

Karl Heinrich Lieser

**Nuclear and Radiochemistry:  
Fundamentals  
and Applications**



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# Preface

This textbook gives a complete and concise description of the up-to-date knowledge of nuclear and radiochemistry and applications in the various fields of science. It is based on teaching courses and on research spanning over 40 years.

The book is mainly addressed to chemists desiring sound information about this branch of chemistry dealing with the properties of radioactive matter. Students and scientists working in other branches of chemistry, in environmental science, physics, geology, mineralogy, biology, medicine, technology and other fields will also find valuable information about the principles and applications of nuclear and radiochemistry.

Nuclear science comprises three overlapping fields, nuclear physics, nuclear and radiochemistry, and nuclear technology. Whereas nuclear physics deals with the physical properties of the atomic nucleus and the energetic aspects of nuclear reactions, in nuclear and radiochemistry the chemical aspects of atomic nuclei and of nuclear reactions (nuclear chemistry) and the chemical properties, preparation and handling of radioactive substances (radiochemistry) are considered. The concern of nuclear technology, on the other hand, is the use of nuclear energy, in particular the production of nuclear fuel and the operation of nuclear reactors and reprocessing plants. A well-founded knowledge of nuclear reactions and of nuclear and radiochemistry is needed in nuclear technology. Another related field, radiation chemistry, deals with the chemical effects of radiation, in particular nuclear radiation, and is more closely related to physical chemistry.

Research in nuclear and radiochemistry comprises: Study of radioactive matter in nature, investigation of radioactive transmutations and of nuclear reactions by chemical methods, hot atom chemistry (chemical effects of nuclear reactions) and influence of chemical bonding on nuclear properties, production of radionuclides and labelled compounds, and the chemistry of radioelements – which represent more than a quarter of all chemical elements.

Applications include the use of radionuclides in geo- and cosmochemistry, dating by nuclear methods, radioanalysis, the use of radiotracers in chemical research, Mössbauer spectrometry and related methods, the use of radionuclides in the life sciences, in particular in medicine, technical and industrial applications and investigations of the behaviour of natural and man-made radionuclides, in particular actinides and fission products, in the environment (geosphere and biosphere). Dosimetry and radiation protection are considered in the last chapter of the book.

Fundamentals and principles are presented first, before progressing into more complex aspects and into the various fields of application. With regard to the fact that radioactivity is a property of matter, chemical and phenomenological points of view are presented first, before more theoretical aspects are discussed. Physical properties of the atomic nucleus are considered insofar as they are important for nuclear and radiochemists.

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Endeavours are made to present the subjects in clear and comprehensible form and to arrange them in a logical sequence. All the technical terms used are defined when they are first introduced, and applied consistently. This is a glossary at the end of the text. In order to restrict the volume of the book, detailed derivations of equations are avoided and relevant information is compiled in tables, as far as possible. More complex relations are preferably elucidated by examples rather than by giving lengthy explanations.

For further reading, relevant literature is listed abundantly at the end of each chapter. Generally, it is arranged in chronological order, beginning with literature of historical relevance and subdivided according the subject matter, into general and more special aspects.

I am indebted to many colleagues for valuable suggestions and I wish to thank Mrs. Boatman for reading the manuscript.

Darmstadt, April 1996

K. H. Lieser

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# 1 Radioactivity in Nature

## 1.1 Discovery of Radioactivity

Radioactivity was discovered in 1896 in Paris by Henri Becquerel, who investigated the radiation emitted by uranium minerals. He found that photographic plates were blackened in the absence of light, if they were in contact with the minerals. Two years later (1898) similar properties were discovered for thorium by Marie Curie in France and by G. C. Schmidt in Germany. That radioactivity had not been discovered earlier is due to fact that human beings, like animals, do not have sense organs for radioactive radiation. Marie Curie found differences in the radioactivity of uranium and thorium and concluded that these elements must contain unknown radioactive elements. Together with her husband, Pierre Curie, she discovered polonium in 1898, and radium later in the same year.

Radioactivity is a property of matter and for the detection of radioactive substances detectors are needed, e.g. Geiger–Müller counters or photographic emulsions. It was found that these detectors also indicate the presence of radiation in the absence of radioactive substances. If they are shielded by thick walls of lead or other materials, the counting rate decreases appreciably. On the other hand, if the detectors are brought up to greater heights in the atmosphere, the counting rate increases to values that are higher by a factor of about 12 at a height of 9000 m above ground. This proves the presence of another kind of radiation that enters the atmosphere from outside. It is called cosmic radiation to distinguish it from the terrestrial radiation that is due to the radioactive matter on the earth. By cascades of interactions with the gas molecules in the atmosphere, cosmic radiation produces a variety of elementary particles (protons, neutrons, photons, electrons, positrons, mesons) and of radioactive atoms.

## 1.2 Radioactive Substances in Nature

Radioactive substances are widely distributed on the earth. Some are found in the atmosphere, but the major part is present in the lithosphere. The most important ones are the ores of uranium and thorium, and potassium salts, including the radioactive decay products of uranium and thorium. Uranium and thorium are common elements in nature. Their concentrations in granite are about 4 and 13 mg/kg, respectively, and the concentration of uranium in seawater is about 3 µg/l. Some uranium and thorium minerals are listed in Table 1.1. The most important uranium mineral is pitchblende ( $U_3O_8$ ). Uranium is also found in mica. The most important thorium mineral is monazite, which contains between about 0.1 and 15% Th.

The measurement of natural radioactivity is an important tool for dating, e.g. for the determination of the age of minerals (see section 16.1).

**Table 1.1.** Uranium and thorium minerals.

Mineral	Composition	Conc of U [%]	Conc of Th [%]	Deposits
Pitchblende	$U_3O_8$	60–90		Bohemia, Congo, Colorado (USA)
Becquerelite	$2UO_3 \cdot 3H_2O$	74		Bavaria, Congo
Uraninite		65–75	0.5–10	Japan, USA, Canada
Broeggerite	$UO_2 \cdot UO_3$	48–75	6–12	Norway
Cleveite		48–66	3.5–4.5	Norway, Japan, Texas
Carnotite	$K(UO_2)(VO_4) \cdot nH_2O$	45		USA, Congo, Russia, Australia
Casolite	$PbO \cdot UO_3 \cdot SiO_2 \cdot H_2O$	40		Congo
Liebigite	Carbonates of U and Ca	30		Austria, Russia
Thorianite	$(Th, U)O_2$	4–28	60–90	Ceylon, Madagascar
Thorite	$ThSiO_4 \cdot H_2O$	1–19	40–70	Norway, USA
Monazite	Phosphates of Th and Rare Earths		0.1–15	Brazil, India, Russia, Norway, Madagascar

The radioactive atoms with half-lives  $>1$  d that are found in nature are listed in Table 1.2. The table shows that radioactivity is mainly observed with heavier elements and seldom with light ones (e.g.  $^{40}K$  and  $^{87}Rb$ ).  $^{14}C$ ,  $^{10}Be$ ,  $^7Be$ , and  $^3H$  (tritium) are produced in the atmosphere by cosmic radiation. The production of  $^{14}C$  is about  $22 \cdot 10^3$  atoms of  $^{14}C$  per second per square metre of the earth's surface and about  $2.5 \cdot 10^3$  atoms of tritium per second per square metre of the earth's surface. Taking into account the radioactive decay and the residence times in the atmosphere, this means a global equilibrium inventory of about 63 tons of  $^{14}C$  and of about 3.5 kg of  $^3H$ .

The measurement of the natural radioactivity of  $^{14}C$  or of  $^3H$  is also used for dating. However, interferences have to be taken into account due to the production of these radionuclides in nuclear reactors and by nuclear explosions.

The energy produced by decay of natural radioelements in the earth is assumed to contribute considerably to the temperature of the earth. In particular, the relatively high temperature gradient of about  $30^\circ C$  per 1 km depth observed up to several kilometres below the surface is explained by radioactive decay taking place in the minerals, e.g. in granite.

**Table 1.2.** Naturally occurring radioactive species (radionuclides) with half-lives  $>1$  d (decay modes are explained in chapter 5).

Radioactive species (radionuclides)	Half-life	Decay mode	Isotopic abundance [%]	Remarks
$^{238}\text{U}$	$4.47 \cdot 10^9$ y	$\alpha, \gamma, e^{-}(\text{sf})$	99.276	Uranium family $A = 4n + 2$
$^{234}\text{U}$	$2.45 \cdot 10^5$ y	$\alpha, \gamma, e^{-}(\text{sf})$	0.0055	
$^{234}\text{Th}$	24.1 d	$\beta^{-}, \gamma, e^{-}$		
$^{230}\text{Th}$ (Ionium)	$7.54 \cdot 10^4$ y	$\alpha, \gamma(\text{sf})$		
$^{226}\text{Ra}$	1600 y	$\alpha, \gamma$		
$^{222}\text{Rn}$	3.82 d	$\alpha, \gamma$		
$^{210}\text{Po}$	138.4 d	$\alpha, \gamma$		
$^{210}\text{Bi}$	5.01 d	$\beta^{-}, \gamma(\alpha)$		Actinium family $A = 4n + 3$
$^{210}\text{Pb}$	22.3 y	$\beta^{-}, \gamma, e^{-}(\alpha)$		
$^{235}\text{U}$	$7.04 \cdot 10^8$ y	$\alpha, \gamma(\text{sf})$	0.720	
$^{231}\text{Th}$	25.5 h	$\beta^{-}, \gamma$		
$^{231}\text{Pa}$	$3.28 \cdot 10^4$ y	$\alpha, \gamma$		
$^{227}\text{Th}$	18.72 d	$\alpha, \gamma, e^{-}$		
$^{227}\text{Ac}$	21.77 y	$\beta^{-}, \gamma, e^{-}(\alpha)$		
$^{223}\text{Ra}$	11.43 d	$\alpha, \gamma$		Thorium family $A = 4n$
$^{232}\text{Th}$	$1.405 \cdot 10^{10}$ y	$\alpha, \gamma, e^{-}(\text{sf})$	100	
$^{228}\text{Th}$	1.91 y	$\alpha, \gamma, e^{-}$		
$^{228}\text{Ra}$	5.75 y	$\beta^{-}, \gamma, e^{-}$		
$^{224}\text{Ra}$	3.66 d	$\alpha, \gamma$		
$^{190}\text{Pt}$	$6.0 \cdot 10^{11}$ y	$\alpha$	0.013	
$^{186}\text{Os}$	$2.0 \cdot 10^{15}$ y	$\alpha$	1.58	
$^{187}\text{Re}$	$4.6 \cdot 10^{10}$ y	$\beta^{-}$	62.60	
$^{174}\text{Hf}$	$2.0 \cdot 10^{15}$ y	$\alpha$	0.16	
$^{176}\text{Lu}$	$3.6 \cdot 10^{10}$ y	$\beta^{-}, \gamma, e^{-}$	2.60	
$^{152}\text{Gd}$	$1.1 \cdot 10^{14}$ y	$\alpha$	0.20	
$^{147}\text{Sm}$	$1.06 \cdot 10^{11}$ y	$\alpha$	15.0	
$^{148}\text{Sm}$	$7 \cdot 10^{15}$ y	$\alpha$	11.3	
$^{144}\text{Nd}$	$2.1 \cdot 10^{15}$ y	$\alpha$	23.80	
$^{138}\text{La}$	$1.06 \cdot 10^{11}$ y	$\varepsilon, \beta^{-}, \gamma$	0.09	
$^{123}\text{Te}$	$1.3 \cdot 10^{13}$ y	$\varepsilon$	0.908	
$^{115}\text{In}$	$4.41 \cdot 10^{15}$ y	$\beta^{-}$	95.7	
$^{113}\text{Cd}$	$9.3 \cdot 10^{15}$ y	$\beta^{-}$	12.22	
$^{87}\text{Rb}$	$4.80 \cdot 10^{10}$ y	$\beta^{-}$	27.83	
$^{40}\text{K}$	$1.28 \cdot 10^9$ y	$\beta^{-}, \varepsilon, \beta^{+}, \gamma$	0.0117	
$^{14}\text{C}$	5730 y	$\beta^{-}$		Produced in the atmosphere by cosmic radiation
$^{10}\text{Be}$	$1.6 \cdot 10^6$ y	$\beta^{-}$		
$^7\text{Be}$	53.3 d	$\varepsilon, \gamma$		
$^3\text{H}$	12.33 y	$\beta^{-}$		

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## 2 Radioelements, Isotopes and Radionuclides

### 2.1 Periodic Table of the Elements

The Periodic Table of the elements was set up in 1869 by Lothar Meyer and independently by D. Mendeleev, in order to arrange the elements according to their chemical properties and to make clear the relationships between the elements. This table allowed valuable predictions to be made about unknown elements. With respect to the order of the elements according to their atomic numbers Moseley's rule proved to be very useful:

$$\sqrt{\nu} = a(Z - b)$$

( $\nu$  is the frequency of a certain series of X rays,  $Z$  is the atomic number, and  $a$  and  $b$  are constants).

The Periodic Table initiated the discovery of new elements which can be divided into three phases, overlapping chronologically:

- (a) Discovery of stable elements: The last of this group were hafnium (discovered in 1922) and rhenium (discovered in 1925). With these, the group of stable elements increased to 81 (atomic numbers 1 (hydrogen) to 83 (bismuth) with the exception of the atomic numbers 43 and 61). In addition, the unstable elements 90 (thorium) and 92 (uranium) were known.
- (b) Discovery of naturally occurring unstable elements: Uranium had already been discovered in 1789 (Klaproth) and thorium in 1828 (Berzelius). The investigation of the radioactive decay of these elements, mainly by Marie and Pierre Curie, led to the discovery of the elements with the atomic numbers 84 (Po = polonium), 86 (Rn = radon), 87 (Fr = francium), 88 (Ra = radium), 89 (Ac = actinium), and 91 (Pa = protactinium).
- (c) Discovery of artificial elements: The missing elements 43 (Tc = technetium) and 61 (Pm = promethium) have been made artificially by nuclear reactions. Element 85 (At = astatine) was also first produced by nuclear reaction, and later it was found in the decay products of uranium and thorium.

Of special interest was the discovery of the transuranium elements, because this meant an extension of the Periodic Table of the elements. So far 20 transuranium elements are known, beginning with element 93 (Np = neptunium), 94 (Pu = plutonium), 95 (Am = americium), 96 (Cm = curium), and ending at present with element 112. The first transuranium elements were discovered at Berkeley, California, by G. T. Seaborg and his group, the first reports about elements 104 to 107 came from Dubna, Russia, and the production of the last ones (elements 107 to 112) were first reported at Darmstadt, Germany. With increasing atomic number the stability decreases appreciably to values of the order of milliseconds, and the question whether an "island" of somewhat higher stability may be reached at atomic numbers of about 114 is still open.

The radioactive elements mentioned under (b) and (c) are called radioelements. All radioelements exist only in unstable forms. To-day 31 radioelements are known, with the atomic numbers 43, 61, and 84 to 112.

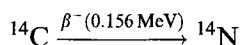
Some artificial elements (group c) have probably been produced in the course of the genesis of the elements and were present on the earth at the time of its formation. The age of the earth is estimated to be about  $4.5 \cdot 10^9$  y. During this time artificial elements of shorter half-life disappeared by nuclear transformations. After waiting for about  $10^6$  y, most of the artificial elements would have decayed again, and after a much longer time (about  $10^{12}$  y) the radioelements U and Th would also not exist any more on the earth in measurable amounts, with the consequence that the Periodic Table of the elements would end with element 83 (Bi = bismuth).

## 2.2 Isotopes and the Chart of the Nuclides

The investigation of the natural radioelements (group b) led to the realization that the elements must exist in various forms differing from each other by their mass and their nuclear properties. In fact, about 40 kinds of atoms with different half-lives were found, for which only 12 places in the Periodic Table of the elements were available on the basis of their chemical properties. The problem was solved in 1913 by Soddy, who proposed putting several kinds of atoms in the same place in the Periodic Table. This led to the term isotope which means “in the same place”. Isotopes differ by their mass, but their chemical properties are the same, if the relatively small influence of the mass on the chemical properties is neglected. Immediately after Soddy’s proposal, the existence of isotopes of stable elements was proved by Thompson (1913) using the method for analysis of positive rays, and with appreciably higher precision by Aston (1919), who developed the method of mass spectrography.

For many elements a great number of isotopes is known, e.g. tin has 10 stable and 18 unstable isotopes. Some elements have only one stable isotope, e.g. Be, F, Na, Al, P, I, Cs.

The various kinds of atoms differing from each other by their atomic number or by their mass are called nuclides. The correct name of unstable (radioactive) nuclides is radionuclides, and the terms radioelements for unstable elements and radionuclides for unstable nuclides are analogous. For identification, the symbol (or the atomic number) and the mass number are used. For example,  ${}^{14}_6\text{C}$  is carbon with the mass number 14 and the atomic number 6. The atomic number can be omitted ( ${}^{14}\text{C}$ ), because it is known by the symbol.  ${}^{14}\text{C}$  can also be written as C-14. For complete information, the kind and the energy of transmutation and the half-life may also be indicated:



It is evident that the Periodic Table of the elements does not have room to include information about all the isotopes of the elements. For that purpose the chart of the nuclides has been designed, which is based on the proton–neutron model of atomic

nuclei. In this model, protons and neutrons are considered to be the elementary particles building up the nuclei of atoms. They are therefore called nucleons. The number of nucleons in the nucleus is equal to the mass number, and the number of protons is equal to the atomic number. By combination of various numbers of protons and neutrons the atomic nuclei are obtained, as shown in Table 2.1 for light nuclei. Stable nuclei result from the combination of about equal numbers of protons and neutrons. The transfer of this information into a diagram in which the number of protons is plotted as ordinate and the number of protons as abscissa gives the chart

**Table 2.1.** Proton–neutron model of the nuclides ( $P$  = number of protons;  $N$  = number of neutrons).

$P$	$N$	Nuclide	Nuclide mass [u]	Natural abundance [%]	Atomic mass [u]	Remarks
1	0	$^1\text{H}$	1.007825	99.985	} 1.00797	Stable
1	1	$^2\text{H}$ (D)	2.014102	0.0155		Stable
1	2	$^3\text{H}$ (T)	3.016049			Unstable
2	1	$^3\text{He}$	3.016030	$\leq 0.0001$	} 4.00260	Stable
2	2	$^4\text{He}$	4.002603	100.00		Stable
2	3	$^5\text{He}$				Unstable
2	4	$^6\text{He}$	6.018891			Unstable
3	2	$^5\text{Li}$				Unstable
3	3	$^6\text{Li}$	6.015123	7.42	} 6.940	Stable
3	4	$^7\text{Li}$	7.016004	92.58		Stable
3	5	$^8\text{Li}$	8.022487			Unstable
3	6	$^9\text{Li}$	9.026790			Unstable
4	3	$^7\text{Be}$	7.016930			Unstable
4	4	$^8\text{Be}$	8.005305			Unstable
4	5	$^9\text{Be}$	9.012183	100.00	9.01218	Stable
4	6	$^{10}\text{Be}$	10.013535			Unstable
4	7	$^{11}\text{Be}$	11.021660			Unstable
5	3	$^8\text{B}$	8.024608			Unstable
5	4	$^9\text{B}$				Unstable
5	5	$^{10}\text{B}$	10.012938	19.6	} 10.811	Stable
5	6	$^{11}\text{B}$	11.009305	80.4		Stable
5	7	$^{12}\text{B}$	12.014353			Unstable
5	8	$^{13}\text{B}$	13.017780			Unstable
6	4	$^{10}\text{C}$	10.016858			Unstable
6	5	$^{11}\text{C}$	11.011433			Unstable
6	6	$^{12}\text{C}$	12.000000	98.892	} 12.0112	Stable
6	7	$^{13}\text{C}$	13.003354	1.108		Stable
6	8	$^{14}\text{C}$	14.003242			Unstable
6	9	$^{15}\text{C}$	15.010599			Unstable
6	10	$^{16}\text{C}$	16.014700			Unstable
7	5	$^{12}\text{N}$	12.018613			Unstable
7	6	$^{13}\text{N}$	13.005739			Unstable
7	7	$^{14}\text{N}$	14.003074	99.635	} 14.0067	Stable
7	8	$^{15}\text{N}$	15.000108	0.365		Stable
7	9	$^{16}\text{N}$	16.006099			Unstable
7	10	$^{17}\text{N}$	17.008449			Unstable

of the nuclides, the first part of which is shown in Fig. 2.1. The atomic number  $Z$  is equal to the number of protons  $P$  ( $Z = P$ ), and the mass number  $A$  is equal to the number of protons  $P$  plus the number of neutrons  $N$  ( $A = P + N$ ). Therefore,  $N = A - Z$ .

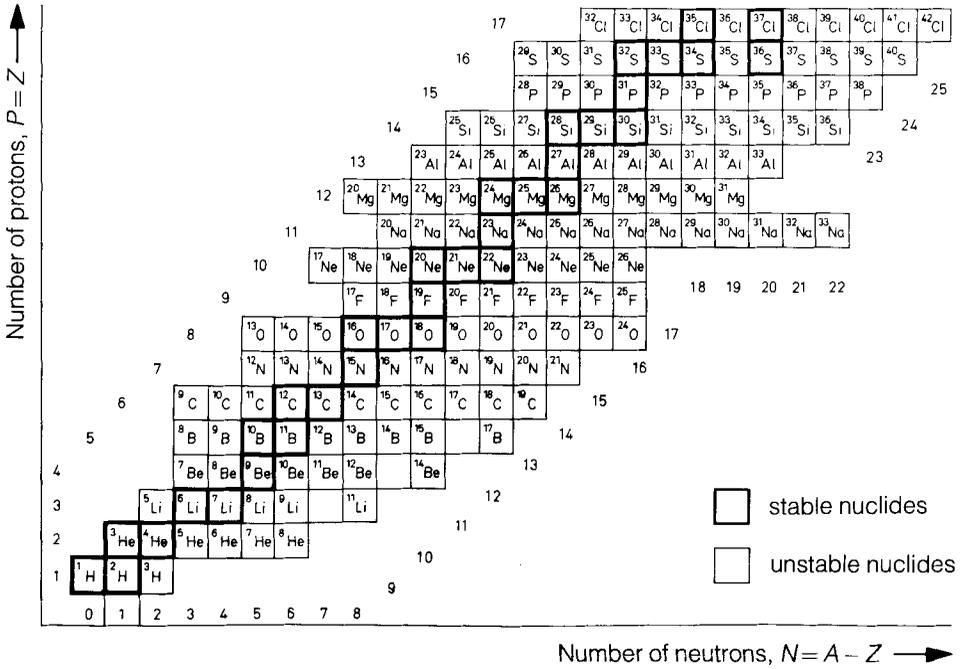


Figure 2.1. First part of the chart of nuclides.

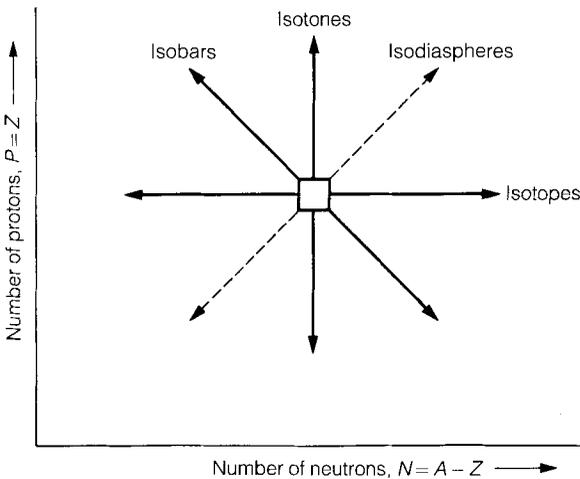
About 2800 nuclides are known. About 340 of these are found in nature and may be subdivided into four groups: (1) 258 are indisputably stable. (2) For 25 nuclides with atomic numbers  $Z < 80$  radioactive decay has been reported, but not confirmed for 7 of these. Many exhibit extremely long half-lives (9 nuclides  $> 10^{16}$  y and 4 nuclides  $> 10^{20}$  y), and radioactivity has not been proved unambiguously. Some have later been reported to be stable, and the 15 nuclides with half-lives  $> 10^{15}$  y may be considered to be quasistable. (3) Main sources of natural radioactivity comprising 46 nuclides are  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$  and their radioactive decay products. (4) Several radionuclides are continuously produced by the impact of cosmic radiation, and the main representatives of this group are  $^{14}\text{C}$ ,  $^{10}\text{Be}$ ,  $^7\text{Be}$  and  $^3\text{H}$ . Radionuclides present in nature in extremely low concentrations, such as  $^{244}\text{Pu}$  and its decay products or products of spontaneous fission of U and Th, are not considered in this list. Radionuclides existing from the beginning, i.e. since the genesis of the elements, are called primordial radionuclides. They comprise the radionuclides of group (2) and  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{232}\text{Th}$  and  $^{244}\text{Pu}$ .

The following groups of nuclides can be distinguished:

- Isotopes:  $Z = P$  equal
- Isotones:  $N = A - Z$  equal
- Isobars:  $A = N + Z$  equal
- Isodiaspheres:  $A - 2Z = N - Z$  equal

The positions of these groups of nuclides in the chart of the nuclides is shown in Fig. 2.2.

For certain nuclides, different physical properties (half-lives, mode of decay) are observed. They are due to different energetic states, the ground state and one or more metastable excited states of the same nuclide. These different states are called isomers or nuclear isomers. Because the transition from the metastable excited states to the ground states is “forbidden”, they have their own half-lives, which vary between some milliseconds and many years. The excited states (isomers) either change to the ground state by emission of a  $\gamma$ -ray photon (isomeric transition; IT) or transmutation to other nuclides by emission of  $\alpha$  or  $\beta$  particles. Metastable excited states (isomers) are characterized by the suffix m behind the mass number  $A$ , for instance  $^{60\text{m}}\text{Co}$  and  $^{60}\text{Co}$  or  $^{103\text{m}}\text{Ru}$ ,  $^{103\text{m}}\text{Ru}$  and  $^{103}\text{Ru}$ . Sometimes the ground state is indicated by the suffix g. About 400 nuclides are known to exist in metastable states.



**Figure 2.2.** Isotopes, isotones, isobars (and isodiaspheres) in the chart of nuclides.

By comparison of the number of protons  $P$  and the number of neutrons  $N$  in stable nuclei, it is found that for light elements (small  $Z$ )  $N \approx P$ . With increasing atomic number  $Z$ , however, an increasing excess of neutrons is necessary in order to give stable nuclei.  $A - 2Z$  is a measure of the neutron excess. For  $^4\text{He}$  the neutron excess is zero. It is 3 for  $^{45}\text{Sc}$ , 11 for  $^{89}\text{Y}$ , 25 for  $^{139}\text{La}$ , and 43 for  $^{209}\text{Bi}$ . Thus, if in the chart of the nuclides the stable nuclides are connected by a mean line, this line starts from the origin with a slope of 1 and is bent smoothly towards the abscissa. This mean line is called the line of  $\beta$  stability (Fig. 2.3).

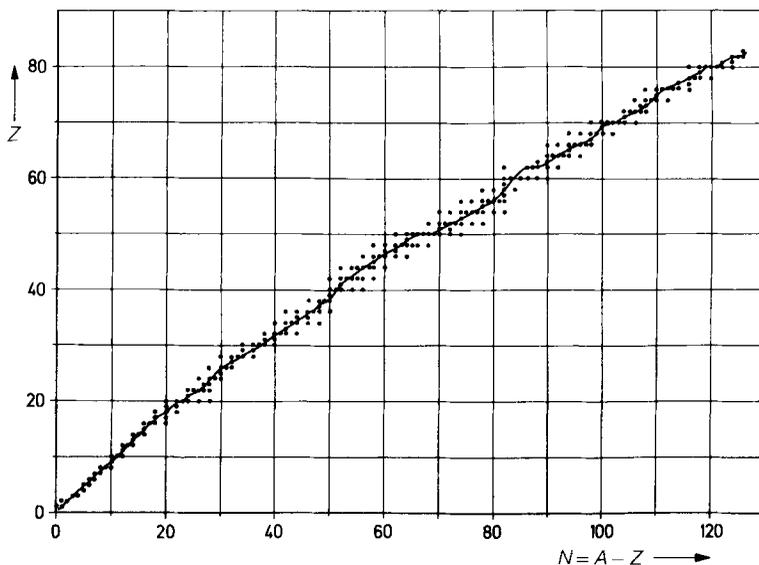


Figure 2.3. Stable nuclides and the line of  $\beta$  stability.

### 2.3 Stability and Transmutation of Nuclides

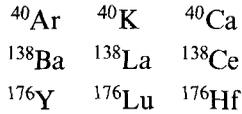
On the basis of the proton–neutron model of atomic nuclei the following combinations may be distinguished:

$P$ even, $N$ even (even–even nuclei)	Very common, 158 nuclei
$P$ even, $N$ odd (even–odd nuclei)	Common, 53 nuclei
$P$ odd, $N$ even (odd–even nuclei)	Common, 50 nuclei
$P$ odd, $N$ odd (odd–odd nuclei)	Rare, only 6 nuclei ( ${}^2\text{H}$ , ${}^6\text{Li}$ , ${}^{10}\text{B}$ , ${}^{14}\text{N}$ , ${}^{50}\text{V}$ , ${}^{180}\text{Ta}$ )

This unequal distribution does not correspond to statistics. The high abundance of even–even nuclei indicates the high stability of this combination. On the other hand, odd–odd nuclei seem to be exceptions. Four of the stable odd–odd nuclei are very light.

Alpha activity is preferably found for heavier elements  $Z = P > 83$  (Bi). Elements with even atomic numbers exhibit mainly  $\beta$  activity or electron capture. In the case of  $\beta$  decay or electron capture, the mass number  $A$  remains constant. Either a neutron is changed into a proton or a proton into a neutron. Thus, odd–odd nuclei are transformed into even–even nuclei – for instance,  ${}^{40}\text{K}$  into  ${}^{40}\text{Ar}$  or into  ${}^{40}\text{Ca}$ .

In finding nuclides of natural radioactivity, the Mattauch rule has proved to be very helpful. It states that stable neighbouring isobars do not exist (exceptions:  $A = 50, 180$ ). For instance, in the following sequences of isobars, the middle one is radioactive:



Detailed study of the chart of nuclides makes evident that for certain values of  $P$  and  $N$  a relatively large number of stable nuclides exist. These numbers are 2, 8, 20, 28, 50, 82 (126, only for  $N$ ). The preference of these “magic numbers” is explained by the shell structure of the atomic nuclei (shell model). It is assumed that in the nuclei the energy levels of protons and of neutrons are arranged into shells, similar to the energy levels of electrons in the atoms. Magic proton numbers correspond to filled proton shells and magic neutron numbers to filled neutron shells. Because in the shell model each nucleon is considered to be an independent particle, this model is often called the independent particle model.

## 2.4 Binding Energies of Nuclei

The high stability of closed shells (magic numbers) is also evident from the binding energies of the nucleons. Just below each magic number the binding energy of an additional proton or neutron is exceptionally high, and just above each magic number it is exceptionally low, similarly to the binding energies of an additional electron by a halogen atom or a noble gas atom, respectively.

Not all properties of the nuclei can be explained by the shell model. For calculation of binding energies and the description of nuclear reactions, in particular nuclear fission, the drop model of the nucleus has proved to be very useful. In this model it is assumed that the nucleus behaves like a drop of a liquid, in which the nucleons correspond to the molecules. Characteristic properties of such a drop are cohesive forces, surface tension, and the tendency to split if the drop becomes too big.

In order to calculate the binding energy ( $E_B$ ) of the nuclei, Weizsäcker developed a semi-empirical formula based on the drop model:

$$E_B = E_V + E_C + E_F + E_S + E_g \quad (2.1)$$

$E_B$  is the total binding energy of all nucleons. The most important contribution is the volume energy

$$E_V = +a_V A \quad (2.2)$$

where  $a_V$  is a constant and  $A$  is the mass number. The mutual repulsion of the protons is taken into account by the Coulomb term  $E_C$ :

$$E_C = -a_C \frac{Z(Z-1)}{A} \quad (2.3)$$

where  $a_C$  is a constant and  $Z$  is the atomic number.  $A^{1/3}$  is a measure of the radius of the nucleus and therefore also of the distance between the protons. With increasing

surface energy a drop of water becomes more and more unstable. Accordingly, in the drop model of the nucleus a surface energy term  $E_F$  is subtracted:

$$E_F = -a_F A^{2/3} \quad (2.4)$$

where  $a_F$  is again a constant and  $A^{2/3}$  is a measure for the surface. Neutrons are necessary to build up stable nuclei. But the excess of neutrons diminishes the total energy of the nucleus. This contribution is called the symmetry energy  $E_S$ :

$$E_S = -a_S \frac{(A - 2Z)^2}{A} \quad (2.5)$$

Finally, the relatively high stability of even–even nuclei is taken into account by a positive contribution to the total binding energy  $E_B$  of the nucleus, and the relatively low stability of odd–odd nuclei by a negative contribution. The following values are taken for this odd–even energy  $E_g$ :

$$E_g = \begin{cases} +\delta(A, Z) & \text{for even–even nuclei} \\ 0 & \text{for even–odd and odd–even nuclei} \\ -\delta(A, Z) & \text{for odd–odd nuclei} \end{cases} \quad (2.6)$$

The value of  $\delta$  is given approximately by  $\delta \approx a_g/A$ , where  $a_g$  is a constant. From the masses of the nuclides the following values have been calculated for the constants:  $a_V \approx 14.1$  MeV,  $a_C \approx 0.585$  MeV,  $a_F \approx 13.1$  MeV,  $a_S \approx 19.4$  MeV,  $a_g \approx 33$  MeV.

As can be seen from the various terms,  $E_B$  plotted as a function of  $Z$  will give parabolas, one parabola for odd mass numbers  $A$  ( $E_g = 0$ ) and two parabolas for even mass numbers  $A$  ( $E_g = \pm \delta$ ). These parabolas give the energetics for a certain row of isobars. Two examples are given in Figs. 2.4 and 2.5. The transmutation into stable nuclei proceeds by  $\beta$  decay or electron capture. As increasing binding energies are drawn towards the bottom, the stable nuclei are at the bottom of the curves. The unstable isobars are transformed stepwise, either by  $\beta^-$  decay from lower to higher atomic numbers or by  $\beta^+$  decay (alternatively by electron capture, EC, symbol  $\epsilon$ ) from higher to lower atomic numbers into stable nuclides. Nuclides at the bottom of the parabolas for odd–odd nuclei have two possibilities,  $\beta^-$  decay and  $\beta^+$  decay (or electron capture). The continuation of these parabolas through the whole chart of the nuclides gives the “valley of stable nuclei” and the “line of  $\beta$  stability”.

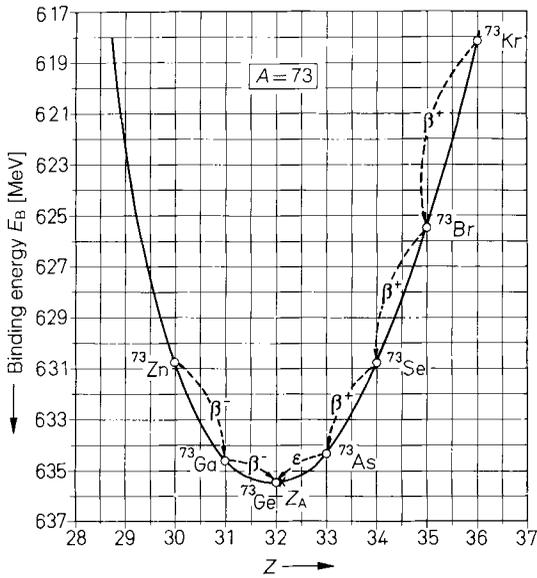


Figure 2.4. Binding energy and transmutation of nuclides with odd mass numbers.

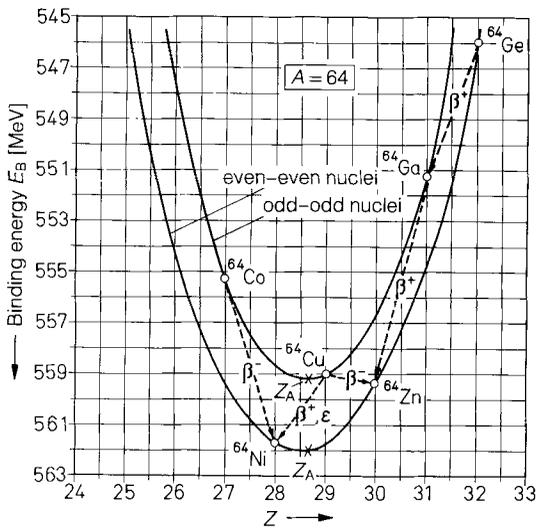


Figure 2.5. Binding energy and transmutation of nuclides with even mass numbers.

## 2.5 Nuclide Masses

The mass number  $A$  is equal to the number of nucleons,  $A = P + N$ , and is always an integer. The nuclide mass  $M$ , on the other hand, is the exact mass of the nuclides in universal atomic mass units  $u$ , and the atomic mass is the mean of the nuclide masses of the stable nuclides in their natural abundance.

The basis of the atomic mass unit  $u$  is the mass of the carbon isotope  $^{12}\text{C}$ :  $M(^{12}\text{C}) = 12.000000$ . Nuclide masses and atomic masses include the mass of the electrons of the neutral atom:  $M = \text{mass of the nucleus} + Zm_e$ , where  $Z$  is the atomic number and  $m_e$  the mass of one electron in atomic mass units  $u$ . One atomic mass unit is equal to  $(1.660566 + 0.000009) \cdot 10^{-24}$  g. The most accurate determinations are made by mass spectrometry with an error varying between  $10^{-8}$  and  $10^{-5}$   $u$ .

The mass  $m$  of particles travelling with very high velocities increases as the velocity approaches the velocity of light  $c$ :

$$m = \frac{m_0}{\sqrt{1 - (v/c)^2}} \quad (2.7)$$

where  $m_0$  is the mass of the particle at rest and  $v$  its velocity. Eq. (2.7) was derived by Einstein in his special theory of relativity. Another result of this theory is the equivalence of mass and energy:

$$E = mc^2 \quad (2.8)$$

The conversion factor is given by the square of the velocity of light  $c$ . Eq. (2.8) is of fundamental importance for all branches of nuclear science. For instance, it allows calculation of the energy that can be gained by conversion of matter into energy in nuclear reactions like nuclear fission or fusion. Since  $1 u = 1.660566 \cdot 10^{-24}$  g and  $c = 2.997925 \cdot 10^8$  m s $^{-1}$ ,  $1 u$  is equivalent to  $1.49244 \cdot 10^{-10}$  J. The energy units mainly used in nuclear science are eV (energy gained by an electron passing in vacuo a potential of 1 V;  $1 \text{ eV} = 1.60219 \cdot 10^{-19}$  J), keV and MeV. By application of these units it follows that

$$1 u = 931.5 \text{ MeV} \quad (2.9)$$

On the basis of the proton–neutron model of atomic nuclei, the following equation can be written for the mass of a nuclide:

$$M = ZM_{\text{H}} + NM_{\text{n}} - \delta M \quad (2.10)$$

where  $M_{\text{H}}$  is the nuclide mass of  $^1\text{H}$  and comprises the mass of one proton as well as that of one electron.  $M_{\text{n}}$  is the mass of the neutron in atomic mass units, and  $\delta M$  is called the mass defect. It is due to the fact that the binding energy  $E_{\text{B}}$  of the nucleons according to eq. (2.8) results in a decrease in the mass compared with the sum of the masses of the individual particles. The effect of the binding energy of the electrons is very small with respect to the binding energy of the nucleons and can be neglected.

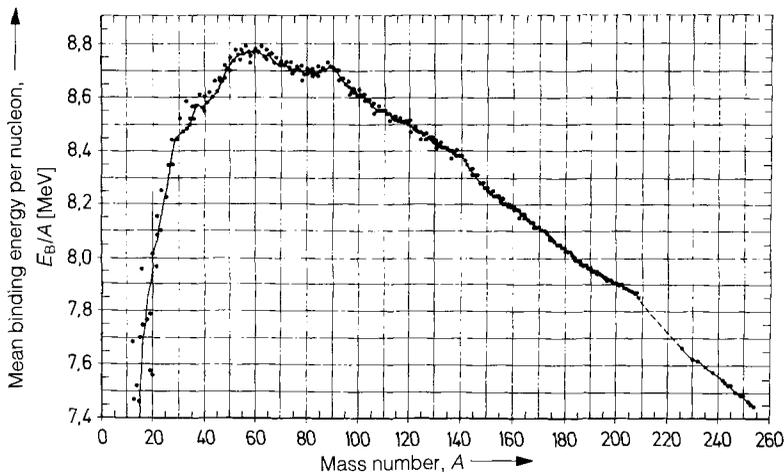
Application of eq. (2.8) gives

$$\delta M = \frac{E_B}{c^2} = ZM_H + NM_n - M \quad (2.11)$$

If  $E_B$  is divided by the mass number, the mean binding energy per nucleon is obtained, which is a measure of the stability of the nucleus:

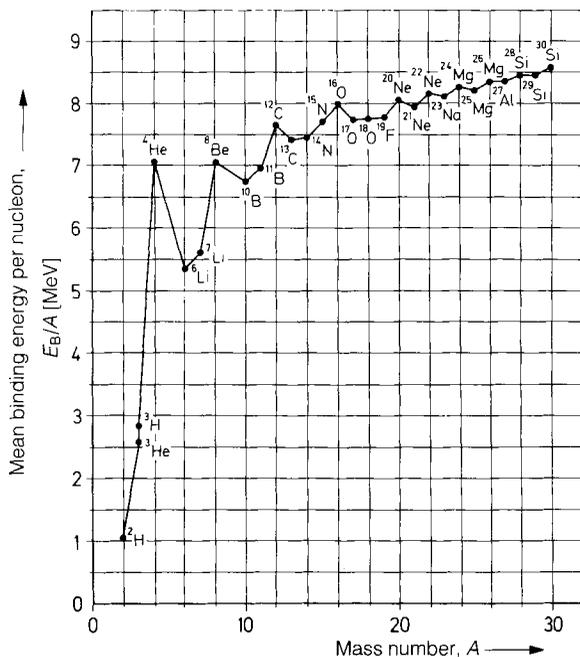
$$\frac{E_B}{A} = \frac{c^2}{A} (ZM_H + NM_n - M) \quad (2.12)$$

The mean binding energy per nucleon is plotted in Fig. 2.6 as a function of the mass number  $A$ . The figure shows that the elements with atomic numbers around that of iron have the highest mean binding energies per nucleon. Above  $Z \approx 90$  the mean binding energy decreases continuously. Thus, it can be deduced immediately from Fig. 2.6 that the fission of heavy nuclei into two smaller ones leads to a gain in energy. For instance, the difference in the mean binding energy for uranium atoms and two nuclides with half the mass number of uranium is about 1 MeV. As uranium contains about 200 nucleons, about 200 MeV should be gained by the fission of one uranium atom into two smaller atoms. This is the energy set free by nuclear fission in nuclear reactors.



**Figure 2.6.** Mean binding energy per nucleon.

On the other hand, in the range of light atoms, the even–even nuclei  ${}^4\text{He}$ ,  ${}^{12}\text{C}$  and  ${}^{16}\text{O}$  have particularly high mean binding energies of the nucleons. Values for light nuclei are plotted separately in Fig. 2.7. It is obvious from this figure that  ${}^4\text{He}$  is an extremely stable combination of nucleons, and very high energies may be obtained by fusion of hydrogen atoms ( ${}^1\text{H}$ ,  ${}^2\text{H}$  or  ${}^3\text{H}$ ) to  ${}^4\text{He}$ . This is the aim of the development of fusion reactors.



**Figure 2.7.** Mean binding energy per nucleon for light nuclides.

As the increase of the curve in Fig. 2.6 in the range of light nuclides is much steeper than the decrease in the range of heavy nuclides, the energy gained per mass unit of “fuel” is much higher for fusion than for fission. In the sun and in the stars the energy is produced mainly by nuclear fusion.

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