NBS TECHNICAL NOTE

428

Activation Analysis:

Cockcroft-Walton Generator
Nuclear Reactor,
LINAC
July 1966 through June 1967

U.S. DEPARTMENT OF COMMERCE
National Bureau of Standards

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NATIONAL BUREAU OF STANDARDS • A. V. Astin, Director

NBS TECHNICAL NOTE 428

ISSUED NOVEMBER 1967

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Activation Analysis Section
Analytical Chemistry Division
Institute for Materials Research

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FOREWORD

The Analytical Chemistry Division was established as a separate division at the National Bureau of Standards on September 1, 1963, and became part of the Institute for Materials Research in the February 1, 1964, reorganization. It consists at present of nine sections and about 100 technical personnel encompassing some 45 different analytical competences from activation analysis and atomic absorption to vacuum fusion and x-ray spectroscopy. These competences, and in turn the sections which they comprise, are charged with research at the forefront of analysis as well as awareness of the practical sample, be it standard reference material or service analysis. In addition it is their responsibility to inform others of their efforts.

Formal publication in scientific periodicals is a highly important output of our laboratories. In addition, however, it has been our experience that informal, annual summaries of progress describing efforts of the past year can be very valuable in disseminating information about our programs. A word is perhaps in order about the philosophy of these yearly progress reports. In any research program a large amount of information is obtained and techniques developed which never find their way into the literature. This includes the "negative results" which are so disappointing and unspectacular but which can often save others considerable work. Of importance also are the numerous small items which are often explored in a few days and which are not important enough to warrant publication--yet can be of great interest and use to specialists in a given area. Finally there are the experimental techniques and procedures, the designs and modifications of equipment, etc., which often require months to perfect and yet all too often must be covered in only a line or two of a journal article.

Thus our progress reports endeavor to present this information which we have struggled to obtain and which we feel might be of some help to others. Certain areas which it appears will not be treated fully in regular publications are considered in some detail here. Other results which are being written up for publication in the journal literature are covered in a much more abbreviated form.

At the National Bureau of Standards publications such as these fit logically into the category of a Technical Note. In 1967 we plan to issue these summaries for all of our sections. The following is the first annual report on progress of the Activation Analysis Section.

W. Wayne Meinke, Chief
Analytical Chemistry Division

PREFACE

As indicated in last years' report, a separate progress report has been made for the Activation Analysis Section.

The Activation Analysis Section has the responsibility to develop measurement techniques for the qualitative and quantitative analysis of materials through the use of nuclear reactions. The technique has high inherent sensitivity, and consequently, it is being used for the analysis of very pure materials.

A basic requirement for the development of such measurement techniques using radioisotopes is the thorough understanding of the chemical, nuclear and physical principles which form the foundation of a new analytical measurement technique. However, it is essential that the developed methods be practical in the sense that they can be used successfully on materials in which science and industry have an interest. Therefore, these developed techniques are applied to the analysis of NBS Standard Reference Materials where considerable cross checking of analytical techniques is required throughout the process of certification. A very important part of the Analytical Chemistry Divisions' activity is the certification of the Standard Reference Material. In many of the Standards considerable effort is expended to remove the systematic errors in the analysis.

Activation analysis is particularly suited for this because it has very few sources of systematic errors that are difficult to evaluate. This is particularly true when the technique is compared to other trace analytical techniques. The single fact that the frequency of presence of an analytical blank is low makes it the method of choice for high accuracy trace analysis.

Standard Reference Materials certified for trace elements with high accuracy can then serve as a suitable reference point for such techniques as the spark source mass spectrometer, and many types of nondestructive activation analysis.

The Activation Analysis Section is composed of three groups that are oriented to a specific source of nuclear radiation; the reactor, LINAC, and Cockcroft-Walton generator groups. This method of division serves the administrative needs. More important is the fact that considerable interaction among the groups is encouraged so that activities of common interest are worked upon jointly in the several groups. In addition, the Radiochemical Analysis Section retains groups in nuclear instrumentation and nuclear chemistry which consult with both sections.

A roster of the groups in this section is listed in Part 6. The National Bureau of Standards has several programs whereby a scientist from the United States or abroad may work in our laboratories for one or two years. It is hoped that by utilizing these programs this section will be able to perpetuate a stimulating environment.

In order to specify adequately the procedures, it has been necessary occasionally to identify commercial materials and equipment in this report. In no case does such identification imply recommendation or endorsement by the National Bureau of Standards, nor does it imply that the material or equipment identified is necessarily the best available for the purpose.

J. R. DeVoe, Chief
Activation Analysis Section
Analytical Chemistry Division

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ACTIVATION ANALYSIS: COCKCROFT-WALTON GENERATOR, NUCLEAR REACTOR, LINAC

July 1966 through June 1967

Edited by James R. DeVoe

ABSTRACT

This is the first separate (not combined with that of Radiochemical Analysis) summary of progress for the Activation Analysis Section of the Analytical Chemistry Division of the National Bureau of Standards.

Pertinent information on the irradiation facilities of the nuclear reactor, Linac and Cockcroft-Walton generator are described. The initial installation of pneumatic transfer tubes is completed. Of great significance is the completion of a clean room that is used for pre-irradiation chemical separation for high sensitivity activation analysis.

A dual-sample biaxial rotating 14 MeV neutron-irradiation assembly has been tested. This assembly is part of a system that is controlled by a relay type of programmer to automate those irradiation steps that adversely affect precision of the analysis. This system was used to improve greatly the precision of analysis for oxygen and fluorine.

As a result of improved precision with the 14 MeV neutron irradiation, a study of systematic errors is now possible. Efforts to establish a reproducible correction factor for neutron and gamma-ray absorption between different matrices within the sample have shown considerable promise.

Standard Reference Material (SRM) steel chip Nos., 3b, 3c, and 30f have been analyzed for vanadium. Preliminary analyses of high purity platinum (a potential SRM) showed silver, copper, gold, palladium, and iridium at the ppm level. Future biological SRM's, (tree leaves, and beef liver) were tested for homogeneity and found satisfactory.

A variety of service analyses were performed. In particular, very pure aluminum was found to contain fractions of a ppm of chromium and scandium.

Additional efforts are being made to monitor reactor neutron fluxes. Additional nuclear instrumentation is being assembled to improve the versatility of the analysis technique.

Performance of the bremsstrahlung target, used with the linear electron accelerator, has been evaluated. Photon intensity distribution and stability was found to be within usable range. Analyses using the (γ,n) or (γ,p) reactions have been developed for magnesium in steels, and carbon in sodium metal.

James R. DeVoe, Chief
Activation Analysis Section
Analytical Chemistry Division

KEY WORDS:

NBS reactor, NBS Linac, Cockcroft-Walton generator, digital computers, activation analysis, Standard Reference materials, photoneutron reactions, flux monitors, vanadium in irons and steels; homogeneity testing, biological, botanical samples, carbon in sodium, self absorption corrections, high precision 14 MeV neutron activation, instrumentation, flux monitors.

1. INTRODUCTION

The usefulness of the activation analysis technique for the certification of Standard Reference Material (SRM) has become decidedly apparent during the past year. Along with this it has been equally apparent that certain inherent difficulties of the technique should properly be understood and then overcome. Probably the most serious of these is the difficulty in approaching the rapid multi-element capability of emission spectroscopy or spark source mass spectrometry.

Past efforts to solve this made use of nondestructive techniques coupled with mathematical resolution of complex spectra. This technique met with only moderate success, and in more than one case lead to serious questioning of the value of the nuclear activation technique.

For the certification of Standard Reference Materials containing $\leq$ 10 μg of element per gram of sample, it is clear that destructive analysis is the only conclusive way of evaluating the systematic errors related to self absorption. For example, the stoichiometric standard of pure element and the element in the sample should be in the same (or at least a known relative) physical and chemical environment for both irradiation and counting. It is of great importance to realize that once these Standard Reference Materials have been certified, the use of the nondestructive technique described above becomes enhanced, because this SRM can take the form of the stoichiometric standard of a pure element used in the destructive technique. In fact, after evaluating correction factors for systematic errors, certification of a given element can proceed by a nondestructive procedure in many cases.

Evaluation of the systematic and random errors has been paramount in the efforts of all the projects in the section. Improvement in the precision of analysis of even macro quantities of the elements has been demonstrated in the 14 MeV neutron activation project. Upgrading the magnitude of neutron flux will be accomplished by purchase of a new Cockcroft-Walton generator in the next few months.

Of particular importance in the research with 14 MeV neutrons is the precise evaluation of systematic errors resulting from self absorption and degradation of neutrons during irradiation and of gamma-rays during counting. This technique may allow extrapolation of correction factors between matrices, and consequently would increase the applicability of the technique.

The reactor neutron activation project continues to suffer from not having neutrons available at the reactor site. Irradiation service at the Naval Research Laboratory has been curtailed in the anticipation of start-up at the National Bureau of Standards Reactor (NBSR). No schedule of start-up is available. As a result of this, irradiations are being performed at other nuclear centers. Design of facilities has proceeded into stages that were planned after irradiation tests were performed at NBSR. We have attempted to gain some experience using other reactors in the country.

Our photonuclear activation project is in full operation for the initial design of target assembly. The most significant result to date is the analysis of carbon at the 10 ppm level in sodium metal. This has been worked on for some time at the Argonne National Laboratory and it appears to both of our laboratories that the $^{12}C(\gamma,n)^{11}C$ reaction is a superior one. Our operating schedule allows one day per week for irradiations of samples supplied by the Mössbauer spectroscopy, and nuclear chemistry groups in the Radiochemical Analysis Section; reactor neutron activation group in this section, and several groups in the Radiation Physics Division in addition to the LINAC activation project staff. Plans are underway to alleviate this burden by providing a new irradiation facility in the vicinity of the drift tube at the end of the 30 MeV acceleration stage. The beam will be magnetically deflected into a target with the capability of at least three simultaneous irradiations. Beam perturbation by multiple irradiations will be minimized in the activation-analysis-rabbit position. This facility will allow us to have several irradiation periods a week with less irradiation time per period. From all of our tests the NBS LINAC is more stable in energy than any other comparable machine of which we are aware.

Most of our laboratory facilities in the Reactor Building are completed. Final design and installation of an elaborate pneumatic tube system with many send-receive stations are in final processing. In order to adequately inform those interested in the extent of our facilities, an NBS Technical Note that describes the facilities and their operational success will be published within several months after the NBSR reaches continuous full power operation.

2. LIAISON ACTIVITIES AND GENERAL FACILITIES

A. Liaison Activities

Liaison with reactor users from other government agencies has been only superficial since the NBSR has not yet gone critical. However, three groups not mentioned in last years' report have expressed an interest in pursuing the possibility of using the reactor facilities. Representatives of the Walter Reed Army Hospital have expressed an interest in the neutron activation analysis of foods.

In addition two nongovernment organizations have expressed an interest in the facilities of the NBSR. The Dupont Winterthur Museum (Mr. Charles F. Montgomery) has expressed an interest in the use of activation analysis for authentication of early Americana, i.e., by compositional analysis to establish dates. They are interested in silver coins, among other items. It has been established that silver made during a given period had characteristic copper content. These people have been considering an NBS Industrial Research Fellowship, but no further word has been received from them since January 1967.

The Smithsonian Institution (Jacqueline Olin) has a project with the Brookhaven National Laboratory (E. V. Sayre) to determine the elemental content of ceramic shards by nondestructive neutron activation analysis (Ge-Li detectors). This project will be completed at about the time our reactor is expected to be operating and Miss Olin has expressed interest in establishing such a project at the NBSR.

B. General Facilities

New facilities that are specific to a particular project will be described elsewhere in this report. There are a number of improvements in facilities that provide an overall increase in the capabilities of all of the projects.

1. Radiochemical Laboratories in B-Wing Reactor Building

Six laboratories (Rooms B121, 122, 140, 143, 153 and

154) have been remodeled in the B-wing of the Reactor Building in addition to those done last year. The improvements made are described in last years' report [1].

2. Radiological Laboratories in the Reactor Building

The modifications to these laboratories described in last years' report are completed. Two radiochemical hood arrangements were designed for each of the two laboratories (C-001 and C-002).

For laboratory C-001 (see floor plan, figure 1) a good utilization of space was obtained by arranging four six foot hoods in the center of the room with laboratory benches and storage space around the outside of the room. Four pneumatic tube receivers are situated in the eight foot hood in one corner of the room. Since this room is intended to be shared with users from groups other than those in the section, a community receiver station was deemed desirable.

Laboratory C-002 was designed for use of the section and the pneumatic tube receivers are placed in each of the four six foot hoods (see figure 2). Consequently, each hood interacts as little as possible with the other three from a radiation hazard and a convenience in operation standpoint (see figure 3). Unfortunately, space considerations would not allow positioning of the high efficiency filter boxes other than in the radiological laboratories. This is contrasted to those laboratories in the B-wing of the Reactor Building where the filter boxes are located near the exhaust fans close to the roof of the building. Changing the filters in the radiological laboratories may constitute a radioactivity contamination hazard.

Operational characteristics of these two types of laboratory designs will be described after practical usage with the NBSR has been achieved.

(Project Leaders, G. W. Smith, and J. R. DeVoe)

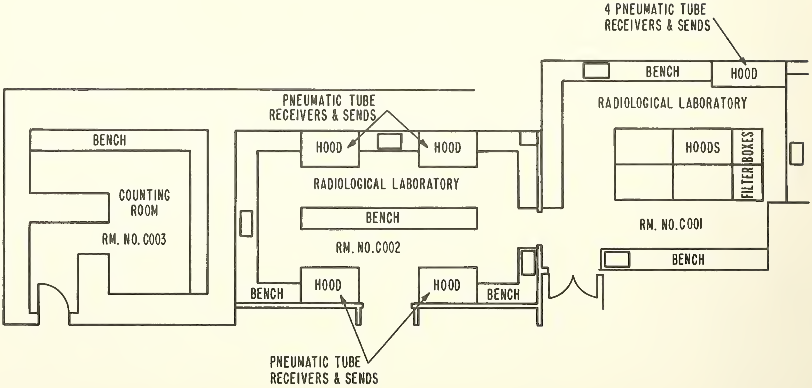

Figure 1. Floor plan for radiological laboratory, C-001 [2].

Figure 2. View of radiological laboratory (C-002).

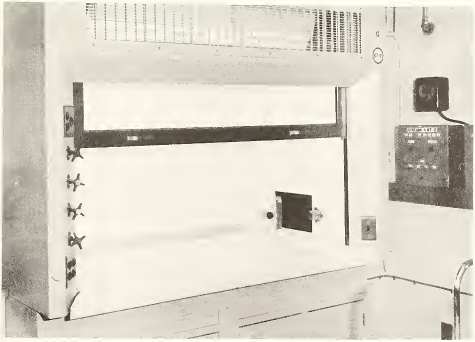

Figure 3. Photograph of typical pneumatic tube receiver built into hood.

3. Pneumatic Tubes

All of the pneumatic tubes presently installed have been made operational. Considerable difficulty was encountered with malfunctioning switches and rough edges at splices. The design characteristics of the system have been described in a previous report [2]. Unfortunately, no operational experience with the NBSR can be reported.

A detailed study of the radiation hazard to be expected from the polyethylene rabbits has been completed. Appropriate masses of material were irradiated at a flux of 8×10^{12} $n.cm^{-2}s^{-1}$ at the Naval Research Laboratory reactor. Of nine compositions from five manufacturers, we have found the Phillips-Marlex Type 5040* polyethylene to produce the least radioactivity from a ten minute irradiation. A beta plus gamma survey meter, for an eight gram sample, detected about 5 mr/h, 20 seconds after irradiation, and 2.5 mr/h, 160 seconds after irradiation. Rabbits from this material are now being fabricated and its operational characteristics will be presented in a later report.

Contracts are soon to be let for pneumatic tube systems from a fast fission flux converter tube, thermal column, and vertical thimble tubes to laboratories in the radiological laboratory and the B-wing. A comprehensive report on the design and operational characteristics of all of these pneumatic tube systems will be published as a future NBS Technical Note.

(F. A. Lundgren, P. D. LaFleur and J. R. DeVoe)

4. Other Irradiation Facilities

Fabrication of a device that will allow insertion of samples into the reactor core for periods exceeding 24 hours is now in progress. Details of the device and operating

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*For disclaimer of equipment and materials see last paragraph of preface.

characteristics will be described in a future report.

(F. A. Lundgren, G. W. Smith, Project
Leaders and J. R. DeVoe)

5. Clean Room

For applications of very high sensitivity activation analysis two basic principles must be strictly followed. The sample must be very carefully handled if nondestructive analysis inducing short lives radioisotopes is to be done, since time for surface etching after irradiation will not be available. In other cases certain interfering elements will have to be removed before irradiation. This procedure compromises one of the primary advantages of the nuclear activation technique, but sensitivities below 10^{-12}g are able to be realized. Consequently, every precaution must be taken to reduce the blank of the analysis. Particulate matter even of microns in diameter can result in many times the desired blank.

We have constructed a clean room that has met a specification of less than 100 particles ($\leqq$ 0.3 microns diameter) per cubic meter of air (Class 100). This clean room uses the laminar air flow principle. One entire wall of the room is filled with "absolute" filter banks. This produces an air layer which sweeps across the total area from ceiling to floor with the velocity vectored parallel to the floor.

The layout of the room is shown in figure 4. Personnel put on protective clothing in the anteroom (see figure 5) which is equipped with a downflow laminar air system. The velocity of this air is 200 ft/m. The doors of the anteroom are suitably interlocked so that only one may be opened at any time.

The velocity in the main room is 100 ft/m. It is obvious from the layout that care must be taken by the personnel to not work upstream from anothers' work area. Of course, the most critical operation should be on the bench next to the wall containing the filter bank (figure 5, top photograph).

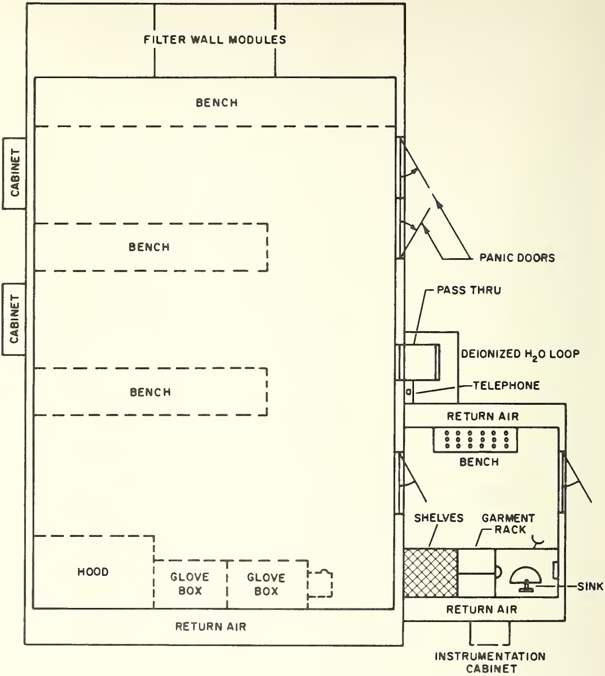
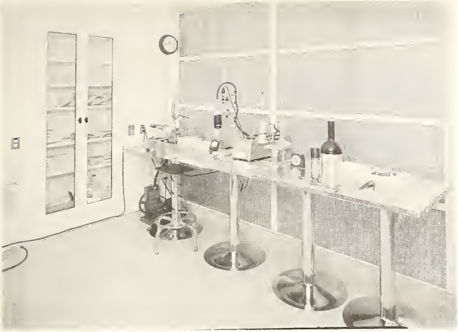


Figure 4. Layout of the clean room.



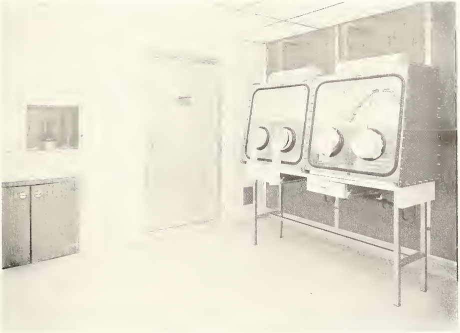


Figure 5. Absolute filter bank on end wall of clean room along with recessed cabinet in side wall (top). Other end of room where two glove boxes reside and door to antechamber (bottom). Pass through is on left.

At the end of the room are a fume hood and two glove boxes (figure 5, bottom photograph). Here a sample can be chemically treated, dissolved, etc. All of the air in this room is recirculated except that exhausted to the outside through the fume hoods. The entire room is kept at 0.15 in water positive pressure with respect to outside the room by adding makeup air to the system. Relative humidity and temperature are controlled to minimize buildup of electrostatic charges and still maintain some degree of comfort for personnel.

All fixtures such as telephone, drawers, and cabinets are recessed into the wall. A small passageway for transfering small items in and out of the clean room is designed on the same principle as the anteroom. Some of the operational experiments are shown in figure 5. Even though some of the equipment could be considered to be dust catchers, the air flowing past the equipment "washes" the loose particulate matter from the equipment within a few hours. The greatest precaution required to maintain cleanliness in these rooms is to minimize careless movements by operating personnel.

This room is built in a modular type of construction so that it can, with a few man weeks of effort be moved to a new location. The cost of this facility is approximately $150\$/ft^2$ including all auxiliary equipment and installation.

(W. P. Reed and J. R. DeVoe)

3. ACTIVATION ANALYSIS WITH 14 MeV NEUTRONS

A. Introduction

The use of 14 MeV neutrons for the activation technique has been used by industry for the analysis of specific elements such as oxygen, nitrogen, fluorine and phosphorous.

Recent interest has occurred in improving analysis precision; and of course, in most cases the analysis for Standard Reference Materials demands high accuracy. The efforts of this project have been to improve the accuracy and precision by whatever means were possible. Special instrumentation such as a sample rotator in the neutron flux, and a programmed automatic operation was instituted. Accuracy has been studied by evaluating systematic errors resulting from sample type and size causing attenuation effects in both the neutron flux and the induced gamma rays entering the detector. In addition, the chances of high accuracy are improved by using automated data handling techniques along with digital computer calculations.

The activation analysis system has been tested on a variety of Standard Reference Materials and other samples. Some of the examples which are particularly interesting will be described. In addition, particular attention is being given to the analysis of macro concentrations (10% by weight) with high precision and accuracy.

B. Instrumentation

It has been necessary to design and fabricate special mechanical and electrical packages suitably interfaced to form one comprehensive activation analysis facility. The neutron generator and detector system has already been described in a previous report [1]. The design work has been directed toward obtaining precise and accurate analyses as well as a high degree of analytical flexibility. The design, fabrication, and operational details of the various units are described below:

1. Dual Sample Biaxial Rotating Assembly

Imprecision in 14 MeV neutron activation analysis is primarily due to inhomogeneity and anisotropy in the usable neutron flux. This fact is pronounced in analyses producing short-lived radioactivity such as in the determination of oxygen by $^{16}O(n,p)^{16}N$ reaction.

A detailed description of a variable speed dual sample rotating assembly has been submitted for publication. This presentation discusses the basic engineering concepts governing the design and fabrication of the assembly. Experiments which test the reproducibility of measurement and general performance are described. Since this assembly also permits the direct determination of neutron and gamma attenuation in dense samples, a high degree of accuracy with precision dictated almost entirely by counting statistics has been achieved.

Detailed engineering drawings are shown in figures 6-8. The principle of the device is to introduce two rabbits, one for the sample and one for the standard into the sample rotator with nitrogen gas pressure. The rabbits are rotated about two axes. One asis is concurrent with the beam axis and the other is the long axis of each rabbit. Suitable arrangement of solenoid operated detents combined with a current reversal braking process in the electric drive motor, stops and aligns the rabbits for pneumatic sequential exit to the detector system.

(F. A. Lundgren and S. S. Nargolwalla)

2. Pneumatic Transfer System Sequence Programmer

Automatic sequence programmers generally used in conjunction with pneumatic transfer systems vary considerably in design, functional capacity and flexibility. The basic design of this programmer has been based on certain analytical criteria requiring both simple actuating mechanisms and rugged operational capability. Considerable flexibility of

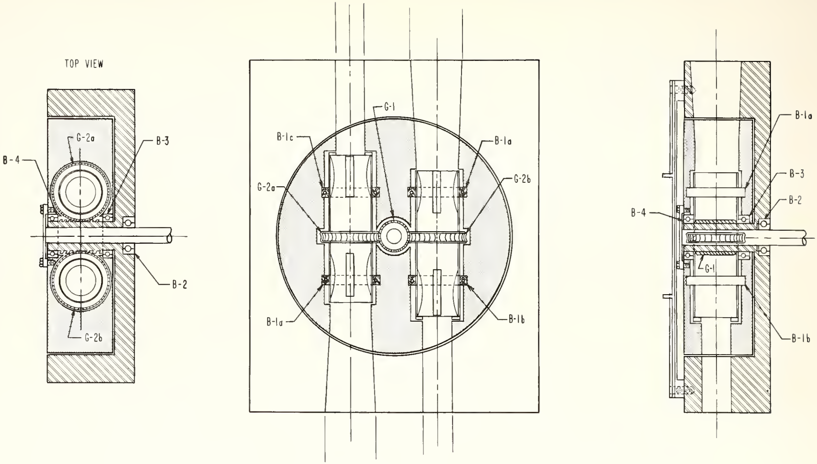


Figure 6. Assembly drawing of the dual sample biaxial rotating assembly.

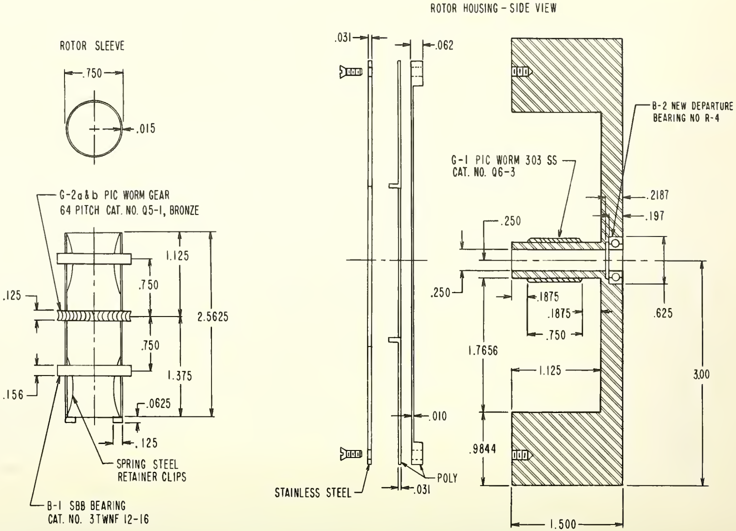


Figure 7. Engineering drawing of the dual sample biaxial rotating assembly.

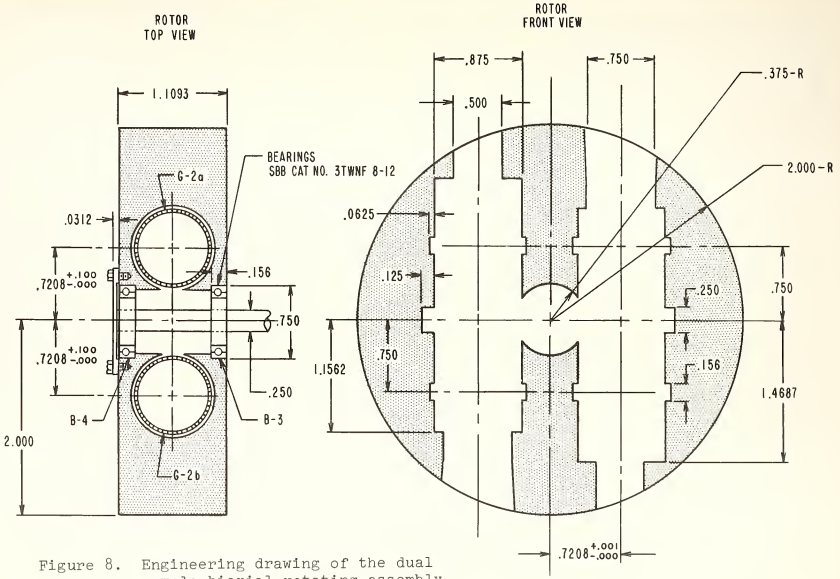


Figure 8. Engineering drawing of the dual sample biaxial rotating assembly.

operational modes has been included so that a wide variety of analyses can be performed without loss of precision. In general, the various functions and method of analysis are such as to reduce errors of timing and of variation in the two independent detector systems to a minimum [1]. The basic method of advancing from one function to the next is done by a stepping relay circuit in the programmer.

The programmer consists of a 9 pole 25 position stepper relay and fourteen 4 pole 2 position auxiliary relays. From the time the samples are manually loaded and the rotating assembly set in motion, the programmer automatically executes all required functions related to irradiation and counting with a timing accuracy of 0.1 second.

Provisions have been made for manually stepping through 25 positions whenever long-lived radioisotopes are being analyzed and the automatic sequence is not necessary. The operation of the programmer through the individual steps can be followed with the aid of the flow diagram shown in figure 9.

a. Method of Operation. Each of the twenty-five steps are indicated to be in operation by individual pilot lights. As an additional aid to understanding the status of the system, a simplified flow diagram similar to that in figure 9 is overlayed by a series of pilot lights which show the location of the particular programmed operation.

Position Home (H). After presetting the gas pressures on the main nitrogen supply, bleed nitrogen, spinner air, irradiation timer, delay timer and total time for multiscaling on the analyzer clock-timer; sample #1 is manually loaded into the receiver. In this position the routing valve is switched to route #1 and the arrester pin above the receiver withdrawn. The manual advance button on the front panel is pressed. This advances the programmer to position #3.

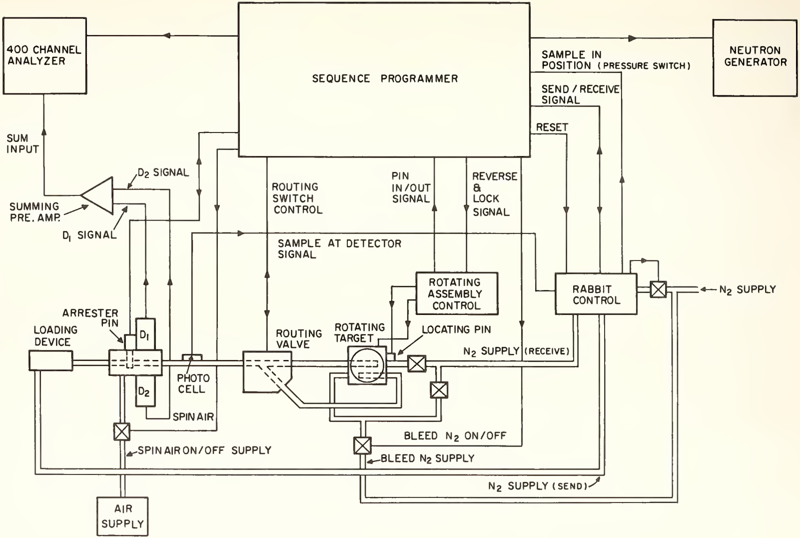


Figure 9. Flow diagram showing general operation of the sequence programmer.

Position #3. Sample #1 is injected into the rotating assembly. The "sample in" situation is sensed by a mercury pressure switch which promptly cuts off the propelling nitrogen gas supply and automatically advances the programmer to position #4. Sample #2 is manually loaded.

Position #4. The routing valve is changed to route #2 and the manual advance steps the programmer to position #5.

Position #5. The rabbit control is reset and the programmer automatically advances to position #6.

Position #6. Sample #2 is injected by push button into the irradiation assembly; the routing valve returns to route #1 and the programmer advances automatically to position #10.

Position #10. In this position the motor-start button on the rotating assembly console is pressed which initiates the rotation. The speed control dial is adjusted for the desired orbital rpm as indicated on the tachometer. (This rarely required adjustment.) The manual advance button is pressed and the programmer advances to position #12. This initiates the analyzing automatic cycle which terminates after both samples have been counted and returned to their individual receptacles in the rotating sample assembly.

Position #12. The neutron beam comes on, the irradiation timer starts; and nitrogen gas is bled into the rotating assembly. Termination of the preset irradiation time initiates a preset delay timer, and the programmer advances to position #13.

Position #13. The beam is turned off, and the rotating assembly is given a stop signal which reverses the motor and indexes the assembly in its proper orientation for rabbit ejection. The irradiation timer is reset, and the bleed nitrogen turned off. When the indexing has been completed, the programmer receives a "pin-in" signal and advances to position #14.

Position #14. The arrester pin is inserted to stop the sample in between the two detectors. Sample #1 is returned, and the photoelectric gas trigger cuts off the return gas and advances the programmer to position #15.

Position #15. The spinner air is turned on. Termination of the delay timer turns on the analyzer, and the sample is counted for a preset time. When counting has been completed the programmer advances to position #16.

Position #15. The analyzer gate is closed, the arrester pin is withdrawn; the sample is dropped by gravity into the receiver and after a one second delay is propelled into its irradiation position. Its arrival there is sensed by the mercury pressure switch which cuts off the gas supply and advances the programmer to position #17.

Position #17. The arrester pin is inserted, and the routing valve switched to route #2 and sample #2 is returned to the counting site. Once again the photoelectric gas trigger cuts off the gas and opens the analyzer gate. At the same time the spinner air is turned on. The programmer advances to position #18.

Position #18. Sample #2 is counted for the time remaining on the preset timer on the analyzer. When counting is stopped the programmer advances to position #19.

Position #19. The arrester pin is withdrawn, and the sample falls into the receiver. After a one second delay it is injected into its irradiation position. Its arrival there is sensed by the mercury pressure switch which cuts off the gas supply and advances the programmer to position #20.

Positions #20 to #25. This ends the fully automatic sequence of operation. Since the samples are in the irradiation assembly, the program can be recycled as often as necessary. However, if it is desired to terminate the experiment the programmer can be stepped from position #20 to position #25 during which both samples can be returned for removal from the receiver.

It is possible to operate the system either in multiscaler or in pulse height mode. The programmer allows either manual or semiautomatic operation. The sequence programmer has proved to be extremely rugged and accurate in its performance. There have been practically no breakdowns. The stepper relay system appears to have a great degree of flexibility and can be easily adapted to the experiment of interest.

(B. Bettum, P. Thomas and R. Shideler,
Radiochemical Analysis Section, and
F. A. Lundgren and S. S. Nargolwalla)

3. Pneumatic Transfer Construction

The general design of the pneumatic tube system closely approximates that presented in an earlier report [1]. Aluminum alloy tubing 7/8 inch O.D. was used throughout. Special polyethylene rabbits* (Olympic Plastics, Los Angeles) containing 0.2 - 0.3 mg total oxygen are used for all oxygen analyses. These 2 dram capacity vials were molded under special contract and are found to be superior not only in terms of lower oxygen content but also in the variability of oxygen content from capsule to capsule.

A system of solenoid valves controlled by the sequence programmer actuates the send and receive gas, bleeds nitrogen gas into the rotating sample assembly during the irradiation step and injects air into the spinner assembly located just below the detectors in the counting shield.

A gas trigger console* (Reactor Experiments Inc., Belmont, California) is used to perform all send and receive operations (see B-3 above).

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*For disclaimer of equipment and materials see last paragraph of preface.

4. Photoelectric Gas Trigger

In view of the coarse response of the mercury pressure switch in the existing commercially purchased system for return gas cut off, a very sensitive photoelectric trigger was designed to perform this function. The photocell and lamp assembly are located about 1 foot above the counting station. The trigger is programmed in such a way as to cut off the return gas supply whenever the first capsule ejected from the irradiation site intercepts the light beam. In the case of the second sample, the trigger is also programmed to open the analyzer gate so that counting is initiated. This trigger can sense a change in light intensity even when a polyethylene rabbit passes the light beam at speeds approaching 100 ft/s. Circuit drawings with electronic details are described below.

The circuit drawings are shown in figures 10 and 11. When a rabbit passes between the lamp and photocell, the bias on the base of T1 goes positive (dark) and then negative (light). On the negative transition, T1 and T2 are turned on. When T2 conducts, it triggers the tunnel diode discriminator which consists of D1 and T3. The pulses from T3 are amplified and shaped by T4 and T5 so that a positive 10 volt pulse occurs at the output of T5 every time the rabbit interrupts the light beam. The pulses from T5 are used to trigger a monostable into its metastable condition. The contacts of the relay are closed thus causing the appropriate relay in the automatic sequence programmer to latch.

(F. C. Ruegg, Radiochemical Analysis
Section, S. S. Nargolwalla)

5. Summing Preamplifier

The 500 μA beam capacity of the neutron generator places a great restriction on the sensitivity of analysis. In order to improve the sensitivity two detectors were used and their individual responses integrated by means of a summing

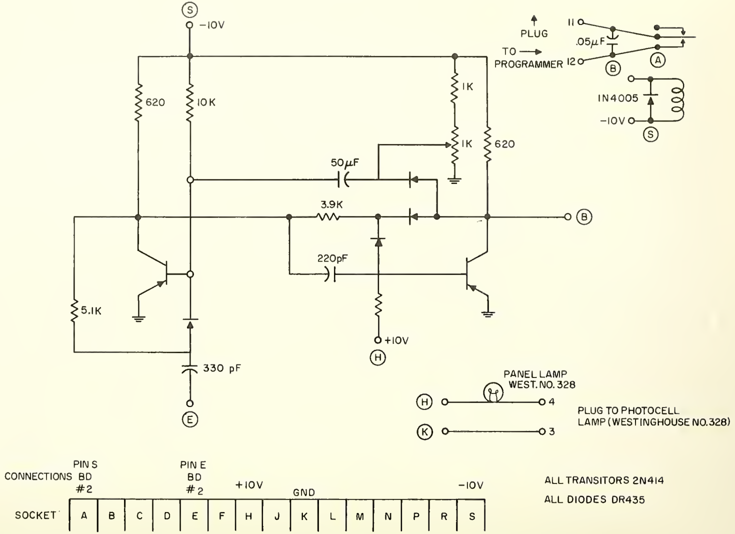

Figure 10. Circuit drawing of the photoelectric gas trigger - Board No. 1 photocell pulse generator board

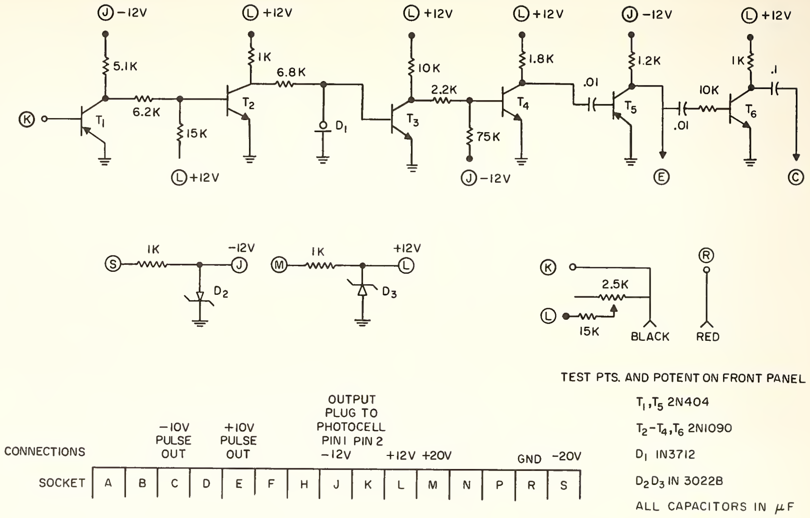

Figure 11. Circuit drawing of the monostable for the photoelectric gas trigger. Board No. 2.

preamplifier. This preamplifier has the capacity of accepting pulses from each or both detectors and also a test pulse from a precision pulse generator. The two detectors are balanced to equivalent output (e.g. from ^{137}Cs .66 MeV) by inserting a dummy capacitance that matches that detector which was switched out of the circuit (see figure 21, reference [1]). The only changes made were the addition of a test input for a pulser.

The performance of the unit shows a reproducible and quantitative summing process within the limits of counting statistics with less than 0.5% loss in overall gain whenever both detectors are being used simultaneously.

(F. C. Ruegg, Radiochemical Analysis Section)

C. High Precision 14 MeV Neutron Activation Analysis

The purpose of this extensive study was to investigate the major sources of systematic errors which directly affect accuracy of analysis. In order to make a significant contribution to the state of the art, a critical evaluation and determination of correction factors were necessary. At the same time it was necessary to maintain a high degree of flexibility so that the method developed could be easily adapted for both routine analyses and research. A comprehensive manuscript describing the approach and method of analysis is presently being prepared for publication. In this report, only a cursory treatment will be given with emphasis on certain interesting examples.

By and large this study has been prompted by the need for a rapid, precise and accurate analytical technique for oxygen in metals and metal alloys. Standard Reference Materials certified for oxygen have been in great demand by industrial laboratories for quality control, apparatus calibration and interlaboratory comparison.

A proper assessment of the state of the art must include the standard non-nuclear techniques of vacuum and inert gas fusion. These techniques can determine less than 10 µg of oxygen. However, the destructive nature of these techniques, incomplete recovery of oxygen from hard-to-decompose matrix oxides, loss of oxygen by the gettering effect, and limited sample size are some of the undesirable factors leading to inaccuracies in the analysis.

During the last decade the 14 MeV neutron activation technique has been rigorously used for the interference free determination of oxygen in a wide variety of matrices. Considerable improvement in both precision and accuracy has resulted from studies [3-8] carried out under carefully controlled conditions. Development of sample rotation systems has helped to improve precision. Today, the technique is recognized for its rapid, nondestructive and interference free nature for routine determinations.

Our approach to the problem is based on an attempt to recognize all of the nuclear and instrumental problems which directly affect the precision and accuracy of the final result. The sample size, matrix effects and sample inhomogeneity are factors which must be considered in the analysis of the practical sample. The ultimate aim was to achieve unbiased analyses with a precision governed only by counting statistics. In order to meet these stringent requirements great care has been taken in sample preparation, sample selection and treatment, and in reproducibility of sample placement both at the irradiation and counting sites.

The statistical treatment of raw data has been facilitated by the use of the computer programs, NGEN, NGENB, NGEN-1 and NGEN-2. These programs permit a comparison between the precision due to counting only and the overall experimental precision. A number of analyses indicate that the precision resulting from counting statistics is by far the predominant one. This has made it possible to determine correction factors resulting from differences in the attenuating characteristics of the sample and standard.

In a special case of metal samples, the possible effect of the environment on the shelf life of a standard is under study. Special cleaning methods and packaging procedures are also under study.

The general technique has been applied to the analysis of O, F, N, P, S and Si. Analysis of macro constituents in biological samples has been successfully performed with a high degree of precision and accuracy. Specific examples testing the established method are described below.

1. Elemental Analysis of Proteins

This work was performed in conjunction with the Chemistry Department of the Catholic University of America, Washington, D. C. A published summary of the presentation to be made before the American Nuclear Society follows:

> "ELEMENTAL ANALYSIS OF PROTEINS BY 14 MeV NEUTRON ACTIVATION", by M. Crambes, S. S. Nargolwalla and L. May (Catholic University of America)

As part of a program to evaluate the usefulness of macro constituent nondestructive 14 MeV neutron activation analysis of biological materials, the amino acids, glycine and cystine, and the protein casein were analyzed for nitrogen, oxygen, sulfur and phosphorous. The effects of the irradiation on the sample were determined by measuring the infrared spectrum before and after irradiation. The cystine provides an opportunity to observe the effect of fast neutrons on the disulphide linkage. The infrared spectra show no detectable bond alterations. The necessary experimental parameters for the analysis of each element (table 1), shows that at a flux of $10^9 n.cm^{-2}s^{-1}$, the minimal amounts of amino acids or proteins that could be used were 10 milligrams for a precision

Table 1. Experimental parameters and sensitivity chart

Element	Reaction Used	Time			Radiation Measured	Standard Used	Minimum Weight of Element for Desired Precision mg.			
							1σ		2σ	
		Irrad	Dec	Count			1%	5%	1%	5%
O	$^{16}O(n,p)^{16}N$	45s	3s	20s	γ(4.8-8 MeV)	$(COOH)_2$	8	0.3	31	1.2
P	$^{31}P(n,\alpha)^{28}Al$	2m	1.5m	2m	γ(1.78 MeV)	$Ca_3(PO_4)_2$	0.5	0.02	2	0.1
N	$^{14}N(n,2n)^{13}N$	10m	20m	10m	γ(0.51 MeV)	NH_4NO_3	2	0.06	6	0.3
S	$^{32}S(n,p^{32}P$	1h	24h	15h	Brems. (1.7 MeV β)	S	13	3	28	5

of 5% (one standard deviation) and 100 milligrams for a precision of 1% (two standard deviations). The determination of carbon by 35 MeV photon activation analysis shows that the minimum sample weights would be adequate for comparable statistics. The results of the analyses (table 2) for N, O, P and S agree within 0.5 to 4% at the 95% confidence level with the analyses performed by standard chemical methods.

The general analytical procedure is based on a highly precise and accurate technique developed for the determination of oxygen. The sample and a standard are irradiated simultaneously in a dual sample-biaxial assembly and sequentially counted between two 4 in x 3 in NaI(Tl) scintillation detectors coupled to a 400-channel pulse height analyzer system. Experimental correction factors for both neutron and gamma attenuation were determined and necessary corrections made to the final results.

An appraisal of this study indicates the feasibility of nondestructive total elemental analysis of proteins without any measurable effect from radiation damage. The methods developed serve a useful purpose in the field of biochemistry, particularly where a prior knowledge of the elemental composition is necessary for subsequent destructive biological tests.

2. Analysis of Oxygen in Steel

This SRM No. 163 is a type 52100 steel which had been submitted for certification of Cu, Al and O. Approximately 800 lbs of this material had been obtained from powdered metallurgy processes by atomization of the molten metal in argon gas to form the maximum amount of particles between 20 and 100 mesh. By selecting a 60-100 mesh sieve fraction it was possible to obtain adequate homogeneity. The aimed oxygen content was 500 ppm.

A sample of approximately 34 gm was carefully prepared in the irradiation capsule under dry nitrogen and analyzed

Table 2. Analysis of amino acids and protein by 14 MeV neutron activation analysis

Number of Determinations	Compound	Weight Percent		
		Activation[a]		Chemical
	Glycine			
6	O	42.40	0.51	42.63
1	N	17.71	0.36	18.66
	Cystine			
6	O	27.10	0.42	26.63
1	N	11.37	0.26	11.66
1	S	27.56	0.53	26.68
	Casein			
6	O	24.17	0.60	-
1	N	11.25	0.28	14.5[d]
1	S	Not Detectable		0.8[b]
1	P	1.06	0.07	0.59[d] 0.9[c]

[a] The error given expresses the total experimental uncertainty at the 95% confidence level using the standard deviation of the mean.

[b] Kleiner, I. S. and Orten, J. M., "Biochemistry", Mosby, C. V., St. Louis, 1966.

[c] Carter, H. E., ed., "Biochemical Preparations, Vol. 1", John Wiley, New York, 1949.

[d] Haurowitz, F., "Chemistry and Biology of Proteins", Academic Press, New York, 1950.

according to the developed method described in the above sections of this report. A sample of oxalic acid served as the standard. Both samples were simultaneously irradiated in the rotating sample assembly for 30 seconds. The steel sample was pneumatically ejected and counted for 20 seconds, with a 4 second time lapse after termination of irradiation. The standard was counted for 30 seconds. After background subtraction the sample activity was corrected for attenuation. By comparison of the corrected activity with that resulting from the standard, the oxygen content of the steel sample was found to be 1498 ± 39 ppm (2σ)* for a series of six determinations. The counting error was 35 ppm (2σ), and is indicative of the high degree of experimental precision achieved.

3. Analysis of Oxygen and Nitrogen in Be_3N_2

A sample of Be_3N_2 on which some heat capacity measurements had been made was submitted for oxygen and nitrogen analysis in order to determine if its composition had been altered in any way due to sample heating in the calorimeter.

Five determinations for oxygen were made on a single sample of about 6 g taken from the bulk sample of about 60 g. The weight percent of oxygen was found to be 2.384 ± 0.041% (2σ).

Nitrogen was determined by counting the 0.51 MeV annihilation radiation from the decay of ^{13}N from the $^{14}N(n,2n)^{13}N$ reaction. By multiscaling of the counts and by comparing with a NH_4NO_3 standard, the nitrogen content from five determinations was found to be 40.35 ± 0.21% (2σ). Although this result was highly reproducible, a considerable discrepancy existed when compared to the stoichiometric amount (49%) determined by Kjeldahl method. In this case about

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*2σ indicates that the stated precision represents twice the standard deviation of the average.

0.5 gm samples were analyzed. This disagreement between the results prompted further investigation.

A possible matrix effect of Be in the sample was studied by analyzing for oxygen in pure BeO. It was found that the presence of Be did not affect the oxygen result.

The NH_4NO_3 standard was rechecked and cross-checked against a fresh standard. No anomalous behavior was observed. Past analyses for nitrogen in amino acids indicated absence of systematical errors.

The original sample of Be_3N_2 was divided into approximately a 1:2:3 weight ratio and each fraction was admixed with BeO in order to maintain constant sample density. Nitrogen values from 37-49% were observed with no correlation between the amount of Be_3N_2 added and the observed percent nitrogen content in the admixture.

Finally, a fresh sample of Be_3N_2 was analyzed and the result confirmed the original value of 40.3%. The entire sample was carefully ground to a fine powder, well mixed and reanalyzed. A value of 49.25 ± 0.32% (2σ)* was obtained for six determinations.

These results may indicate an unusual type of homogeneity problem. In view of the limited amount of work done, no detailed conclusions can be drawn. Further work is in progress to determine the nature of this inhomogeneity. These experiments will include a cross-sectional sampling of the entire sample and analysis for nitrogen with and without the grinding operation on each sample.

4. Analysis of Fluoride in Fluorspar

A feasibility study is being made in order to establish the limits of accuracy and precision with 14 MeV neutron activation analysis as applied to the analysis of fluoride in fluorspar.

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*2σ indicates that the stated precision represents twice the standard deviation of the average.

Two samples each of finely ground fluorspar [NBS SRM Nos. 79(a) and 79(b)] were analyzed using the $^{19}F(n,2n)^{18}F$ reaction. After a ten minute irradiation in the rotating sample assembly the sample and standard were sequentially counted for ten minutes each with a delay of 1 minute between samples. Multiscaling of the 0.51 MeV annihilation radiation and comparison with a NaF standard, sample 79(a) gave a fluoride content of 47.82 ± 0.48% (2σ)*, and sample 79(b) 47.03 ± 0.46% (2σ). The errors given were almost five times greater than the desired precision. Since the flux used was very low, a better precision can be expected by irradiating in a higher flux and by increasing the irradiation time by a factor of two.

On the basis of the preliminary results it is estimated that under the best experimental conditions, it would be possible to improve the precision to about 0.4% (2σ) at the 48% fluorine concentration.

(M. Crambes and S. S. Nargolwalla)

D. Effect of Sample Size and Gamma-Ray Energy on the Attenuation Factors

Throughout all of the analyses performed so far, the difference in the attenuation of 14 MeV neutrons and the pertinent gamma-ray energy of the irradiated sample and standard have been determined. The importance of these factors has been evident from the comprehensive study relating to the determination of oxygen. It was necessary to study the change in the correction factors as a function of sample diameter and gamma-ray energy of the irradiated sample. It was anticipated that positron emitters would not behave in an identical manner and some correction would be necessary to account for variation in the energy of the positron.

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*2σ indicates that the stated precision represents twice the standard deviation of the average.

This work is now approaching completion and will be incorporated in a manuscript being prepared for publication. This manuscript will contain details relating to the solution of problems in 14 MeV neutron activation analysis with evidence of the effect of key variables on precision and accuracy. Experiments for the evaluation of these correction factors and empirical equations describing the attenuation process will also be included.

Our experience in the past has confirmed the absolute necessity of such studies. The usefulness of our approach has been evident in the application of the developed technique for the analysis of a variety of SRM's for a number of elements in different matrices. The understanding of the attenuation processes has provided a large measure of flexibility to the facility for application in research and routine analysis. The certification of new standards with a high degree of speed, precision and accuracy will also be facilitated.

(S. S. Nargolwalla and M. R. Crambes)

4. ACTIVATION ANALYSIS WITH REACTOR NEUTRONS

A. Facilities

Much of those facilities described in the section entitled "General Facilities" are being used by the personnel in this project. Specific use will be made of the radiological laboratories (C-001 and C-002) along with the associated counting room (C-003). A photograph of the receiver for the small sample shuttle from laboratory (C-002) to the counting room is shown in figure 12. Sample preparation of moderately short-lived radioactivities will be performed in the laboratory and the counting sample will be then quickly transferred to the counting room by the shuttle where another experimenter will insert the sample onto the detector. In addition to speed, radioactive contamination problems may be reduced.

Figure 13 shows a photograph of some of the instrumentation, such as a Northern Scientific* 1,024 channel pulse height analyzer, Technical Measurements Corporation* 400 channel analyzer and a NaI(Tl) gamma-ray detector.

Our facility at the Naval Research Laboratory which was described in last years' report remains essentially unchanged [1]. This facility will be moved to the counting room when the NBSR reaches full power continuous operation.

Additional counting facilities are ready for installation, but until the reactor goes critical, components of these systems are being used elsewhere.

(P. D. LaFleur and J. R. DeVoe)

B. Analysis of Standard Reference Materials

1. Nondestructive Analysis for Vanadium

Three Standard Reference Material steel chip standards Nos. 3b, 3c and 30f were analyzed for vanadium content.

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*For disclaimer of equipment and materials see last paragraph of preface.

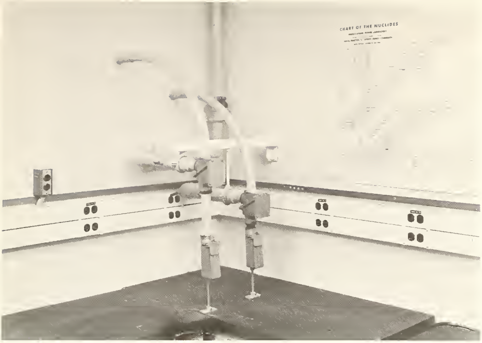

Figure 12. Small sample shuttle receiver in counting room, C-003.

Figure 13. Photograph of counting room, C-003.

This element has been analyzed previously by neutron activation analysis personnel in Standard Reference Materials stainless steel No. 73c [1] and could be done nondestructively. These steel chips were very similar in composition to each other and also to another Standard Reference Material, steel chip No. 3a. The elemental composition of Standard Reference Material Nos.3a and 30e is given in table 3. The significant difference between the previous SRM 73c and the 3a, 3b, and 3c samples is the lower vanadium content of 60 ppm compared to about 320 ppm for the 73c. No. 30f has considerably more vanadium than the others. Since some difficulty had been encountered in the previous vanadium analysis when comparing the activation analysis results to wet chemical results (see page 53 reference [1]) it was decided to analyze all three SRMs in the series, 3a, 3b and 3c even though 3a had already been certified.

a. Experimental

Equipment and Facilities - The equipment used for this analysis was a 3" x 3" NaI(Tl) detector in conjunction with a 400 channel pulse height analyzer. The analysis was done at the Naval Research Laboratory Nuclear Reactor using their pneumatic tube irradiation system. These facilities have been described elsewhere (see page 14 reference [1]).

Due to the high manganese levels present in the three samples and the low vanadium contents, some interference was expected from the manganese Compton-scattered radiation. Therefore, several preliminary experiments were done which showed that subtraction of the long-lived manganese-56 from the short-lived vanadium would eliminate any possibility of interference due to the manganese. This was accomplished in a manner similar to that used previously for the analysis of aluminum (see page 52 reference [1]) in which the entire manganese contribution was subtracted from the spectrum after a specified decay time. An example of this classical tech-

Table 3. Composition of SRM white iron 3a[a]

Element	Composition (%)
Carbon	2.30
Manganese	0.317
Phosphorus	0.118
Sulfur	0.083
Silicon	1.12
Copper	0.121
Nickel	0.017
Chromium	0.048
Vanadium	0.006
Molybdenum	0.006
Nitrogen	0.008

Composition of SRM chromium-vanadium steel No. 30e[b]

Carbon	0.505
Manganese	0.786
Phosphorus	0.026
Sulfur	0.036
Silicon	0.269
Copper	0.094
Nickel	0.027
Chromium	0.934
Vanadium	0.149
Molybdenum	0.007
Nitrogen	0.007

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[a] Also aim composition for renewal SRM Nos. 3b and 3c.

[b] Also aim composition for renewal SRM No. 30f.

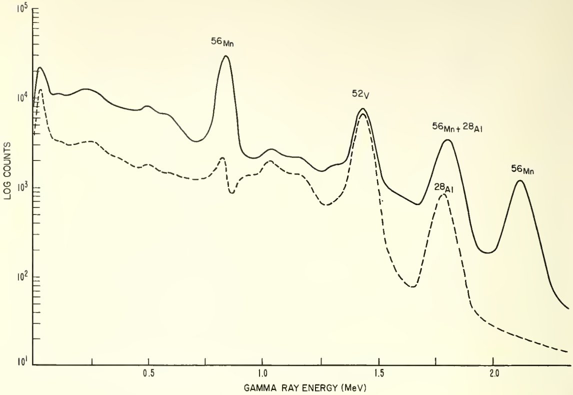

Figure 14. Gamma-ray spectrum illustrating removal of a long-lived contaminant by subtraction.

nique is shown in figure 14.

Samples were used as received. 100 mg samples of each SRM were taken and sealed in polyethylene snap-cap vials. Solution standards of vanadium (99.9+% vanadium, obtained from NRL, (see page 53 reference [1])) bracketing the concentration present in the sample were pipetted onto filter paper inside of polyethylene snap-cap vials and evaporated. The vials were heat sealed. In order to monitor the neutron flux during the irradiations, copper foil flux monitors (see page 39 reference [1]) were attached to all samples and standards.

The procedure used is as follows: (a) irradiate the sample (or standard) for exactly 4.0 min in the north pneumatic tube facility; (b) return the rabbit to the radiochemistry laboratory after irradiation and allow exactly 2.5 min decay before commencing counting (during this decay time the flux monitor is removed and the sample is positioned exactly on the detector; peak position is confirmed and dead time of the detector system must not vary significantly from previously established limits); (c) the sample is then counted for 4.0 min live time in the add mode; (d) the vanadium is then allowed to decay out and the manganese-56 is subtracted by accumulating, without disturbing the sample from the previous count, for 4.4 min live time in the subtract mode starting the count exactly 25.5 min after starting the first count. This technique results in a pure vanadium spectrum as shown in figure 14. (The relatively small amount of aluminum-28 seen in the vanadium spectrum is due to an impurity in the sample container. This impurity did not interfere with the analysis since the sample and standard containers were identical.) At some later time, the copper foil flux monitors are counted and the samples and standard normalized to the same conditions.

For SRM No. 30f, five 100 mg samples were weighed as received, and each was sealed in a polyethylene snap-cap vial with flux monitor attached. Standards were accurately measured aliquots of a standard vanadium solution, prepared from high purity vanadium. The analysis procedure was the same as in the previous report except the irradiation time was 1.0 min, the decay time was 1.0 min, the samples were counted for 2.0 min live time and there was no subtraction of the manganese activity.

The data accumulated using the above technique were reduced by computer integration of the peak areas with background subtraction and calculation of peak counts per unit weight. Corrections included flux normalization as stated above and neutron self-shielding corrections which were small for ~100 mg iron samples. The gamma-ray attenuation was calculated to be negligible for these conditions.

b. Results and Conclusions. The analysis results for SRMs may be found in table 4. The certified value of 60 ppm for the 3a sample lies within the confidence limits for the value as determined in this analysis. The individual values for all three SRMs showed significant variations, indicating perhaps an inhomogeneity in the SRM itself for ~100 mg samples. This type of inhomogeneity has been demonstrated to be present in NBS SRM steel alloys by electron microprobe studies of steel SRMs [9].

No complement subtraction procedure was required for the high vanadium concentration in SRM No. 30f. Replicate samples agree very well with each other and there seems to be no evidence of vanadium inhomogeneity within the limits of precision of this analysis.

(D. A. Becker and P. D. LaFleur)

2. Activation Analysis of Platinum

Two platinum Standard Reference Materials were analyzed; one with very high purity (total concentration ~1 ppm) and a doped platinum (future SRM Nos. 680 and 681, respectively).

The procedure used for the analysis of the elements silver, copper, gold, palladium, and iridium is as follows:

Nondestructive neutron activation analysis of impurities in platinum are often limited to those elements having half-lives of $\gtrsim$ 1 month because of the activation of the platinum matrix (see table 5). Since platinum is difficult to dissolve (several hours are required), destructive analysis after irradiation is limited to those impurity elements whose half-life is greater than about 2 hours.

In the analysis of SRM 680 and SRM 681 these factors, and the low neutron capture cross-sections of several of the trace impurities, limited the applicability of neutron activation analysis to Ir, Au, Pd, Cu, and Ag. Two platinum

Table 4. Results of vanadium analysis of SRM 3a, 3b, 3c and 30f

SRM Sample	Results from this Analysis		Results from other Analytical Techniques		Certified Value[b]
	Individual	Average[a]	NBS Wet Chem.	NBS Co-operators	
3a-1	0.0069	0.0065 ±0.0003			0.006
3a-2	0.0067				
3a-3	0.0060				
3b-1	0.0060	0.0057 ±0.0002	0.0055	0.005 0.008	
3b-2	0.0056				
3b-3	0.0054				
3c-1	0.0075	0.0077 ±0.0001	0.0063	0.005 0.008	
3c-2	0.0076				
3c-3	0.0080				
30f-1	0.192	0.192 ±0.0004	0.182	0.189 0.184 0.182 0.181	
30f-2	0.193				
30f-3	0.193				
30f-4	0.192				
30f-5	0.192				

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[a] value is the standard deviation of a single determination

[b] Certified by NBS Certificate of Analysis. This value was available only for 3a; the other samples were not yet certified.

Table 5. Platinum radioisotopes induced by neutron activation analysis

Isotope	Main Nuclide Produced	Half-Life	Principal Radiation and Energy in MeV
^{190}Pt	^{191}Pt	3d	E.C. γ: 0.54, .36, .44, etc.
^{192}Pt	^{193m}Pt	4.4d	I.T. .13, .013
^{194}Pt	^{195m}Pt	3.5d	I.T. .13, .099
^{195}Pt	^{196}Pt	Stable	
^{196}Pt	^{197}Pt	18H	β^- γ .279, .19, .077
^{198}Pt	^{199}Pt	30 min	$\beta^- \rightarrow {}^{199}$Au 3.1d half life
			β^- γ: .158, .208, .05

materials were available, one of low impurity, SRM No. 680, and one of intermediate impurity, SRM No. 681. The trace element content in SRM 680 and SRM 681 is shown in table 6.

Table 6. Estimated impurity content in ppm

Impurity	Pt SRM No. 680	Pt SRM No. 681
Ir	<1.0	9.7
Rh	0.5	4.5
Pd	<1.0	4.9
Ag	< .5	4.3
Au	<1.0	4.5
Ca	0.2	8.0
Pb	<1.0	12.0
Mg	1.0	5.5
Cu	<1.0	3.5
Fe	4.0	~5.0
Si	2.0	~5.0
Al	<1.0	~5.0

The activation characteristics of the elements determined are shown in table 7. The Ir was determined nondestructively after approximately 6 weeks cooling, and the others after dissolution of the Pt matrix and subsequent separation.

Table 7. Radioisotopes induced by neutron activation analysis of the trace elements in platinum

Element	Isotope Used	Relative Abundance %	Nuclide Produced	Principal Radiation and Energy (MeV)
Au	^{197}Au	100	^{198}Au	β^-, γ 0.41
Pd	^{108}Pd	26.71	^{109}Pd	$\beta^- \rightarrow \text{Ag}^{109m} \gamma .088$ & R.X.
Cu	^{63}Cu	69.09	^{64}Cu	β^-, β^+(γ annihilation.511)
Ag	^{109}Ag	48.65	^{110m}Ag	β^-, γ 0.66 0.89 ...
Ir	^{191}Ir	37.3	^{192}Ir	β^- γ .32 .47 ...

a. Apparatus and Reagents. Two chromatographic columns; 10 mm in diameter and 30 cm long are needed for this separation. The different solutions used are:

(1) 1% dimethylglyoxime solution in ethanol

(2) 100 ppm dithizone solution in CCl_4

(3) 0.5 M Tetrahexyl ammonium iodide (THAI) solution in methyl butyl ketone (Hexone).

(4) The two ion exchange resins used here, were Dowex 1 x 8 (100-200 mesh) and Dowex 50 x 8 (200-400 mesh) - the resin beds were about 9 cm high.

b. Procedure.

(1) Sample preparation and irradiation. Samples of 0.5 mm Pt wire weighing approximately 100 mg were irradiated in the glory hole facilities of the Naval Research Laboratory Reactor at a neutron flux of approximately

$8 \times 10^{12} n.cm^{-2}s^{-1}$. Standards of Au, Cu, Pd and Ag foils weighing about 1 mg each were irradiated simultaneously. Copper flux monitors were used to correct for flux gradients due to the sample position in the glory tube.

(2) Sample dissolution and standard preparation. After irradiation the samples were etched with aqua regia and washed with distilled water. They were then placed in a small round bottom flask fitted with a reflux condenser along with 10 ml of aqua regia and 100 μg each of Au, Cu and Pd carrier. After refluxing for 5-8 hours the samples were completely dissolved. The solution was evaporated to dryness. A few drops of concentrated HNO_3 were added to the residue to insure the +3 oxidation state of the Au and the residue was dissolved with 5 ml of concentrated HCl.

The standards were dissolved in aqua regia or nitric acid and diluted to volume in a volumetric flask. Aliquots of these solutions (1/1000 of the Au and 1/500 of the Ag, Cu and Pd) were treated as follows:

The Au was retained on a column containing Dowex-50. The Cu was extracted into dithizone-CCl_4, and the Pd was extracted into $CHCl_3$ as the dimethylglyoximate, and Ag was extracted into hexone with THAI. This allowed the sample fractions and the standards to be counted under the same conditions of composition and geometry.

(3) Separations.

(a) Gold. The solution of the irradiated Pt wire was passed through a Dowex-50 resin bed which had been prewashed with concentrated HCl, at a flow rate of 0.25 ml $min^{-1}cm^{-2}$. Under these conditions only the Au is retained on the column. The column was washed with 30 ml of concentrated HCl to remove completely the other activities.

(b) Palladium. The solution that had been passed through the Dowex-50 column and the wash, were evaporated to dryness and the residue dissolved in 10 ml of

1N HCl. The solution was transferred quantitatively to a separatory funnel and 2 ml of the alcoholic solution of dimethylglyoxime was added. The solution was extracted twice with 5 ml portions of $CHCl_3$. The $CHCl_3$ extracts were combined with evaporated to dryness, the palladium dimethylgyoximate was destroyed by oxidation with nitric acid, and the extraction into $CHCl_3$ as the palladium dimethyldyoximate was repeated to increase the decontamination from Pt.

(c) Copper. The aqueous phase from the first palladium dimethylglyoximate extraction was evaporated to dryness and the residue dissolved with 2-3 drops of concentrated HNO_3 and 5 ml of 1N HCl. The solution was passed through a column of Dowex-1 which had been pre-equilibrated with 1N HCl at a flow rate of about 0.3 ml min^{-1} cm^{-2}. Pt and Ag are retained on the column and Cu passes through. The column was washed with 20 ml of 1N HCl and the Cu solution (including the wash) was extracted twice with 5 ml portions of dithizone in CCl_4.

(d) Silver. Silver was eluted from the Dowex-1 column with 30 ml of 10N HCl at a flow rate of approximately 0.3 ml min^{-1} cm^{-2}. The eluate was diluted with 30 ml of H_2O, and the Ag was extracted with tetrahexyl ammonium iodide into hexone.

(4) Counting. The activities in the various samples after separation were measured using a 3" x 3" NaI(Tl) crystal and a 400 channel pulse height analyzer. The 410 keV transition of ^{198}Au was used for the Au analysis, and the 22 keV Ag x-ray (from ^{109}Ag) was used for the Pd analysis. The 511 keV annihilation radiation from the decay of ^{64}Cu was used for the copper analyses and the 660 keV transition in the decay of ^{110m}Ag was used for the silver analysis. Counting was begun about 28 hours after the end of the irradiation.

(5) Results. The results are tabulated in table 8. Corrections were made for flux gradients.

Table 8. Results of trace analysis in platinum[a]
(all values in ppm)

	Ultra Pure		Doped	
Silver		<~0.6	1.56	
			1.97	average = 1.76
			2.07	± 0.15
			1.46	
	Aim composition	<0.5		4.3
Copper	0.0834		4.90	
	0.0818	average = 0.0871	4.84	average = 5.00
	0.0979	± 0.004	5.23	± 0.08
	0.0855		5.04	
	Aim composition	<1.0		3.9
Gold		<~0.5	2.48	
			2.65	average = 3.22
			4.08	± 0.36
			3.66	
	Aim composition	<1.0		4.5
Palladium	0.130		5.86	
	0.122	average = 0.127	5.76	average = 5.58
	0.130	± 0.003	5.49	± 0.14
	0.241[b]		5.20	
	Aim composition	<1.0		4.9
Iridium	65		11.78	
	68	average = 67.3	11.74	average = 11.49
	95[b]	± 1.3	11.11	± 0.14
	69		11.42	
	Aim composition	<1.0		9.7

[a] ± value is the standard deviation of the average.

[b] Rejected by Dixon criterion and not included in average. See NBS Handbook No. 91, pg. 17-3.

c. Discussion of Results.

The analysis of Au is complicated by the production of ^{199}Au from the decay of ^{199}Pt. However, Au can be determined in Pt at the level of a few tenths of a ppm [26]. Although the long-lived ^{110m}Ag is produced, a separation of the Ag fraction was performed since the 74d ^{192}Ir interfered with the nondestructive analysis.

Checks on the efficiency of the radiochemical separation were made in great detail. Table 9 presents the yield and decontamination factors observed.

Table 9. Yield and decontamination for radiochemical separations

Element	Extraction in %	Average & Standard Deviation of the Average		Decontamination[a] Factor
Au	99.49			
	99.94	99.75	0.14	$\simeq 10^6$
	99.82			
Pd	98.50			
	97.91	98.11	0.24	$> 10^6$
	97.92			
Cu	99.85			
	99.73	99.73	0.16	$> 10^6$
	99.48			
	99.86			
Ag	99.58			
	99.12	99.45	0.16	$> 10^6$
	99.66			

[a] Amounts of contaminants used are given in table 6.

In general, the ultrapure platinum exhibited less than 1 ppm of the analyzed elements. The results of these analyses for the doped platinum do not agree with the aim composition. However, adding impurities constitutes a very difficult process in such matrices as platinum.

The unexpectedly large imprecision in the silver (27%) and the told (38%) indicate possible inhomogeneity. Subsequent information received from conductivity measurements by NBS Boulder Laboratories has confirmed this. Further work is to be done on these materials.

(M. R. Crambes, R. R. Ruch and G. W. Smith)

3. Activation Analysis of Biological and Botanical Materials.

The work reported here describes an initial investigatory series of analyses to determine if it is feasible to produce a biological or botanical Standard Reference Material. Our primary concern was homogeneity of these materials. Several laboratories have worked on such materials. Professor Kenworthy [10] has produced a variety of these materials and Professor Bowen has also produced some samples which have been widely circulated [11]. Mr. Robert Keenan has supplied us with some beef liver which has exhibited great shelf life at his laboratory [12].

Our findings during this study are summarized in the following abstract of a paper presented at the Amsterdam Symposium on Nuclear Activation Techniques in the Life Sciences, May 8-12, 1967.

THE PREPARATION OF AN NBS BIOLOGICAL STANDARD REFERENCE MATERIAL FOR TRACE ELEMENTAL ANALYSIS,

by G. W. Smith and D. A. Becker

A survey of leading U. S. biological, medical and forensic laboratories confirms the need for well characterized botanical, soft mammalian tissue, bone tissue and blood standards. The only sources of dried and sterilized tissue and blood known to the authors are certain U. S. drug concerns, whereas H. J. M. Bowen's kale has only been available since 1964 as a botanical comparison sample.

Before reliable analytical data can be obtained, the reference material must be considered homogeneous and tests for inhomogeneity should be performed. Examples are given of this procedure, which is carried out routinely by the NBS Standard Reference Materials. Homogeneity of Bowen's kale for chlorine (~ 4000 ppm), manganese (~ 14 ppm) and copper (~ 6 ppm) as well as for manganese in beef liver (~ 10 ppm) citrus leaf (~ 28 ppm) and oak leaf (~ 1000 ppm) are given. Measurements were made by neutron activation analysis both with and without chemical separation, using a Ge(Li) semiconductor detector. This technique was also used for the quantitative determination of the trace constituents chlorine and manganese, nondestructively. In addition, a destructive method was used for the elements arsenic and copper.

Analyses of variance, using the activation analysis data, show for all of the biological materials except the citrus leaves no evidence of inhomogeneity. Therefore, preparation under contract, of large lots of deciduous leaves and beef liver, will begin. Tests for inhomogeneity and elemental content certification will be made by NBS activation analysis and other analytical competencies.

4. General Approach.

It was at the Institute of Materials Research Symposium on Trace Characterization held in October 1966 that our section first attempted to present the general approach which we expect to use for Standard Reference Material calibration of trace element concentration in pure materials. Implicit within the abstract to follow is the need for radiochemical separations so that self absorption corrections can be made. It should be emphasized that certified Standard Reference Materials for trace elements will provide a considerable advantage for the use of nondestructive activation analysis methods.

DETERMINATION OF TRACE ELEMENTS IN STANDARD REFERENCE MATERIALS BY NEUTRON ACTIVATION ANALYSIS

by G. W. Smith, D. A. Becker, G. J. Lutz, L. A. Currie and J. R. DeVoe

Neutron activation analysis with its high sensitivity and accuracy in trace analysis is being used at the NBS for analysis of Standard Reference Materials. Problems affecting precision and accuracy have been encountered and solutions to four problems are discussed. First, the positive bias introduced by induced radioactivity of similar gamma-ray energy was found in the determination of interstitial argon in ultrapure silicon. A decay-curve resolution technique was used to compensate for the error. Secondly, errors due to differences in geometrical location between sample and standard during irradiation were observed. Flux gradients were determined by copper foil flux monitoring to give necessary means of correction. Thirdly, errors are caused by differences in neutron self-shielding between sample and standard during irradiation. An empirical correction method was used and a computer program written for calculation. Examples are given. Finally, the problem of gamma-ray attenuation during counting of sample and standard has been explored. A theoretical and experimental study gives the necessary corrections. It is concluded that careful study is vital to assure accurate analyses by neutron activation analysis on trace elements in complex matrices such as many Standard Reference Materials.

C. Analysis for Impurities in Pure Reagents.

The National Bureau of Standards Separation and Purification Section of the Analytical Chemistry Division is presently doing research in the field of preparation and purification of analytical reagents. Some work is going in to the internal preparation of very pure reagents, while other work is going into the characterization and evaluation of presently available reagents, including commercially available reagents of special purities.

The problem of reagent purity is an important one in chemistry since a great deal of effort is expended in the critical areas of very pure material preparation and evaluation. In most methods of chemical analysis, reagents must be used in the analysis, so that reagent blanks are always present. Thus, for trace analysis (concentration of 1 to 100 ppm) a reagent blank is usually significant, while for ultratrace analysis (concentration of < 1 ppm) the reagent blank may well be the major portion of the signal.

In this situation, the analytical technique of neutron activation analysis is an extremely valuable one, since it has no reagent blank for nondestructive analysis, and then has only a "radioactive blank" for a destructive analysis. This "radioactive blank" is zero for all short-lived radioisotopes ($T\ 1/2 \lesssim 100$ days) which makes up virtually all of the radioisotopes used in reactor activation analysis. Thus, with essentially no reagent blank, neutron activation analysis can play an extremely important role in the determination of reagent purities.

Some of the impurity elements expected to be present in purified water are sodium, manganese, copper, bromine, and gold. The reagents other than water (e.g. HNO_3, HCl, NH_4OH, NaOH, etc.) will tend to have higher impurity levels than water and may tend to obscure the impurities present if the matrix becomes radioactive unless chemical separations are carried out.

Even though neutron activation analysis has essentially no reagent blank problem, there still are many problems inherent in very pure material analysis, especially liquids. The first and foremost one is contamination before or during analysis. This is a problem faced by all purified reagent users; e.g. that of containment without contamination. Virtually all containment materials are less pure than the specially purified material of interest, and thus introduce impurities simply by storage previous to and during analysis. The contamination process can be slowed during storage by cooling or freezing. This procedure may even possibly be used to reduce contamination during irradiation also by irradiating the sample of reagent in the solid (frozen) state and then washing the exposed surface to leave only the uncontaminated inner material for the analysis. A second and perhaps more reasonable approach is to make the storage materials themselves out of specially purified materials. However, this involves a great deal of expense and time but it still may be the best possible solution. Etching and leaching of standard containment materials (such as polyethylene or pure quartz) may reduce some of their impurities to acceptable levels.

The neutron activation analysis technique has additional containment problems during the irradiation that most other techniques do not have. Some of the effects of nuclear bombardment such as nuclear recoil and gamma heating of the sample and container may serve to introduce foreign matter or increase the kinetic rate of exchange of the impurities between sample and container. In addition, since polyethylene and other plastics become damaged by excess radiation, most trace analysis work must be done with such resistant containment materials as high purity quartz. This introduces the problem of removing the sample after irradiation while exposing as little fresh, unetched surface of the quartz to the sample as possible. One way of minimizing this is by providing a breakseal tip to the quartz and using a plastic disposable hypodermic syringe to remove the sample and quickly transfer it immediately after irradiation for counting or chemical separation.

The problem of material decomposition is also a serious one for neutron activation analysis, especially concerning liquid reagents. Virtually all liquids partially decompose during irradiation to form gases. These gases can build up great pressure within the containment vessel and eventually rupture the vessel, possibly even vessels made from quartz. Fortunately, few irradiations require such doses, and the primary danger is the pressure release accompanying the opening of the containment vessel. This can be especially serious if the reagent is an acid or strong caustic. Therefore, precautions must be taken to shield the vulnerable areas from splashing as well as radiation during removal and opening of the vessel. Particular notice should be given to the danger inherent in irradiating hydrochloric acid. Upon irradiation the acid dissociates and recombines to form hydrogen and chlorine, which then reacts explosively upon exposure to visible radiation.

Preliminary work on this study was initiated in the spring of 1965 by one of the authors (G. W. Smith) on the basis of work done by Ralph E. Thiers of Harvard on very pure water and analytical reagent acids and bases. Analyses of the water and reagents were quoted and recommendations made for preparative methods and containers.

In March 1966, a survey of the possible uses of neutron activation analysis for determining subtrace levels of elements in ultrapure water and reagents was made. Based upon expected concentrations, Al, Au, Br, Cl, Cu, Mn, Na and Sr can be analyzed in water at the ppb range and below. Ultrapure quartz (Suprasil from Amersil Quartz Division* of Englehard Industries, Inc.) was obtained for encapsulation and irradiation, which was claimed and found by neutron

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*For disclaimer of equipment and materials see last paragraph of preface.

activation to contain a total impurity level, except for chlorine (37 ppm), of less than 1 ppm.

Some exploratory analysis of several ultrapure reagents are given in table 10. Corrections for blanks were made for each reagent and element. These corrections amounted to $\leq$ 15% of the result.

(D. A. Becker and G. W. Smith)

Table 10. Preliminary analyses of purified reagents

	Concentrations of Impurities (ppb)				
Reagent	Na	Mn	Cu	Br	Au
Water	104 170	0.14			0.014
Nitric Acid	16		7.1	7.0	0.006
Hydrochloric Acid	4.0		2.1	2.6	<0.001

D. Analysis of Impurities in Semiconductor Silicon and Germanium (preliminary work).

The role of semiconductors in the electronic age needs no emphasis. However, the role of unwanted impurities in semiconductor materials may be less well known. The applicability of semiconductors to electronics depends upon the proper introduction of selected "impurities" (dopants) in order to produce a specified result. The presence of unknown or undesirable impurities in the starting materials can and has played havoc with the production of useful electronic components. Neutron activation analysis has been an important technique for the analysis of semiconductor materials [13,14] and will continue to be. Indeed, with the advent of semiconductor particle and gamma-ray detectors made of silicon and germanium, activation analysts can take a very personal interest in the advance of semiconductor techniques.

The Electronic Devices Section of the Instrumentation Division at NBS has been involved in the production and evaluation of various semiconductor devices, including lithium drifted germanium gamma-ray detectors. They, as well as other groups around the country, have had difficulty in the correlation and evaluation of resistivity measurements with detector properties and/or impurity levels. They have asked the Activation Analysis Section to examine the impurity levels of some silicon and germanium samples. They are especially interested in the metals copper, silver and gold, since these impurities seem to affect strongly the carrier lifetimes which is an important quantity in the function of semiconductor devices.

At present, only very preliminary work has been done on this problem, which has consisted primarily of nondestructive analysis using sodium iodide detectors. A great deal of additional work is planned including nondestructive analyses with semiconductor detectors and analyses with chemical separations.

1. Experimental.

The facilities and equipment used for this work have been described earlier in this report, and consisted basically of a 3 in x 3 in NaI(Tl) detector in conjunction with a 400 channel pulse height analyzer.

The irradiations were carried out at the NRL Reactor for 3 hours at a thermal neutron flux of $\sim 6 \times 10^{12}$ n.cm^{-2}s^{-1}. The samples were allowed to decay for at least 16 hours. The surface contamination was removed by etching in a mixture of hydrofluoric and nitric acids before starting the counting. The sample sizes varied up to 1.3 grams for the silicon and 2.5 grams for the germanium. Most samples required considerable additional decay periods before counting could proceed, due to high matrix activities. The counting times varied up to 24 hours depending upon the sample activities. A copper foil flux monitor was irradiated with each sample. The impurity concentrations were then calculated from the copper radioactivity and the nuclear constants of the impurity elements.

2. Results and Discussion.

The preliminary results are presented in table 11. The estimated random error of 20% (standard deviation of the average) is due in large measure to the errors involved with such calculations using nuclear constants.

(D. A. Becker)

Table 11. Preliminary results from the nondestructive analysis of impurities in semiconductor silicon and germanium.

	Concentrations (ppb)[a]		
Sample	Gold	Sodium	Antimony
Silicon (S-1)	1	--	--
Silicon (S-2)	0.8	4	--
Germanium (G-1)	4	--	--
Germanium (G-2)	12	--	93

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[a] Random Error = 20% (standard deviation of the average)

E. Service Analyses.

1. Analysis for Silver in Lead.

The Reactor Radiations Division of NBS had obtained some lead to use as shielding for an experiment. It was pointed out to them when they obtained this lead that it had a probable silver impurity of about 0.8 weight-percent. Since silver absorbs neutrons to form long-lived radioactive products, they wished to confirm the presence and concentration of the silver impurity before going ahead with their proposed usage of this material. The Activation Analysis Section was asked to confirm the presence of the silver and for an estimate (±10%) of the silver concentration. The sample as submitted was a solid chunk about 1/2" in diameter with a dull oxidized surface.

The nuclear reaction $^{109}Ag(n,\gamma)^{110}Ag$ was used for the analysis. Silver-110 has a half-life of 24 sec with a prominant gamma ray of 660 keV. This would allow more than adequate sensitivity for a small sample and excellent specificity due to the very short irradiation time required. The lead matrix does not become significantly radioactive upon a short neutron bombardment. The combination of these factors allowed a simple, rapid, nondestructive analysis to be performed.

a. Experimental.

The facilities and equipment used for this analysis have been described earlier in this report. The standards were aliquots of a freshly prepared silver nitrate solution, encapsulated in polyethylene snap cap vials. The samples were small slivers of the freshly exposed interior portion, taken with a specially cleaned stainless steel knife. Counting of the sample previous to irradiation showed no gamma-ray radioactivity above approximately 200 keV. The sample sizes were about 50 mg each to eliminate gamma-ray self-absorption corrections. The samples were also sealed in polyethylene snap cap vials. The polyethylene vial showed no significant gamma rays in the region of interest after irradiation as described below.

The following procedure was used: each standard or sample was irradiated for 30 seconds at a thermal neutron flux of 8×10^{11} $n.cm^{-2}s^{-1}$, allowed to decay for 45 seconds, and then counted for 1.0 minute clock time (live time varied from ~0.9 to ~0.7 minutes). Additional counts were taken for specific samples to verify the half-life of the silver-110 radioisotope.

b. Results and Discussion.

A plot of the gamma-ray spectrum (counts vs channel number) confirmed that the energy of the prominent gamma ray was 660 keV. Half-life studies verified that the sample and standard half-lives were the same, and closely approximated the literature value of 24 seconds. The silver concentration, as calculated from the data, was 0.74% and 0.73%.

(D. A. Becker and G. W. Smith)

2. Nondestructive Analysis for Impurities in Ultrapure Aluminum Using a Semiconductor Detector (preliminary work).

The NBS Service Analysis coordinator, Dr. John K. Taylor, sent two samples of ultrapure aluminum to the Activation Analysis Section for analysis. They had been submitted to him by Dr. Vincent Arp who is interested in obtaining extremely pure aluminum metal for use as resistivity standards [15].

Application of the resistivity measurement technique to these aluminum samples which were obtained from a commercial concern indicated a higher concentration of impurities present than thought desirable and also indicated a significant variability of the aluminum purity between the samples. If possible, we were to resolve the question of impurities in these aluminum samples and, hopefully, correlate our results with the resistivity measurements.

The aluminum samples were produced by extensive zone refining of a highly purified aluminum starting material. The quoted purity for the aluminum starting material was 99.9999%, and the samples underwent twenty passes of zone refining. Special precautions were taken to prevent contamination before and after the zone refining. These included taking only a selected portion of the refined rod for the measurements. The two samples submitted, (labelled Al-I and Al-II) were in fact somewhat different from each other. Sample Al-II had been melted under vacuum in order to outgas the metal, while sample Al-I had not been outgassed.

The samples came to us with the request to see if we could detect any impurities in the aluminum, and if so, to give a rough estimate (±20% to 30%) of the amount present. Consideration of the nuclear reactions which would result from the neutron bombardment of the aluminum matrix revealed that a large amount of sodium-24 radioactivity would be produced from the fast neutron reaction $^{27}Al(n,\alpha)^{24}Na$ in the irradiation positions available. The sodium-24 radioisotope has a 15.0 hour half-life with 1.37 and 2.75 MeV gamma rays. These high energy gamma rays effectively screen from observation all impurity elements with a half-life of several days or less. This permits only relatively long-lived activation products to be determined nondestructively. In the future, the high pure thermal neutron flux of the NBSR thermal column should allow the nondestructive analysis for many additional elements of shorter half-life. In this case, the limiting factor will be the thermal neutron activation of the aluminum matrix to produce the 2.3 minute aluminum-28.

The use of the semiconductor detector with its inherent high resolution for the gamma-ray counting permits the determination of the gamma-ray energies with high accuracy. This aids materially in the unambiguous identification of the impurities present, as well as permitting good separation of gamma rays with nearly identical energies. At present, we have available for use only a rather small detector (~4cc.) which results in low detection efficiencies. Therefore, after peak identification and assurance that only one gamma ray is present in the region of interest, a 3 in x 3 in sodium iodide (thallium activated) scintillation detector was used for maximum detection sensitivity.

a. Experimental. The samples were received as 2.5 in diameter rods, about 10 in long. A thin disk of about 3 mm thick was cut off one end of each of the samples with a jewelers saw. Samples were then thoroughly etched in concentrated hydrochloric acid, washed several times in distilled, deionized water, and accurately weighed. They were prepared for irradiation by sealing separately in 4 dram polyethylene snap cap vials.

The samples were irradiated several weeks apart to allow for counting one sample before irradiating the second. Each sample was irradiated for 2 hours in the NRL glory tube number 1-A. The thermal neutron flux in this position is about 6×10^{12} $n.cm^{-2}s^{-1}$ with a gold cadmium ratio of about 2.5.

After irradiation and etching to remove surface contamination, the samples were closely observed to determine when the matrix radioactivity had decayed sufficiently to obtain useful information on the impurities present. The actual counts were started after a decay of about 4 days and were taken for 40 days. The length of each individual count was governed by the sample radioactivity, up to a maximum of ~24 hours. A 24 hour background count showed no significant peaks except the potassium-40 peak.

Difficulties were encountered during the counting which precluded the accurate determination of half-life information. These included electronic difficulties, redrifting of the lithium drifted germanium detectors, and construction in the temporary counting laboratories at the NBSR. After ~30-40 days decay, a sodium iodide detector was used to obtain sufficient counts for a quantitative determination of the impurity concentrations of the long-lived impurities found.

b. Results and Conclusions. The results found under the above described conditions are given in table 12. The concentrations quoted are estimated to be ±30% absolute and ±5%

relative to each other. Since it was not known a priori which elements would be found as impurities, no standards were irradiated with the samples. In future work with these aluminum samples, standards of both chromium and scandium will be irradiated along with the samples. Consequently, the high error estimates result primarily from calculations using the nuclear constants.

Table 12. Preliminary results from the nondestructive analysis of ultrapure aluminum

Sample	Element	Conc. (ppm)	Half-life[a]	Gamma-ray Energies(Mev) Experiment	Gamma-ray Energies(Mev) Literature[b]
Al-I	Chromium	0.6	27.8d	0.32	0.32
	Scandium	0.2	84d	0.89 1.11	0.89 1.12
Al-II	Chromium	2	27.8d	0.32	0.32
	Scandium	0.1	84d	0.89 1.12	0.89 1.12

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[a] Half-life quoted is literature value-difficulties with counting equipment precluded determination of experimental half-lives other than approximate values.

[b] Literature values taken from G. E. Chart of the Nuclides, March 1965 edition.

The significantly less chromium found in sample Al-I compared to sample Al-II seems to correlate quite well with the resistivity measurements [15]. The resistivities for samples Al-I and Al-II were quoted as 9000 Ω cm and 200 Ω cm, respectively. The scandium impurity content of both samples is less than the chromium and does not seem to vary between the two samples as much as the chromium. Other than these two elements, sample Al-I had no additional gamma-ray peaks while Al-II had several additional gamma-ray peaks which could not be positively identified due to lack of half-life information.

Additional work on these and/or subsequent samples will likely be necessary. The coming operation of the NBSR and its relatively high flux thermal column will permit much additional work to be done nondestructively, while some destructive analyses may be done if circumstances warrant the additional effort. A further sample of very high resistivity (ca. 24,000 Ω cm) may become available for analysis to aid in the evaluation of the resistivity criterion.

(D. A. Becker and P. D. LaFleur)

F. Miscellaneous.

1. Determination of Neutron Flux Distribution in the NRL In-core Irradiation Facilities.

The need for knowledge of the irradiation positions available for neutron activation analysis in terms of the neutron flux and neutron energy spectrum is sufficiently well accepted to preclude any justification. Previously, in our work at the NRL nuclear reactor this information had been determined for the north pneumatic tube facility (see pages 41-8 ref [2]). Recently much more use has been made of the in-core irradiation positions (glory tubes), so it was deemed advisable to examine these positions for relative neutron flux and flux variability within a single position. our work with the pneumatic tube system had shown that its neutron energy distribution followed closely the fission neutron spectrum with significant thermalization occurring due to its position. The position of the glory tubes within the core grid itself would verify the higher gold-cadmium ratio reported by NRL of 2.5 (compared to ~11 in the north pneumatic tube). With that assumption, no further work was done on the neutron energy distribution. The main object was to determine 1) the relative thermal neutron flux between the four glory tubes, and 2) the flux variation between the top and bottom of a single glory tube. It seemed a reasonable assumption that all four glory tubes would have approximately the same flux gradient within each position.

a. Experimental. Copper-lead alloy foil flux monitors were used for all measurements (see page 39 ref [1]). These foils had approximately 0.1% copper, were from the same batch, and were homogeneous to within ± 1% (see page 39 ref [1]). The foils were placed at the bottom of each irradiation position in a polyethylene bucket. In addition, foils were placed at the middle and top of glory tube No. 1A; which is the position that we use most often. Since the samples and standards in any irradiation could be kept vertical with one another, it was not considered necessary to determine the side-to-side variation in neutron flux. However, any other sample orientation could result in difficulty.

All irradiations were for exactly 15.0 minutes, with the majority of the four irradiations occurring at the identical time. The annihilation radiation from the positron decay of copper-64 was counted on a 3 in x 3 in sodium iodide (thallium activated) detector and 400 channel pulse height analyzer. The 0.511 MeV peak was then integrated, background subtracted, and corrected to time zero. In all cases at least 200,000 counts were accumulated in the peak.

b. Results and Conclusions. The results may be found in table 13. It is immediately apparent that there is significant difference between all four in-core irradiation positions, with position number four having the highest flux at its base of approximately 6.6×10^{12} $n.cm^{-2}s^{-1}$. However, it may also be seen that within a single irradiation position, the vertical neutron flux gradient is quite small, with a maximum in the center of the sample holder and a flux drop-off of about 1% in each direction.

(D. A. Becker)

Table 13. Relative and absolute thermal neutron fluxes in the NRL in-core irradiation positions.

Irradiation Position	Relative Neutron Flux[a]	Absolute Neutron Flux[b]
1A Bottom	1.000	$6.13 \times 10^{12} n.cm^{-2}s^{-1}$
1A Middle[c]	1.013	$6.21 \times 10^{12} n.cm^{-2}s^{-1}$
1A Top[d]	0.997	$6.11 \times 10^{12} n.cm^{-2}s^{-1}$
2 Bottom	0.668	$4.09 \times 10^{12} n.cm^{-2}s^{-1}$
3 Bottom	0.887	$5.44 \times 10^{12} n.cm^{-2}s^{-1}$
4 Bottom	1.082	$6.63 \times 10^{12} n.cm^{-2}s^{-1}$

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[a] Relative to 1A Bottom = 1.000

[b] Calculated from nuclear constants for 1A Bottom with the following conditions: σ_{th} of $^{63}Cu(n,\gamma)$ $^{64}Cu = 4.5b$; neglecting $^{65}Cu(n,2n)$ ^{64}Cu $^{65}Cu(\gamma,n)$ ^{64}Cu; detector calibrated with 0.511 MeV position annihilation radiation from NBS calibrated ^{22}Na point source; ^{65}Cu B^+ decay fraction = 0.19; ^{64}Cu half-life = 12.9h.

[c] 1A Middle is 26 mm above bottom of position

[d] 1A Top is 52 mm above bottom of position

2. Modifications in the Metal Foil Flux Monitor Encapsulation.

The advantages of using a copper foil or copper-lead alloy foil as a flux monitor have been described previously (see page 39 ref [1]). The procedure as described used the foils taped to a cardboard or plastic base with cellulose tape. It has become apparent that with a moderate irradiation dose the cellulose tape disintegrates with resultant loss of monitor identification and/or monitor positioning. This new technique to be described permanently seals the foil monitor inside of two thicknesses of approximately 20 mil polyethylene. This flux monitor encapsulation technique provides much greater radiation resistance, can be permanently marked, and results in significantly lower background radiation due to the encapsulation material.

a. Encapsulation Technique. Obtain a supply of thin-walled polyethylene tubing of ~1/2 inch in diameter [16]. This material should then be cut into pieces about 1 inch long, and leached in nitric acid-deionized water for 2-3 hours. Subsequently, the tubing pieces should be thoroughly rinsed in several changes of deionized water with about 1/2 to 1 hour for each change. This procedure removes most of the impurities which become radioactive upon neutron irradiation. Air dry the tubing pieces on clean tissue.

One end of the tubing is sealed to receive the flux monitor foil. To do this, hold the end to be sealed in the flame from a microburner with clean stainless steel or plastic forceps. When both sides of the polyethylene become translucent, remove and quickly squeeze the end together with a pair of stainless steel crucible tongs. (The serrations should be filed off the tong tips to give smooth surfaces). The result is a clean flat even seal with no entrapped air.

This forms a pocket in which the previously weighed foil is placed for sealing inside the polyethylene. The same procedure is followed with the foil inside the polyethylene. It must be remembered to start the sealing from the bottom, in order that the air be expelled as the seal works up. This prevents air pockets from forming around the flux monitor and elsewhere. The result should be a completely sealed irregular flat piece of polyethylene within the flux monitor located somewhere near the center.

To prepare for use, the polyethylene should be trimmed to a convenient size. Since most of our work is done with the samples in 2/5 dram polyethylene snap cap vials, we use 3/16 inch diameter punched foil disks and 3/8 inch diameter punched polyethylene sealed foil flux monitors. The 3/32 inch border surrounding the foil inside the polyethylene is sufficient for a good positive seal. However, no restriction need be set on the foil size or shape, or the finished sealed flux monitor shape. It may be trimmed to any size desired.

The polyethylene may be marked for future identification by notching the edge, using a stain or ink of some sort, or by writing in the polyethylene itself with a sharp, pointed instrument or hot iron. Care should be exercised that the marking method does not add significant contamination to the flux monitor package.

Our experience with this type of flux monitor has been very good. We have used a number of them many times for short irradiations (i.e., several minutes at $\sim 8 \times 10^{11}$ $n.cm^{-2}s^{-1}$) with no apparent degradation of the polyethylene encapsulation.

(D. A. Becker)

3. Gamma-Gamma Coincidence System.

In a previous report from this laboratory a sum-coincidence system was described which used a 400 channel pulse height analyzer (PHA), a coincidence resolver and routing unit (CRRU), and the normal preamplifiers and sodium iodide detectors (see page 19-28 ref [1]). This system was of limited use because both detector windows were controlled from the PHA and thus had to be identical. In this article a coincidence system is described which uses exactly the same equipment but provides effectively a single channel analyzer (SCA) in coincidence with the PHA. In addition, by simply changing two connectors, the SCA window can be seen on the CRT of the PHA and calibrated for the gamma-ray energy region of interest. The SCA uses the discriminators of the CRRU, which leaves the PHA discriminators available for use during the analysis. This means that, if desired, two very narrow windows may be in coincidence to count only a specific radioisotope. With this system the equipment can be quickly switched from normal PHA mode, to gamma-gamma sum-coincidence, to straight gamma-gamma coincidence.

The pictorial diagram of the equipment is shown in figure 15. With the B detector disconnected, the C detector may be connected as shown by the dashed lines to calibrate the window of the SCA. Otherwise the output of detector C is left disconnected. The output of the detector B is preamplified and goes directly into the PHA amplifier input. This provides the standard PHA spectrum. Meanwhile the tenth dynode output of detector C goes to both inputs B and D of the CRRU. This output must go to both inputs because both a coincidence route and an anticoincidence route must be available from the same detector. Each input route, B and D, has a ten turn pot which acts as a lower level discriminator for that particular route. The lower level discriminator of the CRRU route B is set at an energy of, say, 0.80 MeV. Then by putting it in coincidence with the output of detector B, all of the gamma rays above 0.80 MeV seen by detector C will open the coincidence gate of the PHA. Thus the PHA collects any pulses seen by detector B while the coincidence gate is open. In addition, the route D of the CRRU is set at, say, 0.90 MeV, and put in anticoincidence with detector B. As a result, all pulses above 0.90 MeV generate a blocking pulse which closes the coincidence gate and thus does not allow the PHA to see information from detector B. This provides, in effect, a SCA with a window between 0.80 and 0.90 MeV. This means that any gamma ray absorbed by detector C between these two energies opens the blocking gate and allows only coincident pulses from detector B to accumulate in the PHA memory. Other connections are shown in figure 15.

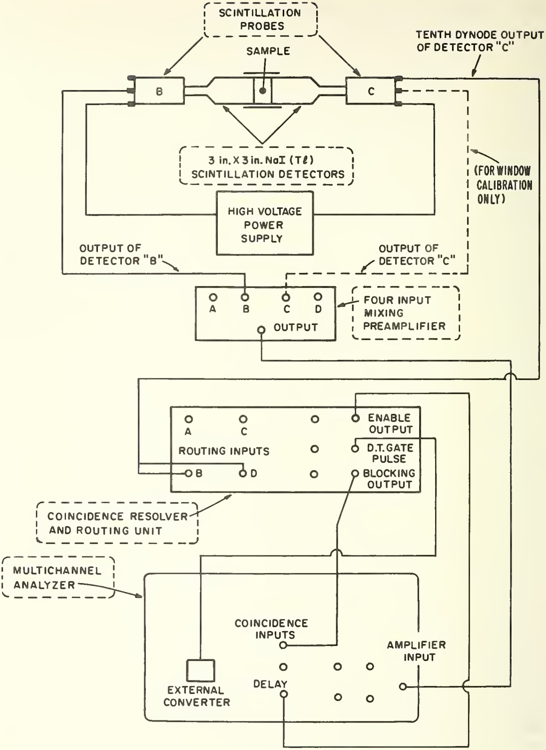

Figure 15. Gamma-gamma coincidence spectrometer.

In order to test this system, the radioisotope bismuth-207 was counted with the SCA set for the 0.569 MeV gamma ray in the presence of large amounts of cobalt-60 and cesium-137 radioisotopes (figure 16). The result shows excellent discrimination between the coincident and non-coincident radiation, and should be very useful in the resolution of complex gamma-ray mixtures. One word of caution, however. The coincidence system is a slow coincidence, and cannot be used with high levels of radioactivity where the random coincidence rate is significant unless the random coincidence rate is measured.

(D. A. Becker and R. W. Shideler)

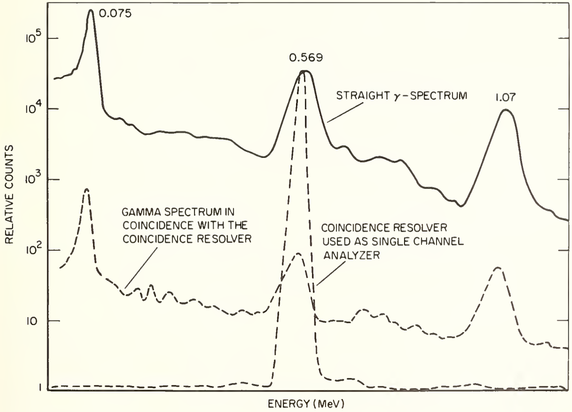

Figure 16. Gamma spectra of Bi^{207} and each of the two coincidence spectra.

5. ACTIVATION ANALYSIS WITH THE ELECTRON LINEAR ACCELERATOR (LINAC)

A. Introduction.

Photon activation analysis is a very useful technique for a variety of elements and samples. There is potential for determining elements such as fluorine, carbon, oxygen, iron and lead at levels below one microgram. Samples with major composition of elements such as lithium, boron, and rare earths, which have very high cross sections for thermal neutron reactions and consequently high self absorption can be analyzed via gamma activation.

The installation of a bremsstrahlung producing target and pneumatic transfer system in the 45 degree arm of the accelerator beam handling system in service area Number 2 has been completed. Experimental efforts have concentrated on carbon, oxygen and magnesium as well as on general aspects of photon activation analysis. These efforts are described below.

B. Facilities.

1. Photon Target Assembly.

The initial design of the photon target assembly is described in the following abstract of a talk presented before the American Nuclear Society meeting at San Diego:

PHOTON ACTIVATION TARGET ASSEMBLY FOR
THE NBS LINEAR ELECTRON ACCELERATOR

by F. A. Lundgren and G. J. Lutz, Trans. Am. Nucl. Soc. June 1967.

One can determine some of the lighter elements with great sensitivity by means of photonuclear reactions induced by the high energy bremsstrahlung from an electron accelerator. This paper describes the activation target assembly for the NBS linear electron accelerator.

Figure 17 shows a cutaway view of the target, rabbit terminal, and beam dump as it appears before the beam. The electron beam enters the target assembly through a .004"

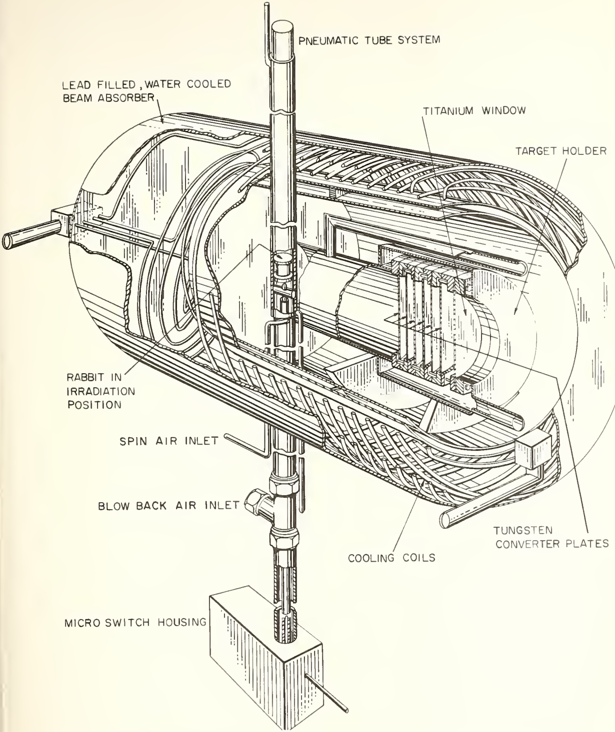

Figure 17. Cutaway view of the target, rabbit terminal, and beam dump as it appears before the beam.

titanium window 5/8" in front of the first converter plate. The converter plates consist of six 0.008 inch thick tungsten foils coated with about 100 microinches of gold to prevent the tungsten from flaking off into the cooling water. For ease of cooling, several thinner target foils are used instead of one thicker plate. Furthermore, the failure of one or two target foils would still allow a good photon flux. The entire target assembly is cooled by flowing water. Three inches of water behind the last target element absorbs any unconverted electrons, thereby preventing heating and bremsstrahlung production in the sample.

The beam dump is designed to contain most of the scattered radiation produced in the target and absorb the photon beam after it passes the terminal. The inside surface is a 1/4 inch cooling jacket which supplies two separate sets of cooling coils in the body of the pump. Two other sets of cooling lines are supplied by the inlet water manifold and are alternately coiled close to the inside surface. The main body of the dump is 19 inches long and 14 inches in diameter filled with chemical grade lead. This gives 3 inches of lead shielding on the sides of the target and 8 inches to the rear.

The sample is packaged in an aluminum rabbit 1.75 inches long and 0.75 inches in diameter. The rabbit can be blown at speeds of up to 200 fps by compressed air into a position directly behind the target assembly. Simultaneously, air is injected tangentially into the rabbit tube at a position about one inch below the rabbit. This spins the rabbit allowing a more homogeneous irradiation and also serves to cool the rabbit.

Flow switches have been incorporated in the exit water lines of the target, beam dump and rabbit tube, and are interlocked into the accelerator trigger circuit so that a flow interruption will turn off the beam to prevent overheating.

The irradiation facility has been tested for about 50 hours, at electron energies in excess of 100 MeV and integrated beam currents up to 300 μA. Examination of the target elements after irradiation showed no visual damage.

A photograph of the target assembly is shown in figure 18. A detailed description of this assembly will be published at a later date. A circular disc of aluminum foil is inserted between the acceleration chamber and the target assembly. The entire disc is coated with a thin layer of zinc sulfide with the exception of a small 30° sector. The phosphor allows location of the beam via a television camera. When the beam is properly located the uncoated sector is

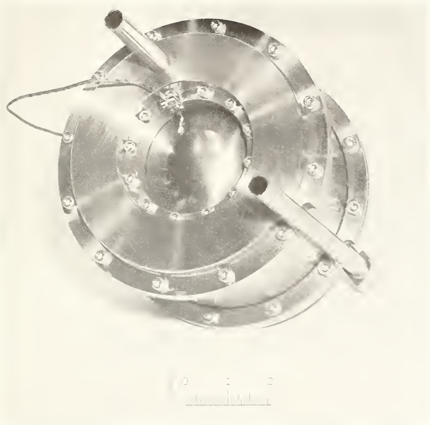

Figure 18. Bremsstrahlung producing target for photon activation analysis.

rotated into the beam. The entire foil is floated above ground potential and a signal is derived which is proportional to current in the beam.

The target assembly produces a large number of neutrons which are moderated in the cooling water and are capable of inducing interfering (n,γ) reactions. About fifty mils of cadmium foil was wrapped around the rabbit terminal to somewhat alleviate this effect.

2. Laboratory Facilities at LINAC.

A control console in our laboratory allows the pneumatic transfer of sample to and from the irradiation facility. The console is shown in figure 19. The aluminum rabbits and inner polyethylene liners along with copper flux monitors used are shown in figure 20. The sending and receiving station for samples is shown in figure 21. Counting facilities consist of a RIDL nanolyzer*, NaI crystal, teletype output and plotter. These are shown in figure 22.

C. Preliminary Evaluations for Elemental Analyses.

1. Reproducibility.

Since it is axiomatic to the performance of an activation analysis by the comparator method that the relative amount of irradiation of sample and standard be known, a large number of experiments have been performed to assess the reproducibility of irradiations. Although over a period of several hours, the intensity of the photon beam may vary, the position and energy are very stable. This is shown in Table 14. The samples were magnesium standards in which ^{24}Na had been induced via the reaction ^{25}Mg$(\gamma,p)^{24}$Na. The flux monitors were copper discs. All irradiations were for two minutes and they were carried out over a period of about four hours. It will be noted in columns one and two (table 14) that the beam

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*For disclaimer of equipment and materials see last paragraph of preface.

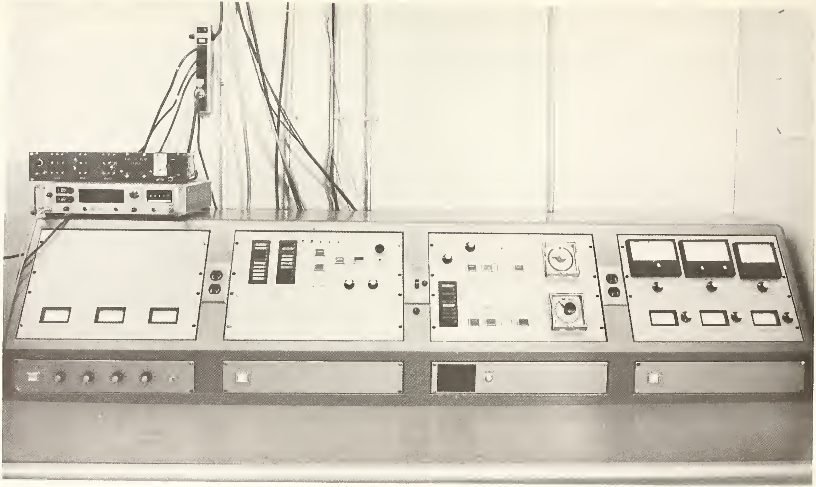

Figure 19. Operating console in service area number 2 of the Radiation Physics Building.

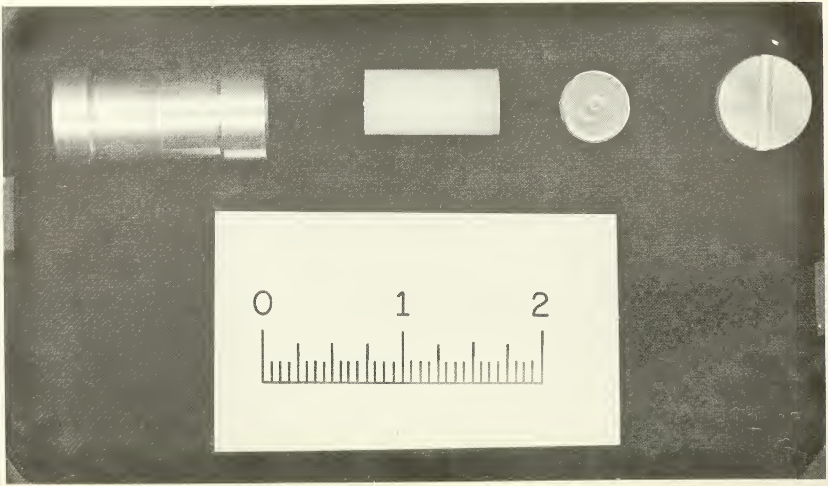

Figure 20. Aluminum rabbit with polyethylene liner and copper flux monitor. Scale in inches.

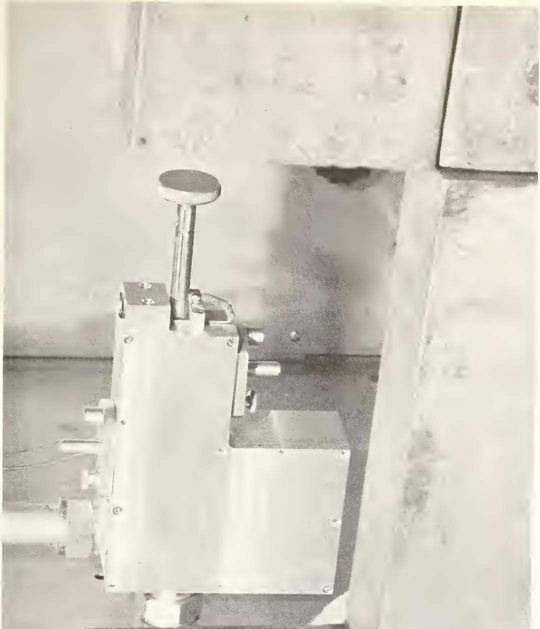

Figure 21. Sending and receiving station for samples.

Figure 22. Counting facility in service area 2 of the Radiation Physics Building.

Table 14. LINAC reproducibility runs.

I cpm/g[a] of Mg at end of irradiation	II cpm/mg[b] of flux monitor at end of irradiation	III Ratio of I to II
6982	99.8	69.9
7027	100.8	69.7
7795	116.7	66.8
5568	82.7	67.3
5044	72.1	70.0
4513	64.2	70.3
	Average	69.0
Standard Deviation of Average		1.4

---

[a] Counts per minute per gram

[b] Counts per minute per milligram

intensity varied substantially over the period of the irradiations. However, column three, which is the ratio of column one to column two, is quite constant.

2. Beam Distribution Over Length of Rabbit.

Experiments were conducted to determine the distribution of the photon beam over the length of the rabbit. Polyethylene discs, 1/16 inch thick and of approximately the inside diameter of the aluminum rabbit were stacked to fill the rabbit, irradiated and counted. Table 15 shows the distribution along the length of the rabbit. Counting rates were corrected for decay and for slight variations in weight of the discs, and were normalized to 100 for the disc with the highest specific activity. The data are for 34 MeV electron energy. Normally at the beginning of each irradiation period a check of contour is made and the beam is slightly repositioned to exhibit the form of table 15.

Table 15. Distribution of photon beam produced by 34 MeV electrons

	Position No.	Intensity Index
TOP	1	85
	2	89
	3	91
	4	94
	5	94
	6	97
	7	98
	8	100
	9	99
	10	98
	11	97
	12	97
	13	95
	14	94
BOTTOM	15	91

3. Magnesium.

Magnesium can be determined via the reaction $^{25}Mg(\gamma,p)$ ^{24}Na. Interferences include sodium via the reaction $^{23}Na(n,\gamma)$ ^{24}Na and aluminum via the reaction $^{27}Al(n,\alpha)^{24}Na$. The extent of these interferences with our current target assembly is shown in Table 16. Since the ratio of ^{24}Na produced from the interferences to that produced from magnesium varies with irradiation energy, irradiations at different energies will allow a magnesium determination in the presence of sodium or aluminum if they are not present in excessively large amounts. Typical results obtained are those of National Bureau of Standards SRM 342, a nodular cast iron. Magnesium is certified in this material at 0.053%. Triplicate analyses by photon activation gave 0.053, 0.051 and 0.052%. We are currently working on the certification of magnesium in a number of Standard Reference Material irons.

4. Oxygen.

The $^{16}O(\gamma,n)^{15}O$ reaction yields an isotope with a half-

Table 16. Relative sodium-24 production from magnesium, aluminum and sodium

Electron Energy (MeV)	Ratio of ^{24}Na induced in Al to that induced in Mg	Ratio of ^{24}Na induced in Na without Cd shielding to that induced in Mg	Ratio of ^{24}Na induced in Na with 30 mils Cd shielding to that induced in MG
25	8.66×10^{-3}	5.62×10^{-1}	2.99×10^{-1}
30	1.31×10^{-2}	1.94×10^{-1}	7.94×10^{-2}
35	1.71×10^{-2}	1.09×10^{-1}	2.56×10^{-2}
40	2.30×10^{-2}	1.05×10^{-1}	2.38×10^{-2}
47	4.55×10^{-2}	7.50×10^{-2}	7.57×10^{-3}

NOTE: All measurements on equal weight basis.

life of 124 seconds, which decays by β^+ emission. Many elements, upon photon irradiation, yield positron emitters so the measurement of trace amounts of ^{15}O in a typical plethora of other activities is generally not practical.

An attempt was made to determine oxygen using the reaction $^{16}O(\gamma,2n)^{14}O$. This has a threshold energy of 28.9 MeV. The cross section is at a maximum at 40 MeV. The product isotope has a half-life of 72 seconds and decays mainly by positron emission in cascade with a 2.3 MeV gamma ray.

It was hoped that one could make oxygen determinations by following the half-life of the activity under the 2.3 MeV photo peak.

Irradiations were made at 55 MeV and a beam current of about 10 μamps. For a 30 second irradiation about 120 total counts per mg of oxygen can be obtained "counting the sample to death". Substantial interference from the sample matrix was also observed causing coincidence losses and pulse pile up.

5. Carbon.

Early preliminary experiments with the linear accelerator indicated that the intrinsic sensitivity for carbon would be very low--about 10^{-7} grams. The reaction involved would be $^{12}C(\gamma,n)^{11}C$. ^{11}C has a half-life of 20.5 minutes and decays by positron emission. This means that in most samples a chemical separation would be required.

Our first efforts in carbon analysis have been determinations of carbon in sodium. Analysis of sodium for carbon is of interest in fast breeder reactor technology.

The main irradiation products of sodium are given in table 17. The last reaction is induced by neutrons produced in the target.

The separation technique used in similar to that described by Kallman and Liu [17]. The sodium sample, after irradiation, is etched in an ethanol-water mixture to remove

Table 17. Nuclear reactions of interest
of carbon and sodium.

Reaction	Half-life	Threshold Energy (MeV)
$^{12}\mathrm{C}(\ ,n)^{11}\mathrm{C}$	20.5 min	18.7
$^{23}\mathrm{Na}(\ ,n \ )^{18}\mathrm{F}$	110 min	20.9
$^{23}\mathrm{Na}(\ ,n)^{22}\mathrm{Na}$	2.58 y	12.1
$^{23}\mathrm{Na}(n, \ )^{24}\mathrm{Na}$	15h	

surface contamination. After scraping the bulk of the alcohol residue from the sample, it is placed in a silica crucible with about 100 mg of Na_2CO_3. The crucible is placed in a quartz combustion flask connected to a gas absorption bottle. The sample is heated in a stream of oxygen to about 800°C. The sodium is converted to sodium oxide and the carbon to sodium carbonate. After cooling, 50 ml of 3.5N sulfuric acid is added to the combustion flask and the liberated Co_2 is absorbed in a potassium hydroxide solution in the gas absorption bottle. After diluting the solution to a constant volume, the ^{11}C radioactivity is measured with a sodium iodide detector and multichannel analyzer.

Experiments with irradiated sodium carbonate and graphite added to inactive sodium indicate that the separation is quantitative. The half-life of the separated activity was found to be in good agreement with the value generally accepted.

(G. J. Lutz, F. A. Lundgren
and D. A. DeSoete)

6. PERSONNEL AND ACTIVITIES

A. Personnel Listing

Activation Analysis Section
- J. R. DeVoe, Section Chief (1)
- P. A. Snyder, Secretary (1)

Nuclear Reactor Project
- P. D. LaFleur, Project Leader (1/2)
- E. D. Anderson (1)
- D. A. Becker (1)
- B. S. Carpenter - Transferred (1/2)
- T. E. Gills (2/3)
- W. D. Kinard (1)
- F. A. Lundgren (1)
- L. T. McClendon (2/3)
- W. P. Reed (1)
- G. W. Smith (1)

Cockcroft-Walton Generator Project
- S. S. Nargolwalla, Project Leader (1)
- P. Black (1/3)
- M. R. Crambes (1)
- J. E. Suddueth (1/12)

Linear Accelerator Project
- G. J. Lutz, Project Leader (1)
- D. A. DeSoete (2/3) 9 months NATO Fellowship,
  3 months NBS contract
- D. M. Setlock (1/4)

B. Publications

G. W. Smith, D. A. Becker, G. J. Lutz, L. A. Currie and J. R. DeVoe, "Determination of Trace Elements in Standard Reference Materials by Neutron Activation Analysis", Anal. Chim. Acta, 38, 333-340 (1967).

C. List of Talks

J. R. DeVoe, Chairman, Session on Nuclear Methods, Institute for Materials Research Symposium on Trace Characterization, October 1966.

J. R. DeVoe, "Neutron Activation Analysis", American Section of Society for Applied Spectroscopy, Cincinnati, Ohio, November 1966.

D. A. Becker, "Neutron Activation Analysis for Aluminum, Selenium and Vanadium in NBS Standard Reference Material Steel Alloys Utilizing Short Lives Radioisotopes", American Chemical Society, Miami, Florida, April 1967.

G. W. Smith, "The Preparation of an NBS Biological Standard Reference Material for Trace Elemental Analysis", International Atomic Energy Agency Symposium on Nuclear Activation Techniques in the Life Sciences, Amsterdam, The Netherlands, May 1967.

S. S. Nargolwalla, "Applicability of 14 MeV Neutron Activation Analysis for Biological Materials", International Atomic Energy Agency Symposium on Nuclear Activation Techniques in the Life Sciences, Amsterdam, The Netherlands, May 1967.

G. J. Lutz, "Photon Activation Target Assembly for the NBS Linear Electron Accelerator", American Nuclear Society, San Diego, California, June 1967.

S. S. Nargolwalla, "Elemental Analysis of Proteins by 14 MeV Neutron Activation", American Nuclear Society, San Diego, California, June 1967.

7. ACKNOWLEDGMENTS

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We wish to express our thanks to Dr. C. O. Muehlhause and his staff for their cooperation in making arrangements for our use of the new facilities in the NBS Reactor Building. Further the assistance of Dr. J. E. Leiss and staff of the Radiation Physics Division for arrangements to use the LINAC facilities is gratefully acknowledged. The assistance of Mr. Goetz, Acting Chief, as well as Mr. H. J. Rouse, Mr. T. B. McKneely, Jr., and Mr. M. F. Kelly of the Plant Division NBS is also acknowledged. We also wish to thank J. Cameron and D. Hogben for assistance in the statistical analysis of experimental data.

We also wish to express our appreciation to Dr. Laverlochere, AEE, Grenoble, France and Professor J. Hoste, University of Ghent, Belgium for making possible the visit of Doctors Crambes amd DeSoste, respectively.

This section acknowledges Mrs. R. S. Maddock for her assistance in preparing this report.

Special appreciation is expressed to Mrs. P. Snyder who typed the entire report and whose tireless efforts as section secretary has made our entire operation more efficient and effective.

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