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Activation Analysis with Charged Particles

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Activation Analysis with Charged Particles

by

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Tuxedo, New York

Subcommittee on Radiochemistry
National Academy of Sciences—National Research Council

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FOREWORD

The Subcommittee on Radiochemistry is one of a number of subcommittees working under the Committee on Nuclear Science within the National Academy of Sciences - National Research Council. Its members represent government, industrial, and university laboratories in the areas of radiochemistry and nuclear chemistry. Support for the activities of this and other subcommittees of the Committee on Nuclear Science is provided by a grant from the National Science Foundation.

The Subcommittee has concerned itself with preparation of publications, encouraging and supporting activities in nuclear education, sponsoring symposia on selected current topics in radiochemistry and nuclear chemistry, and investigating special problems as they arise. A series of monographs on the radiochemistry of essentially all the elements and on radiochemical techniques is being published. Initiation and encouragement of publication of articles on nuclear education in various subject areas of chemistry have occurred, and development and improvement of certain educational activities (e.g., laboratory and demonstration experiments with radioactivity) have been encouraged and assisted. Radioactive contamination of reagents and materials has been investigated and specific recommendations made.

This series of monographs has resulted from the need for comprehensive compilations of radiochemical and nuclear chemical information. Each monograph collects in one volume the pertinent information required for radiochemical work with an individual element or with a specialized technique. The U. S. Atomic Energy Commission has sponsored the printing of the series.

Comments and suggestions for further publications and activities of value to persons working with radioactivity are welcomed by the Subcommittee.

N. E. Ballou, Chairman
Subcommittee on Radiochemistry

ABSTRACT

A review of charged-particle activation analysis is presented.

Charged particles are compared and contrasted with uncharged particles for activation analysis and a brief discussion is given of the machines available for particle acceleration. Thick and thin targets are compared followed by the specific use in activation analysis of protons, deuterons, helium-3 ions, alpha particles, tritons and recoil particles in a nuclear reactor.

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Activation Analysis with Charged Particles

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1. Introduction

Activation analysis is a method of elemental analysis in which nuclei are made radioactive by irradiation in a suitable source of activating particles and subsequent (or simultaneous measurements are made of the induced activities). Any means of inducing radioactivity (neutron, photon or charged particle irradiation) can be used for activation analysis. By far the most extensive use of activation analysis has been made by application of nuclear reactor produced thermal neutrons. This is due partly to the large cross-sections and high specificity of (n, γ) reactions, and the availability in nuclear reactors of a high neutron flux which gives rise to the high sensitivity of the method. The success of neutron activation analysis is also due in part to the fact that the neutron, being uncharged, passes relatively freely through most samples. Thus, large amounts of sample can be analyzed for trace constituents distributed throughout the sample matrix.

A serious limitation of activation analysis using reactor neutrons is that the method does not work well for elements lighter than neon (atomic number, $Z=10$). This is because elements of Z less than 10 have small cross-sections for (n, γ) reactions, or the reactor induced activities are either too short lived or too long lived to be measured conveniently. Other methods of inducing radioactivity include irradiation with high energy (e.g. 14 MeV) neutrons, photons or charged particles.

This monograph will confine itself to activation analysis using charged particles. For applications using high energy neutrons or photons, reference may be made to review articles by Sayre (1) or Bowen and Gibbons (2). Previous workers who have used high energy neutrons, photons, or charged particles have concentrated on elements

of low atomic number, which are difficult or impossible to determine by reactor neutrons. It should be remembered, however, that activation by high energy charged particles is by no means restricted to the low atomic number elements; in fact these reactions tend to have larger cross-sections with increasing atomic number. The application of charged-particle activation analysis to elements of high atomic number has been successfully demonstrated in a few striking examples that will be described later.

There have been several previous review articles and discussions of specific aspects of charged particle activation analysis: Odeblad (3), Albert (4), Sippel (5), Gill (6), Winchester (7), Bowen and Gibbons (2), Heslop (8), and Sayre (1). Bate (9) has reviewed the nuclear methods of oxygen analysis and Koch (10) has listed data on possible methods of charged particle or photon activation analysis.

2. ACTIVATION ANALYSIS IN GENERAL

For the nuclear reaction

where M_1 is a stable nuclear isotope and M_2 is the radioactive product that decays with a decay constant λ , the net rate of production of M_2 over a small time interval dt is given by

$$\frac{dN_2}{dt} = R - N_2 \lambda \quad (1)$$

where N_2 is the number of atoms of M_2 at any time during the irradiation, $N_2 \lambda$ is the rate of decay of M_2 and R is the rate of production of M_2 from M_1 . For a machine such as a nuclear reactor or cyclotron,

$$R = N_1 \sigma \phi$$

where N_1 is the number of target atoms, σ is the cross-section for the nuclear reaction $M_1 \longrightarrow M_2$ and ϕ is the flux of particles striking the target. N_1 , σ and ϕ must all remain essentially constant for R to be a constant.

For a nuclear reactor the neutron flux is measured as neutrons/cm².sec. and N_1 is the total number of target atoms. For a cyclotron the particle flux is usually measured as beam current in microamperes which may be converted to particles/sec since $1 \mu A = 6.24 \times 10^{12}$ positive charges/sec, and N_1 is expressed as the number of target atoms/cm².

Rearranging, equation (1) becomes

$$\frac{dN_2}{dt} + N_2\lambda = R \quad (3)$$

This is a first order linear differential equation whose solution is (ref. 11):

$$N_2 e^{\lambda t} = \frac{R}{\lambda} (e^{\lambda t} - 1) \quad (4)$$

$$\text{or} \quad D_2^t = N_1 \sigma \Phi (1 - e^{-\lambda t}) \quad (5)$$

where $D_2^t (= \lambda N_2)$ is the disintegration rate of M_2 at any time t during the irradiation. After the irradiation, the activity of M_2 at a decay time T is given by

$$D_2 = N_1 \sigma \Phi (1 - e^{-\lambda t}) e^{-\lambda T} \quad (6)$$

where t is the duration of the irradiation.

An examination of the saturation factor $(1 - e^{-\lambda t})$ shows that it is usually unprofitable to irradiate a material for more than one or two half-lives of the product activity. Irradiation for two half-lives gives 0.75 of the maximum activity possible while irradiation for seven half-lives gives 0.992 of the maximum activity, an increase in activity of only 32%. The relationship between induced activity and irradiation time is approximately linear for irradiations of less than one half-life.

To avoid measuring the cross-section (σ) and the flux (Φ) during an irradiation for the purpose of activation analysis, it is usual to irradiate the sample and a standard in the same irradiation

position. The counting rates of a particular radionuclide induced in the sample and the standard are then compared for the same conditions of irradiation and decay. The weight of element in the sample W_x is calculated from the relation

$$W_x = W_s \cdot \frac{C_x}{C_s} \quad (7)$$

where W_s is the weight of the element in the standard and C_x and C_s are the measured counting rates of the sample and the standard respectively.

The activation equation (6) shows that the sensitivity of the method is directly proportional to the activation cross-section and to the flux. The half-life of the radioactivity produced affects the sensitivity in two ways. Firstly it predetermines the length of irradiation necessary because it occurs in the saturation factor $(1-e^{-\lambda t})$. Secondly, it limits the time available after bombardment for chemical separations or other manipulations before the radioactivity is measured.

3. Charged Versus Uncharged Particles for Activation Analysis

Neutrons and photons, being uncharged, penetrate deeply into matter and, in most cases, activate quite large samples of material. This property increases the sensitivity of the method with respect to charged particle activation analysis, assuming the same cross-section. However, it can also cause the production of large amounts of interfering or unwanted activity. Gram-sized samples irradiated in nuclear reactors, or with 14 MeV neutrons from the deuterium-tritium reaction, are subject to the serious effects of self-shielding. This can be partially overcome, by spinning the targets (both sample and standard) (12). In thermal neutron activation, there is also the problem of trace impurities having very high cross-sections. Charged-

particle reaction cross-sections in cyclotrons (where the energy resolution of the beam is poor) do not show serious resonances, and the cross-sections are about the same. Charged particle reactions using a Van de Graaff accelerator do show resonances for the light nuclei, but not as large as reactor neutron resonances. Also, the reactor flux changes from position to position both with respect to total flux and the flux spectrum.

Charged particles are stopped in small thicknesses of solids. The depth of penetration can best be calculated from tabulations given by Sternheimer (13) or Bichsel (14), and Figure I shows the ranges of charged particles of energies from 10 to 40 MeV in aluminum. The range decreases with increasing charge of the incident particle and with increasing atomic number of the stopping material. At the same velocity (not energy) the range of heavy ions in a particular material is nearly proportional to m/Z^2 where m is the mass and Z the charge of the ion. It is a simple matter to approximate the range-energy relation for He^3 or alpha particles from that for protons in the same material (16). This small penetration (0.002 inches for an 8 MeV He^3 ion in aluminum) can be advantageous for analyzing surfaces, and presents no problem if the sample is homogeneous. Materials like aluminum which have a superficial oxide layer may be etched after irradiation to remove the surface layer, thus allowing the oxygen content of the sub-surface material to be measured without interference from the high concentration of oxygen on the surface.

The small depth of penetration and relatively high kinetic energies of the bombarding particles results in the production of large amounts of heat when the charged particles are stopped. A 10 MeV beam of 10 microamp will deposit 100 watts of power in a thick target. Thus

FIGURE I. CHARGED PARTICLE RANGES IN ALUMINUM (15)

targets should be preferably non-volatile solids, good thermal conductors and not easily decomposed by heat, or must be suitably cooled. Water cooling is adequate in most cases. Metals and refractories make the best targets for charged particle activation. However, Fleckenstein, et.al. (17) have used 4 MeV protons to analyze for O^{18} in complex organic molecules by cooling with solid CO_2 , and Sile (18) has developed a technique using deuterons to analyze organic materials for carbon.

In general, increasing the energy of the bombarding particle increases the number of possible nuclear reactions. This is true for charged or uncharged particles. For charged particles approximately mono-energetic beams are available, and by controlling the energy of the bombardment the products of the reaction can be controlled. Uncharged particles of high energy are more difficult to obtain as mono-energetic beams.

4. Machines for Particle Acceleration

There are a number of methods of accelerating charged particles. It is not intended to give here anything more than a few comments on the use of these machines for activation analysis. The reader is referred elsewhere for detailed descriptions of these particle accelerators such as the excellent review of McMillan (19).

The Van de Graaff machine can be used to accelerate charged particles of all types up to a maximum voltage, which is ultimately limited by the voltage breakdown of the insulating gas. This voltage is about 10 million volts for a single and 20 million volts for a tandem machine. It can be operated at any energy up to the maximum, and the energy definition of the beam can be controlled within very small limits (a few keV).

The cyclotron can be used to accelerate a wide range of charged particles over a wider range of energies than the Van de Graaff machine, but the energy definition is not as good (about ± 2 MeV at 50 MeV). The simple cyclotron has an upper energy limit of about 30 MeV for protons; quite high enough for charged-particle activation analysis or isotope production, and has a much larger beam current than the synchro-cyclotron, which is used for producing higher energy particles. A large beam current is an advantage providing heating effects do not become troublesome. Irradiations can be performed in the internal circulating beam of the cyclotron by inserting the target between the "dees" on the end of a probe. The target can be irradiated with particle beams of different energy by varying the distance of the target from the center of the cyclotron. In addition, irradiations can be performed external to the cyclotron (outside the vacuum chamber) by causing the beam to be deflected from its circular path along an evacuated beam tube and through a window onto the target. The beam current is reduced to between 1% and 25% compared to that in internal irradiations and the energy of the external beam can be degraded by means of absorbers.

The linear accelerator can be used to accelerate ions of the same charge-to-mass ratio (e.g. H^2 , He^4 , Li^6 , B^{10} , C^{12} , N^{14} , O^{16} , etc.), when an appropriate ion source is available and with proper adjustment of the radiofrequency system, although with suitable adjustment "HILACS" can be made to accelerate almost anything. The energy of a particular particle beam is fixed for a given machine. Lower energies can be achieved by using absorbers to degrade the energy of the particle beam, but this reduces the sharp energy definition of the beam, or a portion of the machine can be turned off and particles allowed to drift through.

Irradiations are usually performed through a thin window of metallic foil.

The three charged-particle accelerators mentioned above may have fluctuations in the beam current of as much as 20%, so that it is essential to monitor the beam during the irradiation. This may be done by means of a Faraday cup; but, for activation analysis, it is better to irradiate a monitor containing a known amount of the element to be measured at the same time as the sample of unknown composition. The monitor and sample can then be treated in the same way after the irradiation, and the ratio of activities can be used to determine the composition of the sample. The monitor must be either thin enough to reduce the energy of the particle by only a small amount if it is to be interposed between the sample and the beam, or the monitor (and/or the target) can be rotated through the beam a known number of times.

The simple cyclotron is the most useful for charged-particle activation analysis, from the point of view of versatility and economy, and proposals have been made to design and build a small (30 inch diameter) machine for activation analysis (57). Such a machine could also be used as a high energy neutron or photon source, because high energy charged particles produce high energy neutrons and bremsstrahlung radiation when stopped in matter.

5. THICK VERSUS THIN TARGETS

A thin target may be defined as one that reduces the energy of the bombarding particle beam by a negligible amount. Such targets are ideal for charged-particle activation analysis. They may be foils or be prepared by vacuum evaporation of materials onto a non-reacting backing material (such as tantalum metal) or by evaporation of solutions on a suitable support. Oxide or other films on metals are also excellent as thin targets for charged particle activation analysis.

Thick targets are those that significantly reduce the energy of the bombarding particles or completely absorb the bombarding particle beam. The energy of the bombarding particle beam varies with depth of penetration, and the cross-section for the nuclear reaction of interest varies with bombarding energy. In general, the excitation functions for charged particle reactions have the features seen in figure 2. The cross-section increases from zero (or a very small value) at the threshold energy (E_t) to a maximum as the kinetic energy approaches the Coulomb barrier energy. The threshold of the reaction is thermodynamically equal to the Q value, which may be positive or negative. The cross-section for a particular nuclear reaction (σ_a) then decreases as more complex nuclear reactions compete. However, the total cross-section for all reactions (σ_t) continues to increase. In some cases of low-energy nuclear reactions, resonances occur when the compound nucleus is formed at an excitation energy in the vicinity of a nuclear energy level. Therefore, the composition and density of thick targets used for charged-particle activation analysis, both sample and standard, should be as much alike as possible.

FIGURE 2. GENERAL FEATURES OF THE VARIATION OF CROSS-SECTION WITH BOMBARDING ENERGY FOR A CHARGED PARTICLE REACTION

Sippel and Glover (5), and Odeblad (3), have discussed the effect of stopping power of thick targets in charged-particle activation analysis. The change in energy E with distance of penetration x (i.e. the stopping power), in matter for non-relativistic charged particles is given by the expression (20)

$$-\frac{dE}{dx} = \frac{4\pi e^4 z^2}{m_0 V^2} NZ \ln \frac{2m_0 V^2}{I} \quad (8)$$

$$= (k/E) \ln(E/I) \quad (9)$$

where ez is the charge of the bombarding particle, V is its velocity, m_0 is the rest mass of the electron, N and Z are the number and charge of the target atoms, and I is the mean ionization potential of the target atoms. This equation is strictly valid only for high-energy particles. At low energies, corrections must be made for interaction with atomic electrons. However, the expression is reasonably accurate for protons down to 0.1 MeV. The mean ionization potential, I , is obtained experimentally and varies from 15.6 eV for hydrogen to 276 eV for copper. Calculations of the stopping power of different elements have been made for charged particles (13, 14). These data may be used to calculate the stopping power of compounds by summing the stopping powers of the component elements, taking into account the partial density of each elemental component.

Sippel and Glover found by calculation that the mass stopping powers of the common minerals in sedimentary rocks for 2, 1, and 0.75 MeV protons, were constant to within 5% for each bombarding energy, and concluded that the effects of stopping power need not be considered further in connection with sedimentary rock analysis. They found that it was necessary for the rocks to be finely crushed, so that a representative homogenous sample could be taken (5).

Ricci and Hahn (21) have introduced an average cross-section, $\bar{\sigma}$, in considering the activation of thick targets by charged-particle bombardments. They define

$$\bar{\sigma} = \frac{\int_E^0 \sigma_e (dx/dE) dE}{\int_E^0 (dx/dE) dE} \quad (10)$$

where σ_e is the energy dependent cross-section, and insert the expression for stopping power for the non-relativistic case (equation 9). Then

$$\bar{\sigma} = \frac{\int_E^0 \sigma_e E [k \ln(E/I)]^{-1} dE}{\int_E^0 E [k \ln(E/I)]^{-1} dE} \quad (11)$$

For a given target, I is constant, so the term $\ln(E/I)$ varies slowly with energy, or $k \ln(E/I)$ is approximately constant.

Therefore:

$$\bar{\sigma} \approx \frac{\int_E^0 \sigma_e E dE}{\int_E^0 E dE} \quad (12)$$

To a good approximation, $\bar{\sigma}$ is independent of any properties of the target material, and is constant for a given nuclear reaction and a fixed bombarding energy. Ricci and Hahn tested these approximations for the $O^{16} (He^3, p) F^{18}$ reaction by calculation from the experimental results of Markowitz (22) and Demildt (23), and found $\bar{\sigma}$ changed by only 8% from $Z=4$ to $Z=95$, and changed by only 3% for Z values from 4 to 57. These findings, Ricci and Hahn claim, reduce charged particle activation analysis to the simplicity of neutron activation analysis.

6. Charged-Particle Activation

General Discussion - The binding energy of a neutron or proton , in all except the lightest elements (up to boron), is approximately $8\frac{1}{2}$ 0.5 MeV). The lighter nuclei have smaller binding energies. Thus, if a nucleus absorbs a particle containing x nucleons, it will increase its energy by $8x$ MeV less the binding energy of the bombarding particle. In this way we can crudely calculate the energy gain of the target nucleus when it takes up a charged particle; this is shown in Table I.

From this table, we see that the most efficient charged particles for activation are deuterons and tritons as these give the maximum excitation energy to the target nucleus for the smallest bombarding energy. He^3 and Li^7 nuclei provide slightly more energy but require twice and three times as much kinetic energy to overcome the Coulomb barrier to entering the nucleus, which is proportional to the nuclear charge of the bombarding particle.

The above method of calculating the energy gain of the target nucleus is only a rough approximation. For specific target elements it is possible to calculate the Q of the reaction from the mass difference of reactants and products, and tables of reaction Q values have been published (24). An example of the Q values of some simple nuclear reactions on oxygen-16 is given in Table II (the small numbers are the errors associated with the Q values). This data confirms the conclusion reached above that the most efficient charged particles for activation are deuterons, tritons, and helium-3 ions, since the reactions they induce have higher positive Q values. The reactions involving the emission of a gamma-ray from the compound nucleus are not as useful as might be thought from an examination of this table, even though the "gamma-out" reactions have the highest Q values. This is because the

TABLE I - APPROXIMATE ENERGY GAIN OF TARGET NUCLEUS
for CHARGED PARTICLE NUCLEAR REACTIONS (Ref.2)

Particle	No. of Nucleons	Nuclear Charge	Binding Energy (MeV)	Energy Gain of Target Nucleus (MeV)
p	1	1	-	8
d	2	1	2.2	13.8
t	3	1	8.5	15.5
He ³	3	2	7.7	16.3
α	4	2	28	4
Li ⁷	7	3	39	17
C ¹²	12	6	91	5
N ¹⁴	14	7	105	7
O ¹⁶	16	8	127	1

TABLE II - REACTION Q VALUES IN MEV FOR NUCLEAR
REACTIONS ON OXYGEN-16, Ashby and Catron (23)

OUT IN	γ	N	P	D	T	He ³	He ⁴
γ		-15.6533 .0031	-12.1109 .0045	-20.7216 .0039	-25.0102 .0062	-22.7702 .0057	- 7.1504 .0041
N	4.1471 .0039		- 9.6029 .0133	- 9.8848 .0052	-14.4633 .0053	-14.6004 .0053	- 2.2022 .0046
P	.6004 .0042			-13.4272 .0058	-20.3865 .0088	-15.2267 .0053	- 5.2056 .0052
D	7.5432 .0094	- 1.6257 .0049	1.9210 .0048		- 9.3950 .0069	- 6.6160 .0065	3.1152 .0044
T	11.6868 .0082	1.2849 .0101	3.7371 .0101	- 2.1112 .0062		-10.3663 .0144	7.6937 .0063
He ³	8.4270 .0095	- 2.9550 .2001	2.0483 .0101	- 4.8945 .0063			4.9147 .0067
He ⁴	4.7564 .0092	-12.1410 .0089	- 8.1178 .0075	-16.2936 .0096	-19.2042 .0060	-16.4209 .0059	
TABLE NO. 35				MASS EXCESS & ERROR	00.0000 .0000	TARGET NUCLIDE	8 16 Z A

Note: These data are calculated on the atomic mass scale

O¹⁶ = 16 atomic mass units. New convention is to take

C¹² = 12 atomic mass units.

Coulomb barrier to the entry of a charged particle requires that the compound nucleus be formed in an excited state, so that sufficient energy is available for the emission of a proton or an alpha particle; gamma-ray emission at this degree of excitation is improbable due to the high angular momentum transfers required, and the strong competition from particle emission.

The Coulomb barrier (in MeV) to the charged particle entering the nucleus is given by the expression (see appendix and (25)).

$$B = \frac{0.96 Z_1 Z_2}{A_1^{1/3} + A_2^{1/3}} \quad (13)$$

where Z is the nuclear charge and A is the mass number and the subscripts 1 and 2 refer to the projectile and target respectively. To determine the minimum projectile energy for the reaction to proceed, we must add to this barrier potential the fraction of the kinetic energy retained by the products according to the principle of conservation of momentum. Charged-particle reactions do proceed at bombarding particle energies less than those required to overcome the Coulomb barrier, although with smaller cross-section. This is explained in quantum mechanics by "tunneling" through the potential barrier. A few specimen calculations of the Coulomb barrier for some typical charged particle reactions are given in the Appendix. If a charged particle reaction has a large positive Q value, this does not mean that the reaction will proceed at very low energy as is the case for neutron reactions, but once formed, the products will have high kinetic energy.

Figures 3 and 4 are schematic representations of some charged particle reactions. Figure 3 shows the energetics involved in some of the nuclear reactions induced by He³ bombardment of O¹⁶. The Coulomb barrier to the entry of a He³ particle into the O¹⁶ nucleus is 4.60 MeV and is represented by the dashed curve. The excited compound nucleus formed

Figure 3. Schematic Representation of the
Energetics of the Reactions Possible
With He^3 on O^{16} .

is Ne^{19} . The excitation energy of the Ne^{19} nucleus is greater than that required to overcome the Coulomb barrier to the emission of a proton (2.76 MeV), to form F^{18} , or the emission of an alpha particle to form O^{15} . The emission of a gamma-ray to form Ne^{19} is possible, but unlikely because of the high excitation of the compound nucleus. At higher He^3 bombarding energies, the formation of F^{17} becomes energetically possible.

Figure 4 shows the competition between some of the possible nuclear reactions that may be induced by He^3 bombardment of C^{12} . The $\text{C}^{12}(\text{He}^3, \alpha)\text{C}^{11}$ reaction is most favored because it has a positive Q value. The next most probable reaction is $\text{C}^{12}(\text{He}^3, n)\text{O}^{14}$, with competition from the reactions $\text{C}^{12}(\text{He}^3, d)\text{N}^{13}$ and $\text{C}^{12}(\text{He}^3, \gamma)\text{O}^{15}$. These diagrams are constructed from the more detailed energy level diagrams given in reference (26).

Charged particles of moderately high energies can give rise to as many as nine separate nuclear reactions. For 20 MeV protons these are:

(p,n)	(p, α)
(p,pn) or (p,d)	(p,t)
(p,2n)	(p, γ)
(p,2p)	(p, He^3)

The cross-sections of these reactions are about the same (in the range 10 to 200 mb) except for the last two, which are several orders of magnitude smaller. This complexity of nuclear products is advantageous, since it increases the probability of producing a conveniently detectable radionuclide which makes an activation analysis possible; but it is a disadvantage in that it increases the chance of interference to the analysis of one element by the production of the same radionuclide from

Figure 4. Competition Between Nuclear Reactions Produced By He^3 Bombardment Of C^{12} .

other elements, or by general complexity due to the large number of radioactive species. The beam energy, however, can be controlled and reduced to eliminate interfering reactions in very many cases.

A characteristic of simple charged particle reactions is that the cross-section increases from zero at the energetic threshold, passes through a maximum about 10 MeV above the threshold energy, and then decreases as the next higher energy reaction competes. Examples of this behavior are shown in figures 5 and 6. Figure 5 shows the excitation functions for the bombardment of bismuth with protons. It can be seen that the cross-section for the $(p,2n)$ reaction increases from zero at the energetic threshold (~ 10 MeV), passes through a maximum at about 20 MeV, and then decreases to a small constant value as the next competing reaction $(p,3n)$ occurs. Similar behavior is observed in figure 6, which shows excitation functions for proton reactions (p,pn) , $(p,p2n)$, (p,n) and $(p,2n)$ on copper. Thus, if an irradiation is made with particles of energy above that for maximum cross-section, the largest amount of radioactive product will be produced at a distance beneath the surface of the target. This fact could be used to analyze for components beneath the surface of a sample, when the surface is contaminated.

Table III lists the nuclear reactions of the target elements lithium to fluorine with projectiles p, d, t, He^3 , and α particles, which are important for activation analysis using charged particles. Using this table, we can compare the various charged particle reactions that may be useful for the activation analysis of an isotope.

FIGURE 5: EXCITATION FUNCTIONS FOR (p,xn) REACTIONS ON BISMUTH (REF 27)

FIGURE 6: EXCITATION FUNCTIONS FOR PROTON REACTIONS ON COPPER (REF.28)

TABLE III
Nuclear Reactions for Charged-Particle Activation Analysis
of the Low Atomic Number Nuclides

Target Nuclide	Reaction	Product Nuclide	Half Life of Product	Reaction Q Value (MeV)	Coulomb Barrier (MeV)	Reaction Cross-section at Various Energies (millibarns) at (MeV) (ref.29)	References to Experimental use in Activation Analysis
Li ⁷	(p,n)	Be ⁷	53d	-1.6	1.1	500 at 2.25	
Li ⁷	(d,2n)	Be ⁷	53d	-3.9	1.1		
Li ⁷	(He ³ ,t)	Be ⁷	53d	-0.9	2.4		
Li ⁷	(α,γ)	B ¹¹	stable	+7.54	2.6		
Be ⁹	(p,t)	Be ⁷	53d	-12.1	1.4	12 at 18.5	
Be ⁹	(He ³ ,n)	C ¹¹	20m	+7.6	2.9		
	(α,γ)	C ¹³	stable	+10.65	3.0		
B ¹⁰	(p,γ)	C ¹¹	20,	+8.7	1.7	0.0075 at 1.2	
B ¹⁰	(p,α)	Be ⁷	54d	+1.15	3.0*	200 at 1.1	
B ¹⁰	(d,n)	C ¹¹	20,	+6.5	1.9	>200 at 4	Süe (18)
B ¹⁰	(t,2n)	C ¹¹	20m	+0.21	1.9		
B ¹⁰	(He ³ ,γ)	N ¹³	10m	+21.6	3.4		
B ¹⁰	(He ³ ,d)	C ¹¹	20m	+3.2	3.4		
B ¹⁰	(α,n)	N ¹³	10m	+1.06	3.6		
B ¹¹	(p,n)	C ¹¹	20m	-2.8	1.8	100 at 5	Gill (6), Busch (31)
B ¹¹	(d,2n)	C ¹¹	20m	-5.0	1.8		
B ¹¹	(He ³ ,n)	N ¹³	10m	+10.2	3.3		
B ¹¹	(He ³ ,t)	C ¹¹	20m	-2.0	3.3		
C ¹²	(p,γ)	N ¹³	10m	+1.9	1.9	0.1 at 0.456	Point(37), Pierce(38), Riezler(40)
C ¹²	(p,d)	C ¹¹	20m	-16.5	1.9	80 at 30 (30)	
C ¹²	(d,n)	N ¹³	10m	-0.28	1.9	>55 at 4	{Winchester(7), Süe(18)
C ¹²	(d,t)	C ¹¹	20m	-12.4	1.9		{Curie(41), Sippel(5)
C ¹²	(t,2n)	N ¹³	10m	-6.5	1.9		
C ¹²	(He ³ ,γ)	O ¹⁵	2.1m	+12.1	3.1		
C ¹²	(He ³ ,n)	O ¹⁴	71s	-1.15	3.1		
C ¹²	(He ³ ,d)	N ¹³	10m	-3.5	3.1	100 at 15	Markowitz (42)
C ¹²	(He ³ ,α)	C ¹¹	20m	+1.85	3.1	300 at 10	Markowitz (42)
C ¹²	(α,n)	O ¹⁵	2.1m	-8.5	3.1		

TABLE III (cont'd)

Target Nuclide	Reaction	Product Nuclide	Half Life of Product	Reaction Q Value (MeV)	Coulomb Barrier (MeV)	Reaction Cross-section at Various Energies (millibarns) at (MeV) (ref.29)	References to experimental use in Activation Analysis
O^{13}	(p,n)	N^{13}	10m	-3.0	2.6		
O^{13}	(d,2n)	N^{13}	10m	-5.2	1.8		
O^{13}	(He ³ ,n)	O^{15}	2.1m	+7.1	3.7		
O^{13}	(He ³ ,t)	N^{13}	10m	-2.2	3.7		
N^{14}	(p, γ)	O^{15}	2.1m	+7.35	2.1	400 at 2.5	
N^{14}	(p,d)	N^{13}	10m	-8.3	2.1	>6 at 18.7	
N^{14}	(p, α)	C^{11}	20m	-2.9	4.1	68 at 6.15	
N^{14}	(d,n)	O^{15}	2.1m	+5.12	2.1	>200 at 4.5	S ¹⁸ (18)
N^{14}	(d,t)	N^{13}	10m	-4.3	2.1		
N^{14}	(He ³ , γ)	F^{17}	66s	+15.8	4.2		
N^{14}	(He ³ ,d)	O^{15}	2.1m	+1.8	4.2		
N^{14}	(He ³ , α)	N^{13}	10m	+10.0	4.2		
N^{14}	(α ,t)	F^{17}	66s	-4.7	4.4		
N^{14}	(α ,t)	O^{15}	2.1m	-12.5	4.4		
N^{15}	(p,n)	O^{15}	2.1m	-3.5	2.1		
N^{15}	(p,t)	N^{13}	10m	-12.9	2.1		
N^{15}	(d,2n)	O^{15}	2.1m	-5.8	2.0		
N^{15}	(He ³ , γ)	F^{18}	110m	+14.1	4.1		
N^{15}	(He ³ ,n)	F^{17}	66s	+5.0	4.1		
N^{15}	(He ³ ,t)	O^{15}	2.1m	-2.8	4.1		
N^{15}	(α ,n)	F^{18}	110m	-6.4	4.2		
O^{16}	(p, γ)	F^{17}	66s	+0.6		{ 40 at 8.5 (30)	
O^{16}	(p, α)	N^{13}	10m	-5.2	2.3	{ 55 at 11.3 (30)	
O^{16}	(d, γ)	F^{18}	110m	+7.5	2.3		Amiel and Peisach (56)
O^{16}	(d,n)	F^{17}	66s	-1.6	2.3	>80 at 3.5	S ¹⁸ (18)
O^{16}	(d,t)	O^{15}	2.1m	-9.4	2.3		
O^{16}	(t,n)	F^{18}	110m	+1.2	2.3	100 at 2.13	Born(51), Bailey (52)
O^{16}	(t,2n)	F^{17}	66s	-7.9	2.3		
O^{16}	(He ³ , γ)	Ne^{18}	18s	+8.4	4.6		
O^{16}	(He ³ ,n)	Ne^{18}	1.46s	-3.0	4.6		Markowitz(22),Demildt(23)
O^{16}	(He ³ ,p)	F^{18}	110m	+2.02	4.6		
O^{16}	(He ³ ,d)	F^{17}	66s	-4.9	4.6		
O^{16}	(He ³ , α)	O^{15}	2.1m	+4.9	4.6		

TABLE III (cont'd)

Target Nuclide	Reaction	Product Nuclide	Half Life of Product	Reaction Q Value (MeV)	Coulomb Barrier (MeV)	Reaction Cross-section at Various Energies (millibarns) at (MeV) (ref.29)	References to experimental use in Activation Analysis
O^{17}	(p,He ³)	O^{15}	124s	-8.5	5.0*		
O^{17}	(d,p)	F^{18}	110m	+6.9	2.3		
O^{17}	(d,n)	Ne^{18}	1.43s	+1.9	2.3		
O^{17}	(d, α)	O^{15}	124s	+9.8	4.8*		
O^{17}	(t,p)	F^{19}	18s	+11.1	2.2		
O^{17}	(t,d)	F^{18}	110m	+2.9	2.2		
O^{17}	(He ³ ,pn)	F^{18}	110m	+1.4	4.5		
O^{18}	(p,n)	F^{18}	110m	-2.45	2.2	40 at 4.65	Fogelstrom-Fineman (32)
O^{18}	(d,2n)	F^{18}	110m	-4.7	2.2	10 at 2.77	Fleckenstein (17)
O^{18}	(t, γ)	F^{21}	4.6s	+14.2	2.2	5 at 2.95	Thompson(34)Winchester(7)
O^{18}	(t,n)	F^{20}	11sec.	+6.1	2.2		
O^{18}	(t,d)	O^{19}	21sec.	-2.3	2.2		
O^{18}	(t, α)	N^{17}	4.14s	+3.75	2.2		Amiel (53)
O^{18}	(He ³ ,p)	F^{20}	11s	+6.9	4.4		
O^{18}	(He ³ ,t)	F^{18}	110m	-1.7	4.4		
O^{18}	(He ³ ,2n)	Ne^{19}	18s	-3.8	4.4		
F^{19}	(p,n)	Ne^{19}	18s	-4.0	2.5	>25 at 5.7	
F^{19}	(p,d)	F^{18}	110m	-8.2	2.5	120 at 20 (30)	
F^{19}	(p,t)	F^{17}	66sec.	-11.1	2.5		
F^{19}	(d,t)	F^{18}	110m	-4.2	2.4	4 at 8.65	
F^{19}	(He ³ , γ)	Na^{22}	2.58y	+18.6	4.9		
F^{19}	(He ³ ,n)	Na^{21}	23s	+7.5	4.9		
F^{19}	(He ³ ,t)	Ne^{19}	18s	-3.3	4.9		
F^{19}	(He ³ , α)	F^{18}	110m	+10.1	4.9		

* Coulomb barrier for emission of particle

Specific Use in Activation Analysis

6(a) Proton Activation

Gill (6), in a study of the use of protons in activation analysis for the elements from helium to silicon, used proton activation analysis for the determination of boron in silicon. Silicon samples, sheathed in 2 mil tantalum foil were irradiated in a cyclotron with 20 MeV protons and the following reactions were induced: $B^{11}(p,n)C^{11}$ ($t_{1/2} = 20.4$ min, a positron emitter) and $Si^{30}(p,n)P^{30}$ ($t_{1/2} = 2.5$ min, also a positron emitter). The latter reaction was used as an internal monitor of the intensity of the proton beam. A chemical separation was made and the ratio of the C^{11} to P^{30} activities measured. Gill concluded that proton activation could determine boron in silicon with a sensitivity of 3 ppb. The theoretical limit of sensitivity was 0.3 ppb using 20 MeV protons from the synchro-cyclotron at a beam current of 2 μ a. If a fixed frequency cyclotron were used, this limit could in theory be extended by a factor of 100 to 500 with the greater beam current available. However, problems associated with the dissipation of the additional heat produced in the target would have to be overcome.

Other reactions of possible use in proton activation analysis are suggested by Gill:

$Li^7(p,n)Be^7$	} -----	54d	E.C.	$\gamma = 0.48$ MeV
$Li^9(p,t)Be^7$				
$C^{12}(p,pn)C^{11}$		20.4m	β^+	
$N^{14}(p,pn)N^{13}$	} -----	10.1m	β^+	
$O^{16}(p,\alpha)N^{13}$				
$F^{19}(p,pn)F^{18}$		1.87h	β^+	

Busch, et.al. (31) have also used protons to determine boron in silicon by means of the reaction $B^{11}(p,n)C^{11}$, which has a threshold energy of 2.8 MeV. By using 4 MeV protons, they were able to eliminate the interfering reaction $N^{14}(p,\alpha)C^{11}$ which occurs above 4 MeV.

Fogelström-Fineman, et.al. (32) used proton activation to detect O^{18} in the course of photosynthesis studies using enriched oxygen. Fleckenstein, et.al. (17,33) used 4 MeV protons to analyze for O^{18} in labeled adenosine diphosphate, adenosine triphosphate and creatine phosphate in studies of the metabolism of living cells. Small samples (a few micrograms) of these compounds were separated and purified by paper chromatography and then deposited on platinum foils that were bombarded with 4 MeV protons from a cyclotron. The nuclear reaction $O^{18}(p,n)F^{18}$ (half-life = 110 min) was used, and a gamma-ray spectrometer measured the 511 keV annihilation radiation from F^{18} . The samples were allowed to decay for 2 hours before measurements were made since N^{13} (half-life 10 min) was formed by the (p,α) reaction on O^{18} or by the (p,n) reaction on C^{13} . The platinum foils were cooled with solid carbon dioxide during bombardment. A thin monitor foil containing a known amount of O^{18} (which degraded the energy of the proton beam a negligible amount) was used to monitor the proton beam during each analysis. The ratio of F^{18} activity in the phosphorus compound to that in the monitor foil was used to calculate the O^{18} in the phosphorus compound.

Thompson (34) used the same nuclear reaction to measure the thickness of oxide films on metals. The lower and upper limits of measurement were 1 and 10^5 Angstroms respectively. Elements interfering with the measurement, when no chemical separations were used, were copper, nickel, zinc and titanium.

Tomita and Saisho (35) described the use of 14.5 MeV protons for the activation analysis of niobium in tantalum. They used the reaction $\text{Nb}^{93}(\text{p},\text{n})\text{Mo}^{93\text{m}}$, and 6.9h - $\text{Mo}^{93\text{m}}$ was chemically separated in 1.5 hours. A sensitivity of 0.1 ppm was obtained by this method. They said that neutron activation analysis was not satisfactory in this case. Neutron activation of niobium produces principally 6.6 minute $\text{Nb}^{94\text{m}}$, which is best measured by means of the niobium K x-ray at 16.6 keV (36). Tantalum produces 16 minute $\text{Ta}^{182\text{m}}$ and 115 day Ta^{182} , the gamma-rays from which would obscure the 16.6 keV niobium x-ray if tantalum were present in large excess. A chemical separation would, therefore, be required to separate the small amount of niobium from the tantalum, and the separation must be performed in about six minutes. To add to the problem, niobium and tantalum are difficult to separate chemically, having almost identical covalent atomic radii, because of the lanthanide contraction. However, rapid radiochemical separations of niobium from ore samples have been performed (36).

Point (37) used 0.6 MeV protons and a beam current of 150 μA to determine carbon in iron by the reaction $\text{C}^{12}(\text{p},\gamma)\text{N}^{13}$, which occurs at a resonance energy 0.46 MeV. The N^{13} (10.1m half-life) was detected without chemical separation by coincidence measurement of the positron-annihilation radiation using two scintillation detectors. The method is very specific and interfering nuclear reactions are negligible. The penetration of the protons into the target at this low energy was only 3 microns (0.7 mil). The limit of sensitivity was reported as 3 parts-per-million. Point used iron discs 40 mm in diameter and 1 mm thick, and cleaned the surface before irradiation by grinding on a carbon-free millstone. The rear face of the iron disc was cooled with water during

the irradiation. The proton beam was monitored by rotating a disc of tungsten carbide into and out of the beam 50 times per minute. The amount of carbon in the iron disc was then calculated from the ratio of N^{13} activities in the target and monitor.

Pierce, Peck and Henry (38) have reported the determination of carbon in steels by bombarding with 0.8 MeV protons from a Van de Graaff accelerator using the nuclear reaction $C^{12}(p,\gamma)N^{13}$. Instead of measuring the 10 minute N^{13} after the irradiation, they measured the 2.3 MeV prompt gamma-ray which is emitted during the nuclear reaction. A series of standard samples of steel, of known carbon content, were irradiated to the same constant proton dose, and a linear relationship was obtained between the gamma counts under the 2.3 MeV peak and the carbon content of the steels.

6(b) Deuteron Activation - Winchester (7) has used deuterons from a cyclotron to determine carbon, oxygen and silicon by means of the reactions:

The carbon was determined using 8 MeV deuterons, while 15 MeV deuterons were used for the oxygen and silicon analyses. He discussed the possible reactions of charged particles available at the MIT cyclotron useful for determining carbon, oxygen and silicon; namely 7.5 MeV protons, up to 15 MeV deuterons and 30 MeV alpha particles, and explained why the above three reactions were chosen. As little as 0.01% carbon in SiO_2 was

detected without chemical separation. The stoichiometry of SiO_2 was determined to a precision of a few per cent by simultaneous beta and gamma-ray measurement of the irradiated sample. The determination of trace amounts of oxygen in beryllium metal and natural galena minerals required chemical separation of the induced fluorine-18 activity.

Winchester (7) also gives a good review of the earlier work on charged-particle activation analysis, particularly using deuterons for the determination of carbon in steel. Ardenne and Bernhard (39) used 0.8 MeV deuterons to detect 0.05% carbon in steel, measuring N^{13} (half-life 10 min) without a chemical separation. Riezler (40) showed that 4 MeV is the optimum deuteron energy for this determination, since the cross-section for the $\text{C}^{12}(\text{d},\text{n})\text{N}^{13}$ reaction is a maximum and the interfering reaction $\text{Fe}^{54}(\text{d},\text{n})\text{Co}^{55}$ is small. Curie (41) described a method for the determination of 0.1% carbon in steel, by deuteron activation followed by autoradiography. She (18) investigated the activation analysis of several light elements by deuteron bombardment from a cyclotron. Microgram quantities of boron, carbon, nitrogen and oxygen were detected by their (d,n) products. Sulphur was detected at a level of 10 micrograms by the (d, α) reaction, giving 2.5 minute P^{30} .

6(c) He^3 Activation

Markowitz and Mahony (22) discussed the activation analysis of oxygen and other elements by helium-3 induced nuclear reactions. The helium-3 nucleus has a low binding energy (7.7 MeV), so that many of the simple nuclear reactions which it undergoes are exoergic compared to those of protons and helium-4 ions. Although the deuteron is a commonly accelerated ion of low binding energy (2.2 MeV) and will

produce some exoergic (d,p) and (d,n) reactions, the high neutron background surrounding deuteron accelerators induce (n, γ) reactions that give the same product as (d,p) reactions. This complication is not present for helium-3 ions. The authors measured down to 0.61% oxygen in thorium metal, but justifiably claimed an ultimate sensitivity of fractions of a part-per-billion for oxygen analysis.

Markowitz and Mahoney (42) have also discussed the analysis of carbon by the $C^{12}(He^3, \alpha)C^{11}$ reaction, and have determined carbon and oxygen in gold, silicon, and plastic fibers. They also measured excitation functions for the reactions $C^{12}(He^3, \alpha)C^{11}$ and $C^{12}(He^3, d)N^{13}$.

Demildt (23) used He^3 particles from a linear accelerator to analyze for oxygen in the actinide elements. Oxygen was determined at a concentration of 0.5% with a detection limit of 0.001% in a few hundred micrograms of matrix material. Interference by activation of other elements was restricted by control of the bombarding energy. Other methods of activation analysis were not possible; for example, fast neutron activation would have produced interfering fission products and proton, deuteron, alpha particle or photon activation were either inapplicable or insensitive.

Tilbury and Wahl (43) have used 5 MeV He^3 particles to test the sensitivity of He^3 activation analysis with a beam current of 0.5 microamperes. The following elemental sensitivities were measured for irradiations of 10 minutes.

Boron	0.05 ppm	0.002 $\mu g/cm^2$
Carbon	0.3 "	0.01 "
Nitrogen	1.0 "	-
Oxygen	0.5 "	0.01 "
Fluorine	-	0.1 "

By properly selecting the bombarding energy (above 5 MeV), beam current, and the length of irradiation, these sensitivities would be in the parts-per-billion range or below.

Tilbury and Wahl concluded from their experiments that the greatest single problem in using He^3 activation for elemental analysis is the possibility of interference of other elements to the analysis of a particular element. Table IV shows the elements that gave rise to the radionuclides detected in this work. In some cases, three different elements gave rise to the same radioactive product. Thus it would be difficult to analyze for oxygen in the presence of fluorine and nitrogen, or for carbon in the presence of beryllium or boron. In many cases, the interfering element(s) will be absent in the sample, and no problem will exist. Samples containing carbon, nitrogen, and oxygen could be analyzed for all three elements, there being only a small interference to the measurement of nitrogen due to carbon-13, which has a natural isotopic abundance of 1.1%. However, certain mixtures of elements would be very difficult to analyze, since both boron and carbon yield C^{11} ; boron, carbon, and nitrogen give N^{13} ; and carbon and nitrogen give oxygen-15. Theoretically, it would be possible to analyze for any mixture of elements, by irradiating standards containing known amounts of the elements to be determined. The concentrations of each element in the sample (B), (C), (D),... could be found by solving a number of simultaneous linear equations of the type

$$A = k_1(B) + k_2(C) + k_3(D) + \dots$$

where A is the total activity of a particular product radionuclide (e.g. N^{13}) and k_1 , k_2 , k_3 are the activation rate constants.

If one were not limited to a shortest half-life of 2.1 minutes, carbon could be detected by the reaction $\text{C}^{12}(\text{He}^3, n) \text{O}^{14}$ half-life = 71 sec. and an easily detectable gamma-ray at 2.31 MeV.

TABLE IV - RADIOACTIVITIES DETECTED FROM DIFFERENT
ELEMENTS BY He³ ACTIVATION (Ref. 43)*

<u>Element</u>	<u>Radioactivities Detected</u>
Be	C ¹¹
B	C ¹¹ N ¹³
C	C ¹¹ (N ¹³) O ¹⁵
N	N ¹³ O ¹⁵ (F ¹⁸)
O	O ¹⁵ F ¹⁸
F	F ¹⁸

() indicates that activity is produced from isotope of small natural abundance e.g. C¹³.

* Limited to shortest half-life of 2.1 min O¹⁵.

It would be better of course to eliminate interferences, or make them negligible, by altering the nuclear reaction by change in bombarding particle or bombarding energy. For example, if one wished to analyze for boron and carbon in iron, C^{12} could be determined by $C^{12}(He^3, n) O^{14}$. B^{10} or B^{11} can give no O^{14} , and iron would not be activated by He^3 since the irradiation can be performed below the Coulomb barrier for He^3 particles on iron (9 MeV). To determine boron by He^3 reaction, simultaneous equation solution would be needed if C^{12} is present in larger or comparable amounts, so it would be better to determine boron by $B^{10}(p, \alpha) Be^7$ reaction at about 2 MeV since carbon could not give Be^7 with 2 MeV protons.

6(d) Alpha Particle Activation - Alpha particles have been used to determine the concentrations of Pb^{204} , Pb^{206} and Pb^{208} in one-gram sample of meteorites. Sensitivities down to a part-per-billion Pb^{204} were obtained (Cobb (44)). King and Henderson (45) have used 16 MeV alpha particles to determine about 0.05% copper in silver.

Nozaki, et.al. (46) have reported the use of 40 MeV alpha particles to determine 0.3% oxygen in silicon by the nuclear reaction $O^{16}(\alpha, pn) F^{18}$. The F^{18} was separated chemically prior to measurement.

Odeblad and Nati (47) found that alpha particles from 160 millicuries of polonium-210 would activate B^{10} , F^{19} , Na^{23} , Mg^{25} , and Al^{27} , giving 10 min N^{13} , 2.6 year Na^{22} , 65 sec Al^{26m} , 2.3 min Al^{28} , and 2.6 min P^{30} respectively. The number of radioactive atoms produced were between 0.2 and 2.2 per 10^6 alpha particles from polonium-210. They used this method to measure sodium in urine and aluminum in the peritoneal cavity.

Gold (48) used the Po-Be reaction to detect air-borne beryllium dust. Samples were collected on filter paper, exposed to Po^{210} alpha particles, and the resulting prompt gamma radiation was measured to indicate the beryllium concentration.

6(e) Triton Activation

On irradiating lithium with neutrons in a nuclear reactor, the reaction ${}^6\text{Li}(n,\alpha){}^3\text{H}$ occurs, and the tritons produced can be used to induce secondary nuclear reactions with materials in intimate contact with the lithium. The use of lithium in this way was first proposed by Smales (49) and was applied to the analysis of oxygen in beryllium (50). The range of the high-energy tritium particle is 6 cm in air and 30 microns in solids and liquids, so the beryllium metal powder was mixed with lithium fluoride powder and irradiated. The tritons reacted with oxygen as follows: $\text{O}^{16}(t,n)\text{F}^{18}$ and the activity of the F^{18} was measured after a chemical separation. Born and Riehl (51) used this method for the determination of oxygen in iron, and the method gave a detection limit of 10 parts-per-billion.

Bailey and Ross (52) have used the method for the determination of oxygen in thin wafers of gallium arsenide irradiated with lithium metal between the layers. They claim the maximum sensitivity of the method to be 7×10^{-9} gram of oxygen in gallium arsenide, or 5 parts-per-million in the small samples used. The oxygen actually determined was 72^{+18}_{-18} parts-per-million.

The energy of the tritons formed in the reaction $\text{Li}^6(n,\alpha)\text{H}^3$ is equal to 2.74 MeV, so the tritons formed will react with any nuclide up to an atomic number of 10 or 11, since the Coulomb barrier for the

reaction would be less than the bombarding energy of the tritons. For elements with atomic number greater than eleven, the reaction proceeds only by quantum mechanical tunneling through the Coulomb barrier, so that the reaction cross-sections are small.

Amiel and Welwart (53) have used the method of triton activation in a nuclear reactor for the analysis of lithium, lithium-6, and oxygen-18. They measured the delayed neutrons of N^{17} produced by the reactions $Li^6(n,\alpha)t$ and $O^{18}(t,\alpha)N^{17}$. Nitrogen-17 emits neutrons in its decay (half-life 4.14 sec) and these neutrons can be measured easily, even in samples of high beta and gamma activity.

6(f) Recoil Protons and Deuterons in a Nuclear Reactor

An enriched uranium, water-moderated nuclear reactor produces a high flux of fast neutrons near its core that, by striking protons in ordinary water and deuterons in heavy water, can produce correspondingly large fluxes of recoil protons or deuterons. The neutron flux spectrum approximates the Watt curve (54), and Glickstein and Winter (55) have calculated the proton flux from the neutron flux at any energy. The flux of recoil protons decreases exponentially with increasing energy. Glickstein and Winter detected approximately 10^6 counts/sec of C^{11} due to the reaction $B^{11}(p,n)C^{11}$ in H_3BO_3 irradiated to saturation. This gives a sensitivity for boron analysis of about 40 parts-per-million, and represents their most sensitive result. Thus, the sensitivity is low for activation analysis, using recoil protons or deuterons, and the high ^{thermal} neutron flux will produce large amounts of unwanted activities in most target materials.

Amiel and Peisach (56) have used the reaction $O^{16}(d,n)F^{17}$, using recoil deuterons in a heavy-water-moderated nuclear reactor, to measure the deuterium concentration over a range of 0.5 to 94.5% D_2O content. The 66 sec. half-life positron emission of F^{17} was measured.

7. Conclusions

Because high-energy-particle bombardments (be they neutrons, photons, or charged particles) are capable of producing radioactive isotopes on the neutron-deficient side of the line of beta stability in the chart of the nuclides, it seems evident that the use of such particles in both activation analysis and isotope production will become increasingly important. The use of only low energy (reactor-produced) neutrons for these purposes generally restricts one to the neutron excess radionuclides, thus making use of about one-half of those potentially available.

The application of charged-particle bombardments to the activation analysis of certain low-atomic-number elements (beryllium, boron, carbon, nitrogen and oxygen) has been amply demonstrated. Charged-particle activation analysis has also been applied with remarkable success to the determination of certain elements and isotopes of high mass number where other methods of analysis were unknown or of low sensitivity (e.g. niobium and the isotopes of lead). The extension of charged-particle activation analysis to other elements or other isotopic assays is imminent.

Charged-particle activation analysis offers great versatility when the type of bombarding particle and the energy of the beam can be selected. In this way, the products of the reaction can be controlled and interferences can be minimized. Unfortunately, the cross-sections for charged-particle reactions are known for only a few reactions and the detailed excitation functions are known in even fewer cases. Ideally, excitation functions should be determined for all the stable nuclides over a range of 0 to 30 MeV for all bombarding particles. Experimental determination would be a mammoth task, and the present state

of nuclear theory is such that these results can not be calculated in sufficient detail. If such information were available, it would be possible to choose the best bombarding particle and the optimum energy for the reaction to minimize other interfering reactions. At the moment, however, the choice of conditions depends on a combination of experience, analogy with other elements, and a few test experiments.

Appendix

Specimen calculations of some Coulomb barriers (B) and "thresholds" for the formation of the compound nucleus ($E_{\min}$).

$$\text{The Coulomb barrier, } B = \frac{Z_1 Z_2 e^2}{R_1 + R_2} \quad (\text{ref. 25})$$

$$\text{if } R = 1.5 \times 10^{-13} A^{1/3} \text{ cm}$$

$$\text{then } B = \frac{Z_1 Z_2 (4.8 \times 10^{-10})^2 10^{-6}}{1.5 \times 10^{-13} (A_1^{1/3} + A_2^{1/3}) 1.6 \times 10^{-12}}$$

$$B = 0.96 \frac{Z_1 Z_2}{A_1^{1/3} + A_2^{1/3}} \quad \text{MeV}$$

For the reaction ${}^2\text{He}^{3++}$ on ${}^{16}\text{O}$,

$$B = \frac{0.96 \times 2 \times 8}{3^{1/3} + 16^{1/3}} = 3.88 \text{ MeV}$$

To take account of the conservation of momentum

$$E_{\min} = 3.88 \times \frac{19}{16} = \underline{4.60 \text{ MeV}}$$

For the reaction ${}^1\text{H}^{1+}$ on ${}^{92}\text{U}^{238}$

$$B = \frac{0.96 \times 1 \times 92}{1^{1/3} + 238^{1/3}} = 12.26 \text{ MeV}$$

To conserve momentum

$$E_{\min} = 12.26 \times \frac{239}{238} = \underline{12.32 \text{ MeV}}$$

For the reaction He^{4++} on N^{14} e.g. $\text{N}^{14}(\alpha, p) {}^{17}\text{O}$

$$B = \frac{.96 \times 2 \times 7}{4^{1/3} + 14^{1/3}} = 3.4 \text{ MeV}$$

To take account of the conservation of momentum

$$E_{\min} = 3.4 \times \frac{18}{14} = \underline{4.4 \text{ MeV}}$$

Note that there is no "threshold" for an exo-ergic reaction, i.e. for reactions with positive Q value. For reactions with negative Q values, the minimum kinetic energy for the reaction to proceed is

$$E = -Q \frac{(M+m)}{M}$$

Where M = target mass and m = projectile mass

The Coulomb barrier energy corrected for conservation of momentum may not be enough to induce a negative Q reaction.

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