RADIOISOTOPE LABORATORY TECHNIQUES

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by

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With a foreword by

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and 4 half-tone plates

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FOREWORD

By Dr. Henry Seligman

Head of the Isotope Division, A.E.R.E. Harwell.

Many books have appeared recently which deal with the use of radioisotopes in different fields. Whilst this has been of great importance owing to the rapid spread of isotope uses in research laboratories, factories, and hospitals, there did not exist a comprehensive introduction telling the scientist or technologist how to use this new and important tool.

The authors have been closely connected for many years with the Harwell Isotope School, one as its Head, and they have had to supervise the training of students of many countries in isotope techniques. By their long experience in this field, they are therefore extremely well qualified to put together all the essentials which a worker needs to know when working with isotopes.

But this book is not only for the newcomer in the field; it should also give a wealth of new information to the scientist or technologist which will be useful to him.

The chapters are so written that they do not demand more than a basic scientific knowledge in order to give all technologists an opportunity to follow the subjects easily.

The well selected material is comprehensive, and all relevant information can be found in this volume.

I warmly welcome the appearance of this book, and believe that it fulfils an urgent need.

H. S.

PREFACE

We have tried in this book to make a practical approach to the use of radioisotopes. There are a number of excellent text-books on various aspects of radioactivity, in many of which the theoretical aspect is covered in some detail. This book is intended primarily as a manual for the man in the laboratory, rather than a text-book. We have avoided frequent reference to original literature, but where we have felt it advisable, have made suggestions for additional reading.

For many practical aspects of the work, we have drawn on our own experience in the Isotope School at Harwell and elsewhere, and we have included quite a number of original suggestions. We may be accused of incompleteness in some quarters, but we have deliberately tried to confine ourselves to the most probable approach to each problem in order to avoid confusion. In some places we have referred to specific manufacturers. This is no attempt at gratuitous advertisement, but we realise that a major problem with some readers is where to obtain specialised equipment, and we felt it our duty to include some information of this kind. We have found that the products we have mentioned have a place in a radioactive laboratory —there may well be others.

Our acknowledgements are due to those manufacturers who have supplied information regarding their products.

We should also like to record our thanks to Miss Rose Millett, Dr. Ray Allen, and Mr. Brian Smith for information prepared for the 4th Catalogue of Radioactive Materials published by the Isotope Division, A.E.R.E. We are also indebted to Dr. Henry Seligman, Head of the Isotope Division, for reading the manuscript and for his helpful comments.

R.A.F.
B.H.P.

INTRODUCTION

ARTIFICIALLY produced radioactive materials have been available in quantity for only a few years, but in that time they have been used as tools in many fields of research and technology.

Let us consider the implications of the term "Radioactive Isotope".

RADIOACTIVE implies that radiations are emitted. As will be seen in the next chapter, these may be alpha, beta or gamma, and they have very different properties. They cause ionisation, either directly or from secondary effects, and this is the basis of their detection. Radiation may also cause damage to the body, so it is necessary to set limits to the maximum levels of internal and external radiation, and to see that these are not exceeded. Gamma radiation may be used to bring about chemical effects, such as polymerisation, vulcanisation and other processes, and these are likely to be of very considerable technical value as large sources of radiation become available as by-products from the nuclear power programme. Other applications of large gamma sources are therapy, sterilisation, pest control, and the production of desirable mutations in plants. The rays are penetrating, and so may be used for various types of non-destructive testing, such as thickness gauging and radiography. Because of this penetration, it is necessary to consider the matter of shielding so as to control the level of external radiation reaching the body.

ISOTOPE means "same place", and this implies that isotopes of an element have the same chemical properties. Hence a "radioactive isotope" will follow the same chemical processes as the corresponding stable element, but will act as a radioactive label and may allow the course of a reaction, the uptake of an element, or a metabolic process to be traced. As a physical label it may be used to check the efficiency of mixing in a batch or continuous process, to measure flow rates, locate leaks, and for a whole host of other things. Consideration of the feasibility of these applications requires a knowledge of the physical properties of radioactive materials, and of their methods of detection. Since readily detectable amounts of radiation may be associated with very small masses, tracer techniques are potentially very sensitive and convenient analytical tools, and can often extend conventional limits of detection by several orders of magnitude.

The book is divided broadly into four sections. The first few chapters are devoted to basic considerations of nuclear physics, isotope production and radiological protection. These are followed by chapters on laboratory design, hazard control and waste disposal. The various methods of detection and measurement are treated from a practical aspect and include sections on statistics and on the choice of equipment. In the final chapters, a number of radioisotope techniques and applications are reviewed, and a chapter is included on the calculation of the feasibility of using isotopes in a particular system.

This cannot be a complete guide, although as far as possible suggestions have been given for further reading to enable specific subjects to be pursued if desired. The aim has been to indicate the principles to be followed, and to give as much factual information as possible.

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CHAPTER 1

ELEMENTS OF NUCLEAR PHYSICS

Definitions. Structure of the atom. The nucleus. Stable and unstable nuclei. Modes of disintegration. Disintegration schemes. Rate of disintegration. Radioactive decay.

Definitions

Before we can discuss the occurrence, properties and uses of radioactive isotopes, the structure of the atom must first be considered. At this stage, some of the units and terms used in Nuclear Physics will be defined.

ATOMIC MASS UNIT (a.m.u.)

This is one sixteenth of the mass of the Oxygen-16 atom

$$=1{\cdot}6603\times10^{-24}\text{ gram.}$$

(Note that this is the physical unit of atomic weight, and is smaller than the chemical unit which ignores the presence of the heavier isotopes of oxygen and fixes the atomic weight of oxygen as 16·000 units.) The unit of atomic mass is the reciprocal of Avogadro's Number, $6{\cdot}0247\times10^{-23}$, which is the number of atoms of any element in a gram-atom (defined as the atomic weight expressed in grams).

ENERGY

A convenient unit is the ELECTRON-VOLT, (eV), which is the energy acquired by an electron in falling through a potential difference of one volt. $1\text{ eV} = 1{\cdot}6\times10^{-12}$ erg.

MASS/ENERGY RELATION

Einstein has shown that mass and energy are related by the equation

$E = mc^2$ where E = energy in ergs
m = mass in grams
c = velocity of light in cm/sec ($2{\cdot}99792\times10^{10}$).

As a consequence of this a particle's rest mass has an equivalent energy, and a moving particle has a greater mass than a stationary one. From this equation, 1 a.m.u. = 931 MeV (million electron volts).

SUB-ATOMIC PARTICLES

Electron.—Mass 0·00055 a.m.u. Negative charge equal to $1{\cdot}6 \times 10^{-19}$ coulombs (ampere-seconds).

Proton.—Mass 1·00759 a.m.u. Positive charge equal in magnitude to that of the electron.

Neutron.—Mass 1·00898 a.m.u. Uncharged.

These three are the fundamental particles which go to make up the structure of the atom. There are other sub-atomic particles such as mesons, neutrinos, and anti-protons, but they need not concern us here, and the reader is referred to works on nuclear physics for a discussion of their significance.

Structure of the Atom

The Rutherford-Bohr model of the atom fits modern ideas of atomic structure very well, giving as it does a reasonable explanation of the phenomena of atomic physics, chemical combination and valency, and the properties of the nucleus. The structure supposes a central core (called the *nucleus*) consisting of protons and neutrons, and a cloud of electrons moving round the nucleus in defined orbits. Each orbit contains only two electrons, but orbits are grouped together to form shells (Fig. 1).

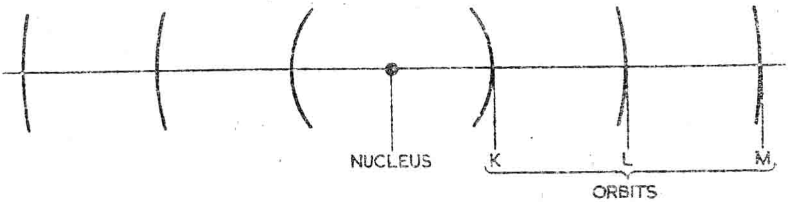

FIG. 1. Electron Orbits around the Nucleus of an Atom

The electrons in the outermost shells are the valency electrons and take part in chemical combination. As the atomic structure is built up by filling the shells according to a periodic pattern, the

"Periodic Table of the Elements" of Mendeleef is shown to follow closely modern ideas of the atom.

Each orbit represents a definite energy level, and movement of electrons from one orbit to another involves energy changes, which result in the giving out of definite quanta of energy. Movements of electrons between the innermost shells cause the production of X-rays whose wavelength is characteristic of the shell from which they come, and of the element concerned. Movements in outer shells are associated with the production of light, and give the characteristic spectral lines.

The phenomena associated with radioactivity are concerned with the nucleus, and are studied under the heading of "Nuclear Physics". "Atomic Physics" is concerned with extra-nuclear phenomena. There are large differences in the energies involved in these two subjects, e.g. the force between two atoms held by a chemical bond is of the order of electron-volts, X-ray production involves thousands of electron-volts, but the changes occurring within the nucleus evolve, or are brought about by millions of electron-volts in most cases.

The Nucleus

The diameter of an atom is about 10^{-8} cm, whereas that of the nucleus is about 10^{-12} cm which is about ten thousand times smaller. Since the nucleus is composed of protons and neutrons, each of mass about 1 a.m.u., and the electron weighs only 0·00055 a.m.u. (about 1/1800 of the mass of the proton), virtually all the mass of an atom is contained in the nucleus. From the relative sizes it follows that the density of the nucleus is of the order 10^{14} grams per cm^3, which gives a clue that the forces which hold together the nuclear particles are very different from those met with outside the nucleus. For detailed consideration of the implications of this, and for a discussion of the present state of knowledge about the nucleus, the references at the end of this chapter should be consulted. For the present treatment, certain characteristics of the nucleus will be assumed, and no attempt will be made to justify them.

ATOMS AND NUCLEI

Since the atom is electrically neutral, the total positive charge on the nucleus must equal the total negative charge on the orbital electrons. The only charged particle in the nucleus is the proton with charge equal in magnitude to that of the electron, so it follows that there must be the same number of protons in the nucleus as electrons in the various orbits. This number, Z, is the ATOMIC NUMBER of the element, and determines the place of the element in the periodic table of the elements. Thus Z characterises an element as does its chemical symbol.

The other important number is A, the MASS NUMBER. This is the sum of the number of protons and neutrons in the nucleus, and is the nearest integer to the ATOMIC MASS or the EXACT MASS, which is the sum of the masses of the nucleus and the associated electrons measured in a.m.u. (The chemical atomic weight is the relative weight of an atom, referred to that of oxygen which is taken as 16·000).

We may now define a few terms used in discussing the relationships between nuclei.

Nuclide.—An atom with specific nuclear characteristics, e.g. phosphorus of mass number 32 and atomic number 15, and cobalt of mass number 60 and atomic number 27 are NUCLIDES.

Isotope.—A series of nuclides having the same value of Z, but with different values of A are said to be ISOTOPES of the element in question. Thus phosphorus of mass number 32, and phosphorus of mass number 31 are ISOTOPES of phosphorus. The term "Isotope" is often loosely used where strictly the proper term is "Nuclide", and for instance one would talk of using the "isotopes" Cobalt-60 and Phosphorus-32 in an investigation. Although this is incorrect we ourselves cannot venture criticism against modern usage since we have called this book "Radioisotope Laboratory Techniques", instead of the correct, but rather pedantic title "Radionuclide Laboratory Techniques".

TABLE I

	Z	A	N	Examples
Isotope	Same	Different	Different	${}^{31}_{15}P$ and ${}^{32}_{15}P$
Isobar	Different	Same	Different	${}^{32}_{15}P$ and ${}^{32}_{16}S$
Isotone	Different	Different	Same	${}^{2}_{1}H$ and ${}^{3}_{2}He$

The following terms are sometimes encountered, but are of secondary importance:

Isobars.—Nuclides having the same mass number, but different charge.

Isotones.—Nuclides with the same number of neutrons.

These relationships are summarised in Table 1.

NOTE ON THE SYMBOLIC REPRESENTATION OF NUCLIDES

There are various ways of indicating the characteristics of nuclei, and there is often divergence of opinion on this matter between chemists and physicists. In this book we shall use the convention:

$${}^{Z}_{A}\text{ Chemical Symbol} \qquad \text{e.g. } {}^{32}_{15}\text{P}.$$

The advantage of this method is that the nuclear characteristics are kept to one side, leaving the right hand side free for indications of valency and molecular state e.g. ${}^{35}_{17}\text{Cl}'$ indicates a radioactive chloride ion and ${}^{2}_{1}\text{H}_2$ indicates a molecule of heavy hydrogen. The alternative convention is to put the mass number on the right e.g. ${}_{15}\text{P}^{32}$. The reader may prefer this, and it is certainly easier to write, but we shall adhere to the other form. Since the charge is implicit in the chemical symbol, it may be omitted, and the symbol written as ${}^{32}\text{P}$ or ${}^{3}\text{H}$. The full form is however useful in nuclear equations when it is wished to balance both the masses and the charges on both sides.

Other common alternatives, which we have already used in this book, are to write the name or the symbol of the element followed by a hyphen and the mass number. Thus Carbon-14 or C-14. Unless they introduce clumsiness of phrase, they are quite acceptable, and are often used in describing labelled compounds.

Stable and Unstable Nuclei

Consider Table II, in which are listed the values of Z, N (the number of neutrons) and A, for some typical nuclei.

It will be seen that in this table the ratio of neutrons to protons varies. Some of these ratios give stability, and the isotopes are referred to as "Stable isotopes", whereas other ratios lead to instability and give rise to "Radioactive isotopes" (or "nuclides"!). Consideration of the stability of nuclei involves a study of the "binding energy" of the nucleus, which is the difference between

TABLE II

Element	Z	N	A	
Hydrogen	1	0	1	stable
	1	1	2	,, (heavy hydrogen)
	1	2	3	unstable (tritium)
Carbon	6	4	10	unstable
	6	5	11	,,
	6	6	12	stable
	6	7	13	,,
	6	8	14	unstable

the sum of the masses of the protons, neutrons and electrons associated with the atom, and the exact mass of the nuclide. This is dealt with at length in standard works on nuclear physics to which reference is made at the end of this chapter.

Modes of Disintegration

An unstable nucleus, such as that of Carbon-14, has an excess of energy. In order to achieve stability it undergoes a random rearrangement, during which energy is given out in the form of particles or radiation. Carbon-14 has one too many neutrons for stability. The nuclear rearrangement may be represented as the change of an uncharged neutron into a positively charged proton.

$$n \rightarrow p+\beta^-.$$

The positive charge is balanced by the formation of a negatively charged electron, which is emitted as beta radiation having maximum energy dependent on the change in binding energy resulting from the nuclear reaction. In many cases the nucleus still has an excess of energy after losing a beta particle. This is dissipated as gamma radiation, a form of electromagnetic radiation.

Since in this type of nuclear rearrangement an extra proton has been formed, the nuclide will change to one with an atomic number one unit higher. Thus:

$$^{14}_{6}\mathrm{C} \rightarrow {}^{14}_{7}\mathrm{N}+\beta^-.$$

Initially this nuclide will be formed as an ion, since there will be a deficit of one orbital electron, but it will gain one from its surroundings. The process is commonly referred to as BETA DECAY, (or preferably as BETA DISINTEGRATION).

The heaviest nuclides have such an excess of energy that they commonly lose larger units than beta particles in the disintegration process. These units are ALPHA PARTICLES, having a mass of four units, and a positive charge equal to two units. They are in fact the nuclei of helium atoms. An example of this type of disintegration is that of Radium-226. Thus:

$$^{226}_{88}\mathrm{Ra} \rightarrow {}^{222}_{86}\mathrm{Rn} + {}^{4}_{2}\mathrm{He}.$$

Since alpha particle emitters are not widely used as radioactive tracers, very little will be said about them in this book. It may be mentioned that there are four decay series associated with the alpha-emitting nuclides, both natural and artificial, in which there is a sequence of alpha and beta disintegrations terminating in stable isotopes. Three series end in lead, and the artificial series ends in ^{205}Tl. Further information about this subject will be found in the standard works on Nuclear Physics mentioned at the end of this chapter.

A further method of disintegration that occurs in a very few nuclides with an excess of neutrons, is SPONTANEOUS FISSION. Fission products will be mentioned in the section on the choice of isotopes. It is appropriate here to show diagrammatically (Fig. 2) the yield of the various nuclides formed in the fission of ^{235}U, that for ^{239}Pu being almost identical, but displaced two units to the right.

Turning to neutron-deficient nuclides, of which Carbon-11 is an example, there is an entirely different type of nuclear transformation. In this case a proton changes to a neutron, and a particle of the same mass as the electron but with a positive charge, is emitted. This is a POSITRON. The mechanism of this has been the subject of much recent study but we can express the reaction simply by an equation:

$$p \rightarrow n + \beta^{+}.$$

Positrons tend to interact with electrons giving annihilation radiation. This consists of two gamma rays of 0·51 MeV corresponding to the energy equivalent of two electron masses.

The other disintegration process occurring in nuclides having a neutron excess is ELECTRON CAPTURE. An electron is captured from an inner electron orbit, usually the K-shell, in which case the process is called "K-capture". Commonly the vacant orbit is filled by an electron from an outer orbit, giving rise to the emission of an X-ray, which is then the only external indication that a transformation has taken place.

Other processes are (a) INTERNAL CONVERSION, in which the energy of transition from an excited state is communicated to an orbital electron which is then ejected from the atom, (b) ISOMERIC TRANSITION in which a nucleus has a metastable state with a finite half-life, and falls to a lower energy level with the emission of a gamma ray or by some other process, such as internal conversion.

Before mentioning the diagrammatic representation of disintegration schemes it might be helpful to summarise the main modes of disintegration in a table.

TABLE III

PROCESS	RADIATION			EFFECT ON NUCLIDE	
	Type	Charge	Mass	Charge Z	Mass A
α-emission	α particle	+2	4	−2	−4
β-emission	β^- ,,	−1	0	+1	0
	β^+ ,,	+1	0	−1	0
γ-emission	γ-ray	0	0	0	0
Electron capture	X-rays	0	0	−1	0

Disintegration Schemes

The losses of energy and the changes in Z resulting from disintegration can conveniently be represented by a diagram called a DISINTEGRATION SCHEME. In the diagrams which follow, vertical distances represent energy, movement to the right represents a gain

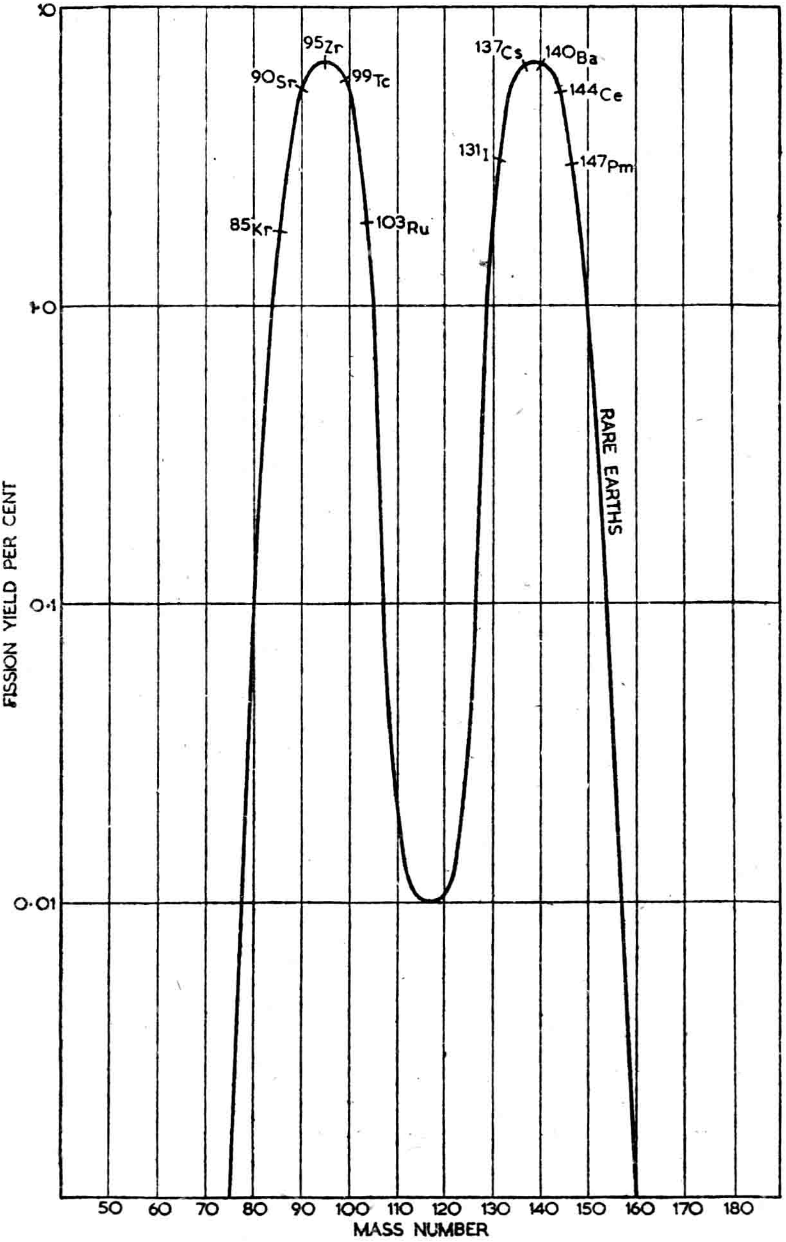

FIG. 2. Fission Yield from Uranium-235