is

$$\phi_i = \phi_o - x, \quad i = 2,4,6, \quad (D.21b)$$

Therefore, using Eq. D.17, D.20, and D.21, ϕ_1 and ϕ_2 are

$$\phi_1 = \phi_o + \left[\frac{M}{2} (20 + 0) + B \right] = \phi_o (1.047) , \quad (D.22a)$$

$$\phi_2 = \phi_o - \left[\frac{M}{2} (20 + 0) + B \right] = \phi_o (0.953) , \quad (D.22b)$$

Also, ϕ_3 and ϕ_4 are

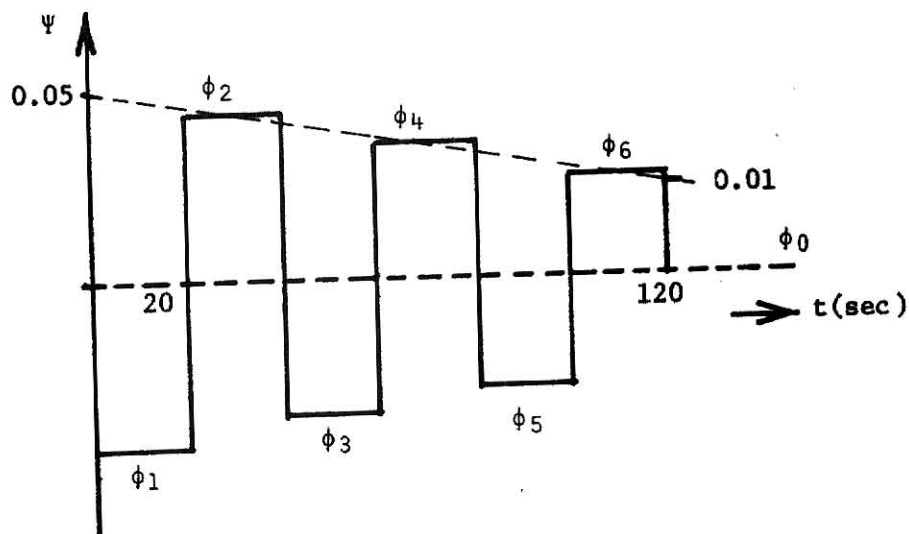
$$\phi_3 = \phi_o + \left[\frac{M}{2} (60 + 40) + B \right] = \phi_o (1.033) , \quad (D.23a)$$

$$\phi_4 = \phi_o - \left[\frac{M}{2} (60 + 40) + B \right] = \phi_o (0.967) , \quad (D.23b)$$

and Eq. (D.7) is

$$\begin{aligned} \psi(t) = \phi_o (e^{\lambda 20} - 1) [1.047 + 0.953 e^{\lambda 20} + 1.033 e^{\lambda 40} + 0.967 e^{\lambda 60} \\ + 1.020 e^{\lambda 80} + 0.98 e^{\lambda 100}] . \end{aligned} \quad (D.24)$$

Case b



For this case, the same steps performed in the case a, may be applied.

Thus ϕ_1 , ϕ_3 and ϕ_5 are given by Eqs. (D.22b), (D.23b), and (D.23d) respectively.

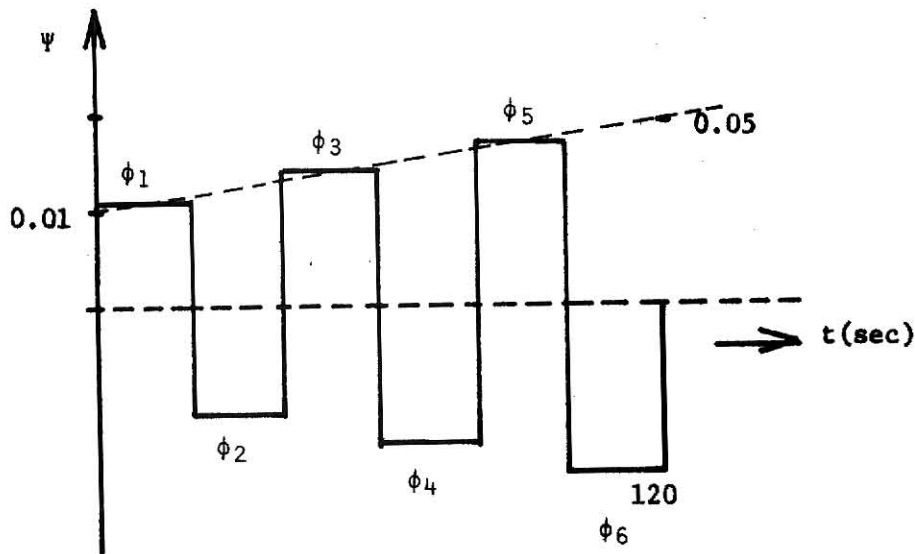
ϕ_2 , ϕ_4 , and ϕ_6 are given by Eqs. (D.22a), (D.23a) and (D.23c) respectively.

Thus, Eq. (D.24) becomes

$$\begin{aligned} \psi(t) = \phi_0 (e^{\lambda 20} - 1) [0.96 + 1.04 e^{\lambda 20} + 0.973 e^{\lambda 40} + 1.027 e^{\lambda 60} \\ + 0.987 e^{\lambda 80} + 1.013 e^{\lambda 100}] . \end{aligned} \quad (D.25)$$

5. For an increasing damping wave.

Case a



The slope and the intercept of the line are now

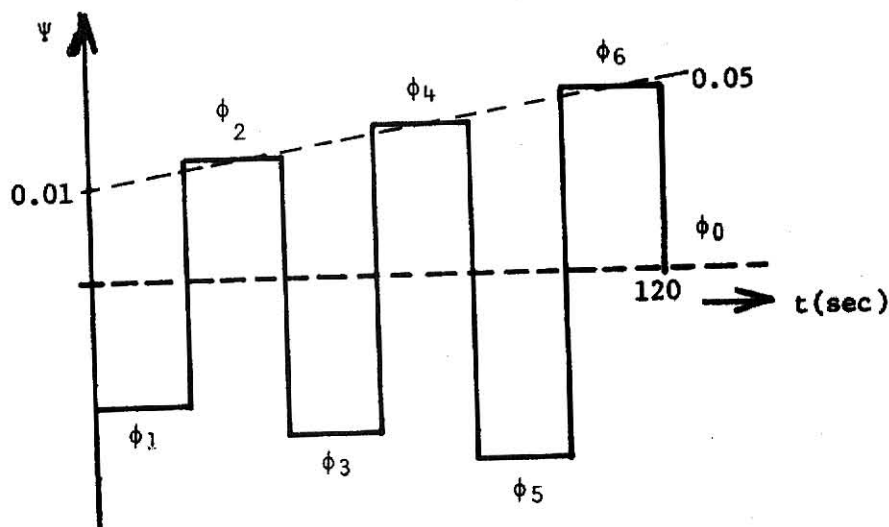
$$M = \frac{(0.05 - 0.04) \phi_0}{t_D - 0}, \quad (D.26)$$

$$B = 0.01 \phi_0, \quad (D.27)$$

From Eqs. D.20, D.21 and D.24, $\psi(t)$ is

$$\begin{aligned} \psi(t) = \phi_0 (e^{\lambda 20} - 1) [1.013 + 0.987 e^{\lambda 20} + 1.027 e^{\lambda 40} + 0.973 e^{\lambda 60} \\ + 1.04 e^{\lambda 80} + 0.96 e^{\lambda 100}] . \end{aligned} \quad (D.28)$$

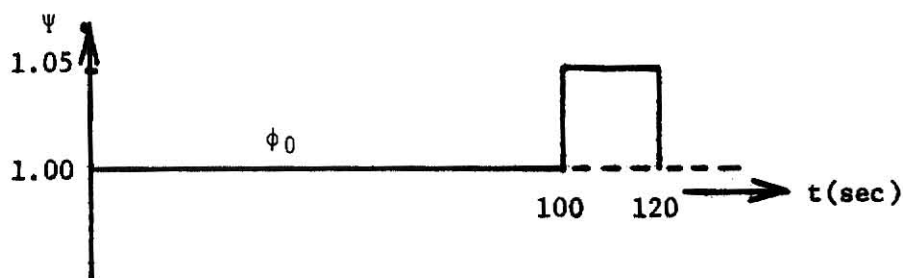
Case b



Equations D.20, D.21, and D.24, apply again, therefore $\psi(t)$ is

$$\begin{aligned} \psi(t) = \phi_0 (e^{\lambda 20} - 1) [0.98 + 1.020 e^{\lambda 20} + 0.967 e^{\lambda 40} + 1.033 e^{\lambda 60} \\ + 0.953 e^{\lambda 80} + 1.047 e^{\lambda 100}] . \end{aligned} \quad (D.29)$$

6. For step function at the end of irradiation.

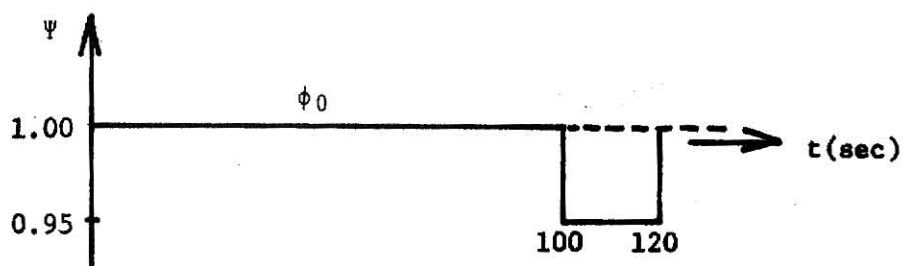
Case a

The flux is ϕ_0 , constant, from $t=0$ to $t=100$ sec and represented as $\phi_0(e^{\lambda 100} - 1)$, for the step function, one can get from Eq. (D.6) for $i=6$, the following

$$\phi_0(1.05)(e^{\lambda 120} - e^{\lambda 100}) . \quad (D.30)$$

Thus, $\psi(t)$ is now

$$\psi(t) = \phi_0(e^{\lambda 100} - 1) + \phi_0(1.05)(e^{\lambda 120} - e^{\lambda 100}) . \quad (D.31)$$

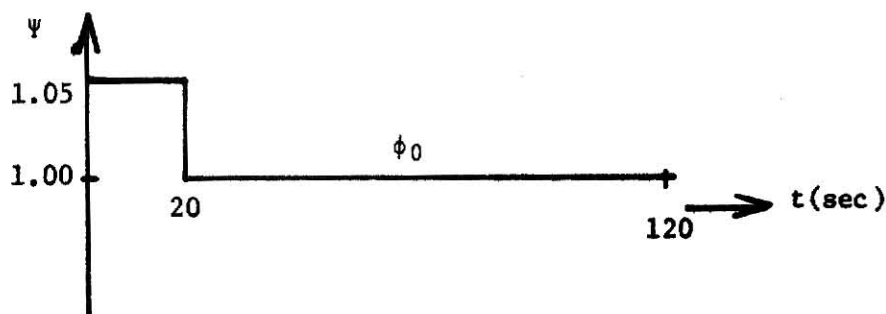
Case b

For a negative step, $\psi(t)$ is

$$\psi(t) = \phi_0(e^{\lambda 100} - 1) + \phi_0(0.95)(e^{\lambda 120} - e^{\lambda 100}) . \quad (D.32)$$

7. For a step function at the beginning of irradiation

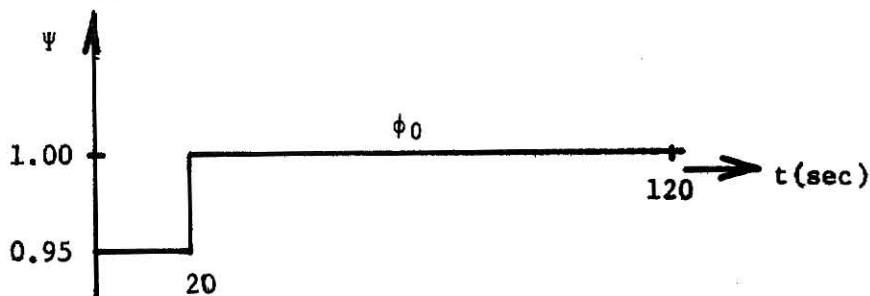
Case a



By similar deduction as in case 6a, we find

$$\psi(t) = (1.05)\phi_0(e^{\lambda 20} - 1) + \phi_0(e^{\lambda 120} - e^{\lambda 20}) . \quad (D.23)$$

Case b



For the negative step, $\psi(t)$ is

$$\psi(t) = (0.95)\phi_0(e^{\lambda 20} - 1) + \phi_0(e^{\lambda 120} - e^{\lambda 20}) . \quad (D.24)$$

By using all the equations for $\psi(t)$ described for the seven different wave-types proposed for the neutron flux, the activities of oxygen and iron samples were calculated. In this way the samples normalized for neutron flux were simulated and the next step was to find the molecular ratio of sample to sample.

Because of the complexity of the possible combinations to produce such samples and such mole ratios, a computer program was developed to perform calculations. The main idea of this study was to find the variations in the percentage error due to variations in neutron flux. Computer results are presented in Tables 5.9 and 5.10 of Section 5 of this work. The computer program is given in Appendix E.

APPENDIX E
Typical Gamma-ray Spectra

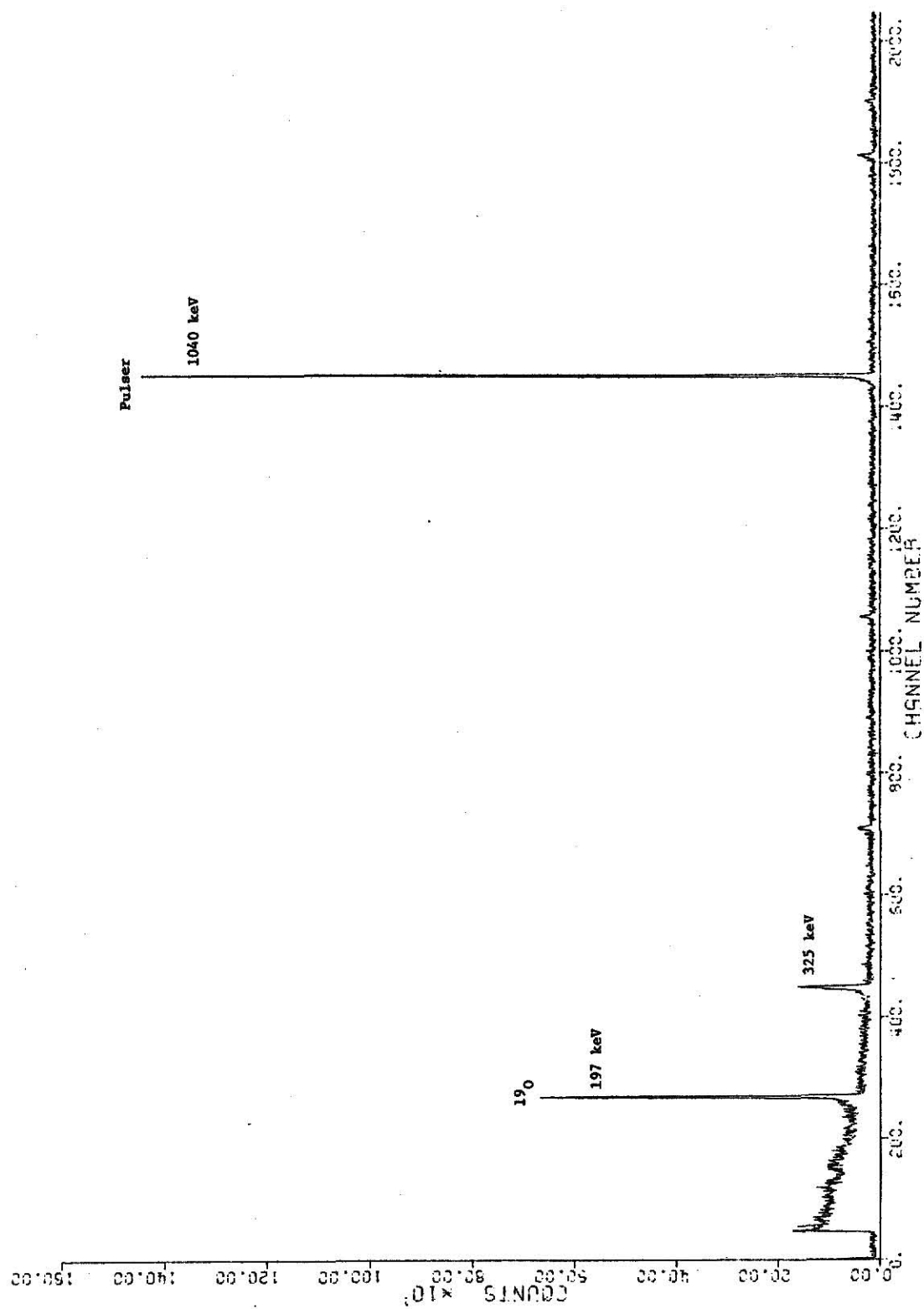


Fig. E.1 Gamma-ray spectrum for α - $\underline{\underline{D}}$ glucose.

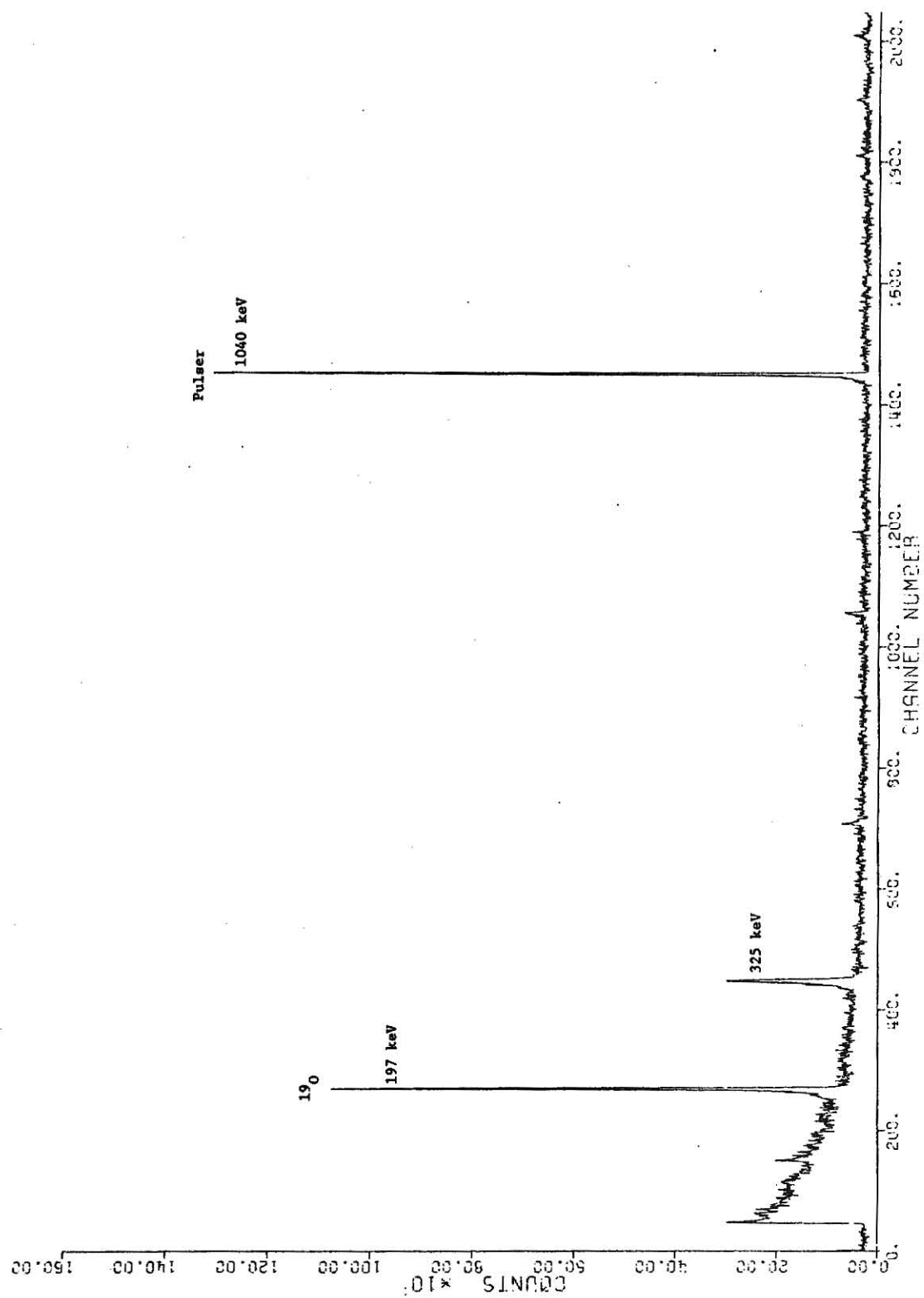


Fig. E.2 Gamma-ray spectrum for water-alcohol mixture.

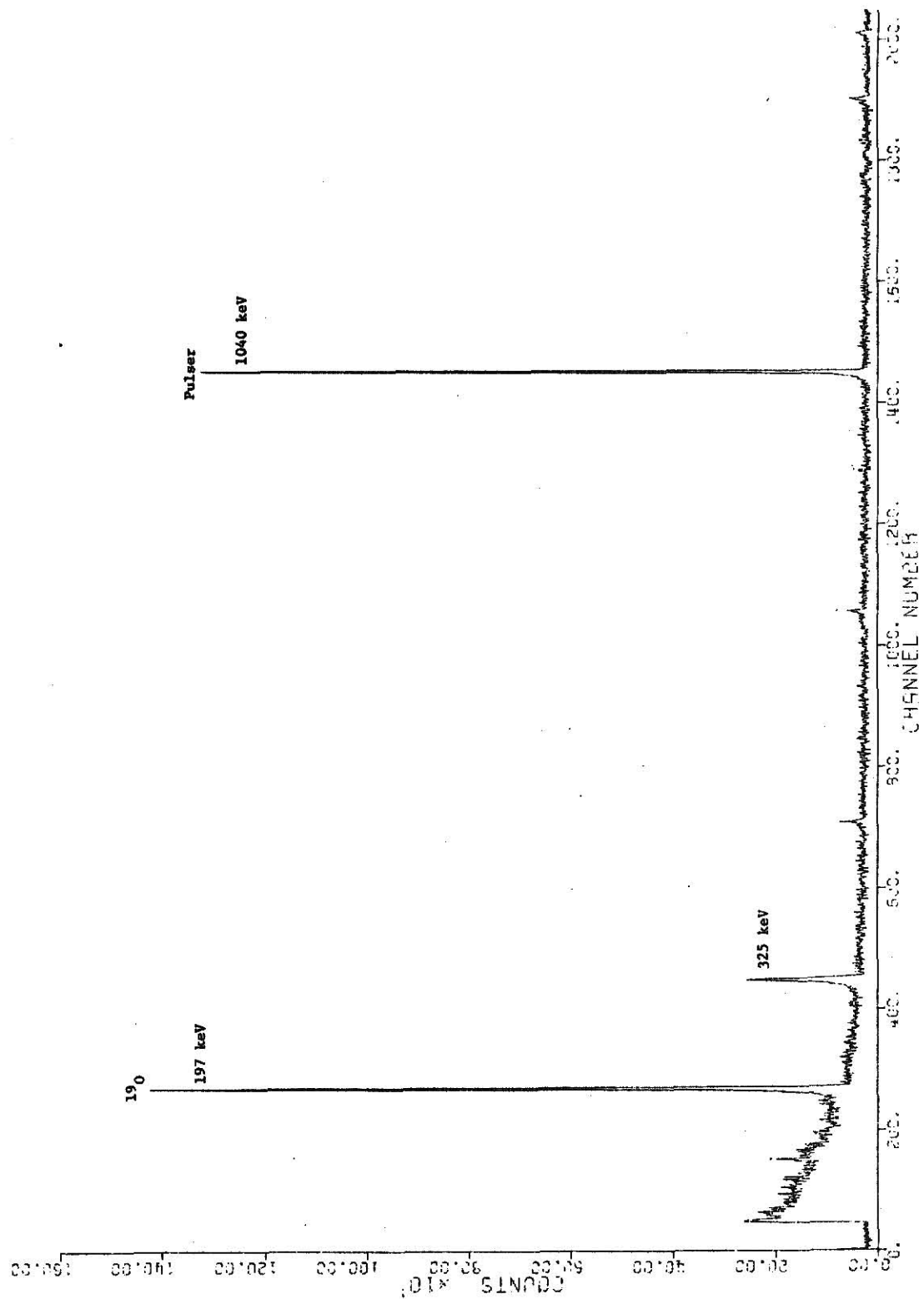


Fig. E.3 Gamma-ray spectrum for distilled water.

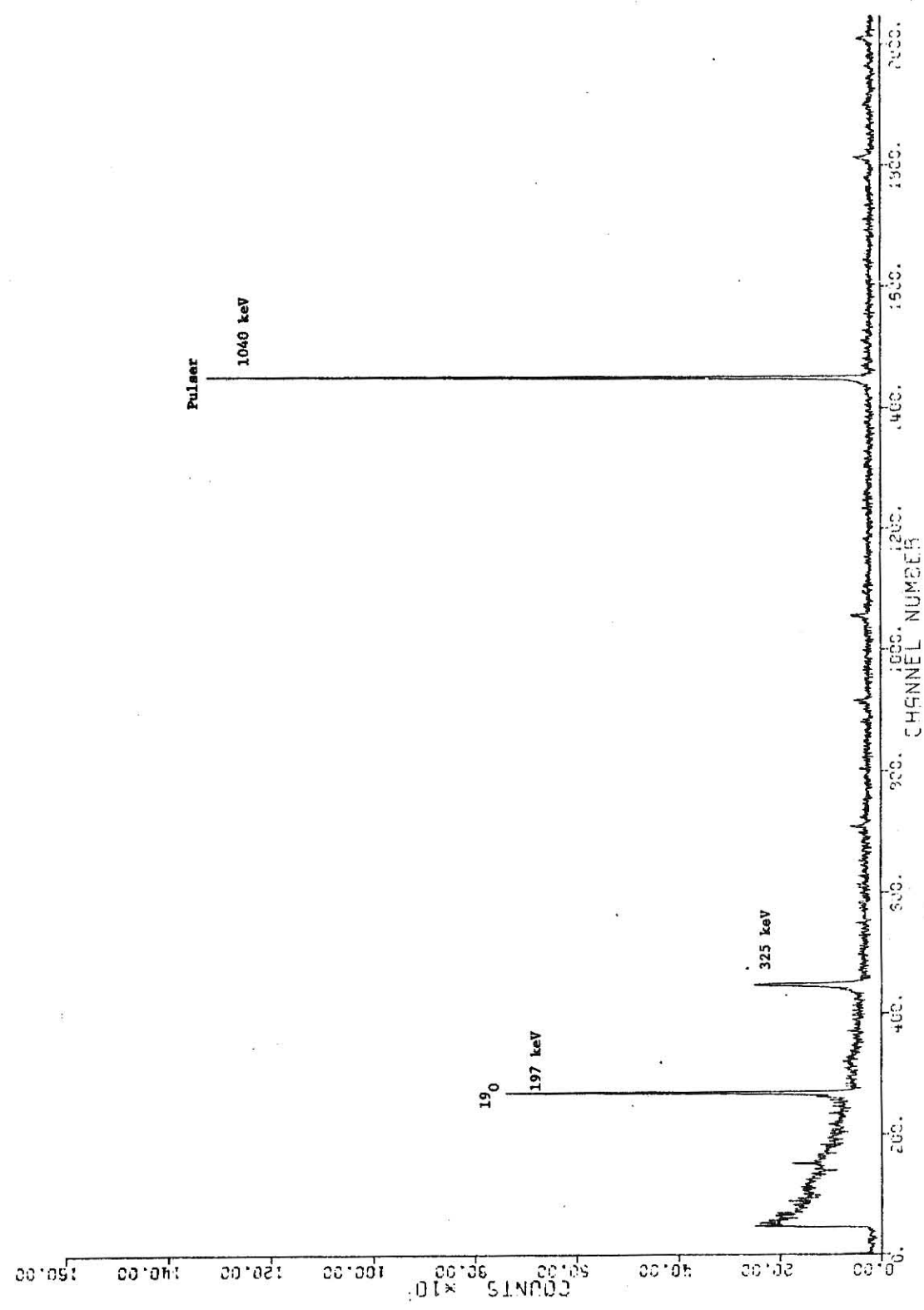


Fig. E.4 Gamma-ray spectrum for α - D fructose.

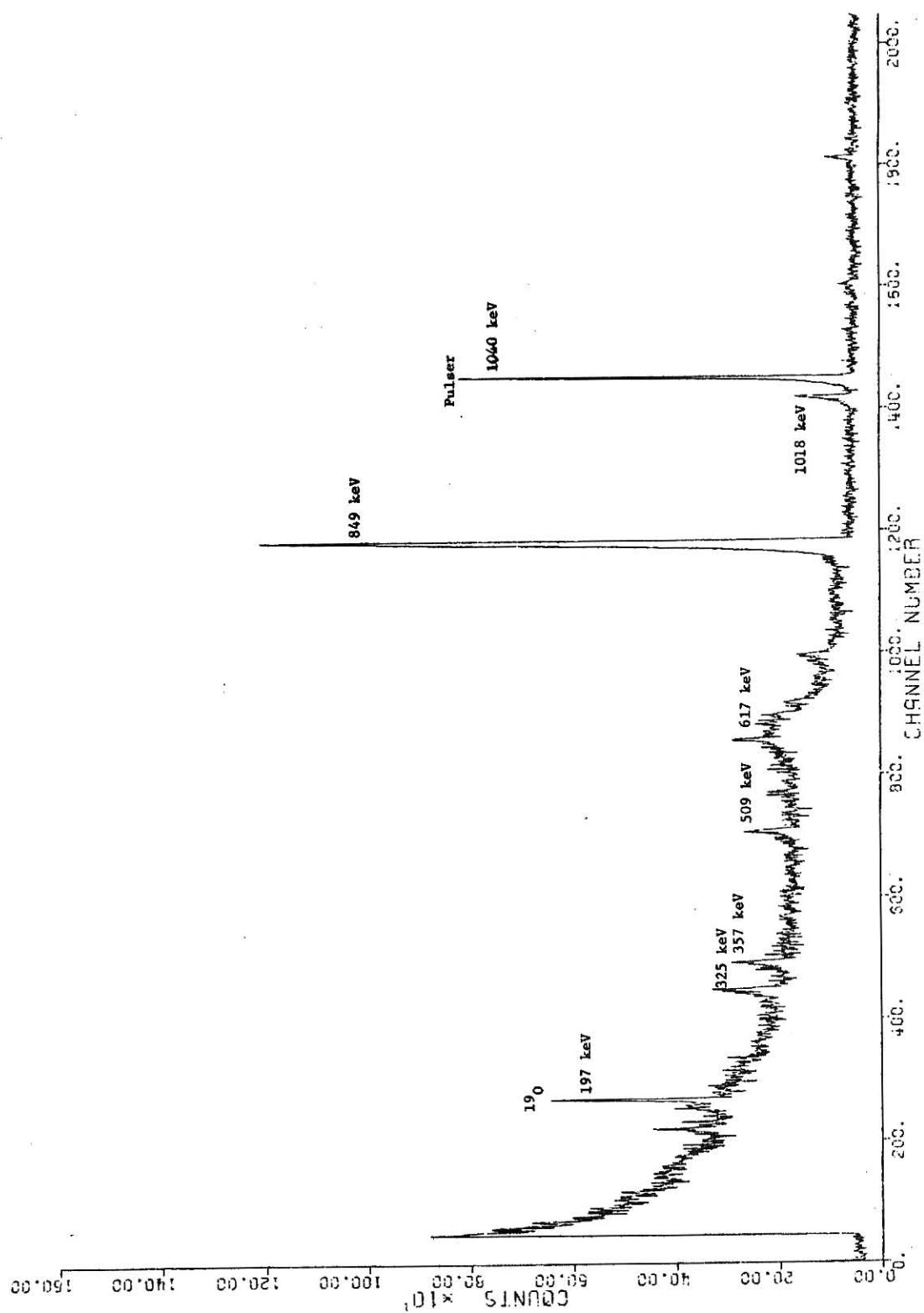


Fig. E.5 Gamma-ray spectrum for standard wheat.

APPENDIX F
Printouts for Computer Programs

```

$JOB          HDM,TIME=(,59)
C *****
C *
C *      MOLE RATIO CALCULATION FOR OXYGEN CONTENTS      *
C *
C *****
1  DIMENSION APEA(4),TAINV(200),TRATIO(200),MOLERA(200)
2  DIMENSION TREG(200),TGDS(200),APK(4),WOLE(200),ZIGMA(200)
3  DIMENSION XNORM(100),XI(4),YI(4),YNORM(100)
4  DIMENSION NAME(100)
5  DIMENSION KNAME(50)
6  DIMENSION KWHICH(50)
7  DIMENSION XM(2),T2(2)
8  DIMENSION KPK(2)
9  DIMENSION SIGMA(200),COUNTS(1024)
10 INTEGER CHANN,COUNTS
11 CHARACTER KNAME*15
12 CHARACTER NAME*15
13 REAL MW,MWSAM,MOLERA,MOL
14 1 READ 50,KPK(1),KPK(2),ITOTL,FELAMD
15 OXLAMD=0.693/26.8
16 READ 52,KKKEY,KAREA2
C *****
C ***** KKKEY - NORMALIZATION *****
C ***** KAREA2 - DESIRED AREA METHOD *****
C *****
17 IF(KKKEY.EQ.1)KEND=2
18 IF(KKKEY.GT.1)KEND=4
C *****
C ***** DATA AND SPECTRA ARE READ *****
C *****
19 DO 500 J=1,ITOTL
20 READ 1000,XM(1),XM(2),T2(1),T2(2)
21 READ 62,NAME(J)
22 DO 300 I=1,KEND
23 READ 200,(COUNTS(I),L=1,36)
24 IF((I.GT.2).AND.(KKKEY.EQ.3))GO TO 300
25 IF(I.GT.2)GO TO 2
26 KPEAK=KPK(I)
27 K=KPEAK-5
28 CHANN=KPEAK+5
C *****
C ***** LOCATION OF CENTROID *****
C *****
29 CALL FINMAX(COUNTS,K,CHANN,IPEAK)
30 2 IF(I.GT.2)IPEAK=22
C *****
C ***** AREAS ARE CALCULATED BY TWO METHODS *****
C ***** ONE IS THE FIXED MINIMA AND THE OTHER TO BE SELECTED *****
C *****
31 CALL AREA(COUNTS,IPEAK,SUMG)
32 IF(I.LE.2)APEA(I)=SUMG
33 IF(I.GT.2)XI(I)=SUMG
34 IF(KAREA2.EQ.1)CALL GREG(COUNTS,IPEAK,SUMG)
35 IF(KAREA2.EQ.2)CALL AREA1(COUNTS,IPEAK,SUMG)
36 IF(I.LE.2)APK(I)=SUMG
37 IF(I.GT.2)YI(I)=SUMG
38 300 CONTINUE
C ***
C *** ACTIVITY/GM AND NORMALIZED FOR NEUTRON FLUENCE ***

```



```

C      ***
39      3 TARM5=(APEA(1)*XM(2))/(APEA(2)*XM(1))
40      Y1=T2(1)*OXLAMD
41      Y2=T2(2)*FELAMD
42      Y1EXP=EXP(Y1)/EXP(Y2)
43      TRATIO(J)=TARM5*Y1EXP
44      TGREY=(APK(1)*XM(2))/(APK(2)*XM(1))
45      TGREG(J)=TGREY*Y1EXP
46      4 TGDS(J)=(1./APK(1))+(1./APK(2))
47      TAINV(J)=(1./APEA(1))+(1./APEA(2))
48      IF(KKKEY.NE.2)GO TO 500
49      XNORM(J)=XI(3)/XI(4)
50      YNORM(J)=YI(3)/YI(4)
51      TRATIO(J)=TRATIO(J)/XNORM(J)
52      TGREG(J)=TGREG(J)/YNORM(J)
53      TGDS(J)=TGDS(J)+(1./YI(3))+(1./YI(4))
54      TAINV(J)=TAINV(J)+(1./XI(3))+(1./XI(4))
55      500 CONTINUE
56      40 READ 51,KREF
57      GO TO(3000,510,510,1,2000),KREF
58      510 READ 60,KTOT,(KWHICH(I),I=1,KTOT)
C      ***
C      *** MOLE RATIO FOR SAMPLE/WATER OR SAMPLE/SAMPLE
C      ***
59      READ 61,(KNAME(I),I=1,KTOT)
60      IF(KREF.EQ.2)GO TO 520
61      INIT=0
62      LAST=0
C
C
C      **** LOOP FOR ALL AND EACH ONE OF SELECTED REFERENCES
63      520 DO 1500 I=1,KTOT
64      PRINT 401
65      PRINT 70,KNAME(I)
66      IF(KKKEY.EQ.2)PRINT 75
67      PRINT 405
68      IF(KREF.EQ.2)PRINT 400
69      IF(KREF.EQ.3)PRINT 403
70      PRINT 405
71      PRINT 410
72      IF(KAREA2.EQ.1)PRINT 423
73      IF(KAREA2.EQ.1)PRINT 424
74      IF(KAREA2.EQ.2)PRINT 420
75      IF(KAREA2.EQ.2)PRINT 421
76      PRINT 405
77      K=KWHICH(I)
78      IF(KREF.EQ.3)GO TO 550
79      INIT=0
80      LAST=0
81      550 READ 100,KEY,NEXT,DESIRE,MWSAM
82      IF(KREF.LT.3)MW=MWSAM/18.01534
83      IF(KREF.EQ.3)MW=1.
84      IDESIR=IFIX(DESIRE)
85      PRINT 750,IDESIR
86      ASIG=0.0
87      AMOLR=0.0
88      XAMOL=0.0
89      XASIG=0.0
90      XTOT=FLOAT(NEXT)
91      IF(KEY.EQ.0)XTOT=XTOT-1.

```

```

52      INIT=LAST+1
93      LAST=LAST+NEXT
94      DO 900 J=INIT,LAST
C
C      *** MOLE RATIO FOR EACH SAMPLE OF SAME GROUP ***
C
55      IF(J.EQ.K)GO TO 900
96      MOLERA(J)=(TRATIO(J)/TRATIO(K))*MW
97      AINV=TAINV(J)+TAINV(K)
98      SIGM=SQRT(AINV)
99      SIGMA(J)=MOLERA(J)*SIGM
100     WOLE(J)=(TGREG(J)/TGREG(K))*MW
101     ASIGM=TGDS(J)+TGDS(K)
102     ZIGMA(J)=(SQRT(ASIGM))*WOLE(J)
103     IF(DESIREEQ.6.0)XK=1.063
104     IF(DESIREEQ.6.0)XK=1.0
105     MOL=XK*MOLERA(J)
106     ZIGM=XK*SIGMA(J)
107     XMOL=XK*WOLE(J)
108     YZIGM=XK*ZIGMA(J)
C
C      **** IF MOLE RATIO IS KNOWN, % ERROR IS CALCULATED ***
C
109     IF(DESIREEQ.0.0)GO TO 575
110     XERROR=((DESIREE-MOL)*100.0)/DESIREE
111     ZERROR=((DESIREE-XMOL)*100.0)/DESIREE
112     IF(DESIREEEQ.6.0)PRINT 450,NAME(J),MOLERA(J),SIGMA(J),
113     1WOLE(J),ZIGMA(J),MOL,ZIGM,XMOL,YZIGM,XERROR,ZERROR
114     IF(DESIREEEQ.6.0)PRINT 451,NAME(J),MOLERA(J),SIGMA(J),
115     1WOLE(J),ZIGMA(J),XERROR,ZERROR
116     575 AMOLR=AMOLR+MOLERA(J)
117     W1=SIGMA(J)**2
118     ASIG=ASIG+W1
119     XAMOL=XAMOL+WOLE(J)
120     W2=ZIGMA(J)**2
121     XASIG=XASIG+W2
122     IF(DESIREEEQ.0.0)GO TO 600
123     GO TO 900
124     600 PRINT 405
125     PRINT 1300,NAME(J),MOLERA(J),SIGMA(J),WOLE(J),ZIGMA(J)
126     900 CONTINUE
C
C      **** AVERAGE VALUES ****
C
127     AMOLR=AMOLR/XTOT
128     ASIGMA=(SQRT(ASIG))/XTOT
129     BMOLB=XAMOL/XTOT
130     BZIGMA=(SQRT(XASIG))/XTOT
131     IF(DESIREEEQ.0.0)GO TO 910
132     XMOL=XK*AMOLR
133     XZIGM=XK*ASIGMA
134     YERROR=((DESIREE-XMOL)*100.0)/DESIREE
135     YMOL=XK*BMOLB
136     ZZIGM=XK*BZIGMA
137     WERROR=((DESIREE-YMOL)*100.0)/DESIREE
138     910 PRINT 407
139     IF(DESIREEEQ.0.0)PRINT 710,AMOLR,ASIGMA,BMOLB,BZIGMA
140     IF(DESIREEEQ.0.0)GO TO 920
141     IF(DESIREEEQ.6.0)PRINT 705,AMOLR,ASIGMA,BMOLB,BZIGMA
142     1,YERROR,WERROR

```

```

140      IF (DESIRE.EQ.6.)PRINT 700,AMOLR,ASIGMA,BMOLB,
141      1BZIGMA,XMOL,XZIGM,YMOL,ZZIGM,VEERROR,WERROR
142      PRINT 407
143      PRINT 405
144      IF(LAST.EQ.ITOTL)GO TO 1500
145      950 IF(KREF.EQ.2)GO TO 550
146      1500 CONTINUE
147      GO TO 40
148      3000 READ 60,KTOT,(KWHICH(I),I=1,KTOT)
149      C      *****
150      C      ***** EACH SAMPLE WITH ITS OWN REFERENCE *****
151      C      ***** GROUPS OF SAMPLE AND REFERENCES REQUIRED ****
152      C      *****
153      PRINT 401
154      IF(KKKEY.EQ.2)PRINT 75
155      PRINT 405
156      PRINT 400
157      PRINT 406
158      PRINT 405
159      PRINT 410
160      IF(KAREA2.EQ.1)PRINT 423
161      IF(KAREA2.EQ.1)PRINT 424
162      IF(KAREA2.EQ.2)PRINT 420
163      IF(KAREA2.EQ.2)PRINT 421
164      PRINT 405
165      DO 4000 I=1,KTOT,2
166      3200 K=KWHICH(I)-1
167      L=KWHICH(I+1)-1
168      READ 100,KEY,NEXT,DESIRE,MWSAM
169      MW=MWSAM/18.01534
170      IDESIR=FIX(DESIRE)
171      PRINT 750,IDESIR
172      XTOT=FLOAT(NEXT)
173      ASIG=0.0
174      AMOLR=0.0
175      XAMOL=0.0
176      XASIG=0.0
177      DO 3300 J=1,NEXT
178      MOLER(J)=(TRATIO(J+K)/TRATIO(J+L))*MW
179      ASIGM=TAINV(J+K)+TAINV(J+L)
180      SIGM=SQRT(ASIGM)
181      SIGMA(J)=MOLER(J)*SIGM
182      WOLE(J)=(TGREG(J+K)/TGREG(J+L))*MW
183      ASIGM=TGDS(J+K)+TGDS(J+L)
184      ZIGMA(J)=(SQRT(ASIGM))*WOLE(J)
185      IF(DESIRE.EQ.6.0)XK=1.063
186      IF(DESIRE.NE.6.0)XK=1.0
187      MOL=XK*MOLER(J)
188      ZIGM=XK*SIGMA(J)
189      XMOL=XK*WOLE(J)
190      YZIGM=XK*ZIGMA(J)
191      IF(DESIRE.EQ.0.)GO TO 3250
192      XERROR=((DESIRE-MOL)*100.0)/DESIRE
193      ZERROR=((DESIRE-XMOL)*100.0)/DESIRE
194      IF(DESIRE.EQ.6.)PRINT 450,NAME(J+K),MOLER(J),SIGMA(J),
195      1WOLE(J),ZIGMA(J),MOL,ZIGM,XMOL,YZIGM,XERROR,ZERROR
196      IF(DESIRE.NE.6.)PRINT 451,NAME(J+K),MOLER(J),SIGMA(J),
197      1WOLE(J),ZIGMA(J),XERROR,ZERROR
198      3250 AMOLR=AMOLR+MOLER(J)
199      W1=SIGMA(J)**2

```

```

193      ASIG=ASIG+W1
194      XAMOL=XAMOL+WOLE(J)
195      W2=ZIGMA(J)**2
196      XASIG=XASIG+W2
197      IF(DESIRED.EQ.0.0)GO TO 3400
198      GO TO 3300
199 3400 PRINT 405
200      PRINT 1300,NAME(J+K),MOLERA(J),SIGMA(J),WOLE(J),ZIGMA(J)
201 3300 CONTINUE
202      AMOLR=AMOLR/XTOT
203      ASIGMA=(SQRT(ASIG))/XTOT
204      BMOLB=XAMOL/XTOT
205      BZIGMA=(SQRT(XASIG))/XTOT
206      IF(DESIRED.EQ.0.0)GO TO 3700
207      XMOL=XK*AMOLR
208      XZIGM=XK*ASIGMA
209      YERROR=((DESIRED-XMOL)*100.0)/DESIRED
210      YMOL=XK*BMOLB
211      ZZIGM=XK*BZIGMA
212      WERROR=((DESIRED-YMOL)*100.0)/DESIRED
213 3700 PRINT 407
214      IF(DESIRED.EQ.0.0)PRINT 710,AMOLR,ASIGMA,BMOLB,BZIGMA
215      IF(DESIRED.EQ.0.0)GO TO 3800
216      IF (DESIRED.EQ.6.0)PRINT 700,AMOLR,ASIGMA,BMOLB,
18ZIGMA,XMOL,XZIGM,YMOL,ZZIGM,YERROR,WERROR
217      IF(DESIRED.NE.6.0)PRINT 705,AMOLR,ASIGMA,BMOLB,BZIGMA
1,YERROR,WERROR
218 3800 PRINT 407
219 4000 CONTINUE
220      GO TO 40
221 2000 PRINT 405
222      PRINT 401
223      50 FORMAT(3(2X,I3),F10.7)
224      51 FORMAT(11)
225      52 FORMAT(11,2X,11)
226      60 FORMAT(13,10I3)
227      61 FORMAT(5A15)
228      62 FORMAT(A15)
229      70 FORMAT(12X,'REFERENCE IS',10X,A15)
230      75 FORMAT(5X,'(DATA NORMALIZED FOR COUNTING TIME BY A PULSER)')
231      100 FORMAT(11,2X,13,2X,F3.0,2X,F7.3)
232      200 FORMAT(6X,12I6)
233      400 FORMAT(30X,'RATIO OF ACTIVITY OF SAMPLE TO DISTILLED WATER ('
1'GM-MOLE BASIS)')
234      401 FORMAT('1')
235      403 FORMAT(36X,'RATIO OF ACTIVITY SAMPLE TO SAMPLE ('
1,'GM-MOLE BASIS)')
236      405 FORMAT(' ')
237      406 FORMAT(17X,'EACH SAMPLE HAS ITS OWN REFERENCE')
238      407 FORMAT(17X,2('**',19X,'***',20X),'**',14X,'***')
239      410 FORMAT(17X,'**',10X,'OBSERVED MOLE RATIO',12X,'**',10X,
1'CORRECTED MOLE RATIO',11X,'**',11X,'% ERROR')
240      420 FORMAT(3X,'S A M P L E',3X,2('**',3X,'BY FIX. MIN. ',3X,'***',4X,
1'BY MINIMA ',4X),'**',1X,'BY FIX. MIN.',1X,'***',1X,
2'BY MINIMA ')
241      421 FORMAT(17X,2('**',6X,'7 POINT',6X,'***',6X,'VARIABLE',6X),
1'***',3X,'7 POINT',4X,'***',3X,'VARIABLE')
242      423 FORMAT(3X,'S A M P L E',3X,2('**',3X,'BY FIX. MIN. ',3X,'***',4X,
1'BY GREGORY S',4X),'**',1X,'BY FIX. MIN.',1X,'***',1X,
2'BY GREGORY S')

```

```

243 424 FORMAT(17X,2('**',6X,'7 POINT',6X,'***',6X,'EQUATION',6X),
      1**',3X,'7 POINT',4X,'***',3X,'EQUATION')
244 450 FORMAT(2X,A15,2('**',1X,F7.3,1X,'('',1X,F7.3,')***',2X,F7.3,
      11X,'('',1X,F7.3,')')',**',3X,F7.3,4X,'***',3X,F7.3)
245 451 FORMAT(2X,A15,'**',1X,F7.3,1X,'('',1X,F7.3,')***',2X,F7.3,
      11X,'('',1X,F7.3,')**',41X,'**',3X,F7.3,4X,'***',3X,F7.3)
246 700 FORMAT(7X,'AVERAGE ='',1X,2(2X,F7.3,1X,'('',1X,F7.3,')',4X,
      1F7.3,1X,'('',1X,F7.3,')')',4X,F7.3,9X,F7.3)
247 705 FORMAT(7X,'AVERAGE ='',1X,2X,F7.3,1X,'('',1X,F7.3,')',4X,
      1F7.3,1X,'('',1X,F7.3,')',46X,F7.3,9X,F7.3)
248 710 FORMAT(7X,'AVERAGE ='',1X,2X,F7.3,1X,'('',1X,F7.3,')',4X,
      1F7.3,1X,'('',1X,F7.3,')')
249 750 FORMAT(2X,'EXPECTED MOLE RATIO ='',2X,I3)
250 1000 FORMAT(2(F7.4,3X),F6.0,3X,F7.0)
251 1300 FORMAT(2X,A15,'**',1X,F7.3,1X,'('',1X,F7.3,')***',2X,F7.3,1X,
      1('',1X,F7.3,')',3X,'(FRACTION OF OXYGEN)')
252 STOP
253 END

C
C SUBROUTINE TO FIND CHANNEL OF MAXIMUM
C

254 SUBROUTINE FINMAX(COUNTS,K,CHANN,IPEAK)
255 DIMENSION COUNTS(1024)
256 INTEGER COUNTS, CHANN
257 LARGE=COUNTS(K)
258 IPEAK=K
259 DO 6 KK=K,CHANN
260 IF(COUNTS(KK).LE.LARGE)GO TO 6
261 LARGE=COUNTS(KK)
262 IPEAK=KK
263 6 CONTINUE
264 RETURN
265 END

C ***** AREA USING GREGORY'S EQUATION *****

266 SUBROUTINE GREG(COUNTS,IPEAK,SUMG)
267 DIMENSION COUNTS(1024)
268 DIMENSION FACTR(4),DELT(1024)
269 INTEGER COUNTS
270 DATA FACTR/.083333,.041667,.026389,.01875/
271 SUMG=0.0
272 MINX=IPEAK-7
273 MAXX=IPEAK+2
274 NP2=MAXX-MINX-1
275 NP1=MAXX-MINX
276 DO 25 LL=MINX,MAXX
277 IX=LL-MINX+1
278 DELT(IX)=COUNTS(LL)
279 25 CONTINUE
280 DO 100 JJ=MINX,MAXX
281 PTS=COUNTS(JJ)
282 IF((JJ.EQ.MINX).OR.(JJ.EQ.MAXX))PTS=PTS/2.0
283 SUMG=SUMG+PTS
284 100 CONTINUE
285 IF(NP2.GT.4)NP2=4
286 DO 150 JQ=1,NP2
287 DO 125 JP=1,NP1
288 125 DELT(JP)=DELT(JP)-DELT(JP+1)
289 SIGN=(-1.0)**JQ

```

```

290      SUMG=SUMG-FACTR(JQ)*(DELT(NP1)+SIGN*DELT(1))
291      NP1=NP1-1
292      150 CONTINUE
293      RETURN
294      END

```

```

C      *****
C      ***** AREA BY FIXED MINIMA METHOD *****
C      *****

```

```

295      SUBROUTINE AREA(COUNTS,IPEAK,SUMG)
296      DIMENSION COUNTS(1024)
297      INTEGER COUNTS
298      INTEGER S6435,SUM43
299      SUMG=0.0
300      N6=IPEAK-5
301      N4=IPEAK-3
302      N3=IPEAK+3
303      N5=IPEAK+5
304      S6435=0
305      SUM43=0
306      DO 400 K=N6,N4
307      400 S6435=S6435+COUNTS(K)
308      DO 500 K=N3,N5
309      500 S6435=S6435+ COUNTS(K)
310      DO 600 K=N4,N3
311      600 SUM43=SUM43+COUNTS(K)
312      SUMG=SUM43-(7.*S6435)/6.
313      IF (SUMG.LT.0.0) SUMG=SUM43
314      RETURN
315      END

```

```

C      *****
C      ***** AREA BY VARIABLE MINIMA METHOD *****
C      *****

```

```

316      SUBROUTINE AREA1(COUNTS,IPEAK,SUMG)
317      DIMENSION COUNTS(1024)
318      INTEGER COUNTS
319      L1=IPEAK-10
320      IF (L1.LE.0) L1=1
321      L2=IPEAK-3
322      ISERIE=0
323      DO 50 I=L1,L2
324      50 ISERIE=ISERIE+COUNTS(I)
325      SERIE=FLOAT(ISERIE)
326      SERIE=SERIE/8.
327      SERIE=SERIE+SQRT(SERIE)
328      L1=L1+3
329      L3=IPEAK-2
330      N4=L1
331      DO 51 I=L1,L3
332      51 IF (COUNTS(I).GT.SERIE) GO TO 52
333      N4=I
334      51 CONTINUE
335      52 N6=N4-2
336      L1=IPEAK+3
337      L2=IPEAK+10
338      IF (L2.GT.36) L2=36
339      ISERIE=0
340      DO 60 I=L1,L2
341      60 ISERIE=ISERIE+COUNTS(I)

```

```

342      SERIE=FLOAT(I SERIE)
343      SERIE=SERIE/8.
344      SERIE=SERIE+SQRT(SERIE)
345      L2=L2-3
346      L3=IPEAK+2
347      N3=L3
348      DO 61 I=L3,L2
349      IF(COUNTS(I)).LE.SERIE GO TO 62
350      N3=I
351  61 CONTINUE
352  62 N5=N3+2
353      TN=N3-N4+1
354      BSUM=0.0
355      SUM=0.0
356      DO 70 K=N6,N4
357  70 BSUM=BSUM+COUNTS(K)
358      DO 71 K=N3,N5
359  71 BSUM=BSUM+COUNTS(K)
360      DO 72 K=N4,N3
361  72 SUM=SUM+COUNTS(K)
362      SUMG=SUM-(TN*BSUM)/6.
363      IF(SUMG.LT.0.0) SUMG=SUM
364      RETURN
365      END

```

\$ENTRY

```

$JOB
C
C *****
C ***** THEORETICAL NEUTRON FLUX BEHAVIOR *****
C *****
C
1  DIMENSION L(100),M(100)
2  DIMENSION D1(7),D2(7),F1(7),F2(7),RATIO(100)
3  REAL NOX,NFE
4  DATA OXFE,DEFE,AVOG/0.2E-27,2.7E-24,6.023E23/
5  DATA A,B,FLUX/0.01,0.05,1.0E12/
6  AOX=17.59416
7  ALOX=2.586E-02
8  ALFE=7.46E-05
9  AFE=55.934934
10 SM=1./((100.*120.))
11 NOX=AVOG/AOX
12 NFE=AVOG/AFE
C
C *** DATA FOR OXYGEN AND IRON ***
C ***
13 ACTOX=((DEFE*NOX)/ALOX)*(EXP(-ALOX*120.))
14 ACTFE=((DEFE*NFE)/ALFE)*(EXP(-ALFE*120.))
C
C ** ACTIVITY/GM **
C **
15 DO 300 K=1,2
16 IF(K.EQ.1)Y=ALOX
17 IF(K.EQ.2)Y=ALFE
18 DO 200 J=2,3
19 IF(J.EQ.2)X=A
20 IF(J.EQ.3)X=B
C
C **
C ** FLUX TYPE SQUARE WAVE 2 AND 3 ARE GENERATED **
C **
21 CALL SUM1(X,Y,FA,FB)
22 IF(K.EQ.2)GO TO 100
23 O1(J)=FA
24 O2(J)=FB
25 GO TO 200
26 100 F1(J)=FA
27 F2(J)=FB
28 200 CONTINUE
29 300 CONTINUE
30 DO 500 K=1,2
31 IF(K.EQ.1)Y=ALOX
32 IF(K.EQ.2)Y=ALFE
33 S4=-SM
34 DO 500 J=4,5
35 IF(J.EQ.4)X=B
36 IF(J.EQ.5)X=A
37 IF(J.EQ.5)SM=-SM
C
C ***
C *** FLUX TYPES DECREASING (4) AND INCREASING (5) DAMPING ***
C ***
38 CALL SUM2(X,Y,SM,FA,FB)
39 IF(K.EQ.2)GO TO 400
40 O1(J)=FA
41 O2(J)=FB
42 GO TO 500

```



```

43 400 F1(J)=FA
44 F2(J)=FB
45 500 CONTINUE
46 600 CONTINUE
C ***
C *** ACTIVITY/GM FOR FLUX TYPE 1 (CONSTANT) AND 6 AND 7 (STEP) ***
C ***
47 Q1(1)=ACTOX*(EXP(ALOX*120.)-1.)
48 Q1(1)=Q1(1)*FLUX
49 F1(1)=ACTFE*(EXP(ALFE*120.)-1.)
50 F1(1)=F1(1)*FLUX
51 Q1(6)=FLUX*((EXP(ALOX*100.)-1.)*(1.+B)*(EXP(ALOX*120.)
1-EXP(ALOX*100.)))
52 Q2(6)=FLUX*((EXP(ALOX*100.)-1.)*(1.-B)*(EXP(ALOX*120.)
1-EXP(ALOX*100.)))
53 F1(6)=FLUX*((EXP(ALFE*100.)-1.)*(1.+B)*(EXP(ALFE*120.)
1-EXP(ALFE*100.)))
54 F2(6)=FLUX*((EXP(ALFE*100.)-1.)*(1.-B)*(EXP(ALFE*120.)
1-EXP(ALFE*100.)))
55 Q1(7)=FLUX*((1.+B)*(EXP(ALOX*20.)-1.)*(EXP(ALOX*120.)-EXP(ALOX
1*20.)))
56 Q2(7)=FLUX*((1.-B)*(EXP(ALOX*20.)-1.)*(EXP(ALOX*120.)-EXP(ALOX
1*20.)))
57 F1(7)=FLUX*((1.+B)*(EXP(ALFE*20.)-1.)*(EXP(ALFE*120.)-EXP(ALFE
1*20.)))
58 F2(7)=FLUX*((1.-B)*(EXP(ALFE*20.)-1.)*(EXP(ALFE*120.)-EXP(ALFE
1*20.)))
59 DO 700 J=2,7
60 Q1(J)=Q1(J)*ACTOX
61 Q2(J)=Q2(J)*ACTOX
62 F1(J)=F1(J)*ACTFE
63 F2(J)=F2(J)*ACTFE
64 700 CONTINUE
65 PRINT 1800
66 PRINT 1900
67 PRINT 1900
68 PRINT 1900
69 PRINT 1630
70 PRINT 1001
71 PRINT 1900
72 PRINT 1000
73 PRINT 1900
74 PRINT 1100
75 PRINT 1630
76 PRINT 1200, Q1
77 PRINT 1300, (F2(J), J=2, 7)
78 PRINT 1400, F1
79 PRINT 1300, (F2(J), J=2, 7)
80 PRINT 1900
81 PRINT 1600
82 PRINT 1900
83 PRINT 1610
84 PRINT 1620
85 PRINT 1630
C
C *** THEOR. SAMPLES WITH NEUTRON FLUENCE CORRECTION ***
C
86 DO 801 J=1,7
87 L(J)=J
88 801 PATIO(J)=Q1(J)/F1(J)

```

```

89      DO 802 J=2,7
90      L(5+J)=6+J
91      802 RATIO(5+J)=O2(J)/F2(J)
92      PRINT 1540,(L(N),N,N,RATIO(N),N=1,4)
93      PRINT 1652,(L(N),N,N,RATIO(N),N=5,7)
94      PRINT 1653,(L(N),N-6,N-6,RATIO(N),N=8,11)
95      PRINT 1654,(L(N),N-6,N-6,RATIO(N),N=12,13)
96      PRINT 1900
C      ***
C      *** THEOR, "MOLE RATIO" ***
C      ***
97      PRINT 1800
98      PRINT 1900
99      PRINT 1700
100     PRINT 1701
101     PRINT 1900
102     PRINT 1720
103     PRINT 1630
104     K=1
105     DO 860 I=1,13
106     DO 850 J=1,13
107     IF(J.EQ.1)GO TO 850
108     FOUND=RATIO(I)/RATIO(J)
109     ERROR=(1.-FOUND)*100.
110     IF(K.EQ.45)GO TO 900
111     IF(K.EQ.87)GO TO 900
112     IF(K.EQ.129)GO TO 900
113     810 PRINT 1750,I,J,FOUND,ERROR
114     K=K+1
115     GO TO 850
116     900 PRINT 1800
117     PRINT 1900
118     PRINT 1900
119     PRINT 2000
120     PRINT 1630
121     GO TO 810
122     850 CONTINUE
123     860 CONTINUE
124     PRINT 1800
125     1000 FORMAT(1X,'PART A: THEORETICAL ACTIVITY AT END OF IRRADIATION')
126     1001 FORMAT(12X,12X,'TABLE 5.9 RESULTS BY ASSUMING WAVE FUNCTION ',
127     '1'BEHAVIOR FOR THE NEUTRON FLUX')
127     1100 FORMAT(6X,'MODE',3X,'FLUX TYPE 1',6X,'TYPE 2',10X,'TYPE 3',
128     '110X','TYPE 4',10X,'TYPE 5',10X,'TYPE 6',10X,'TYPE 7')
128     1200 FORMAT(1X,'OXYG.',2X,'A',7(2X,E14.6))
129     1300 FORMAT(8X,'U',16X,6(2X,E14.6))
130     1400 FORMAT(1X,'IRON ',2X,'A',7(2X,E14.6))
131     1600 FORMAT('0','PART B: ACTIVITY / GM FOR "OXYGEN" SAMPLES',
132     '1' NORMALIZED FOR THE NEUTRON FLUENCE')
132     1610 FORMAT(1X,'RATIO',4(2X,'SAMPLE',2X,'RATIO',15X))
133     1620 FORMAT(1X,'MODE',1X,4(3X,'U',4X,'TYPE',4X,'ACTIVITY',4X))
134     1630 FORMAT(' ')
135     1640 FORMAT(1X,'A:A',4(6X,12,4X,11,' ',11,1X,E14.6))
136     1652 FORMAT(4X,3(6X,12,4X,11,' ',11,1X,E14.6))
137     1653 FORMAT(1X,'B:B',4(6X,12,4X,11,' ',11,1X,E14.6))
138     1654 FORMAT(4X,2(6X,12,4X,11,' ',11,1X,E14.6))
139     1700 FORMAT(3X,'TABLE 5.10 THEORETICAL MOLE RATIO FOR SAMPLE(1)',
140     '1' TO SAMPLE (J).')
140     1701 FORMAT(15X,'EXPECTED VALUE IS 1:1')
141     1720 FORMAT(14X,'1 : J',14X,'RATIO',14X,'% ERROR')

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```

142 1750 FORMAT(IX,4X,I2,2X,':',2X,I2,9X,F9.6,11X,F9.3)
143 1800 FORMAT('11')
144 1900 FORMAT('1-1')
145 2000 FORMAT('1',3X,'TABLE 5.10 (CONTINUED)')
146 STOP
147 END

148 SUBROUTINE SUM1(X,Y,FA,FB)
149 DIMENSION T(10)
150 FLUX=1.8E12
151 T(1)=0.
152 FA=FB=0.0
153 DO 100 I=2,7
154 T(I)=T(I-1)+20
155 Z=FLUX*(EXP(Y*T(I))-EXP(Y*T(I-1)))
156 IF((I.EQ.3).OR.(I.EQ.5).OR.(I.EQ.7))GO TO 50
157 FA=FA+Z*(1.+X)
158 FB=FB+Z*(1.-X)
159 GO TO 100
160 50 FA=FA+Z*(1.-X)
161 FB=FB+Z*(1.+X)
162 100 CONTINUE
163 RETURN
164 END

165 SUBROUTINE SUM2(X,Y,SV,FA,FB)
166 DIMENSION T(10)
167 FLUX=1.8E12
168 T(1)=0.
169 FA=FB=0.0
170 DO 100 I=2,7
171 T(I)=T(I-1)+20
172 Z=FLUX*(EXP(Y*T(I))-EXP(Y*T(I-1)))
173 W=(SV*(T(I)+T(I-1)))/2.+X
174 IF((I.EQ.3).OR.(I.EQ.5).OR.(I.EQ.7))GO TO 50
175 FA=FA+Z*(1.+W)
176 FB=FB+Z*(1.-W)
177 GO TO 100
178 50 FA=FA+Z*(1.-W)
179 FB=FB+Z*(1.+W)
180 100 CONTINUE
181 RETURN
182 END

```

ENTRY

APPENDIX G

Printouts for Nova Computer Programs

```

00050 102440 BEGIN:  SUB# 0,0
00051 042075 STA 0,0ZTIME
00052 020076 LDA 0,ASK
00053 040020 STA 0,20 ;AUTO INCREMENT
00054 022020 LDA 0,020,0
00055 006173 JSR 0,TYPE
00056 024147 LDA 1,EQUAL
00057 106414 SUB# 0,1,SZB ;TYPED "="?
00060 000054 JMP 0,4 ;NO.
00061 126440 AC1: SUB# 1,1
00062 006174 AC2: JSR 0,GET ;GET #
00063 006173 JSR 0,TYPE ;ECHO #
00064 034203 LDA 3,CR
00065 116415 SUB# 0,3,SNR ;END?
00066 000204 JMP 0,FFOT ;YES
00067 034150 LDA 3,060 ;"0"
00070 030151 LDA 2,071 ;"9"
00071 142033 ADCZ# 2,0,SNR ;SKIP IF > 9
00072 116032 ADCZ# 0,3,SZC ;SKIP IF > OR = 0
00073 000165 JMP 0,ERRC ;NO DIGIT OR CR.
00074 000152 JMP 0,FYES
00075 000176 ZTIME: TIME
00076 000077 ASK: 0,ASK
00077 000000 .ASK: 0
00100 000124 "T
00101 000111 "I
00102 000115 "M
00103 000105 "E
00104 000040 40
00105 000111 "I
00106 000116 "N
00107 000040 40
00110 000123 "S
00111 000105 "E
00112 000103 "C
00113 000012 12
00114 000015 15
00115 000050 "C
00116 000105 "E
00117 000116 "N
00120 000104 "D
00121 000040 40
00122 000127 "W
00123 000110 "H
00124 000111 "I
00125 000124 "T
00126 000040 40
00127 000103 "C
00130 000101 "A
00131 000122 "R
00132 000122 "R
00133 000111 "E
00134 000101 "A
00135 000107 "G
00136 000105 "E
00137 000040 40
00140 000122 "R
00141 000105 "E
00142 000124 "T

```

```

00144 000122 "R
00145 000116 "N
00146 000051 "D
00147 000075 EQUAL:  "="
00150 000060 C60:    60
00151 000071 C71:    71
00152 034150 FYES:   LDA      3,C60
00153 162400         SUB      3,0      ;OUT ASCII CODE
00154 152440         SUBO     2,2
00155 034172         LDA      3,LF      ;= 10
00156 054201         STA      3,C7
00157 133000         ADD      1,2      ;CONVERT TO BINARY
00160 014201         DSZ      C7       ;# IN "DECIMAL " POSITION
00161 000157         JMP      .-2
00162 113000         ADD      0,2
00163 145000         MOV      2,1
00164 000062         JMP      AC2      ;NEXT NUMBER
00165 020202 ERROC:   LDA      0,WHAT
00166 006173         JSR      0,TYPE   ; "?"
00167 020172         LDA      0,LF
00170 006173         JSR      0,TYPE
00171 000061         JMP      AC1      ;GET TIME AGAIN
00172 000012 LF:     12
00173 000346 .TYPE:   TYPE
00174 000356 .GET:    GET
00175 177775 THREE:  -3
00176 000000 TIME:    0
00177 000007 BELL:    7
00200 000000 TIME1:   0
00201 000012 C7:      12
00202 000077 WHAT:    "?"
00203 000015 CR:      15
00204 034172 OFFOT:   LDA      3,LF
00205 054201         STA      3,C7
00206 152440         SUBO     2,2
00207 133000         ADD      1,2      ;MULTIPLY BY TEN
00210 014201         DSZ      C7
00211 000207         JMP      .-2
00212 050176         STA      2,TIME   ;COUNTING TIME READY
00213 020177 START:   LDA      0,BELL
00214 030175         LDA      2,THREE
00215 006173         JSR      0,TYPE
00216 151404 INC       2,2,SZR ;RING BELL 3 TIMES
00217 000215         JMP      .-2
00220 020176         LDA      0,TIME
00221 040200         STA      0,TIME1
00222 176520 SUBZL     3,3
00223 060231 NIOS CNTRL
00224 076031 DOP      3,CNTRL
00225 060131 NIOS     CNTRL
00226 063631 SKPEN    CNTRL
00227 000225         JMP      .-2
00230 152440         SUBO     2,2
00231 102440         SUBO     0,0
00232 163000 CLEAR:   ADD      3,0      ;CLEAR NS 636 MEMORY
00233 063030 DOP      0,DAC
00234 071030 DCA      2,DAC
00235 060130 NIOS     DAC
00236 063530 SKPEZ    DAC

```

```

00240 014341 DSZ      CHANN      ;=4096
00241 000232 JMP      CLEAR
00242 060231 NI OC    CNTRL
00243 020342 LDA      0, CHANL
00244 040341 STA      0, CHANN
00245 102440 SUBC     0, 0
00246 105400 INC      0, 1
00247 062031 DOP      0, CNTRL
00250 067031 DOP      1, CNTRL
00251 065014 DOA      1, RTC
00252 063731 SKPDZ    CNTRL      ;SKIP NEXT INSTRUCTION UNTIL AN EXTERNAL
00253 000255 JMP      .+2        ;STARTS THE NS-636 ACCUMULATING
00254 000252 JMP      .-2
00255 060114 NIOS     RTC        ;SIGNAL HAS ARRIVED
00256 063614 SKPDN    RTC
00257 000256 JMP      .-1
00260 014200 DSZ      TIMEI      ;DECREMENT TIME
00261 000255 JMP      .-4
00262 060231 NI OC    CNTRL
00263 060077 NIO      CPU
00264 060077 NIO      CPU
00265 063632 SKPDN    MISS       ;MAG. TAPE AVAILABLE?
00266 000265 JMP      .-1
00267 060333 GAP:     NIOP       MPER      ;WRITE FILE GAP
00270 063632 SKPDN    MISS
00271 000270 JMP      .-1
00272 030344 LDA      2, INIT    ;FIRST TAGWORD
00273 102520 SUBZL     0, 0
00274 024337 LDA      1, TAG8E
00275 062031 DOP      0, CNTRL
00276 063531 SKPBZ    CNTRL
00277 000276 JMP      .-1
00300 071030 DOA      2, DAC
00301 066030 DOP      1, DAC
00302 060077 NIO      CPU
00303 060077 NIO      CPU
00304 060231 NI OC    CNTRL
00305 060077 NIO      CPU
00306 020343 LDA      0, MSGOUT
00307 062131 DOPS     0, CNTRL
00310 063531 SKPBZ    CNTRL
00311 000310 JMP      .-1
00312 060131 NIOS     CNTRL
00313 063631 SKPDN    CNTRL
00314 000312 JMP      .-2
00315 063731 SKPDZ    CNTRL
00316 000315 JMP      .-1
00317 060231 NI OC    CNTRL
00320 060077 NIO      CPU
00321 060222 NI OC    NSMG
00322 060077 NIO      CPU
00323 020344 LDA      0, INET
00324 024340 LDA      1, ENDTG   ;=170000
00325 106415 SUB#     0, 1, SNR
00326 000333 JMP      BIGTW
00327 024345 LDA      1, TIP
00330 107000 ADD      0, 1
00331 044344 STA      1, INIT
00332 000213 JMP      START      ;NEXT SPECTRUM

```

```

00334 102440 SUBO      0,0
00335 040344 STA      0,INIT
00336 000213 JMP      START
00337 000000 TAGS1:   0
00340 170000 ENDTG:   170000
          000031 CNTRL=31
          000030 DAC=30
          000022 NSMG=22
          000032 MISS=32
          000033 MPER=33
00341 010000 CHANN:   10000
00342 010000 CHANL:   10000
00343 000005 MGCUT:   5
00344 010000 INIT:    10000 ;SYMBOLIC 1 (FIRST TAGWORD)
00345 010000 TIP:     10000 ;SAME
00346 054354 TYPE:    STA    3,EXIT1
00347 063511 SKPBZ    TTO
00350 000347 JMP      .-1
00351 061111 DOAS     0,TTO
00352 002354 JMP      *EXIT1
00353 000177 MASK1:   177
00354 000000 EXIT1:   0
00355 000000 EXIT3:   0
00356 054355 GET:     STA    3,EXIT3
00357 060110 NIOS     TTI
00360 063610 SKPDN    TTI
00361 000360 JMP      .-1
00362 060610 DIAC     0,TTI
00363 034353 LDA      3,MASK1
00364 163400 AND      3,0
00365 002355 JMP      *EXIT3
          000050      .END    BEGIN

```


AC2	000062
ASK	000076
BEGIN	000050
BELL	000177
BIGTW	000333
C60	000150
C7	000201
C71	000151
CHANL	000342
CHANN	000341
CLEAR	000232
CNTRL	000031
CR	000203
DAC	000030
ENDTG	000340
EQUAL	000147
ERROC	000165
EXIT1	000354
EXIT3	000355
EYES	000152
GAP	000267
GET	000356
INIT	000344
LF	000172
MASK1	000353
MCOUT	000343
MISS	000032
MPER	000033
NSMG	000022
OFFOT	000204
START	000213
TAG81	000337
THREE	000175
TIME	000176
TIME1	000200
TIP	000345
TYPE	000346
WHAT	000202
ZTIME	000075
.ASK	000077
.GET	000174
.TYPE	000173

APPENDIX H

"A Nova Computer Code to Determine the Concentration of Calcium in a Sample Activated by Thermal Neutrons."

"This work originally included an On-line activation analysis using the Nova minicomputer. This work is presented here because it represents an approximately six months of work. Unfortunately, it was not possible to finalize due to the limitations of programming complex operation procedures in a minicomputer like the Nova. We strongly suggest that the Nova computer has to be used only for the purpose of controlling peripheral equipment. Therefore, we discourage any work directed to development of a complex program like the one presented here."

Abstract

Determination of calcium by neutron activation analysis has been successfully used by the Nuclear Engineering Department at Kansas State University. A computer code has been developed for Nova computers which may improve direct calculations when calcium is determined via the comparator method. Therefore, this work results in a useful tool for rapid and efficient analysis of such a mentioned element.

Important capabilities of this code are versatility (it can handle spectra from magnetic tape or accumulate spectra by the multichannel analyzer) and simplicity (only requires little information for adequate use). Also, the user has options of selecting pre-programmed data of reference samples and flux monitors and using other information to calculate the concentration of other elements if desired.

The main limitation of the code is in the analysis of a spectrum. Only one photopeak, which represents the element of interest, can be analyzed. Complex spectra cannot, therefore, be analyzed if more than one peak analysis is desired. If other than calcium is desired, the photopeak has to be defined before starting the analysis and results can be limited by the presence of high background.

1. Introduction

This code has been designed for use with the Nova line of computers which are equipped with a real time clock, teletype, and interfaced with the Northern Scientific NS440 Computer NS 646 series memory unit, and NS408 magnetic tape control. It is written in an absolute mode and must be used along with the extended floating point interpreter (tape #091-0000-13-07) (31,32,33,34).

The code itself has three important parts. The first gives instructions for using the program and for requesting pertinent data. The second part calibrates the multichannel analyzer. The last part performs all the calculations, finds the desired concentration, and prints the results.

Although neutron activation analysis is a well known technique, it is necessary to give some background for completion in developing the code. Detailed information is found in references 9 and 10.

Neutron activation analysis is a procedure in which a sample of unknown composition is exposed to a flux of neutrons. The neutron flux causes a small fraction of atoms of the sample to become radioactive through neutron absorption and nuclear transmutation. The radiation, e.g., gamma-rays, from the transmuted nuclei is detected by a high resolution spectroscopy system, e.g., Ge(Li) system, capable of discerning between the different energies of the radiation.

The area under characteristic energy peaks of the spectrum are directly proportional to the weight of the elements from which the radiation is produced. In the "Comparator Method" the area from the unknown sample is compared to that from a peak produced in a reference sample where the composition is known; thus, the unknown composition can be calculated.

Then, in this method sample and reference were treated identically, allowing the equalization of several common parameters, e.g., cross section, even if these parameters are not precisely known.

Thus, the number of target atoms of the unknown is given by (see Appendix A)

$$N(\text{Unknown}) = N(\text{Known}) \frac{\phi(\text{Known})}{\phi(\text{Unknown})} \frac{A_o(\text{Unknown})}{A_o(\text{Known})} , \quad (\text{H.1})$$

where A_0 is the production rate (activity/gram) and is determined:

$$A_0 (\text{activity/gram}) = \frac{C\lambda}{f_m} [(1-e^{-\lambda t_1})e^{-\lambda t_2} (1-e^{-\lambda t_3})^{-1}], \quad (\text{H.2})$$

ϕ , the neutron flux, is monitored by elements which characteristics are well-known and is given by

$$\phi = \frac{A_0}{N_0 \sigma}, \quad (\text{H.3})$$

λ = decay constant (for desired element and for flux monitor),

f = overall efficiency of the counting system,

m = mass of sample (sample, reference, and flux monitors),

t_1 = irradiation time,

t_2 = cooling time (or transfer time),

t_3 = counting time,

C = true area under photopeak of interest,

N_0 = atom density for flux monitor,

σ = microscopic cross section for flux monitor.

Because in the comparator method equal conditions of sample and reference must be carefully maintained, flux monitors are required to allow the canceling of N_0 and σ when substituted into Eq. (H.1), leaving the rest of the parameters to be measured easily and used in the code. It has been estimated that the accuracy of results will be as precise as C , the area under photopeak, could be determined. In general, the mass and times are simple to measure with good precision.

2. Code Description

The computer code can be separated into three parts: 1. instructions, 2. calibration, and 3. calculations.

The entire program requires 3500_{10}^* storage locations, and together with the floating point interpreter occupies over 5000_{10} locations. This limits its use with other program which may require substantial storage.

2.1 Instructions

All operating instructions that are to be required for use in the code are preprogrammed. In this part of the computer code several data are requested, as well as, options of using peripheral equipment according to the wishes of the user. Thus, it could be decided to use the magnetic tape unit or to couple directly to the analyzer. Also pre-programmed data, useful to calculate calcium concentrations available as well as the type of flux monitor (pre-programmed data for Fe and Cu are available) and reference (data for CaCO_3 and CaO). Also the code allows the change of pre-stored data to calculate concentration of any other element than calcium. Main purpose of this part of the code is to type messages (instructions) and accept characters (data or decisions); and it represents the relation between the computer and user.

This part is the simplest of the entire program, however it has the disadvantage of occupying a large part of the storage of the Nova memory, due principally to the large number of characters that a message - to be printed out - could contain. Normally, two characters (maximum) can be stored in one memory location (31,32). Although a special subroutine (TYPE 2) was developed in order to allow up to three characters per memory location, while the messages were reduced to the minimum to be understandable, they still use a large part of useful memory locations (about 600_{10}).

*The Nova memory capacity is often given in octal notation. Here we use decimal notation.

3.2 Calibration

Because the identification of a desired element (isotope) is based on the location of a gamma-ray photopeak of a specific energy in a given spectrum, this part of the code is of real importance, and together with the true calculation of the area under the photopeak, it is formed by most of the subroutines that were developed for the code.

The steps to calibrate any multichannel analyzer are the following:

a) set the spectroscopy system at the desired, gain and specify the other characteristics, b) place on the detector a calibrated source, c) accumulate a calibration spectrum, d) assign a channel to each photopeak of known energies (Kev/channel) assuming a linear relation between channel and energy, e) check the desired energy of the photopeak which represents the element being analyzed, f) correct the system gain and/or change parameters if required. Facilities of Nova computer allow one to pre-program steps "c" through "e", leaving the remaining steps (a and b) to be performed by the user.

Since Co-60 is widely used as a calibration source, it is suggested that it be used; however, there is an option for other sources to be used as desired. Pertinent data are requested for up to five energies. The only limitations are that some of the photopeaks are not easily differentiated from background, or if they are not separated by at least twelve channels (this value was arbitrarily chosen by experimentation).

The program includes a scan of the spectrum data (9) to determine peaks over 12 channels and evaluate them as photopeaks. Several runs, simulating the Nova code by a Fortran program (on the IBM 370/158), were made in order to determine the best method to scan such a spectrum keeping as the only condition the elimination of the Compton edge and "small" peaks

which do not represent the very well defined photopeaks of standard sources.

For a peak with a maximum in channel n the condition to be satisfied is:

$$C_{n-1} < C_n > C_{n+1} , \quad (H.4)$$

where C_i is the counts in channel i .

The peak is retained as a true photopeak if

$$\sigma_s < S/2 , \quad (H.5)$$

where σ_s is the standard deviation of area S , after subtraction of the background, which is calculated by (see Appendix B)

$$S = \sum_{h=n-4}^{n+3} C_h - \frac{9}{6} \left[\sum_{k=n-6}^{n-4} C_k + \sum_{\ell=n+3}^{n+5} C_\ell \right] , \quad (H.6)$$

and the standard deviation is calculated by

$$\sigma_s = \left[\sum_{h=n-4}^{n+3} C_h + \frac{81}{36} \left(\sum_{k=n-6}^{n-4} C_k + \sum_{\ell=n+3}^{n+5} C_\ell \right) \right]^{1/2} . \quad (H.7)$$

The area, S , was arbitrarily chosen to satisfy the requirements of the code. It is important to note that the area under photopeak used to perform calculations and to find the desired results is not calculated by the same equation, therefore, Eqs. (H.6) and (H.7) were used only to scan photopeaks.

Once the photopeaks have been found, the calibration is made assuming a linear relation between energy and channel. If a calibration source with only two photopeaks was used, the basic equation for a straight line is used to compute the slope and intercept. These are defined by

$$M = \frac{y_1 - y_2}{x_1 - x_2} , \quad (H.8)$$

and

$$B = y_1 - Mx_1 . \quad (H.9)$$

where x means channel number and y the corresponding energy.

If more than two photopeaks appear the data are fit to a straight line by the least squares method:

$$M = \frac{N \sum_{i=1}^N x_i y_i - (\sum_{i=1}^N x_i)(\sum_{i=1}^N y_i)}{N \sum_{i=1}^N x_i^2 - (\sum_{i=1}^N x_i)^2} , \quad (H.10)$$

and

$$B = \frac{1}{N} \sum_{i=1}^N y_i - \frac{1}{N} (\sum_{i=1}^N x_i) M , \quad (H.11)$$

The expected channel for the desired photopeak energy of the element is checked to determine if the photopeak lies within the total channels of the analyzer. If an "out of range" is obtained, a message is printed out suggesting that the parameters be changed and the calibration is performed again.

The procedure followed that described above is not absolute; and it has, probably, many limitations if it is to be used for more sophisticated calculations. However, it satisfies the necessities of the problem attempted in this work. The limitations are due principally to the limitations of the Nova computer. This code is only one part, but an important part, of the entire code.

3.3 Calculations

Once the system has been calibrated, operations can be performed to compute the concentration of the desired element in the unknown sample in a simple way, since all constants, but S are easily determined. The accuracy

the photopeak area has been determined to consist of the way in which S the true area under photopeak may be found.

Several methods can be used to calculate S (see Appendix B). The one used in this code is Gregory's formula (13):

$$\begin{aligned} \text{Area} = & h \left(\frac{1}{2} C_0 + C_1 + C_2 + C_3 + C_4 + \frac{1}{2} C_5 \right) - \frac{h}{12} (\Delta C_4 - \Delta C_0) \\ & - \frac{h}{24} (\Delta^2 C_3 + \Delta^2 C_0) - \frac{19h}{720} (\Delta^3 C_2 - \Delta^3 C_0) - \frac{3h}{160} (\Delta^4 C_1 + \Delta^4 C_0), \quad (\text{H.12}) \end{aligned}$$

where h is the separation between a particular reference point (point 1 in this case) and the Δ 's are defined in Appendix B.1.

The basis for use of such an equation is given in Appendix B. It is assumed that the area calculated with the six points around the maximum, inclusive, is proportional to the entire area of the photopeak. The standard deviation of such an area is calculated by the square root of the calculated area value, since the area represents "a count rate" from a radioactive source. Thus the standard deviation of the desired concentration is given by

$$\sigma_c = \text{Concentration} \begin{array}{c} \text{(PPM)} \\ \text{or} \\ \text{(GRMS)} \end{array} \frac{\sigma_a^2(u)}{A^2(u)} + \frac{\sigma_A^2(\text{Feu})}{A^2(\text{Feu})} + \frac{\sigma_A^2(\text{R})}{A^2(\text{R})} + \frac{\sigma_A^2(\text{FeR})}{A^2(\text{FeR})}, \quad (\text{H.13})$$

where A is the area under peak, referred to as the unknown (u), reference (R), flux monitor of unknown (Feu), and flux monitor of reference (FeR). Concentration is the calculated value in PPM or grams.

Equations H.1, H.2 and H.2 which are used to calculate the desired concentration were used precisely, i.e., no approximations were assumed, in order to give more versatility to the code. For example, some samples may require more counting time than others to accumulate adequate statistics, but the main reason is due to the inherent approximation of the Floating Point Interpreter

which was used to perform the complex operations. In this way if a systematic error results, it can easily be corrected.

3.4 Constants

The pre-programmed constants available to the user are given in Table H.1.

Table H.1 Constants used through the Nova Code.

Element	Object	Gamma Energy	Half Life
Co-60	Source calibr.	1.117 MeV 1.3 MeV	-- --
Fe ⁵⁶	Flux monitor	0.847 MeV (95%)	2.582 hrs
Cu	Flux monitor	1.348 MeV (0.5%)	12.74 hrs
CaCO ₃ *	Reference	3.0842 MeV (89%)	8.72 min
CaO**	Reference	3.0842 MeV (89%)	8.72 min

*Concentration of Ca = 0.4004

**Concentration (Ca) = .7147

4. Program Description

This section contains a description of each subroutine used in the computer code.

4.1 FIRST

- a - Instructions:
 - i) ask if calcium concentration is to be determined
 - ii) if NO, provide data for other elements
- b - Data:
 - i) energy of peak which represents desired isotope
 - ii) half life of same isotope
- c - Perform: i) substitute data into normal locations to perform calculations
- d - Critical location in this subroutine is TH11S which can be:
 - 1 = Calcium is desired
 - 2 = Other than calcium

4.2 USER (Main Subroutine)

- a - Instructions for use:
 - i) type of flux monitor
 - ii) type of reference sample
 - iii) magnetic tape or NS636
- b - Data requested:
 - i) masses
 - ii) irradiation times
 - iii) total number of channels
- c - Perform:
 - i) call CALIB to calibrate NS636 system
 - ii) call MTAPE or NS636 as selected
 - iii) call FIND for calculations
 - iv) call PRINT to given results
 - v) ask if there are more samples to analyze

4.3 TYPE1

Allow the typing of characters and/or letters, given in a special code number of stored three letters in one memory location.

4.4 TYPE

Type characters given into accumulator Ø via teletype.

4.6 TYPE2

Allow the typing of characters, other than letters, stored by pairs in one memory location.

4.7 PUTZZ

When data prestored are used, clear locations reserved for data not pre-stored.

4.8 WRITE

Subroutine to decide if the message to be printed is:

- a) letters, call TYPE1
- b) others, call TYPE2
- c) when message to start analyzer is printed out, transfer the control to user

4.9 ACCEPT

- a) get decision of user about features of code. Break character is carriage return
- b) integer number are also accepted and converted to binary integer
- c) determines if the character is a digit; if not, it types out "??" and waits

4.10 GET1

- a) store data in Locations called NUMB
- b) accept real data (numerical values) using the facilities of the Floating Point Interpreter (FPI)
- c) data requires confirmation, as in ACCEPT, to allow corrections
- d) format of data is given in general instructions of FPI user's manual (37)

4.11 TTYIN

Subroutine to get characters via teletype.

4.12 CALIB

- a - Instructions: i) type of source used
 - ii) if not Co-60 ask for data
- b - Data: i) energies of photopeaks, in order minimum to maximum
- c - Finish with message "calibration done" if all is correct
- d - Perform: i) calculate channel of centroid of photopeak of desired element
 - ii) check amplification (number of channels) in subroutine GIVE

4.13 PEAKS

- a - Locate photopeaks of a spectrum
- b - Uses subroutine AREA to perform step a
- c - Region of interest is given in MIN = minimum channel and MAX = last channel

4.14 EXTER

Set NS636 analyzer in external mode (readout).

4.15 SUM

- a) Set NS636 analyzer to desired channel
- b) Set DAC to desired channel and get contents of channel of NS636 analyzer
- c) Add contents of channel in a desired region by steps a and b.

4.16 AREA

- a) Calculate the photopeak area S after subtracting of background by using Eq. H.6.
- b) Neglect peak if area is zero or negative.
- c) Accept peak if $S/2$ is less than standard deviation of S .

4.17 GIVE

- a) Assign each peak a given energy as in subroutine CALIB.
- b) If Co-60 is used to following equation applies $y = mx+b$.
- c) If more than two peaks, fit to a straight line by the least squares method.
- d) Either case slope and intercept are calculated.
- e) Check amplification used. If wrong, a message is printed out and the control is transferred to start CALIB subroutine.

4.18 MTAPE

- a) Control of magnetic tape unit.
- b) Instructions.
- c) Data: i) counting times
 ii) tagwords of recorded spectra.
- d) Locate peaks and calculate true area by calling subroutine LOCTE.
- e) If a given tagword is not found, print message to transfer data, from tape to NS636 memory, manually by user.

4.19 CHPK

Calculate the centroid of photopeak for the selected flux monitor data.

4.20 ENROL

Rewinds magnetic tape.

4.21 NS636

Control of multichannel analyzer in ADC input mode and ADD mode.

Cycles of 0.1 sec are used by setting the real time clock to a time stored in accumulator zero.

4.22 LOCTE

- a) Locate photopeaks by calling subroutine PEAKS.
- b) Calculate area of photopeak by calling subroutine AREAL (true area).
- c) If no peak was located (low activity in sample) area is calculated by using a centroid calculated by the slope and the intercept of a linear relationship between channel and energy.
- d) If more than one peak is found the one closest to the true energy is accepted for the area determination procedure. (Region of interest is about twenty channels therefore, if more than one peak is present in such region an error could result).

4.23 PRINT

Print final results: i) concentration and standard deviation, in PPM and grams.

4.24 (Reserved locations for the floating point interpreter.)

4.25 AREAL

- a) Calculate the true photopeak area by Gregory's formula for a given photopeak. The centroid is stored in accumulator zero.

4.26 (Reserved locations for data requested.)

4.27 FIND

- a) Calculate the concentration of desired isotope in parts per million and in grams.
- b) Get data in accordance with instructions given in previous subroutines.

4.28 ERRSM

Accumulate standard deviations of calculated data (Propagation of errors).

4.29 WHICH

Determines the fraction of calcium or desired isotope according with reference used.

4.30 CONSC

Convert times to seconds. Times given in hours, minutes and seconds.

5. Results and Conclusions

Results can not be presented because we were unable to finish the Nova Code. The major reason is the basic sequence of operations normally used to program a minicomputer like the Nova. To illustrate better the problems, we refer to Figure H.5.1 which is a basic programming flow-chart. Thus, a source program is produced first on paper tape by using the Tape Editor (Tape number 091-000001-07); second, the Extended Paper Tape assembler (Tape # 091-000017-07) is used to translate the source program into machine language, resulting into the another tape (the object tape); third, the program is tested for symbolic errors, and if any error is found, the correction of it forces one to repeat the first two steps again. If no errors are detected, a listing and a binary tape are obtained.

All the basic steps mentioned above require the use of the Teletype. This machine is a slow device; therefore, debugging the program results in ten or more continuous hours for each correction. Figure H.5.1 we indicate that a loading of a Debug Tape must be performed. This tape allows correction of errors without repeating the two steps described above. Unfortunately, the Debug Tape requires a considerable memory (1000_{10} locations) which includes some locations to be reserved specifically, and our program was large enough that the memory capacity of the Nova (8k) was not sufficient to allow its storage and that of the Floating Point Interpreter (FPI). Therefore, such a debugging procedure was not available in our case and we had to test the program by repeating all the steps.

The use of the FPI has also complicated the finalization of the code. The FPI is very important in performing calculations throughout the code. The flow chart in Figure H.5.2 represents the Nova Code which is being developed. The explicit problem in the use of the FPI is mainly that any error in the

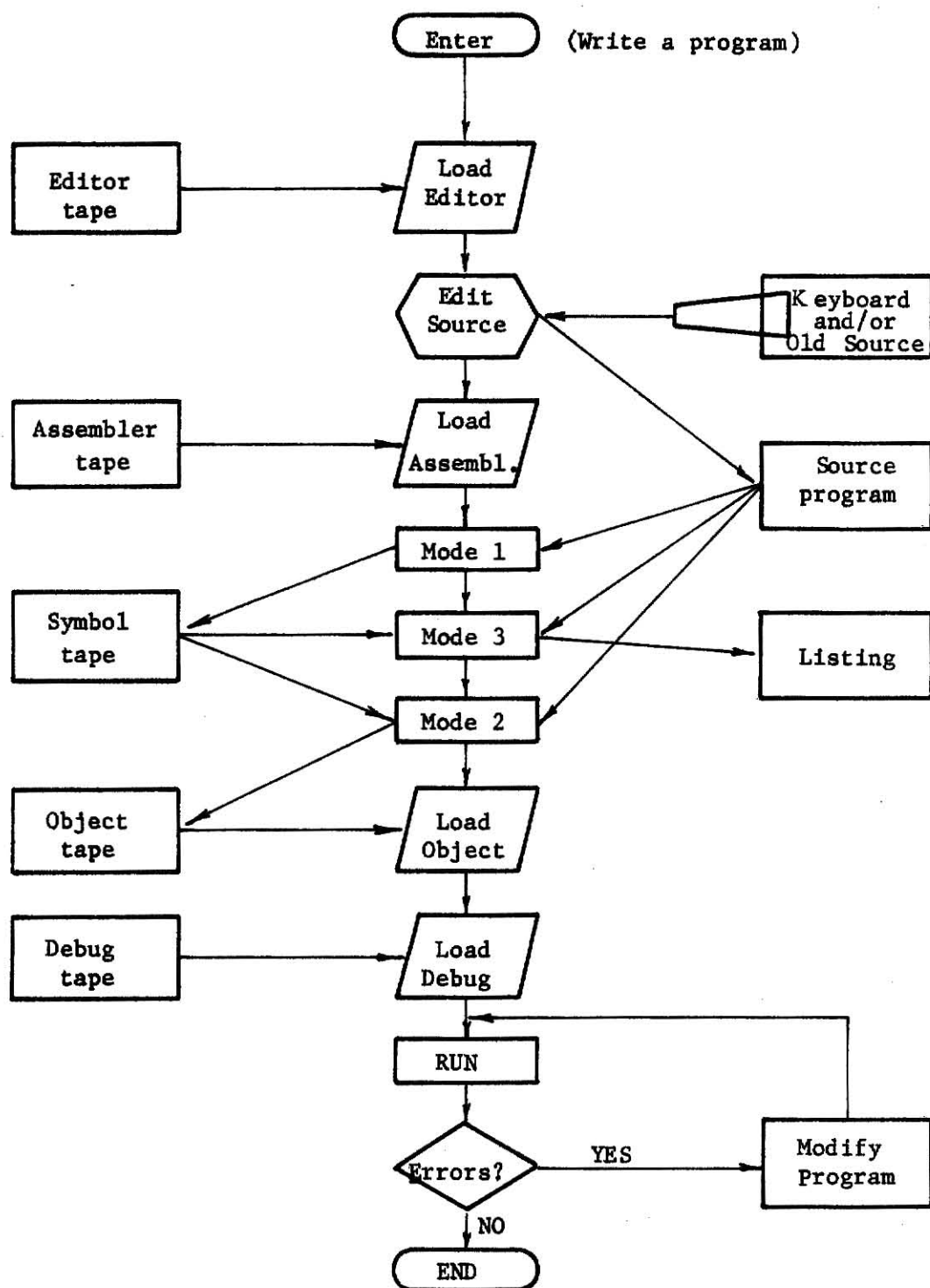


Fig. H.5.1. Nova Basic Programming Flowchart.

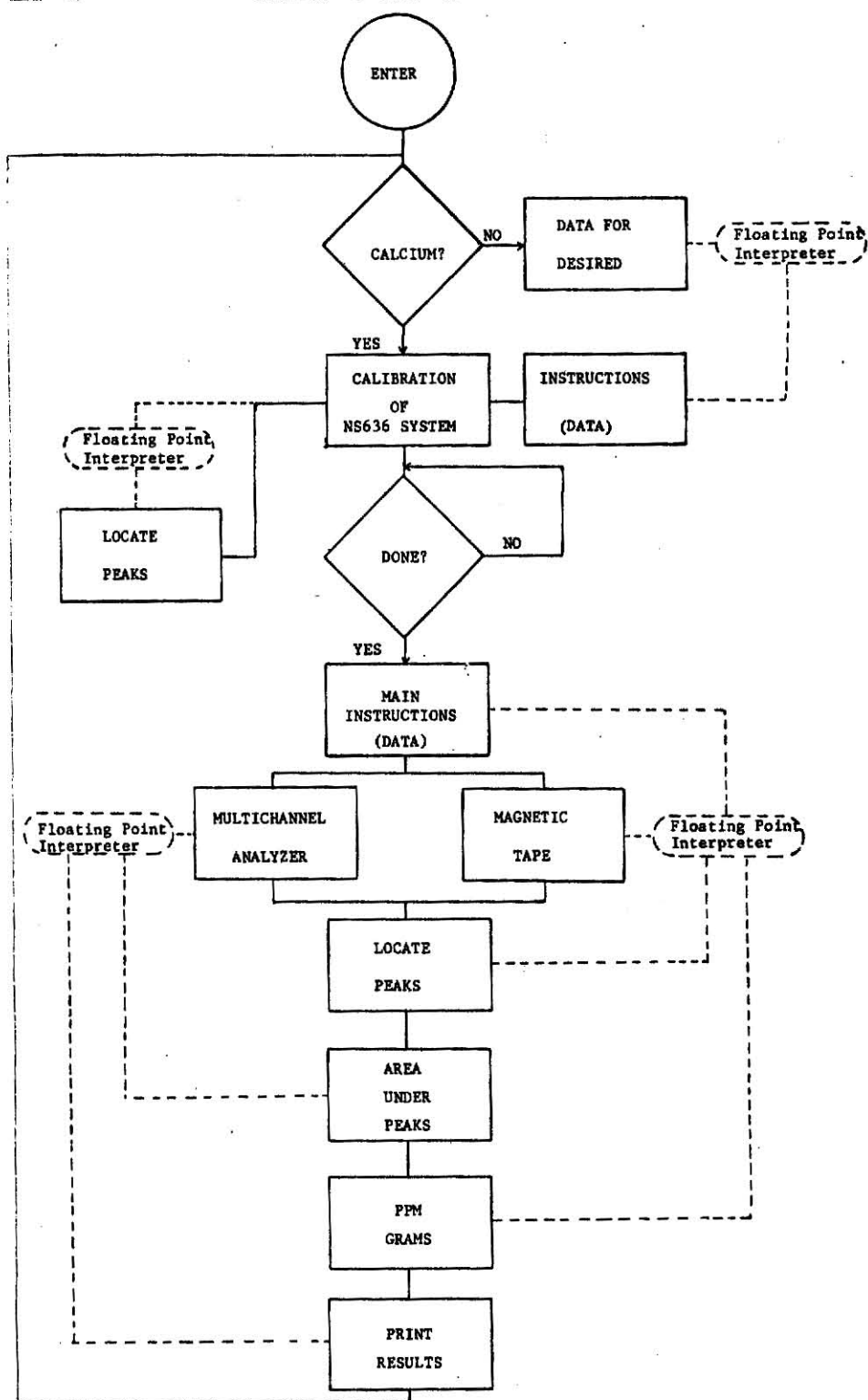


Figure H.5.2 Computer Code Operational Diagram.

code, when the FPI is used, can produce unpredictable loops that destroy the contents of other parts of the Nova Code, or in best situation, the error forces the FPI to stop as an indication of an improper operation. In either case we do not have the facility to determine the location of the error, or the magnitude of the error. Also, most of the subroutines that use the FPI cannot be tested individually, because some operations require jumping from one subroutine to a second and even to a third. The result requires a guess to modify and correct errors. This "method" is time consuming and complicated by the fact that no-obvious errors are not easy to detect.

Four months were devoted to debugging our Nova code. After this period of time we concluded that the Nova Computer system is an excellent device to control peripheral equipment, e.g., the NS636 Multichannel Analyzer, the NS408 Tap unit control, but complex operations which result in a large program are not useful.

We suggest the Nova minicomputer should be used only for the purpose of controlling. In fact, it was successfully used in the oxygen-18 determination part of this work. See Appendix F and also reference 35.

DETERMINATION OF OXYGEN
BY NEUTRON ACTIVATION ANALYSIS

by

HECTOR OCAMPO MANSILLA
B.S., National University of Mexico, 1971

AN ABSTRACT OF A MASTER'S THESIS

submitted in partial fulfillment of the
requirements for the degree

MASTER OF SCIENCE

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KANSAS STATE UNIVERSITY

Manhattan, Kansas 66506

1977

ABSTRACT

The purpose of this work was to develop a neutron activation analysis (NAA) technique to measure the oxygen content in samples of distilled water, α - D fructose, and α - D glucose by utilizing the reaction $^{18}\text{O}(\text{n},\gamma)^{19}\text{O}$. The entire data collection process as well as the analysis process (by the NAA comparator technique) was mainly directed at improving a simple manual transfer method to analyze the short-lived product ^{19}O .

Preliminary results showed a wide distribution in the accuracy of the method when individual measurements were considered. The following possible interferences were investigated: a) inadequate manipulation of samples, b) incorrect weighing of samples and recording of times, c) improper method of area calculation, d) irregularities in position during irradiation and/or counting procedures, e) significant variation in the reactor power level during sample irradiation, and f) instability of the electronic counting system. Experimental results have shown that a and b (gross mistakes) could be easily avoided or, when they occurred, eliminated from record by careful sample preparation. The area calculation was also of significance when a particular method was improperly used. While d and f were the greatest sources of error, significant variations in the power level were unlikely. By using a fixed-position during irradiation and counting and by monitoring the functioning of the electronic system with a precision pulse generator, the method proved to be successful, minimizing possible interferences.

After several experimental runs, we determined that NAA is a precise and accurate method ($\pm 6\%$) to measure ^{18}O concentrations.