Jens-Volker Kratz

# Nuclear and Radiochemistry

Fundamentals and Applications

Fourth Edition

Volume 1



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Volume 1

4th Edition



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Jens-Volker Kratz

Volume 2

4th Edition



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#### Preface

This textbook aims at a complete and concise description of the present knowledge of nuclear and radiochemistry and applications in various fields of the natural sciences. It is based on teaching courses and research spanning several decades. The book is mainly addressed to advanced undergraduate students and to graduate students of chemistry. Students and scientists working in physics, geology, mineralogy, biology, medicine, and other fields will also find useful information about the principles and applications of nuclear and radiochemistry.

Traditionally, nuclear chemistry has been deeply tied to nuclear physics, cooperatively called nuclear science. At the same time, a wide field of applications of nuclear and radiochemistry in other sciences has developed. Therefore, it was considered important to bring together in one textbook a detailed presentation of the physical fundamentals as well as applied aspects of nuclear chemistry ranging from nuclear structure, nuclear masses, nuclear reactions, the production of *radionuclides* and labeled compounds, the chemistry of the *radioelements*, the study of radionuclides in the environment, all the way to the nuclear and radiochemistry needed in nuclear technology. Applications also include the use of radionuclides in analytical chemistry, in geo- and cosmochemistry, dating by nuclear methods, and the use of radionuclides in the life sciences and medicine. For further reading, the relevant literature is listed abundantly at the end of each chapter. Generally, it is arranged in chronological order, beginning with the literature of historical relevance, followed by more recent work subdivided according to the subject matter into general and more specialized aspects.

After the passing of Professor Karl Heinrich Lieser, the younger author, Jens-Volker Kratz, was approached by the Lieser family and by the publisher and was motivated to prepare a generally updated third edition of this textbook. The concept and structure of the book remained largely unchanged; however, new developments and results were incorporated, including the most recent references. These updates concerned the physical properties of atomic nuclei, the nuclear force and nuclear structure, techniques in nuclear chemistry, nuclear reactions, statistical considerations in *radioactivity* measurements, physics and chemistry of the actinides and transactinides, radionuclide mass spectrometry, and modern methods of speciation of radionuclides in the environment. These have been taken from teaching courses held at the Johannes Gutenberg University over the last 30 years.

The third revised edition of Jens-Volker Kratz and Karl Heinrich Lieser "Nuclear and Radiochemistry, Fundamentals and Applications" appeared in late 2013. This successful textbook was close to being sold out in late 2019, why Wiley-VCH re-contacted Jens-Volker Kratz and asked him to prepare a fourth edition with perhaps some 20% of new contents. This was accepted after the development of a suitable concept in April 2020.

The new trend, that was already visible in the third edition, i.e. more weight on the physical aspects of modern nuclear chemistry and less on the traditional fields of radiochemistry, e.g. on radioanalysis and radiotracers in chemistry, as compared to the earlier Lieser editions, has now been even more pronounced: the standard model of particle physics is now enriched by introducing the newly discovered heavy scalar Higgs boson and the omni-present Higgs field, the statistical assessment of lifetimes in the  $\alpha$ -decay chains of odd-Z superheavy elements is illuminated, the important use of recoil momenta and average charge states in gas-filled separators is discussed. the presentation of nuclear heavy-ion reactions is much enlarged, new methods of reprocessing and the treatment of nuclear waste are presented, new insights into the physical and chemical properties of the superheavy elements are discussed, as well as the fascinating news about neutrino masses, to name a few highlights. On the other hand, to avoid an unreasonable increase of the size of the two volumes of the fourth edition, some of the traditional Lieser chapters dealing with fields, in which today, research is no more active, have been omitted. To avoid a wrong impression, the author of the new edition along with the editors of Wiley-VCH want to clarify that the concept and the structure of the book are still basically unchanged, and we deeply appreciate that the roots planted by Karl Heinrich Lieser with the early editions of his book series are still alive and visible.

As in the previous edition, it is my pleasure to thank Mrs. Petra Sach-Muth for help and Mr. Jürgen Hubrath for professionally producing a number of new figures.

Mainz, April 2021

Jens-Volker Kratz

#### **Fundamental Concepts**

Nuclear and radiochemistry cover a wide spectrum of areas such as (i) studies of the chemical and physical properties of the heaviest human-made elements; (ii) studies of nuclear structure, nuclear reactions, and radioactive decay, (iii) studies of nuclear processes in the Universe, such as geochronology and cosmochemistry; and (iv) applications of radioactivity in a vast variety of fields such as radioanalysis, chemistry, life sciences, and industrial applications, and in the geo- and biosphere. Nuclear chemistry has ties to all traditional areas of chemistry. Nuclear chemists are involved in the preparation of radiopharmaceuticals for use in medicine. Radiometric techniques play an important role in analytical chemistry and are often used as references validating other analytical techniques. The study of the actinide and transactinide elements has traditionally involved nuclear chemists studying the limits of nuclear stability and the periodicity of the periodic table of the elements. The physical concepts at the heart of nuclear chemistry have their roots in nuclear physics. Thus, nuclear physics and nuclear chemistry overlap and are cooperatively called nuclear science. However, there are distinctions between these related fields. Besides the close ties to chemistry mentioned earlier, nuclear chemists are studying nuclear problems in different ways than nuclear physicists. Nuclear physics tends to look into the fundamental interactions between subatomic particles and fundamental symmetries. Nuclear chemists have focused on more complex phenomena where statistical properties are important. Nuclear chemists are more involved in applications of nuclear phenomena. For example, the nuclear fuel cycle or the migration of *radionuclides* in the environment is so inherently chemical that they involve nuclear chemists almost exclusively. The other term, radiochemistry, refers to the chemical applications of radioactivity and of related phenomena. Radiochemists are nuclear chemists but not all nuclear chemists are radiochemists. There are many nuclear chemists who use purely instrumental, physical techniques for their research and thus their work is not radiochemistry.

#### 1.1 The Atom

The atom is the smallest unit a chemical element can be divided into without losing its chemical properties. The radii of atoms are on the order of  $10^{-10}$  m (Å). The



Figure 1.1 Schematic representation of the relative sizes of the atom and the nucleus.

atomic nucleus, see Figure 1.1, is a very small object with a radius on the order of  $1-10 \cdot 10^{-15}$  m (femtometer, fm, called fermi) in the center of the atom and contains almost the entire mass of the atom. It contains Z protons, where Z is the atomic number of the element. Being the number of protons, Z is thus the number of positive charges in the nucleus. The nucleus also contains N neutrons, where N is the neutron number. Neutrons are uncharged particles with masses almost identical to the proton mass. Electrons surround the nucleus. Electrons are small negatively charged particles with a mass of 1/1836 of the proton mass. The electrons are bound electrostatically to the positively charged nucleus. In a neutral atom, the number of electrons equals the number of protons in the nucleus. The chemistry of the element is controlled by Z. From quantum mechanics, we know that only certain discrete energies and angular momenta of the electrons are allowed. These quantized states are schematically depicted in Figure 1.1. Later, in Chapter 5, we will see also that nucleons occupy orbits with discrete energies and angular momenta. However, the sizes and energies of atomic and nuclear processes are very different, allowing us to consider them separately.

#### 1.2 Atomic Processes

In the inelastic collision of two atoms, we can anticipate (i) excitation of one or both atoms involving a change in electron configuration or (ii) ionization of one or both atoms, that is, removal of one or more electrons from the atom to form a positively charged ion. For this process to occur, an atomic electron must receive an energy exceeding its binding energy. This energy far exceeds the kinetic energies of gaseous atoms at room temperature. Thus, the atoms must have high kinetic energies as a result of nuclear decay or acceleration to eject electrons from other atoms in atomic collisions. When an electron in an outer atomic electron shell drops down to fill a vacancy in an inner electron shell, electromagnetic radiation called X-rays is emitted. In Figure 1.2, an L-shell electron is shown filling a K-shell vacancy. In the transition, a characteristic K X-ray is emitted. The energy of the X-rays is equal to the difference in the binding energies of the electrons in the two shells, which **Figure 1.2** Scheme showing X-ray emission when a vacancy in an inner electron shell caused by nuclear decay is filled. An L-shell electron is shown filling a K-shell vacancy associated with K X-ray emission.



depends on the atomic number of the element. Specifically, X-rays due to transitions from the L shell to the K shell are called  $K_{\alpha}$  X-rays, while X-rays due to transitions from the M to K shells are termed  $K_{\beta}$  X-rays. Refining further,  $K_{\alpha 1}$  and  $K_{\alpha 2}$  designate transitions from different subshells of the L shell, that is,  $2p_{3/2}$  (L<sub>III</sub>) and  $2p_{1/2}$  (L<sub>II</sub>). X-rays for transitions from M to L are  $L_{\alpha}$  X-rays. For each transition, the change in orbital angular momentum  $\Delta \ell$  and total angular momentum  $\Delta j$  must be  $\Delta \ell = \pm 1$  and  $\Delta j = 0, \pm 1$ .

For a hydrogen-like atom, the Bohr model predicts that the transition energy  $\Delta E$  is

$$\Delta E = E_{\rm i} - E_{\rm f} = R_{\infty} hcZ^2 \left( \frac{1}{n_{\rm i}^2} - \frac{1}{n_{\rm f}^2} \right)$$
(1.1)

where  $R_{\infty}$  is the Rydberg constant, *h* the Planck constant, *c* the speed of light, and *n* the principal quantum number of the electron. The X-ray energy  $E_x = -\Delta E$ , after inserting the physical constants, is

$$E_x = 13.6Z^2 \left(\frac{1}{n_{\rm f}^2} - \frac{1}{n_{\rm i}^2}\right) \,\mathrm{eV}$$
(1.2)

For  $K_{\alpha}$  X-rays from hydrogen-like atoms

$$E_x = 13.6Z^2 \left(\frac{1}{1^2} - \frac{1}{2^2}\right) \text{ eV}$$
(1.3)

and for  $L_{\alpha}$  transitions

$$E_x = 13.6Z^2 \left(\frac{1}{2^2} - \frac{1}{3^2}\right) \text{ eV}$$
 (1.4)

In a realistic atom, Z must be replaced by  $Z_{\text{effective}}$  to take care of the screening of the nuclear charge by other electrons. Henry Moseley showed the frequencies,  $\nu$ , of the  $K_{\alpha}$  X-rays scale as

$$v^{1/2} = \operatorname{const}(Z - 1)$$
 (1.5)

and those of the  $L_{\alpha}$  X-rays scale as

$$v^{1/2} = \text{const}(Z - 7.4) \tag{1.6}$$

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Thus, Moseley showed that the X-ray energies, *hv*, depend on the square of an altered, effective atomic number due to screening. The relative intensities of different X-rays depend on the chemical state of the atom, its oxidation state, complexation with ligands, and generally on local electron density. The relative intensities are, therefore, useful in chemical speciation studies. As will be discussed in Chapter 6, radioactive decays can be accompanied by X-ray production and the latter may be used to identify the decaying nucleus.

#### 1.3 Discovery of the Atomic Nucleus

Before the discovery of radioactivity, elements were considered as unchangeable substances. In 1897, J.J. Thomson discovered the electron and concluded that the atom must have a structure. As the mass of the electron is roughly 1/2000 of the mass of hydrogen, he concluded that most of the mass of the atom must be contained in the positively charged constituents. It was assumed that negative and positive charges are evenly distributed over the atomic volume.

In 1911, Ernest Rutherford studied the scattering of  $\alpha$  particles in thin metal foils. He found that backscattering to  $\theta > 90^{\circ}$  was more frequent than expected for multiple scattering from homogeneously charged atoms. This led Rutherford to postulate the existence of an atomic nucleus having mass and positive charges concentrated in a very small volume. The nucleus was supposed to be surrounded by electrons at the atomic diameter and the electrons do not contribute to the  $\alpha$ -particle scattering. He postulated the following ansatz: the nuclear charge is *Ze*; that of the  $\alpha$  particle is  $Z_{\alpha} = 2e$ . The scattering force is the Coulomb force. The nucleus is at rest in the collision, and the path of an  $\alpha$  particle in the field of the nucleus is a hyperbola with the nucleus at the external focus. From these simplifying geometric properties and from the conservation of momentum and energy, Rutherford derived his famous scattering formula which relates the number  $n(\theta)$  of  $\alpha$  particles scattering angle  $\theta$ 

$$n(\theta) = n_0 \frac{Nt}{16r^2} \left(\frac{ZeZ_{\alpha}e}{\frac{1}{2}M_{\alpha}v_{\alpha}^2}\right)^2 \frac{1}{\sin^4(\theta/2)}$$
(1.7)



**Figure 1.3** Schematic representation of the Rutherford scattering experiment. A collimated beam of  $\alpha$  particles ( $n_0$  number of ingoing  $\alpha$  particles with velocity  $v_{\alpha}$  and rest mass  $M_{\alpha}$ ) hits a gold foil F (thickness *t*, *N* number of target nuclei per cubic centimeter) and is scattered to the polar angle  $\theta$  under which a scintillator S at distance *r* from the target detects  $n(\theta)$  scattered particles.

with  $n_0$  being the number of incident  $\alpha$  particles, *t* the thickness of the target foil, *N* the number of target nuclei per unit volume, and  $M_{\alpha}$  and  $v_{\alpha}$  the mass and initial velocity of the  $\alpha$  particle.

Precision measurements by Hans Geiger and Ernest Marsden soon verified that, for sufficiently heavy scatterers, the number of scattered particles detected per unit area was indeed inversely proportional to the square of the  $\alpha$ -particle energy and to the fourth power of the sine of half the scattering angle. In principle for all, but notably only for light target nuclei, Eq. (1.7) must be modified because the target nucleus is not at rest. This can be accommodated by inserting the center of mass energy instead of the laboratory energy and by using the reduced mass instead of the rest mass. Figure 1.4 shows the apparatus used by Geiger and Marsden. It resembled an exsiccator that could be evacuated. The upper part contained the  $\alpha$ -particle source (in German Emanationsröhrchen, R) in a lead brick. The collimated beam of  $\alpha$  particles passed a gold foil F. The  $\alpha$  particles that, after scattering in F, interacted with the scintillator S were observed through the microscope M. The microscope together with the scintillator could be moved to different scattering angles  $\theta$  by turning the flange (Schliff, Sch). Figure 1.5 shows the results obtained by Geiger and Marsden. They agree in an impressive way over 5 orders of magnitude with the theoretical dependence  $(1/\sin^4(\theta/2))$  for pure Coulomb scattering. This way, it was possible to study systematically the magnitude of the nuclear charge in the atoms of given elements through scattering experiments since the scattered intensity depends on the square of the nuclear charge. It was by the method of  $\alpha$ -particle scattering



**Figure 1.4** Experimental setup by Geiger and Marsden for the observation of Rutherford scattering of  $\alpha$  particles in a gold foil F. The radioactive source R is contained in a lead housing. The scattered  $\alpha$  particles are interacting with the scintillator S that is observed by a microscope M. The microscope together with the scintillator could be turned to variable scattering angles  $\theta$  by turning the flange. Source: Geiger and Marsden (1913), figure 1 (p. 607)/Taylor & Francis.



that nuclear charges were determined and this led to the suggestion that the atomic number Z of an element was identical to the nuclear charge. Further understanding of atomic structure developed rapidly through the study of X-rays and optical spectra, culminating in Niels Bohr's theory of 1913 and Erwin Schrödinger's and Werner Heisenberg's quantum mechanical description of the atom in 1926.

## 1.4 Nuclear Decay Types

*Radioactive decay* involves the spontaneous emission of radiation by an unstable nucleus. While this subject will be discussed in detail in Chapter 6, we present here a general introduction. In Table 1.1, we summarize the characteristics of the various decay types. Three basic decay modes were discovered by Rutherford starting in 1899:  $\alpha$  decay,  $\beta$  decay, and  $\gamma$  radiation. He found that  $\alpha$  particles are completely absorbed in thin metal foils, for example, 15 µm of Al.  $\beta$  particles were found to be largely absorbed only in Al a 100 times thicker. An absorption equation  $I = I_0 e^{-\mu d}$  was found where  $\mu$  is a mass absorption coefficient (cm<sup>-1</sup>) depending on *Z* of the

Decay mode	Symbol	Emitted particle		Decay process and example (in short form)
α decay	α	Helium	${}^{4}_{2}\text{He}^{2+}$	${}^{A}Z \rightarrow {}^{A-4}(Z-2) + {}^{4}_{2}\text{He}^{2+}$
β decay	β-	Electron and antineutrino	$\frac{1}{\overline{\nu}_{e}}^{0}e^{-1}$	${}^{1}_{0}n \rightarrow {}^{1}_{1}p + {}^{0}_{1}e^{-} + {}^{0}_{0}\overline{\nu}_{e}$ (in the nucleus) ${}^{A}Z \rightarrow {}^{A}(Z+1)$ ${}^{14}C(\beta^{-}){}^{14}N$
	$\beta^+$	Positron and neutrino	${}^0_1 e^+$ $ u_e$	${}^{1}_{1}p \rightarrow {}^{0}_{0}n + {}^{0}_{1}e^{+} + {}^{0}_{0}\nu_{e} \text{ (in the nucleus)}$ ${}^{A}Z \rightarrow {}^{A}(Z-1)$ ${}^{11}C(\beta^{+}){}^{11}B$
Electron capture (EC)	ε	Neutrino and X-ray of the daughter nuclide	ν <sub>e</sub>	$ \begin{array}{l} {}^{1}_{1}p(\text{nucleus}) + {}^{0}_{-1}e^{-}(\text{electron shell}) \rightarrow \\ {}^{0}_{0}n + {}^{0}_{0}\nu_{e} \\ {}^{A}Z \rightarrow {}^{A}(Z-1) \\ {}^{37}\text{Ar}(\varepsilon)^{37}\text{Cl} \end{array} $
$\gamma$ transition	γ	Photon	( <i>hv</i> )	Electromagnetic decay of an excited nucleus
Internal conversion (IC)	e <sup>-</sup>	Conversion electron and accompanying processes		Transfer of excitation energy to an electron in the shell <sup>58m</sup> Co(e <sup>-</sup> ) <sup>58</sup> Co
Spontaneous fission	sf	Fission fragments		${}^{A}Z \rightarrow {}^{A'}Z' + {}^{A-A'}(Z - Z')$ ${}^{254}Cf(sf)$
Proton decay	р	Proton	${}_{1}^{1}p$	${}^{A}Z \rightarrow {}^{A-1}(Z-1) + {}^{1}_{1}p$ ${}^{147}\text{Tm}(p)^{146}\text{Er}$
Cluster decay	С	Cluster		$^{223}\text{Ra} \rightarrow {}^{14}\text{C} + {}^{209}\text{Pb}$

**Table 1.1** Characteristics of radioactive decay modes.

absorber and *d* was the thickness in centimeter.  $\gamma$  radiation was found to be almost not absorbed (in aluminum) and a mass absorption coefficient depending on  $Z^5$  was associated with it. Therefore, today, thick bricks of lead are commonly used in radiochemical laboratories for shielding purposes. Recognition of the character of the  $\alpha$ and  $\beta$  rays as high-speed charged particles came largely from magnetic and electrostatic deflection experiments in which  $\beta$  particles were seen to be electrons. From the deflection of  $\alpha$  particles, the ratio of charge to mass was found to be half that of the hydrogen ion. The suggestion that  $\alpha$  particles were <sup>4</sup>He<sup>2+</sup> ions was immediately made. This was proven in 1903 by William Ramsay in an experiment in which  $\alpha$  rays were allowed to pass through a very thin glass wall into an evacuated glass vessel. Within a few days, sufficient helium gas was accumulated in the glass vessel and was detected spectroscopically.  $\gamma$  radiation was found not to be deflected in the magnetic field and was recognized to be electromagnetic radiation. The difference to the atomic X-ray radiation, however, was not clear at that time.

Nuclear  $\beta$  decay occurs in three ways:  $\beta^-$ ,  $\beta^+$ , and electron capture (EC). In these decays, a nuclear neutron or proton changes into a nuclear proton or neutron,

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respectively, with the simultaneous emission of an antineutrino or an electron neutrino and an electron or positron. In EC, an orbital electron is captured by the nucleus changing a proton into a neutron with the emission of a monoenergetic neutrino. Due to the creation of a hole in the electron shell, the subsequent emission of X-rays or Auger electrons occurs. The *mass number A* remains constant in these decays while the atomic number Z is increased by 1 unit in  $\beta^-$  decay and decreased by 1 unit in  $\beta^+$  decay and EC. In  $\beta^-$  and  $\beta^+$  decay, the decay energy is shared between the emitted  $\beta$  particle, the (anti)neutrino, and the recoiling daughter nucleus.

Nuclear electromagnetic decay occurs in two ways:  $\gamma$  emission and internal conversion (IC). A nucleus in an excited state decays by the emission of a high-energy photon or the same excited nucleus transfers its decay energy radiation-less to an orbital electron that is ejected from the atom. As in EC, the creation of a hole in the electron shell causes accompanying processes to occur, such as X-ray emission. There is no change in the number of the nucleons.

In 1940, K.A. Petrzhak and G.N. Flerov discovered spontaneous fission of  $^{238}$ U when they spread out a thin layer of uranium in a large area ionization chamber operated in a Moscow underground train station (to shield against cosmic radiation), observing large ionization bursts much larger than the pulse heights of the abundantly emitted  $\alpha$  particles. A spontaneous fission half-life of  $10^{16}$  years was estimated. It was concluded that the gain in binding energy delivers the decay energy when a nucleus with *A* nucleons splits into two fission fragments of roughly *A*/2.

In 1981, the emission of monoenergetic protons was discovered by S. Hofmann et al. at the GSI Helmholtz Center for Heavy Ion Research, Darmstadt. This proton radioactivity is now a widespread decay mode of very neutron-deficient nuclei. In 1984, H.J. Rose and G.A. Jones discovered cluster radioactivity in the decay of <sup>223</sup>Ra, which emits, with a probability of  $8.5 \cdot 10^{-10}$  relative to the  $\alpha$  particle emission, <sup>14</sup>C clusters and decays into <sup>209</sup>Pb. Heavier clusters are emitted from heavier nuclei with decreasing probabilities: for example, <sup>238</sup>Pu decays by emission of <sup>28</sup>Mg into <sup>210</sup>Pb and by emission of <sup>32</sup>Si into <sup>206</sup>Hg with probabilities of  $5.6 \cdot 10^{-17}$  and  $1.4 \cdot 10^{-16}$  relative to the  $\alpha$ -particle emission.

In 1903 Rutherford was the first scientist to observe the laws of radioactive decay and growth of a radioactive gas emanating from a thorium salt, radon. He used an electroscope, see Figure 1.6, for these radioactivity measurements. In the electroscope, the pointer G, a gold wire, deflected from the central metal bar when the upper part of the condenser was electrically charged relative to the housing. The condenser is discharged by ionizing radiation leading to a decrease in the deflection of the pointer G with a constant speed being a measure of the "saturation current," the *activity*. Figure 1.7 shows schematically the two experiments that Rutherford conducted with 55 seconds <sup>220</sup>Rn. In version (a), the gas inlet and outlet valves in the lower part of the housing are closed. The <sup>228</sup>Th source is placed inside the electroscope and is covered so that only the <sup>220</sup>Rn emanating from the thorium salt can diffuse into the free volume and discharge the condenser, giving rise to a constant activity; see the activity vs. time diagram to the right. At a given time indicated by the arrow, the gas inlet and outlet valves are opened and the lower part of the electroscope and

**Figure 1.6** Electroscope for the measurement of radioactivity. The gold wire G strives against the strut when the upper plate of the condenser is electrically charged relative to the housing. S is an insulator. For charging the condenser, a high voltage is applied to position A. Ionizing radiation is discharging the condenser, visible by a decrease in the deflection of the gold wire from the central metal bar with a constant velocity.





**Figure 1.7** Rutherford observed the growth (a) and decay (b) of a radioactive gas (55 seconds <sup>220</sup>Rn) emanating from a Th source (1.9 years <sup>228</sup>Th).

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causing the activity to fall to zero. Upon closing the valves, new <sup>220</sup>Rn grows from the <sup>228</sup>Th such that the activity discharging the condenser increases until the old saturation activity is reached. This can be repeated over and over again, showing each time the same characteristic time dependence. In version (b), the <sup>228</sup>Th source is placed in a box outside the electroscope and the activity is zero. On opening the valves and flushing <sup>220</sup>Rn into the electroscope with a carrier gas and closing the valves shortly thereafter, the <sup>220</sup>Rn decays with a characteristic time dependence. This can also be repeated over and over again. In the lower right part of Figure 1.7, the logarithm of the activity is plotted vs. time giving a linear decrease with time

$$\ln A(t) = \ln A_0 - \lambda t \tag{1.8}$$

where A(t) is the activity A vs. time  $t, A_0$  is the activity at time zero, and  $\lambda$  is the *decay constant*. In this way, the radioactive decay law

$$A(t) = A_0 \cdot \mathrm{e}^{-\lambda t} \tag{1.9}$$

was discovered. The unit of activity is  $1 \text{ decay s}^{-1} = 1 \text{ Becquerel} = 1 \text{ Bq}$ . The decay constant,  $\lambda$ , is characteristic for each nuclide and is related to the nuclear half-life,  $t_{1/2}$ , by

$$\lambda = \ln 2/t_{1/2} \tag{1.10}$$

The activity is equal to the number of nuclei present, *N*, multiplied by the decay constant  $\lambda$ , that is,  $A = \lambda N$ . Therefore, the number of radioactive nuclei present will also decrease exponentially as

$$N(t) = N_0 \mathrm{e}^{-\lambda t} \tag{1.11}$$

### **1.5 Some Physical Concepts Needed in Nuclear** Chemistry

Some important physical concepts need to be reviewed here because we will make use of them in later discussions.

#### 1.5.1 Fundamental Forces

All interactions in nature are the result of four fundamental forces, see Table 1.2. The weakest force is gravity. It is most significant when the interacting objects are massive, such as stars. The next stronger force is the weak interaction which acts in nuclear  $\beta$  decay. The electromagnetic force is next in strength, while the strong interaction is more than a hundred times stronger than the electromagnetic force. The ranges associated with the four forces are given in Table 1.2 along with their strengths relative to the strong force and with the respective force *carriers* or exchange particles. Among these, gravitons have not yet been observed but are believed to be responsible for gravity, which is not a part of the Standard Model of particle physics, see Section 1.5.6. In Chapter 6, we will see that Glashow, Salam,

Force	Range (m)	Relative strength	Force carrier
Gravitational	$\infty$	10 <sup>-38</sup>	Graviton?
Weak interaction	$10^{-18}$	10 <sup>-5</sup>	$W^{\pm}, Z^0$
Electromagnetic	$\infty$	$\alpha = 1/137$	Photon
Strong interaction	10 <sup>-15</sup>	1	Gluon

 Table 1.2
 Fundamental forces in nature.

and Weinberg introduced a unified theoretical treatment of electromagnetic and weak interactions, the electroweak interaction, in which the photon and the massive vector bosons  $W^{\pm}$  and  $Z^{0}$  emerge from one theory. We note in passing that the free neutron undergoes interactions with all four forces at the same time, see Chapter 8.

#### 1.5.2 Elements from Classical Mechanics

A force is a vector that describes the rate of change of a momentum with time

$$\mathbf{F} = \frac{\mathrm{d}\rho}{\mathrm{d}t} \tag{1.12}$$

For the motion of a particle, the orbital angular momentum of the particle, l, with mass m, relative to the center of mass, is

$$\mathbf{l} = \mathbf{r} \times \mathbf{p} \tag{1.13}$$

**l** is a vector of magnitude *mvr* for circular motion. For motion past a stationary point, the magnitude is *mvb* where *b* is the impact parameter. The relationship between a force **F** and the potential energy *V* is generally

$$\mathbf{F} = \frac{-\partial V}{\partial r} \tag{1.14}$$

Thus, for example, the Coulomb force,  $F_{\rm C}$ , for two charges  $Z_1 e$  and  $Z_2 e$  separated by the distance, r, is

$$F_{\rm C} = \frac{Z_1 Z_2 e^2}{r^2} \tag{1.15}$$

where, for convenience, we set  $e^2 = 1.43998$  MeV fm.

#### 1.5.3 Relativistic Mechanics

When a particle moves with a velocity approaching the speed of light, according to the special theory of relativity by A. Einstein, the mass of the particle changes with speed according to

$$m' = \gamma m_0 \tag{1.16}$$

where m' and  $m_0$  are the masses of the particle in motion and at rest and  $\gamma$  is the Lorentz factor

$$\gamma = (1 - \beta^2)^{-1/2} \tag{1.17}$$

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and

$$\beta = \left(1 - \frac{1}{\gamma^2}\right)^{1/2}$$

where  $\beta$  is v/c, the velocity of the particle relative to the speed of light. The total energy of a relativistic particle is

$$E = m'c^2 \tag{1.18}$$

this being the kinetic energy, T, plus the rest mass energy equivalent  $m_0c^2$ , where

$$T = (\gamma - 1)m_0 c^2 \tag{1.19}$$

For a particle at rest, the total energy is

$$E = m_0 c^2 \tag{1.20}$$

For a massless particle such as the photon,

$$E = pc \tag{1.21}$$

where p is the momentum of the photon. The momentum of a relativistic particle is

$$p = \gamma m \upsilon \tag{1.22}$$

These equations demonstrate why the units  $MeV/c^2$  for mass and MeV/c for momentum are necessary in nuclear calculations.

To give an example, we calculate the velocity, momentum, and total energy of an <sup>40</sup>Ar ion with a kinetic energy of 1 GeV/nucleon. The total kinetic energy is  $40 \times 1$  GeV/nucleon = 40 GeV = 40 000 MeV. The rest mass  $m_0c^2$  is approximately 40 atomic mass units (40 amu) or (40)(931.5) MeV, see Eq. (3.1), or 37 260 MeV. Thus,  $\gamma = T/m_0c^2 + 1 = 1 + 40 000/37 260 = 2.07$ . With Eq. (1.17), we obtain  $\beta = 0.88$ . So the velocity is 0.88c or  $(0.88)(3 \cdot 10^8 \text{ m s}^{-1}) = 2.6 \cdot 10^8 \text{ m s}^{-1}$ . We modify Eq. (1.22) to  $pc = mc/(1 - \beta)^{1/2}$  and obtain (40)(931.5)(0.88)(2.07) = 67.7 GeV, that is, p = 67.7 GeV/c. The total energy, Eq. (1.18), is (2.07)(40)(931.5) = 77.3 GeV.

The space-time coordinates x,y,z,t in a stationary laboratory system are, in the special theory of relativity, related to the space-time coordinates in a system moving along the *x* axis, x',y',z',t', by

$$x' = \gamma(x - \beta ct)$$
  

$$y' = y$$
  

$$z' = z$$
  

$$t' = \gamma[t - (\beta/c)x]$$
(1.23)

This transformation from the stationary to the moving frame is the Lorentz transformation. The inverse Lorentz transformation is obtained by reversing the sign of vgiving

$$x = \gamma(x' + \beta ct')$$
$$y = y'$$
$$z = z'$$

$$t = \gamma[t' + (\beta/c)x']$$
  

$$\Delta t = \gamma[\Delta t' + (\beta/c)\Delta x]$$
  

$$\Delta x = \Delta x'/\gamma$$
(1.24)

For  $\gamma > 1$ , time is slowed down for the scientist in the laboratory, and the distance in the *x* direction is contracted. An example for the relevance of these equations in nuclear chemistry is the decay of rapidly moving particles such as muons in cosmic rays. At rest, the muon has a lifetime of 2.2 µs. At relativistic energies such as in cosmic rays, the lifetime is orders of magnitude longer. Due to this time dilatation, muons can reach the surface of the Earth.

A rule of thumb for the decision of whether the classical expressions or the relativistic expressions are to be used is  $\gamma \ge 1.1$ .

#### 1.5.4 The de Broglie Wavelength

The well-known wave-particle duality says that there is no distinction between wave and particle descriptions of atomic matter; that is, associated with each particle, there is an equivalent description in which the particle is assigned a wavelength, the de Broglie wavelength,

$$\lambda = \frac{h}{p} \tag{1.25}$$

or in rationalized units

$$\lambda = \frac{\hbar}{p} \tag{1.26}$$

with  $\hbar = h/2\pi$ . The relativistic equivalent is

$$\lambda = \frac{\hbar c}{[E_{\rm k}(E_{\rm k} + 2m_0 c^2)]^{1/2}} \tag{1.27}$$

Figure 1.8 shows de Broglie wavelengths for a sample of particles (electron, pion, proton, and neutron, deuteron,  $\alpha$  particle) as a function of kinetic energy. They are largest for the lightest particles at lowest energies. The horizontal bar indicates the order of magnitude where  $\lambda$  becomes larger than the maximum impact parameter *R* for light-particle-induced reactions and from where the wavelength of the projectile influences the nuclear reaction *cross-section*, see Chapter 12.

One can also associate a wavelength to photons

$$\lambda = \frac{c}{\nu} = \frac{hc}{E_{\gamma}} \tag{1.28}$$

where v is the frequency associated with the wavelength  $\lambda$ . A practical form of Eq. (1.28) is

$$\lambda = \frac{1.2397 \cdot 10^{-10}}{E_{\gamma} (\text{MeV})} \text{ cm}$$
(1.29)

Treating photons as particles is useful if they are emitted or absorbed by a nucleus. Here, we have

$$E_{\gamma} = h\nu = pc \tag{1.30}$$