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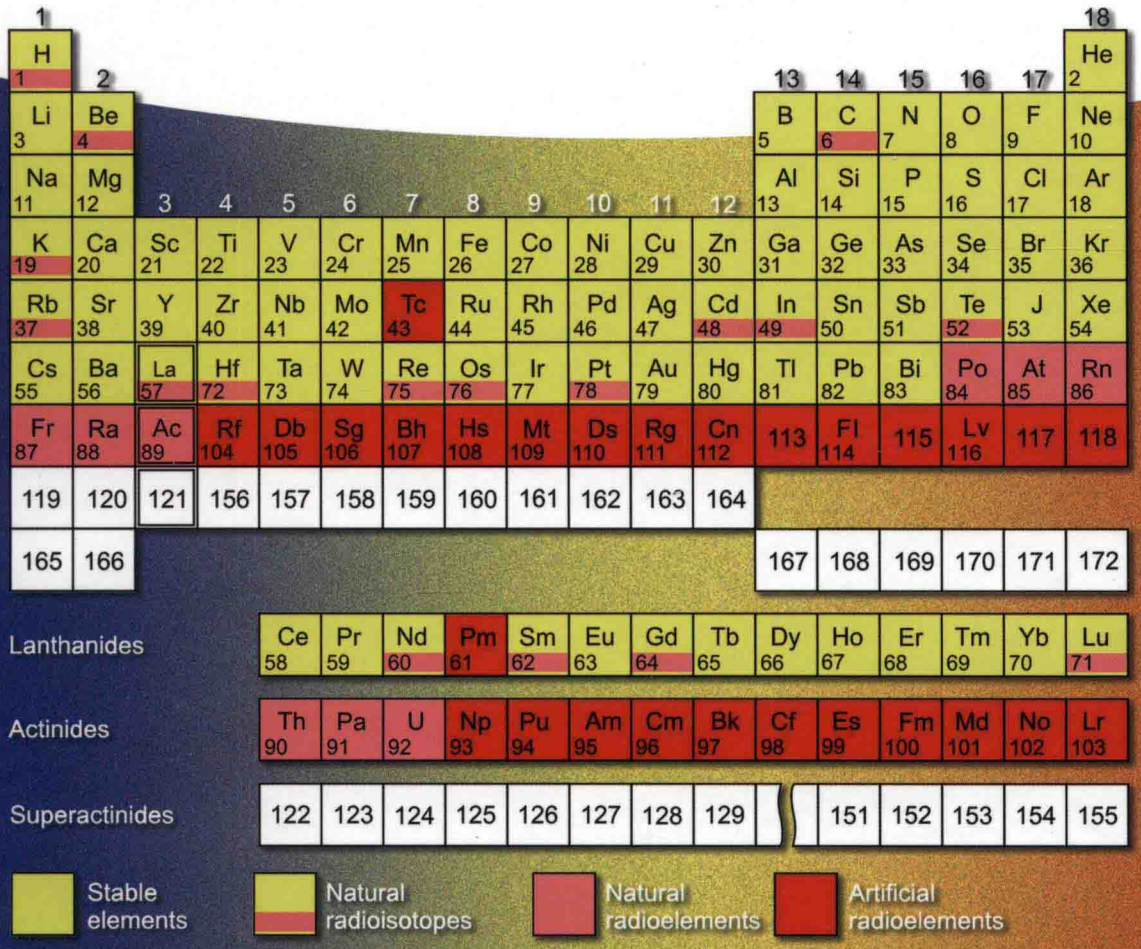
Jens-Volker Kratz, Karl Heinrich Lieser

# Nuclear and Radiochemistry

## Fundamentals and Applications

Volume 2



*Jens-Volker Kratz and Karl Heinrich Lieser*

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## Fundamentals and Applications

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# Nuclear and Radiochemistry

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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 (JVK) 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 remain largely unchanged; however, new developments and results have been incorporated, including the most recent references. These updates concern the physical properties of atomic nuclei, the nuclear force and nuclear structure, techniques in nuclear chemistry, nuclear reactions, statistical considerations in radioactivity measurements, 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.

It is my pleasure to thank Mrs. Petra Sach-Muth for help with the software "wiley-vch.dot" and Mr. Jürgen Hubrath for scanning and impoving a large number of new figures.

Mainz, April 2012 *Jens-Volker Kratz*

## Contents

# 12
# Nuclear Reactions

A nuclear reaction is a process in which a nucleus reacts with a proton or another nucleus, an elementary particle, or a photon to produce one or more other nuclei and possibly other particles. The nuclear states that can be investigated in the decay of radioactive isotopes as discussed in Section 11.9 are limited to a relatively low energy range. A much larger energy range is accessible through nuclear reactions. It is of great principle and practical importance to understand the various reaction mechanisms that occur in nuclear collisions.

The phenomenon of nuclear reactions was discovered by Rutherford in 1919 when he observed that, in the bombardment of nitrogen with the 6.69 MeV $\alpha$ particles of $^{214}$Po, scintillation of a zinc sulfide screen persisted even when enough material to absorb all the $\alpha$ particles was interposed between the nitrogen and the screen. Further experiments showed that the long-range particles causing the scintillation were protons and Rutherford's first reaction may be written

$$^{14}_{7}\mathrm{N} + {}^{4}_{2}\mathrm{He} \rightarrow {}^{17}_{6}\mathrm{O} + {}^{1}_{1}\mathrm{H}$$

with the shorthand notation $^{14}$N($\alpha$, p)$^{17}$O where, as indicated here, atomic numbers are commonly omitted. Most nuclear reactions are studied by inducing a collision between two nuclei where one of the nuclei is at rest, the target nucleus, and the other nucleus, the projectile, is in motion. In a nuclear reaction, there is conservation of the total number of nucleons $A$, charge, energy, momentum, angular momentum, statistics, and parity. Nuclear reactions, like chemical reactions, are always accompanied by a release or absorption of energy, and this is expressed by adding the term to the right hand side of the equation. Thus, a more complete statement of Rutherford's first reaction is

$$^{14}\mathrm{N} + {}^{4}\mathrm{He} \rightarrow {}^{17}\mathrm{O} + {}^{1}\mathrm{H} + Q$$

The quantity $Q$ is the energy of the reaction or simply the reaction $Q$ value. Other than in chemistry, the convention is to assign positive $Q$ values to energy-releasing reactions (exoergic reactions) and negative $Q$ values to energy absorption (endoergic reactions). Another important difference between chemical reactions and nuclear reactions must be pointed out. In chemical reactions, macroscopic amounts of material undergo transmutation and heats of reaction are given per mole of the reactants. In nuclear reactions, single processes are considered and

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the $Q$ values are therefore given per nucleus transformed. For example, the reaction $^{14}N(\alpha, p)^{17}O$ has a $Q$ value of $-1.190$ MeV or $-1.190\,66 \cdot 10^{-6}$ ergs or $-4.56 \cdot 10^{-14}$ calories per $^{14}N$ atom transformed. To convert 1 mole of $^{14}N$ to $^{17}O$ would require an energy of $6.022 \cdot 10^{23} \cdot 4.56 \cdot 10^{-14}$ cal $= 2.75 \cdot 10^{10}$ cal. This is five orders of magnitude larger than the largest values observed for heats of chemical reactions. The $Q$ value is calculated with tabulated mass excesses, see Eq. (3.5), as $Q = \Delta(^{14}N) + \Delta(^{4}He) - (\Delta(^{17}O) + \Delta(^{1}H)) = 2.863 + 2.425 - (-0.811 + 7.289\,03)$ MeV $= -1.190$ MeV. Does that mean that the reaction can actually be produced by $\alpha$ particles whose kinetic energies are just over 1.19 MeV? The answer is no for two reasons. Firstly, in the collision conservation of momentum requires that at least 4/18 of the kinetic energy of the $\alpha$ particle must be retained by the products as kinetic energy. Thus only 14/18 of the $\alpha$ particle's kinetic energy is available for the reaction. The threshold energy of $\alpha$ particles for the $^{14}N(\alpha, p)^{17}O$ reaction is $18/14 \times 1.19$ MeV $= 1.53$ MeV. The second reason why the $\alpha$ particles must have higher energies than the $Q$ value is the Coulomb repulsion between the $\alpha$ particle and the $^{14}N$ nucleus, Eq. (1.15),

$$V_C = 1.44 \frac{Z_1 Z_2}{R_1 + R_2} \text{MeV}$$

Setting $R = 1.5A^{1/3}$ fm, we get a value of about 3.4 MeV for the Coulomb barrier between the $\alpha$ particle and the $^{14}N$ nucleus. Thus, classically, an $\alpha$ particle must have $18/14 \times 3.4 = 4.4$ MeV kinetic energy for the $(\alpha, p)$ reaction to occur even though the energetic threshold for the reaction is only 1.53 MeV. In the quantum-mechanical treatment of the problem, there exists a finite probability for barrier penetration, but the probability is extremely small as we saw in the discussion of $\alpha$ decay in Section 6.2.

## 12.1 Collision Kinematics

Generalizing the above discussion, and using the notation $A_1 + A_2 \rightarrow A_3 + A_4$, we can describe the situation before the collision in the laboratory system by

$$E_{lab} = \frac{m}{2} A_1 v_\infty^2 \tag{12.1}$$

$$P_{lab} = m A_1 v_\infty \tag{12.2}$$

where $m$ is the nucleon mass, $A_1$ is the mass number of the projectile, and $v_\infty$ is the velocity at infinite distance with

$$v_\infty = \sqrt{\frac{2E_{lab}}{mA_1}} = 1.389 \sqrt{\frac{E_{lab}}{A_1}} \text{ cm ns}^{-1} \tag{12.3}$$

In the center-of-mass system, Figure 12.1, before the collision, both ions move toward the center of mass, S, with

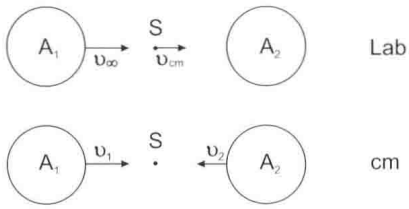


**Figure 12.1** Laboratory and center-of-mass system before the collision.

$$\upsilon_1 = \frac{A_2}{A_1 + A_2}\upsilon_\infty$$
$$\upsilon_2 = \frac{A_1}{A_1 + A_2}\upsilon_\infty \tag{12.4}$$

The kinetic energy and momentum in the center-of-mass system are

$$E_{cm} = \frac{m}{2}\left(A_1\upsilon_1^2 + A_2\upsilon_2^2\right) = \frac{\mu}{2}\upsilon_\infty^2 \tag{12.5}$$
$$P_{cm} = m\left(A_1\upsilon_1 + A_2\upsilon_2\right) = \mu\upsilon_\infty$$

where $\mu$, the reduced mass, equals

$$m\frac{A_1A_2}{A_1 + A_2} = mA_{12}$$

Only the center-of-mass energy $E_{cm}$ and the relative momentum $P_{cm}$ are available for the reaction. The remainder is translational energy (recoil energy) of the total system

$$E_{trans} = \frac{A_1}{A_1 + A_2}E_{lab} \tag{12.6}$$

and

$$E_{cm} = E_{lab} - E_{trans} \tag{12.7}$$

The translational velocity of the center of mass relative to the laboratory system is

$$\upsilon_{cm} = \frac{A_1}{A_1 + A_2}\upsilon_\infty = \upsilon_2 = \sqrt{\frac{2E_{trans}}{m(A_1 + A_2)}} = 1.389\sqrt{\frac{E_{trans}}{A_1 + A_2}}\ \text{cm ns}^{-1} \tag{12.8}$$

Another useful energy variable is the laboratory energy per nucleon

$$\varepsilon = \frac{E_{lab}}{A_1} = \frac{E_{cm}}{A_{12}} = \frac{m}{2}\upsilon_\infty^2 \tag{12.9}$$

After the collision, the situation is as depicted in Figure 12.2. In the center-of-mass system the product $A_3$ is emitted with velocity $\upsilon_3$ under the center-of-mass angle $\Theta$. It is common to look explicitly at the projectile-like product $A_3$; for the target-like fragment, the corresponding variables result from trivial transformation.

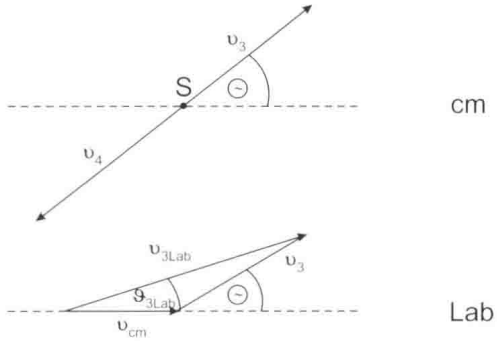


**Figure 12.2** Center-of-mass and laboratory system after the collision.

Figure 12.2 also shows the situation in the laboratory system: vector addition of $\upsilon_3$ and $\upsilon_{cm}$ yields $\upsilon_{3lab}$ under laboratory angle $\vartheta_{3lab}$. The connection between $\vartheta_{3lab}$ and $\Theta_{cm}$ is

$$\tan\vartheta_{3lab} = \frac{\upsilon_3 \sin\Theta}{\upsilon_3 \cos\Theta + \upsilon_{cm}} = \frac{\sin\Theta}{\cos\Theta + \dfrac{\upsilon_{cm}}{\upsilon_3}} \tag{12.10}$$

and vice versa

$$\Theta = \vartheta_{3lab} + \arcsin\frac{\upsilon_{cm}\sin\vartheta_{3lab}}{\upsilon_3}$$

It is customary to introduce the kinematic variable $\gamma_3 = \upsilon_{cm}/\upsilon_3$. For elastic scattering $\upsilon_3 = \upsilon_1$ and $\gamma_3 = A_1/A_2$. For two colliding ions of equal mass, $\gamma_3 = 1$ and $\vartheta_{3lab} = \Theta/2$. In the general case of a two-body reaction, the quantity $\gamma_3$ is given by

$$\gamma_3 = \left(\frac{A_1 A_3}{A_2 A_4}\frac{E_{cm}}{E_{cm}+Q}\right)^{1/2} \tag{12.11}$$

where the $Q$ value can also be determined as the difference of the kinetic energies after and before the reaction as $Q = E_f^{\infty} - E_i^{\infty}$ equal to the total kinetic energy loss $TKEL = TKE_i - TKE_f = -Q$. The $\infty$ sign indicates that the particles need to be separated far enough from each other so that the interaction potential is no longer acting. Because of the conservation of the total energy, the $Q$ value at the same time corresponds to the difference of the rest masses of the initial and final state (Eq. (3.5)).

## 12.2 Coulomb Trajectories

Figure 12.3 shows the classical trajectories of two colliding ions in the center-of-mass system. The distance of closest approach between the two centers of gravity, $D$, and the scattering angle $\Theta$ are related by