Chapter 1

Atoms, nuclides and radionuclides

1.1 Introduction

1.1.1 Radioactivity, from the 1890s to the 1990s

Radioactivity is a characteristic of the nuclei of atoms. The nuclei, and with them the atoms as a whole, undergo spontaneous changes known as radioactive or nuclear transformations and also as decays or disintegrations. The energy released per nuclear transformation and carried away as nuclear radiation is, as a rule, some 10^3 to 10^6 times greater than the energy released per atom involved in chemical reactions.

Radioactivity was discovered in 1896 by the Frenchman H. Becquerel. The discovery occurred while he was experimenting with phosphorescence in compounds of uranium, an investigation aiming only at knowledge for its own sake. However, practical applications of radioactivity appeared not long after its discovery and have multiplied ever since.

Radioactivity could not have been discovered much before 1896 because at naturally occurring intensities it is undetectable by the unaided human senses. The photographic technique which contributed to its discovery was not adequately developed until well into the nineteenth century. But by the end of that century it played a major part in two discoveries which changed the path of science and of history: Röntgen's discovery of X rays in Germany in late 1895, followed by Becquerel's discovery of radioactivity in France in early 1896. These completely unexpected events opened the doors to totally new physical realities, to the emerging world of the nuclei of atoms and the high-energy nuclear radiations emitted by these nuclei.

Following Becquerel's discovery, it took about 35 years of intense scientific work before radioactive atoms could be produced from stable atoms by man-made procedures. Methods have now been developed to produce over 2000

0521553059 - Practical Applications of Radioactivity and Nuclear Radiations: An Introductory Text for Engineers, Scientists, Teachers and Students - G. C. Lowenthal and P. L. Airey

PERIOD	GROUP IA	IIA 2	IIIA 3	IVA 4	VA 5	VIA 6	VIIA 7	VIIA			IB	IIB	IIIB	IVB	VB	VIB	VIIB	VIIIB
1	1 H 1.008																	2 He 4.00
2	3 Li 6.94	4 Be 9.01											5 B 10.81	6 C 12.01	7 N 14.01	8 O 16.00	9 F 19.00	10 Ne 20.18
3	11 Na 22.99	12 Mg 24.31											13 Al 26.98	14 Si 28.09	15 P 30.97	16 S 32.06	17 Cl 35.45	18 Ar 39.95
4	19 K 39.10	20 Ca 40.08	21 Sc 44.96	22 Tl 47.90	23 V 50.94	24 Cr 52.00	25 Mn 54.94	26 Fe 55.85	27 Co 58.93	28 Ni 58.71	29 Cu 63.55	30 Zn 65.38	31 Ga 69.72	32 Ge 72.59	33 As 74.92	34 Se 748.96	35 Br 79.90	36 Kr 83.80
5	37 Rb 85.47	38 Sr 87.62	39 Y 88.91	40 Zr 91.22	41 Nb 92.91	42 Mo 95.94	43 Tc 98.91	44 Ru 101.07	45 Rh 102.91	46 Pd 106.4	47 Ag 107.87	48 Cd 112.40	49 In 114.82	50 Sn 118.69	51 Sb 121.75	52 Te 127.60	53 I 126.90	54 Xe 131.30
6	55 Cs 132.91	56 Ba 137.34	57 La 138.91	72 Hf 178.49	73 Ta 180.95	74 W 183.85	75 Re 186.2	76 Os 190.2	77 Ir 192.22	78 Pt 195.09	79 Au 196.97	80 Hg 200.59	81 Tl 204.37	82 Pb 207.2	83 Bi 208.98	84 Po [209]	85 At [210]	86 Rn [222]
7	87 Fr [223]	88 Ra [226]	89 Ac [227]	104 Rf [261]	105 Db [262]	106 Sg [263]	107 Bh [264]	108 Hs [265]	109 Mt [268]									

58 Ce 140.12	59 Pr 140.91	60 Nd 144.24	61 Pm [145]	62 Sm 150.4	63 Eu 151.96	64 Gd 157.25	65 Tb 158.93	66 Dy 162.50	67 Ho 164.93	68 Er 167.26	69 Tm 168.93	70 Yb 173.04	71 Lu 174.97
90 Th 232.04	91 Pa 231.04	92 U 238.03	93 Np 237.05	94 Pu [244]	95 Am [243]	96 Cm [247]	97 Bk [247]	98 Cf [251]	99 Es [254]	100 Fm [257]	101 Md [258]	102 No [255]	103 Lr [256]

Figure 1.1. The periodic table of the chemical elements as established by the early 1990s listing all elements identified and named by that date. The entries list the *Z* number, the symbol and the atomic weight. The lanthanum and actinide series are parts of periods VI and VII. For further comments see Sections 1.2.1, 1.2.2, 1.5.3.

radioisotopes of the chemical elements listed in the periodic table (Figure 1.1). However, the number of radionuclides with well known characteristics is much lower than that.

Charts of Nuclides published by nuclear research centres (see Section 4.2.1) list some 280 stable isotopes and commonly over 1800 radioisotopes, depending on the criteria used for the selection. Also, the number is increasing with time. There are on average four to eight radioisotopes for elements up to carbon (element 12 in the periodic table), 25 to 35 radioisotopes for each of the heavier of the stable elements, say between tantalum and bismuth (numbers 73 to 83), and there are the isotopes of the naturally occurring and man-made radioelements from polonium upwards. At present between 100 and 200 radionuclides are used on a regular basis, supporting a world-wide industry which continues to grow.

1.1.2 On the scope and content of this text

Clearly, large numbers of radionuclides are available and readers will be advised where to look for their characteristics. Routine applications require knowledge of the relevant physical and chemical properties of rarely more than a few of these radioisotopes, together with basic knowledge about radioactivity as offered in this or similar textbooks. Given these foundations, researchers, technologists, teachers and students will find nuclear radiation applications helpful assets for solving many of their day-to-day problems.

In working through this book experimenters seeking more specialised information will be referred to publications that deal with specific issues in more detail than can be covered in this book. It may be helpful to know in advance that many of these references cite the following publications.

Alfassi, Ed. (1990), *Activation Analysis*
Charlton, Ed. (1986), *Radioisotope Techniques for Problem Solving in Industry*
Debertin and Helmer (1988), *γ and X ray Spectrometry with Semiconducting Detectors*
Heath (1974), *Gamma Ray Spectrum Catalogue*
IAEA (1990), *Guidebook on Radioisotope Tracers in Industry*
L'Annunziata, Ed. (1998), *Handbook of Radioactivity Analysis*
Longworth, Ed. (1998), *The Radiochemical Manual*
Mann *et al.* (1991), *Radioactivity Measurements, Principles and Practice*
NCRP (1985), *A Handbook of Radioactivity Measurement Procedures*
Peng (1977), *Sample Preparation in Liquid Scintillation Counting*
Seltzer and Berger (1982), *Energy Losses and Ranges of Electrons and Positrons.*

Full titles and publishers are listed in the References at the end of this book.

As can be seen in the Contents, Chapters 1 to 6 deal principally with introductory material to the science of radioactivity while Chapters 7 to 9 introduce a large range of applications, briefly describing how they can be used. The two parts are interdependent. Descriptions of applications contain references to the earlier sections of the book where it is pointed out how to use radioactivity, while descriptions of the rules of radioactivity contain references to sections in Chapters 7 to 9 where these rules are put to use.

To conclude this introductory section, data illustrating the extent of radionuclide applications on a national and international level are now presented.

1.1.3 Joining a large scale enterprise

Nuclear power and nuclear radiation applications

Nuclear power for commercial electricity production first became available during the mid-1950s. During the mid-1990s, a total of 470 nuclear power reactors produced 17% of the world's commercially distributed electricity (Hore-Lacy, 1999, Table 6). Nuclear power production could expand as rapidly as it did because of the wide availability of high levels of expertise and training in nuclear science and engineering which led to a similarly high rate of expansion of nuclear radiation applications. It was no accident that nuclear power generation and other nuclear radiation applications advanced together.

Figures from Japan

The figures below were published by members of the Japan Radioisotope Association (Umezawa *et al*., 1996). Quoting from that report, it appears that in 1993 the number of facilities in Japan using radionuclides to a significant extent reached 5509, comprising:

Medical facilities	1385
Educational organisations	411
Research establishments	896
Industrial undertakings	1844
Other organisations	973

Radionuclide applications in industrial undertakings include large radiation processing facilities for food irradiation, the sterilisation of medical supplies and other commercial irradiation plant. There are also the figures referring to the types and quantities of radionuclides employed for non-medical and medical purposes, again for 1993. The total applies to radionuclides from 80 different elements.

First the number of orders, to the nearest thousand, for the purchase of non-medical applications:

Labelled compounds	63 000
Processed radionuclides	7 000

Coming to medical applications, the number of injections of radionuclides made during 1993 for diagnostic purposes exceeded 70 million. Since then most of the stated applications increased in number to a significant extent.

Among the major users of radioisotope applications it appears to be only in Japan where figures are published in such a way that they can be easily and concisely quoted. Other major users of nuclear techniques, notably the USA, produce, import and use radionuclides in a large number of independent installations. To quote these figures, and notably the role played by imported radionuclides, with sufficient accuracy requires more space than can be spared in this book.

The role of research reactors

Radionuclides for industrial and similar applications are not produced in nuclear power reactors, but in so-called research reactors. These were designed as essential tools for research in the nuclear sciences and for the production of radionuclides. The operation of research reactors will be briefly described in Sections 1.4.1 and 1.4.2.

Currently there are 280 research reactors operating in 54 countries (Hore-Lacy, 1999, p. 30), figures which demonstrate the large volume of applications and the fact that facilities for these applications are in constant operation in practically all industrialised countries around the globe.

1.2 An historic interlude: from atoms to nuclei

1.2.1 When atoms ceased to be atoms

During the fifth century BC Greek natural philosophers used the laws of logic to convince themselves and many later philosophers that matter must consist of infinitely small, hard spheres which they called atoms, the Greek word for uncuttable. This hypothesis remained without experimental backing for some 2300 years. It was only early in the nineteenth century that the English chemist John Dalton could revive the Greek model of the atom using experimental evidence to theorise that the atoms of different chemical elements differed in mass by integral multiples. Also, when atoms of different elements combine they do so in ratios of whole numbers and not otherwise.

By the 1860s Dalton's pioneering work had resulted in the discovery of over 60 chemical elements and sufficient new knowledge to lead to the formulation of what became known in due course as the periodic table of the elements proposed by the Russian chemist D.I. Mendeleyev and, independently by a German chemist L. Mayer. Figure 1.1 shows the present state of the table.

To add just a few words on its history, Mendeleyev published his arrangement of chemical elements in 1869. He had ordered them by their atomic weights, which were reasonably well known. Also, it was his idea to take care of the many similarities and differences in the properties of the elements by subdividing them into periods and groups. Beginning with about 60 elements in 1869, subsequent discoveries of elements were greatly aided by Mendeleyev's analysis. However, it was not until the coming of quantum theory during the late 1920s that Mendeleyev's periods and groups could be explained theoretically. A few explanatory comments on the periodic table will follow in Sections 1.2.2 and 1.5.3.

Although the theory behind Mendeleyev and Mayer's periodic table proved remarkably perceptive (Hey and Walters, 1987, Ch. 6), no-one anticipated the radically new knowledge which was to come as the nineteenth century ended.

The first totally inexplicable event occurred in late 1895 when W.C. Röntgen, Professor of Physics at the University of Würzburg, Germany, discovered the aptly named X rays, i.e. rays of unknown origin. The second discovery was equally inexplicable. It was made early in 1896 at the University of Paris, France, when Professor H. Becquerel discovered that the well known element uranium emits mysteriously penetrating radiations similar to X rays, a property later named radioactivity.

With the new century came two, truly revolutionary theories: quantum theory, the first stage of which was published in 1900 by Professor Planck in Berlin, Germany, and the special relativity theory published by Albert Einstein in 1905 and predicting, among others, that energy has mass (Section 1.3.2). These discoveries gave a radically new turn to the physical sciences of the twentieth century.

Following on from Becquerel's work, his student Marie Curie discovered (in 1898) two other radioactive elements she named polonium (after Poland, her country of birth) and radium (the radiator). She also identified the α and β radiations (so named by her) as charged particles emitted by radium and its daughters. A year earlier, in 1897, J.J. Thompson, a professor at Cambridge University, England, had discovered the electron which he called the atom of electricity and which was soon recognised as basic to all electric phenomena.

The most important advances involving radioactivity were made by the New Zealander Ernest Rutherford, then Professor of Physics at Manchester University, England.

Starting their researches during the first decade of the twentieth century, Professor Rutherford and his student-collaborators H. Geiger and E. Marsden used the α particles emitted by radium as projectiles to demonstrate that, although nearly all of them readily penetrated a very thin metal foil, a small fraction was actually backscattered by up to 180 degrees (Hey and Walters, 1987, Ch. 4).

These results were incompatible with the accepted theory of atomic structure due mainly to Professor Thompson, according to which the backscatter of α particles from thin metal foils was ruled out. Rutherford then theorised that the observed very low backscatter rate could be explained if atoms consist of extremely small and dense positively charged particles that he called protons, surrounded, at a relatively very large distance, by an equal number of very light negatively charged electrons (so ensuring electrical neutrality), but otherwise empty of matter. To explain the observed backscatter rate, the ratio of the diameter of the atom to that of its nucleus had to be of order 10^4. Since atomic diameters had been estimated as of order 10^{-10} m, the diameters of the nuclei of atoms would be as small as 10^{-14} m. These results were published in 1911, but they were totally inconsistent with existing knowledge and few researchers were willing to accept them.

Notwithstanding his successes, Rutherford was unable to explain how a nucleus consisting only of positively charged particles could avoid being torn apart by Coulomb repulsion which had to be extremely strong in so tiny a volume. He also could not explain why electrons could remain in stable orbits instead of spiralling towards the oppositely charged protons as demanded by Maxwell's electromagnetic theory, the validity of which had been tested in innumerable experiments.

Rutherford, Bohr, Heisenberg, Chadwick, Dirac and other well known physicists, most of whom were awarded Nobel Prizes, worked hard throughout the 1920s and 1930s until the properties of atoms and of their decay could be explained by the new quantum and relativity theories (Hughes, 2000).

1.2.2 *The atomic nucleus*

The just stated discoveries opened the way to the present knowledge that atomic nuclei consist of protons and neutrons known collectively as nucleons. It is also recognised that each nucleon consists of three smaller particles known as quarks, but the quark structure of nuclei will be ignored in what follows.

The elementary building blocks of nuclei are the positively charged protons (p), the charge being a single elementary unit, 1.602×10^{-19} coulomb (C), and the electrically uncharged neutrons (n). The number of protons in the nucleus determines its chemical nature, i.e. the chemical element to which the atoms belong. It is known as the atomic number of the element, designated Z, and serves as the order number in the periodic table (Figure 1.1).

Neutrons and protons are almost equal in mass (m), the mass ratio m_n/m_p being 1.0014, and they account for 99.95% of the inertial mass of the atom. The atomic electrons account for barely 0.05% of the inertial mass but they move within a volume some 10^{12} times larger than the volume of the nucleus, not along defined orbits as do planets but subject to probability-based relations derivable from quantum mechanics. The volume of the atom is practically empty of inertial matter, but it is permeated by intense electric fields between the charged particles. The force inside the nucleus which holds its components together is of an extremely short range ($\approx 10^{-13}$ m) and is known as the strong nuclear force.

Chemical interactions between elements are governed by forces involving the outer electrons of atoms, i.e. those least strongly bound to their nuclei. In contrast, the laws of radioactivity are determined by intra-nuclear forces which are many orders of magnitude higher than those involved in chemical interactions and well shielded from these interactions. This discovery helped to explain the almost complete independence of radioactive decay rates from all other properties of the radioactive material such as temperature, pressure or state of aggregation.

1.3 Nuclei, nuclear stability and nuclear radiations

1.3.1 The birth of isotopes

Dalton's ideas of integral atomic weights also ran into difficulties. As weighing techniques improved it was discovered that some 20% of the elements have atomic weights that are very nearly integers but not quite. For example, the atomic weight of phosphorus is not 31.00 but 30.97, close to 0.1% smaller than expected. The large majority of atomic weights differ from the nearest integer by amounts which are nearer to half a mass unit than to zero. It was later realised that the large majority of elements did not consist of a single species of atoms but of two and more species, each representing a well defined proportion of the total.

For elements consisting of more than one species of atom, each atom has the same number of protons in its nucleus. However, and this was only clarified with the discovery of the neutron in 1932, these species of atoms differ in the number of neutrons in their nuclei. This difference causes a difference in their atomic mass but has virtually no effect on their chemistry. Species of atoms that belong to the same element but differ in nuclear mass and so in atomic weight were labelled isotopes (derived from Greek and meaning same position) of the element in question.

Only about 20 elements consist of a single stable isotope as does phosphorus. The atoms of the other elements consist of several isotopes, with tin ($Z = 50$) consisting of ten isotopes. The atomic weights of these atoms are the averages of the weights of the isotopes weighted in accordance with their relative abundances. As could be expected, none of these averages is an integer (Figure 1.1).

1.3.2 Mass–energy conversions and the half life

Another characteristic of the nucleus is its mass number, designated A. It is the sum of the number of protons and neutrons in the nucleus with each of them assigned unit mass. The mass of the electrons is disregarded as is the small difference in mass between protons and neutrons. The mass number is then necessarily an integer given by $A = p + n = Z + n$ since $p = Z$ (Section 1.2.2). It is written as the raised prefix to the chemical symbol of the element, ^{A}Z.

Returning to phosphorus, its atoms each contain 15 protons and 16 neutrons, i.e. $A = 15 + 16 = 31$, written ^{31}P. But its atomic weight is 30.97 as noted earlier, a difference which is of fundamental importance.

Measurements of atomic weights are now made relative to the mass of the carbon isotope ^{12}C which is set as equal to 12 atomic mass units (amu). The energy equivalent of an atomic mass unit is calculated using the Einstein equation E (joules, J) $= mc^2$ where m is expressed in kilograms and c is closely equal to 3×10^8 m/s, the speed of light in vacuum. It can then be shown that 1 amu = 932 million electronvolts (MeV) as its energy equivalent. The electronvolt (eV) is related to the joule by 1 eV $= 1.602 \times 10^{-19}$ J.

If the 0.03 mass units which make up the difference between the mass number $A = 31$ and the atomic weight of phosphorus are expressed in energy terms (~30 MeV), one obtains the energy that holds the 31 nucleons in the phosphorus nucleus together. This energy is known as the nuclear binding energy (E_{Bi}) for phosphorus and is obtained at the expense of a small fraction of the inertial mass of the nucleons.

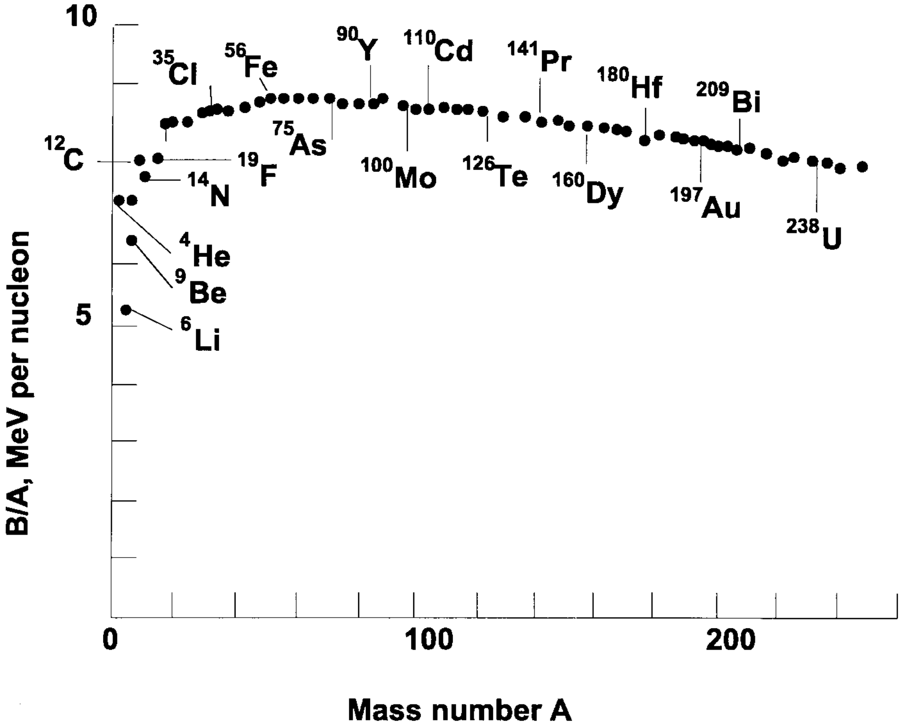

Figure 1.2. The binding energies per nucleon, B/A (B in MeV) as a function of the mass number $A(=p+n)$. Note the strongly bound helium nucleus (α particle). Adapted from Hey and Walters (1987, Ch. 5).

The magnitude of E_{Bi} increases with the mass number. However, the binding energy per nucleon, E_{Bi}/A, increases to a maximum near $Z=26$ (iron, Figure 1.1) where $A \approx 50$. It then decreases, being some 15% smaller near $Z=92$ (uranium) where $A \approx 238$, than near $A \approx 50$ (Figure 1.2).

The release of nuclear binding energy provides the energy for all radioactive transformations (Section 1.5.4). Also, the fact that E_{Bi}/A goes through a maximum near $A=50$ permits the release of very large amounts of energy in two situations: during the fusion of light nuclei, notably of hydrogen nuclei, and during the fissioning of a heavy nucleus, notably that of the uranium isotope ^{235}U. Uranium ($Z=92$) has numerous other isotopes, none of which is stable (Section 1.3.4) and two being of interest here: ^{235}U and ^{238}U with respectively $235-92=133$ neutrons and $238-92=136$ neutrons in their nuclei (Section 1.4.1).

Another nucleus to be noted (Figure 1.2) is that of helium (2p + 2n), better known as an α particle. The figure shows that helium is much more strongly bound than its neighbours, so helping to explain why its protons and