RADIOACTIVATION ANALYSIS AND ISOTOPIC TRACERS

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Contents

Summary

Radioactivation analysis is a nondestructive method that enables one to determine qualitatively and quantitatively the composition of a given sample, by taking advantage of the nuclear properties of the constituent atoms.

When a sample is bombarded with an appropriate beam (consisting, for example, of neutrons, charged particles, or gamma rays), the collision of the incident radiation on the target may give rise to nuclear reactions, in which process the target nuclei might be subjected to transformation. Knowing the type and energy of the incoming projectiles, it is possible to predict the most probable reactions induced on the target constituents and thus relate the outcoming products to the original stable nuclides. Moreover, some of these products may be radioactive and undergo nuclear disintegration, thereby emitting particulate and/or electromagnetic radiation of characteristic energies.

Activation analysis is therefore based on the measurement of the radioactivity induced per activation in a bombarded sample. The measurement of the emitted energies makes it possible to identify the radionuclides produced in the target and, therefore, the corresponding originating stable nuclides. Measurement of the emission intensities is needed for the quantitative determination of the sample components. However, this determination depends on a series of experimental variables usually not known with sufficient accuracy and precision. A relative method is often used to overcome such difficulties.

Activation analysis, as a nondestructive and multi-elemental technique, finds application in a wide range of fields, from material science to health science and from physiology and nutrition to environmental science.

After a brief survey of the specific techniques, two applications are reported as case studies: one related to environmental pollution and the second to elemental biokinetics in humans.

1. Introduction

Activation analysis makes it possible to distinguish different isotopes of the same element on the basis of the radionuclides produced in the activated sample. The nucleus of an atom is an assembly of Z protons and N neutrons, where the atomic number Z identifies the chemical element and the mass number A is equal to Z+N. Of the many possible combinations of Z and N, only a few are energetically stable, whereas most of them are unstable (radioactive) and may undergo a nuclear disintegration with a characteristic half-life, leading toward a stable state. Each chemical element is usually present in nature as a mixture of different stable isotopes (stable combinations of Z and N, with Z being fixed). When a sample is bombarded with an appropriate beam (consisting, for example, of neutrons, charged particles, or gamma rays), the collision of the incident radiation on the target may give rise to nuclear reactions, in which process the target nuclei might be subjected to transformation. Knowing the type and energy of the incoming projectiles, it is possible to predict the most probable reactions of the target constituents and thus relate the outcoming products to the original stable nuclides. Moreover, some of these products may be radioactive and undergo nuclear disintegration, thereby emitting particulate and/or electromagnetic radiation(s) of characteristic energies.

The following sections will introduce the reader to some basic concepts of activation theory and show how it is possible to make qualitative and quantitative determinations based on the measurement of the radioactive decay of radioisotopes produced via nuclear reactions, with particular focus on application in the biological field.

2. Activation Theory

This section provides overview information on types of nuclear reactions and their reaction products. It then presents the various formulas used in describing the activity resulting from nuclear reactions.

2.1. Nuclear Reactions

When a sample is exposed to an energetic particle or photon flux coming from a reactor or an accelerator, nuclear reactions may take place (see *Experimental Nuclear Physics*). A simple nuclear reaction is indicated as

$X + a \rightarrow Y + b$ or $X(a,b)Y$

where *a* is the projectile, *X* the target nucleus, and *Y* and *b* the reaction products.

In the low medium energy range (typically less than 100 MeV), nuclear reactions can be classified mainly into direct reactions or compound nucleus formation, according to the mechanism that governs the process. In direct reactions (DR) the interaction involves only the projectile and one or a few external nucleons of the target, and most of the other nucleons (called spectators) are not implicated.
In compound nucleus (CN) formation all target nucleons are involved in the processes following the collision. It is assumed that projectile and target merge to form a compound nucleus, and that the excess energy is redistributed among all nucleons until enough energy is concentrated by chance on one or more of them, which is/are ejected. The CN may thus decay in a variety of different modes (called "channels") with the emission of photons or particles such as protons, deuterons, tritons, or alphas. The set of final products and their angular distributions are independent of the formation. This complex reaction mechanism characteristically requires more time than direct reactions do (approximately between 10^{-18} s and 10^{-12} s compared with about 10^{-22} s in direct reactions). A compound nucleus reaction is usually indicated as

$X + a \rightarrow CN^{*} \rightarrow Y + b$

where * indicates the excited nucleus.

Although the reaction products created after a compound nucleus formation may be the same as those found in a direct reaction, the time scale and the probabilities of the reaction are different, depending on which mechanism is involved. However, from the point of view of activation analysis, what matters is when they lead to radioactive products, which can then be identified by detecting their characteristic emissions.

In nuclear reactions, energetic aspects and probabilities have to be considered. The difference between the rest energy of the initial participants to the reaction (target nucleus and projectile) and the rest energy of the final products, which is equal to the difference between the final and the initial kinetic energy, is called Q-value. The Q-value may be positive, zero, or negative. A reaction with a positive Q-value (initial mass greater than the final mass, or equivalently final kinetic energy greater than the initial one) is called exothermic: the available energy is greater than what is required for the reaction to proceed, and the excess energy is released as kinetic energy of the final products. Q is zero in the case of elastic scattering.

A reaction with a negative Q-value (initial mass smaller than the final mass, or equivalently final kinetic energy smaller than the initial one) is called endothermic: the deficit in the rest energy requires that some additional energy be supplied to be converted into nuclear mass or binding energy. The target usually being at rest in the laboratory system, the energy needed is provided by the projectile: the minimum projectile energy required for the endothermic reactions to proceed is called threshold energy (E_{th}). In the laboratory system, E_{th} is expressed as:

$$E_{th} = |Q|(1 + m_a / m_X) \quad (1)$$

when the relativistic aspects may be neglected (which is the usual case considered here). A further energetic aspect must be considered in the case of charged projectiles. Charged projectiles are actually positive nucleons, which will thus experience the repulsive Coulomb field because of the protons in the nucleus. Therefore, there is a Coulomb barrier to be overcome before the projectile can come into the range of nuclear forces. Indicative values of the barrier heights are given in Table 1, together with the corresponding Q-values and threshold energies. Whereas no reaction may occur if the projectile energy is less than the threshold energy required by the reaction energetics, there is quantum mechanically a non-negligible chance that the projectile will come into the range of the nuclear force even if its energy is less than the barrier. This chance goes rapidly to zero as the difference between the projectile energy and the barrier height increases.

Reaction	**Maximum cross section (mb)**	**E_{max} (MeV)**	**$T_{1/2}$**	**Main gamma emissions (keV)**
^{95}Mo(p,n)^{95m}Tc	130	12	61 d	204 582 835
^{95}Mo(p,n)^{95g}Tc	400	12	20 h	766
^{96}Mo(p,n)^{96m}Tc	500	12	51.5 min	34 778
^{96}Mo(p,n)^{96g}Tc	60	15	4.28 d	778 812 850

Table 1. Q-values, threshold energies, and Coulomb barrier heights for selected nuclear reactions

Provided that the energy of the incident particle is higher than the eventual threshold value and/or the Coulomb barrier (whichever is the greatest), the probability of a reaction taking place is given by the so-called cross section (σ). Sigma may be interpreted as the effective cross-sectional area that the nuclei *X* present as a target for the incident particles, and it is expressed in units of surface. For a given reaction, the cross section is a function of the projectile energy and of the emission angles of the reaction products. For activation purposes, this latter dependence (a property which is of great importance in nuclear physics) is not relevant, whereas the energy dependence may be important for the optimization of the experimental conditions of activation.

For CN reactions, a typical cross section rises from zero to a maximum, then decreases. The behavior of the cross section as a function of projectile energy (excitation functions) is important, particularly in the case of charged particles impinging on thick targets where, because of the electromagnetic interactions with the electrons of the target atoms, the projectiles constantly lose energy by traversing it.

2.2. Activation Formulas

Let us consider the following situation: a beam passing through a sample of given thickness without significant changes in intensity and energy (a "thin sample"). The production rate of the nuclide Y through the reaction *X(a,b)Y* can be given as:

$$\frac{dN_Y}{dt} = \Phi\ \sigma\ N_X \qquad (2)$$

where Φ is the projectile flux density ($cm^{-2}\ s^{-1}$), that is the number of projectiles impinging on the sample area per unit time, σ is the cross section for the reaction considered (cm^2), and N_X the number of nuclei of the isotope *X* in the sample volume, i.e. the target nuclei.

As indicated in an earlier section (see *Nuclear Reactions*), sample activation occurs when the nuclear reaction gives rise to unstable nuclei. The process of radioactive decay is governed by a stochastic mechanism, where each radioactive nucleus has the same probability per unit time to undergo a transformation, independent of its previous history. So, the decay rate of the radioactive products Y is given by

$$\frac{dN_Y}{dt} = -\lambda N_Y \qquad (3)$$

where the probability constant λ is related to the characteristic mean life $\tau=1/\lambda$.
If the decay process is introduced in Eq. (2), the net production rate of the nuclide Y becomes:

$$\frac{dN_Y}{dt} = \Phi\ \sigma\ N_X\ - \lambda N_Y \qquad (4)$$

Therefore, after an irradiation time t_i, the total number N_Y of radioactive nuclei is obtained by integrating the previous equation. As the quantities Φ and N_X are essentially constant over the period of irradiation t_i, it follows that:

$$N_Y\left(t_i\right) = \frac{\Phi\ \sigma\ N_X}{\lambda}\left(1 - e^{-\lambda t_i}\right) \qquad (5)$$

The activity of nuclide *Y* at time t_i (also called end-of-bombardment (EOB) activity) is given by:

$$A_Y\left(t_i\right) = \lambda N_Y\left(t_i\right) = \Phi\ \sigma\ N_X\left(1 - e^{-\lambda t_i}\right) \qquad (6)$$

The exponential term within brackets takes the name of saturation factor S. Its value ranges from zero, when $t_i=0$, to one, for an infinitely long irradiation time. The S factor grows rapidly with irradiation time t_i, approaching asymptotically the limit value of 1 at $t_i=\infty$. This value corresponds to an EOB activity equal to:

$$A_Y\left(t_i = \infty\right) = \Phi\ \sigma\ N_X \qquad (7)$$

Eq. (7) describes a steady state situation, in which the rate of decay for the radioactive nuclei produced is equal to the rate of formation. In this case, A_Y is independent of the mean life τ of the radionuclide, and represents the maximum activity obtainable given a number N_X of target nuclei and a flux density Φ. Anyway, the EOB activity produced in irradiation periods equal to three τ's or four τ's reaches 95% or 98% of the saturation value, respectively. Therefore, it is not convenient to prolong the irradiation any further.
If after irradiation the sample is left "cooling" out of the beam flux for a time t_c, the number $N_Y(t_i, t_c)$ of nuclei still radioactive at time t_i+t_c is given by:

$$N_Y\left(t_i,t_c\right)=N_Y\left(t_i\right)e^{-\lambda t_c}=\frac{\Phi\,\sigma\,N_X}{\lambda}e^{-\lambda t_c}\left(1-e^{-\lambda t_i}\right) \tag{8}$$

The corresponding activity is expressed through the well-known activity formula:

$$A_Y\left(t_i,t_c\right)=\Phi\,\sigma\,N_X e^{-\lambda t_c}\left(1-e^{-\lambda t_i}\right) \tag{9}$$

which gives the number of disintegrations per unit time of the radioactive isotope *Y* produced via reaction on stable isotope *X* in a sample containing a number N_X of nuclei, which is irradiated by a flux density Φ during a time t_i and then left to decay for a time t_c.

In this formula, the number of nuclei N_X may be substituted for by the corresponding mass M_X of the atoms *X*, remembering that

$$N_X=\frac{N_{Avo}\,M_X}{W_X} \tag{10}$$

(where N_{Avo} is the Avogadro number, and W_X the atomic weight of *X*).

From Eq. (9) it is then possible to obtain the formula of the specific activity A_Y, i.e. the activity per unit mass of the target atoms *X*, by introducing Eq. (10):

$$A_{\ Y}\left(t_i,t_c\right)=\frac{A_Y\left(t_i,t_c\right)}{M_X}=\frac{\Phi\,\sigma\,N_{Avo}e^{-\lambda t_c}\left(1-e^{-\lambda t_i}\right)}{W_X} \tag{11}$$

The preceding discussion considered only one kind of target atom and one specific reaction. However, a projectile of a given energy may induce different reactions, each with its own probability, on the target atoms, and the target is normally composed of different kinds of atoms (particularly complex is the case of biological matrices, which will be considered later in this chapter (see *Practical Applications in Radioactivation Analysis*)). So, the bombardment of a target gives rise to the simultaneous production of several radioactive isotopes, and the total activity of the sample is simply the sum of the activities of all the radioactive reaction products. Only the individual activities for each radionuclide produced on the target nuclei under study, and not the total activity, are the parameters of interest in activation analysis. It is therefore important to be able to detect separately the characteristic emissions of each radionuclide, for example by means of spectroscopic measurements. However, total activity must be taken into consideration, because it can affect the measurements: there might be radiation protection problems in handling a highly active sample, which could additionally determine count losses due to an elevated dead time in the detection system (see *Detectors of Radiation*).

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Biographical Sketches

Marie Claire Cantone, physicist of the Medical Faculty of the University of Milan (Italy) has been a member of the Department of Physics and of the National Institute of Nuclear Physics (INFN) since 1984. She has more than twenty years of expertise in activation analysis and has been a local scientific manager of international and national research projects. She was in charge of the Radiation Protection Unit of the Department of Physics from 1987 to 1991.
Her research activity is mainly conducted in the field of experimental physics, with particular applications to medical and radiation protection problems.

Augusto Giussani graduated in Physics at the University of Milan (Italy) in 1991, and then got his Ph.D. in Natural Sciences from the Faculty of Physics of the University of Tübingen (Germany) in 1997. He worked at the Institute of Radiation Protection of GSF-National Research Center for Environment and Health in Neuherberg (Germany), and since August 1998 in the Physics Department at the University of Milan.
His main research activities are focused on the application of activation analysis and mass spectrometric technique to the detection of single metal isotopes in biological samples, with particular application to samples from tracer kinetic studies in humans, and in the development of compartmental models for elements of interest in the field of nutrition and/or radiation protection.