



普通高等教育“十二五”规划教材

Fundamentals of
Nuclear Chemical Engineering and
Nuclear Fuel Engineering

核化工与核燃料工程基础

刘峙嵘 陈中胜 王云 周利民 主编

中国石化出版社

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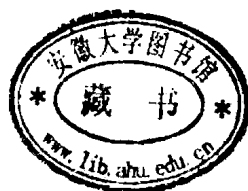


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内 容 提 要

本书是根据核化工与核燃料工程专业英语教学大纲编写的,内容分为两大部分:第一部分为核化学与放射化学基础知识,其中包括7个单元;第二部分为核化学工程与工艺,其中包括6个单元。13个单元的内容涉及核物理、核化学、放射化学、核燃料循环、核化工等。

本书供工科大学核化工与核燃料工程专业或相关专业三、四年级学生使用,也可供相关领域的科技人员使用。

图书在版编目(CIP)数据

核化工与核燃料工程基础:英文 / 刘峙嵘等主编.
—北京:中国石化出版社,2012.3
(普通高等教育“十二五”规划教材)
ISBN 978-7-5114-1370-3

I. ①核… II. ①刘… III. ①核化学-化学工程-高等学校-教材-英文②核燃料-高等学校-教材-英文
IV. ①TL

中国版本图书馆 CIP 数据核字(2012)第 030979 号

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中国石化出版社出版发行

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北京宏伟双华印刷有限公司印刷

全国各地新华书店经销

*

787×1092 毫米 16 开本 8.75 印张 209 千字

2012 年 3 月第 1 版 2012 年 3 月第 1 次印刷

定价:20.00 元

前 言

核能的开发和利用是 20 世纪自然科学和工程技术方面的重大成就。面临越来越严峻的能源问题，人们重新将眼光转向核能，21 世纪核科学与技术将继续发展。核工业是一个庞大的系统，从铀矿开采、铀的提取、纯化、转化、同位素富集、燃料原件制造、乏燃料后处理、高放废物处置，都离不开核化工与核燃料，它是核科学和技术不可或缺的组成部分。

目前，美、英等发达国家在该领域处于领先地位，很多新的研究成果和新技术的介绍都以英文发表，每个从事核科学技术的专业人员都需要阅读大量的英文资料。为了满足越来越多的核化工与核燃料专业人员学习专业英语的需要，编写了本书。

本书是根据核化工与核燃料工程专业英语教学大纲编写的。本书内容分为两大部分：第一部分为核化学与放射化学基础知识，其中包括 7 个单元；第二部分为核化学工程与工艺，其中包括 6 个单元。13 个单元的内容均摘选自核科学与技术相关的原版英文教科书、科技报告和专著。

本书第一部分由陈中胜编写；第二部分由王云编写。全书由刘峙嵘、周利民统稿，刘义保对全书稿进行了审核。

本书在编制和使用过程中得到了东华理工大学化学生物与材料科学学院化工系全体老师的支持和帮助，核化工与核燃料工程专业 2007 级、2008 级同学提出了不少宝贵意见和建议。同时本书的出版得到江西省高水平学科——核科学与技术和江西省重点学科——应用化学的资助。最后，本书得到了中国石化出版社的大力支持，使本书得以顺利出版，在此一并向他们表示深深的感谢。

由于本书涉及的专业面广，加之我们的水平有限，书中不足之处在所难免，恳请读者批评指正。

编者

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Part I

Fundamentals of Nuclear Chemistry & Radiochemistry

Unit 1 → Origin of Nuclear Science

◆ 1.1 Radioactive elements

In 1895, W. Roentgen discovered that when cathode rays (i. e. electrons) struck the wall of an evacuated glass tube, it caused the wall material to emit visible light (fluoresce), while at the same time a very penetrating radiation was produced. The name X-ray was given to this radiation. Learning about this, H. Becquerel, who had been interested in the fluorescent spectra of minerals, immediately decided to investigate the possibility that the fluorescence observed in some salts when exposed to sunlight also caused emission of X-rays. Crystals of potassium uranyl sulfate were placed on top of photographic plates, which had been wrapped in black paper, and the assembly was exposed to the sunlight. After development of some of the photographic plates, Becquerel concluded (erroneously) from the presence of black spots under the crystals that fluorescence in the crystals led to the emission of X-rays, which penetrated the wrapping paper. However, Becquerel soon found that the radiation causing the blackening was not “a transformation of solar energy” because it was found to occur even with assemblies that had not been exposed to light; the uranyl salt obviously produced radiation spontaneously. This radiation, which was first called uranium rays (or Becquerel rays) but later termed radioactive radiation (or simply radioactivity), was similar to X-rays in that it ionized air, as observed through the discharge of electroscopes.

Marie Curie subsequently showed that all uranium and thorium compounds produced ionizing radiation independent of the chemical composition of the salts. This was a convincing evidence that the radiation was a property of the element uranium or thorium. Moreover, she observed that some uranium minerals such as pitchblende produced more ionizing radiation than pure uranium compounds. She wrote, “this phenomenon leads to the assumption that these minerals contain elements which are more active than uranium”. She and her husband Pierre Curie began a careful purification of pitchblende, measuring the amount of radiation in the solution and in the precipitate after each precipitation separation step. These first radiochemical investigations were highly successful-“while carrying out these operations, more active products are obtained. Finally, we obtained a substance whose activity was 400 times larger than that of uranium. We therefore believe that the substance that we have isolated from pitchblende is a hitherto unknown metal. If the existence of this metal can be affirmed, we suggest the name polonium.” It was in the publication reporting the discovery of

polonium in 1898 that the word “radioactive” was used for the first time. It may be noted that the same element was simultaneously and independently discovered by W. Marckwald who called it “radiotellurium”.

In the same year, the Curies, together with G. Bemont, isolated another radioactive substance for which they suggested the name radium. In order to prove that polonium and radium were in fact two new elements, large amounts of pitchblende were processed, and in 1902, M. Curie announced that she had been able to isolate about 0.1 g of pure radium chloride from more than one ton of pitchblende waste. The determination of the atomic weight of radium and the measurement of its emission spectrum provided the final proof that a new element had been isolated.

◆ 1.2 Radioactive decay

While investigating the radiochemical properties of uranium, W. Crookes and Becquerel made an important discovery. Precipitating a carbonate salt from a solution containing uranyl ions, they discovered that while the uranium remained in the supernatant liquid in the form of the soluble uranyl carbonate complex, the radioactivity originally associated with the uranium was now present in the precipitate, which contained no uranium. Moreover, the radioactivity of the precipitate slowly decreased with time, whereas the supernatant liquid showed a growth of radioactivity during the same period (Fig. 1. 1). We know now that this measurement of radioactivity is concerned with only beta- and gamma-radiations, and not with the alpha-radiation which is emitted directly by uranium.

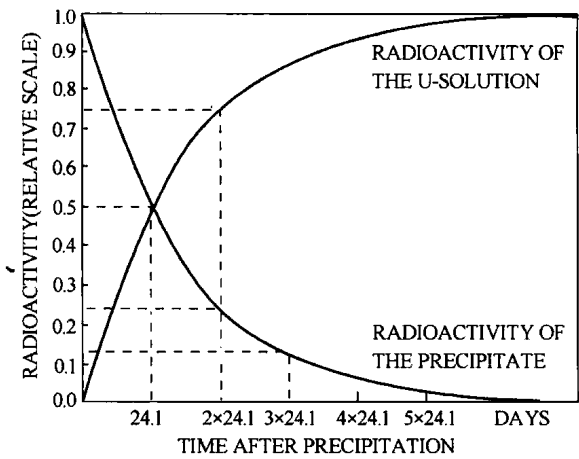


Fig. 1. 1 Measured change in radioactivity from carbonate precipitate and supernatant uranium solution, i. e. the separation of daughter element UX (Th) from parent radioelement uranium

Similar results were obtained by E. Rutherford and F. Soddy when investigating the radioactivity of thorium. Later Rutherford and F. E. Dora found that radioactive gases(emanation) could be separated from salts of uranium and thorium. After separation of the gas from the salt, the radioactivity of the gas decreased with time, while new radioactivity grew in the salt in a manner similar to that shown in Fig. 1. 1. The rate of increase with time of the radioactivity in the salt was found to be completely independent of chemical processes, temperature, etc. Rutherford and Soddy concluded from these observations that radioactivity was due to changes within the atoms themselves. They proposed

that, when radioactive decay occurred, the atoms of the original elements (e. g. of U or of Th) were transformed into atoms of new elements.

The radioactive elements were called radioelements. Lacking in names for these radioelements, letters such as X, Y, Z, A, B, etc. , were added to the symbol for the primary (i. e. parent) element. Thus, UX was produced from the radioactive decay of uranium, ThX from that of thorium, etc. These new radioelements (UX, ThX, etc.) had chemical properties that were different from the original elements, and could be separated from them through chemical processes such as precipitation, volatilization, electrolytic deposition, etc. The radioactive daughter elements decayed further to form other elements, symbolized as UY, ThA, etc. A typical decay chain could be written as: Ra \rightarrow Rn \rightarrow RaA \rightarrow RaB \rightarrow , etc. (shown in Fig. 1. 2).

	81Tl	82Pb	83Bi	84Po	85At	86Rn	87Fr	88Ra	89Ac	90Th	91Pa	92U	93Np
232										Th α 1.79 $\cdot 10^{10}$			
228								MsTh β 5.7y	MsTh β 6.13h	RdTh α 1.90h			
224								ThX α 3.64d					
220						Th α 54.5s				β^- decay $A, Z \rightarrow A, Z+1$			
216				ThA $\alpha\beta$ 0.75s	At α 3.0 $\cdot 10^{-4}$					$A-4, Z-2$ α decay			
212		ThB β 10.9h	ThC $\alpha\beta$ 10.5m	ThC' α 3.0 $\cdot 10^{-7}$									
208	ThC'' β 3.1m	ThD stable											
237													^{237}Np α 2.20 $\cdot 10^6$ y
233												^{233}Pa β 27.4d	^{233}U α 1.82 $\cdot 10^5$ y
229													
225								^{225}Ra β 14.5d	^{225}Ac α 10.0d				
221							^{221}Fr α 48m						
217					^{217}At α 0.018s								
213			^{213}Bi $\alpha\beta$ 47m	^{213}Po α 4.2 $\cdot 10^{-4}$									
209	^{209}Tl β 2.2m	^{209}Pb β 3.2h	stable										
238												U α	
234										UX β	UX β UZ β	U α	
230										Io α			
226								Ra α 1622y					
222						Rn α 3.025d							
218						RaA $\alpha\beta$ 2.05m	^{218}At α 2s						
214		RaB β 28.8m	RaC $\alpha\beta$ 19.7m	RaC' α 1.5 $\cdot 10^{-4}$ s									
210	RaC'' β 132m	RaD β 22y	RaE $\alpha\beta$ 5.0d	RaF α 138.4d									
206	RaE β 4.29m	RaG stable											
235												AcU α 7.13 $\cdot 10^6$ y	
231										UV β 25.6h	Pa α 3.43 $\cdot 10^4$ y		
227								Ac $\alpha\beta$ 22.0y	RdAc α 18.6d				
223						AcK $\alpha\beta$ 21s	AcX α 11.1d						
219						^{219}At $\alpha\beta$ 0.1m	Rn α 2.92s						
215			^{215}Bi β 1m	AcA $\alpha\beta$ 1.83 $\cdot 10^{-5}$ s	^{215}At α 10 $^{-6}$ s								
211		AcB β 35.7m	AcC $\alpha\beta$ 2.14m	AcC' α 0.52s									
207	AcC'' β 4.79m	AcD.D. stable											

Fig. 1. 2 The three naturally occurring radioactive decay series and the man-made neptunium series. Although ^{239}Pu (which is the parent to the actinium series) and ^{244}Pu (which is the parent to the thorium series) have been discovered in nature, the decay series shown here begin with the most abundant long-lived nuclides

A careful study of the radiation emitted from these radioactive elements demonstrated that it consisted of three components which were given the designation alpha (α), beta (β), and gamma (γ). Alpha-radiation was shown to be identical to helium ions, whereas beta-radiation was identical to electrons. Gamma-radiation had the same electromagnetic nature as X-rays but was of higher energy. The rate of radioactive decay per unit weight was found to be fixed for any specific radioelement, no matter what its chemical or physical state was, though this rate differed greatly for different radioelements. The decay rate could be expressed in terms of a half-life, which is the time it takes for the radioactivity of a radioelement to decay to half of its original value. Half-lives for the different radioelements were found to vary from fractions of a second to millions of years; e. g. that of ThA is one tenth of a second, UX 24. 1 days (Fig. 1. 1), and of uranium, millions of years.

◆ 1. 3 Discovery of isotopes

By 1910, approximately 40 different chemical species had been identified through their chemical nature, the properties of their radiation, and their characteristic half-lives. The study of the genetic relationship in the decay of the radioactive species showed that the radioelements could be divided into three distinct series. Two of these originated in uranium and the third in thorium. B. Boltwood found that all the three series ended in the same element - lead.

A major difficulty obvious to scientists at that time involved the fact that while it was known from the Periodic Table that there was only space for 11 elements between lead and uranium, approximately 40 radioelements were known in the decay series from uranium to lead. To add to the confusion was the fact that it was found that in many cases it was not possible to separate some of the radioelements from each other by normal chemical means. For example, the radioelement RaD was found to be chemically identical to lead. In a similar manner, spectrographic investigations of the radioelement ionium showed exactly the same spectral lines that had been found previously to be due to the element thorium.

In 1913, K. Fajans and F. Soddy independently provided the explanation for these seemingly contradictory conditions. They stated that by the radioactive α -decay, a new element is produced at two places to the left of the mother element in the periodic system and by β -decay a new element is produced at one place to the right of the mother element (Fig. 1. 2). The radioelements that fall at the same place in the periodic system are chemically identical. Soddy proposed the name isotopes to account for different radioactive species which have the same chemical identity.

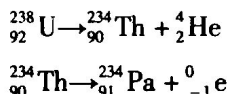
Research by J. J. Thomson soon provided conclusive support for the existence of isotopes. If a beam of positively charged gaseous ions is allowed to pass through electric or magnetic fields, the ions follow hyperbolic paths which are dependent on the masses and charges of the gaseous ions. When these ion beams strike photographic plates, a darkening result is proportional to the number of ions which hit the plate. By using this technique with neon gas, Thomson found that neon consists of two types of atoms with different atomic masses. The mass numbers for these two isotopes

were 20 and 22. Moreover, from the degree of darkening of the photographic plate, Thomson calculated that neon consisted of about 90% of atoms with mass number 20, and 10% of atoms with mass number 22.

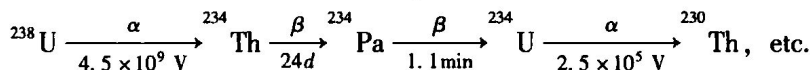
Thus a chemical element may consist of several kinds of atoms with different masses but with the same chemical properties. The 40 radioelements were, in truth, not 40 different elements but were isotopes of the 11 different chemical elements from lead to uranium.

To specify a particular isotope of an element, the atomic number (i. e. order, number, or place in the Periodic Table of elements) is written as a subscript to the left of the chemical symbol and the mass number (i. e. the integer value nearest to the mass of the neutral atom, measured in atomic weight units) as a superscript to the left. Thus the isotope of uranium with mass number 238 is written as ${}_{92}^{238}\text{U}$. Similarly, the isotope of protactinium with mass number 234 is designated as ${}_{91}^{234}\text{Pa}$. For an alpha-particle, we use either the Greek letter α or ${}^4_2\text{He}$. Similarly, the beta-particle is designated either by the Greek letter β or by the symbol ${}^0_{-1}\text{e}$.

In radioactive decay, both mass number and atomic number are conserved. Thus in the decay chain of ${}_{92}^{238}\text{U}$, the first two steps are written as:



Frequently, in such a chain, the half-life ($t_{1/2}$) for the radioactive decay is shown either above or below the arrow. A shorter notation is commonly used:



where the half-lives are given in years (y) and days (d). The three naturally occurring radioactive decay series, which are known as the thorium series, the uranium series, and the actinium series, are shown in Fig. 1. 2. A fourth series, which originates in the synthetic element neptunium, is also shown. This series is not found naturally on earth since all of the radioactive species have decayed away long ago. Both the present symbolism of the isotope as well as the historical (i. e. 'radioelement') symbolism are given in Fig. 1. 2. Note that the rule of Fajans and Soddy is followed in each series so as to cause a decrease in atomic number by two units and mass number by four, whereas β -decay produces no change in mass number but an increase in atomic number by one unit. Moreover, we see a pattern occurring frequently in these series where an α -decay step is followed by two β -decay steps. All known isotopes of elements ${}_{92}\text{U}$ to ${}_{81}\text{Tl}$ are given in associated text.

1. 4 Atomic models

Neither radioactive decay nor the discovery of isotopes provided information on the internal structure of atoms. Such information was obtained from scattering experiments in which a substance, such as a thin metal foil, was irradiated with a beam of α -particles and the intensity (measured by counting scintillations from the scattered particles hitting a fluorescent screen) of the particles scat-

tered at different angles measured. It was assumed that the deflection of the particles was caused by collisions with the atoms of the irradiated material. About one in 8000 of the α -particles was strongly deflected through angles greater than 90° . Consideration of these rare events led Rutherford in 1911 to the conclusion that the entire positive charge of an atom must be concentrated in a very small volume whose diameter is about 10^{-14} meter. This small part of the atom was called the nucleus by him. The atomic electrons had much smaller mass and were assumed to surround the nucleus. The total atom with the external electrons had a radius of approximately 10^{-10} meter in contrast to the much smaller radius calculated for the nucleus.

It was soon shown that the positive charge of the atomic nucleus was identical to the atomic number assigned to an element in the periodic system of Mendeleev. The conclusion, then, was that in a neutral atom, the small and positively charged nucleus was surrounded by electrons whose number was equal to the total positive charge of the nucleus. In 1913, N. Bohr, by using quantum mechanical concepts, proposed such a model of the atom which remains the basis of the modern atomic theory.

Words and Expressions

- radioactive *a.* 放射性的
 cathode *n.* 阴极射线
 electron *n.* 电子
 evacuate *v.* 撤离, 疏散, 排泄, 剥夺
 fluoresce *v.* 发荧光
 penetrating *a.* 有穿透力的, 贯穿的; 渗透的
 spectrum *n.* 光谱
 potassium *n.* 钾, 元素符号为 K
 uranyl *n.* 铀基; 铀酰
 sulfate *n.* 硫酸盐
 photographic *a.* 摄影用的
 wrap *v.* 包, 裹
 erroneously *ad.* 错误地
 spontaneously *ad.* 自发地
 uranium *n.* 铀, 元素符号为 U
 radioactivity *n.* 放射性
 ionize *v.* 电离
 discharge *n.* 放电
 electroscopes *n.* 验电器
 thorium *n.* 钍, 元素符号为 Th
 property *n.* 特性, 属性
 pitchblende *n.* 沥青铀矿
 purification *n.* 提纯, 精炼
 solution *n.* 溶液, 溶剂
 precipitate *n.* 沉淀物
 radiochemical *a.* 放射化学的
 hitherto *ad.* 迄今; 至此
 affirm *v.* 证实; 确认
 polonium *n.* 钋, 元素符号为 Po
 simultaneously *ad.* 同时地
 radiotellurium *n.* 射碲
 radium *n.* 镭, 元素符号为 Ra
 chloride *n.* 氯化物
 decay *n.* 衰变
 carbonate *n.* 碳酸盐
 supernatant *a.* 浮在表面的
 complex *n.* 复合物
 emanation *n.* 射气, 放出物
 atom *n.* 原子
 volatilization *n.* 挥发; 蒸发
 electrolytic *a.* 电解的
 deposition *n.* 处置; 沉积
 neptunium *n.* 镎, 元素符号为 Np
 actinium *n.* 锕, 元素符号为 Ac
 nuclide *n.* 核素
 helium *n.* 氦, 元素符号为 He
 electromagnetic *a.* 电磁的
 half-life *n.* 半衰期
 isotope *n.* 同位素
 approximately *ad.* 大约
 distinct *a.* 不同的, 有区别的
 lead *n.* 铅, 元素符号为 Pb
 Periodic Table *n.* 元素周期表
 ionium *n.* 钍的放射性同位元素
 contradictory *a.* 矛盾的, 对立的
 gaseous *a.* 气体的, 气态的
 hyperbolic *a.* 双曲线的
 neon *n.* 氖, 元素符号为 Ne
 atomic number *n.* 原子序数
 subscript *n.* 下标
 superscript *n.* 上标
 protactinium *n.* 镤, 元素符号为 Pa
 synthetic *a.* 合成的, 人造的
 scattering *n.* 散射
 scintillation *n.* 闪烁; 闪光
 deflect *v.* (使)偏斜, (使)偏转
 diameter *n.* 直径
 nucleus *n.* (原子)核
 radius *n.* 半径
 quantum *n.* 量子

Wilhelm Conrad Röntgen

Born : 27 – Mar – 1845

Birthplace : Lennep, Germany

Died : 10 – Feb – 1923

Location of death : Munich, Germany

Occupation : Physicist

Nationality : Germany

Executive summary : Discovered X-Rays



As a German physicist born in Lennep on the 27th of March 1845, he received his early education in Holland, and then went to study at Zürich, where he took his doctor's degree in 1869. He then became assistant to Kundt at Würzburg and afterwards at Strassburg, becoming privat-docent at the latter university in 1874. Next year he was appointed to be professor of mathematics and physics at the Agricultural Academy of Hohenheim, and in 1876 he returned to Strassburg as extraordinary professor. In 1879 he was chosen ordinary professor of physics and director of the Physical Institute at Giessen, whence in 1885 he removed in the same capacity to Würzburg. It was at the latter place that he made the discovery for which his name was chiefly known, the Röntgen rays.

In 1895, while experimenting with a highly exhausted vacuum tube on the conduction of electricity through gases, he noticed that a paper screen covered with barium platinocyanide, which happened to be lying near, became fluorescent under the action of some radiation emitted from the tube, which at the time was enclosed in a box of black cardboard. Further investigation showed that this radiation had the power of passing through various substances which were opaque to ordinary light, and also of affecting a photographic plate. Its behavior being strange in several respects, particularly in regard to reflection and refraction, doubt arose in his mind whether it was to be looked upon as light or not, and he was led to put forward the hypothesis that it was due to longitudinal vibrations in the ether, not to transverse ones like ordinary light; but in view of the uncertainty existing as to its nature, he called it X-rays.

For this discovery, he received the Rumford medal of the Royal Society in 1896, jointly with Philipp Lenard, who had already shown, as also had Heinrich Hertz, that a portion of the cathode rays could pass through a thin film of a metal such as aluminum. Röntgen also conducted researches in various other branches of physics, including elasticity, capillarity, the conduction of heat in crystals, the absorption of heat-rays by different gases, piezoelectricity, the electromagnetic rotation of polarized light, etc.

Unit 2 Radioactivity and Nuclear Reactions

Following the discovery of radioactivity by Henri Becquerel in 1896, two young scientists, Frederic Soddy and Ernest Rutherford then at McGill University in Canada, set about to investigate the recently discovered phenomena. In 1901, the twenty-four-year-old chemist Soddy and Rutherford were attempting to identify a gas which was being released from samples of radioactive thorium oxide. They believed that this gas they called it an “emanation” was related to the radioactivity of the thorium sample. In order to investigate the nature of this gas, Soddy passed it over a series of powerful chemical reagents, heated white-hot. No reactions took place. Following this discovery, Rutherford and Soddy published nine joint papers between 1902 and 1903 in a period of extremely productive research. In 1902 they described their theory of radioactivity as a spontaneous disintegration of the radioactive element by the expulsion of particles with the result that new elements were formed. This was the ultimate step in the ancient alchemists’ dream of transmutation.

2.1 Simple Radioactive Decay: Half-life and Decay Constant

Radioactive decay is a random process. As such, one cannot state with certainty when an unstable nuclide will decay. The probability that an atom will decay during the time dt is given by kdt where k is the constant of proportionality known as the decay constant. In a system where there are $N(0)$ atoms present initially, the number of atoms decaying in time dt is given by $-dN = kNdt$. In the limit of very small time intervals, this can be expressed as

$$dN/dt = -kN$$

Integration with respect to time gives the number of atoms present at any time t , i. e.

$$N(t) = N(0)e^{-kt}$$

The half-life, τ , is used to denote the time at which the number of atoms has decreased to half of the initial value, i. e. $1/2 = e^{-k\tau}$. Hence the half-life is related to the decay constant through the relation:

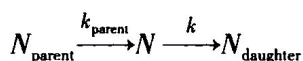
$$\tau = \ln 2/k \approx 0.693/k$$

2.2 Activity

The number of decays per unit time interval, i. e. the activity A , is defined by

$$A = -dN/dt = kN$$

It should be noted in this definition that it is assumed that N is decreasing due to decay. In general the rate equation contains a term for decay (removal) to the daughter and in-growth (production) from the parent, i. e. N_{parent}



for which the rate equation becomes

$$\frac{dN}{dt} = -kN + k_{\text{parent}}N_{\text{parent}}$$

A situation could arise in which $kN = k_{\text{parent}}N_{\text{parent}}$ and thus N is constant, i. e. $dN/dt = 0$. Clearly the activity is not zero. In the definition of A above only decay is considered. In the general case where decay and growth occur, A is given by $A = kN$. Hence A is the number of disintegrations per second even though N may be constant. The unit of activity is the Becquerel, i. e. $1 \text{ Bq} = 1$ disintegration per second.

A technical problem arises in the evaluation of the activity in the case where the half-life is less than 1 s. The activity defined above gives the instantaneous disintegration rate. If the half-life is ≤ 1 s, a significant amount of the material has decayed in the first second. The above definition of the activity will then overestimate the emitted radiation. The difficulty can easily be overcome by defining the activity per integral second, i. e.

$$A_{\text{is}} = \int_0^1 kN dt = N(0)(1 - e^{-k})$$

where $k = \ln 2 / \tau$ (s) and $N(0)$ is the number of atoms at time 0. For the calculation of the specific activity, denoted spA , $N(0)$ is the number of atoms in 1 g i. e. $N(0) = N_a / A'$. Hence

$$\text{spA} = \frac{N_a(1 - e^{-k})}{A'}$$

$$\text{or spA (Bq/g)} = 6.022 \times 10^{23} \cdot \frac{1 - e^{-\frac{\ln 2}{\tau(s)}}}{A'}$$

◆ 2.3 Average (Mean) Lifetime

The half-life of a nuclide is a statistical property and is a valid concept only because of the very large number of atoms involved. Any individual atom of a radionuclide may be transformed at any time, from zero to infinity. For some calculations, it is convenient to use the average life of a radionuclide. The average life is defined as the sum of the lifetimes of the individual atoms divided by the total number of atoms present originally. During a time interval from t to $t + dt$, the total number of transformations is $kNdt$. Each atom that decayed during this time interval had existed for a total lifetime t . The sum of the lifetimes of all atoms that were transformed during the time interval dt , having survived from $t = 0$ is $tkNdt$. The average lifetime is then given by

$$t = \frac{1}{N(0)} \int_0^{\infty} tkNdt$$

It is then straightforward to show that the relationship between the average or mean lifetime and the half-life is given by $t = 1.44 \tau$.

◆ 2.4 Branching Ratios and Number of Decay Modes

Many nuclides have more than one decay mode. Consider a nuclide in which there are two decay modes. The probability that an atom will decay by process 1 in time dt is $k_1 dt$. Similarly, the probability that it will decay by process 2 in time dt is $k_2 dt$. Hence the equation governing the radio-

active decay can be written as

$$\frac{dN}{dt} = -(k_1 + k_2)N$$

The total decay constant for the decay of the parent nuclide is the sum of the partial decay constants i. e. $k = k_1 + k_2$. Hence, the branching ratios for modes 1 and 2 are defined as

$$BR_1 = \frac{k_1}{k}, \text{ and } BR_2 = \frac{k_2}{k}$$

In general, the branching ratio (BR) for a particular decay mode is defined as the ratio of the number of atoms decaying by that decay mode to the number decaying in total, i. e.

$$BR_i = \frac{k_i}{(k_1 + k_2 + \dots + k_i + \dots)} = \frac{k_i}{k}$$

Alternatively, given the total decay constant, the “partial” decay constant is given by

$$k_i = BR_i \cdot k$$

Number of Decay Modes

There are a number of ways in which a nuclide can decay. Usually the number of decay modes is one or two. There are nuclides, however, which have many decay modes. In Table 2.1, the seven decay modes of the nuclide ^{11}Li are listed.

Table 2.1 Decay modes, branching ratios, and daughters of ^{11}Li

Decay mode	Branching ratio	Daughters
β^-	8.07×10^{-2}	^{11}Be
β^-, d	1.30×10^{-4}	^9Li
β^-, α	1.00×10^{-2}	^7He
β^-, n	8.49×10^{-1}	^{10}Be
$\beta^-, 3n$	1.90×10^{-2}	^8Be
β^-, t	1.40×10^{-4}	^8Li
$\beta^-, 2n$	4.10×10^{-4}	^9Be

2.5 Decay Chains

It is very often the case that the daughter product of a nuclear decay is also radioactive. In such cases one speaks of radioactive decay “chains”. As an example, consider the decay chain $N_1 \rightarrow N_2 \rightarrow N_3 \rightarrow \dots$ in which the starting or “parent” nuclide N_1 decays to the “daughter” N_2 , this daughter in turn is radioactive and decays to N_3 . More generally, each nuclide in the decay chain N_i can “branch”, with branching ratio kN_i/N_j , to more than one daughter. In addition, there may be an external source term S_i for the production of N_i (apart from the decay of the parent).

The situation for successive radioactive decay is shown schematically in Fig. 2.1. This general process of radioactive decay was first investigated systematically by Bateman.

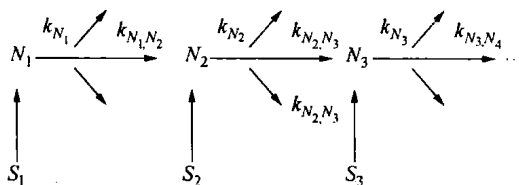


Fig. 2.1 Successive radioactive decay with branching and source terms

The differential equations governing the above processes can be written as:

$$\begin{aligned}\frac{dN_1}{dt} &= S_1 - k_{N_1} \cdot N_1 \\ \frac{dN_2}{dt} &= S_2 + k_{N_1 \cdot N_2} \cdot N_1 - k_{N_2} \cdot N_2 \\ \frac{dN_i}{dt} &= S_i + k_{N_{i-1} \cdot N_i} \cdot N_{i-1} - k_{N_i} \cdot N_i \\ \frac{dN_n}{dt} &= S_n + k_{N_{n-1} \cdot N_n} \cdot N_{n-1} - k_{N_n} \cdot N_n\end{aligned}$$

where N_n is the number of atoms of species n present at time t , k_n is the decay constant (total removal constant) for species n ($k = \ln 2 / \tau$), $k_{n,n+1}$ is the partial decay constant (partial removal constant) and is related to the branching ratio $BR_{n,n+1}$ through the relation $k_{n,n+1} = BR_{n,n+1} \cdot k_n$. The solution to this system of equations is

$$N_n(t) = \sum_{i=1}^{i=n} \left[\left(\prod_{j=1}^{j=n-1} k_{j,j+1} \right) \times \sum_{j=i}^{j=n} \left(\frac{N_i(0)e^{-k_j t}}{\prod_{\substack{p=i \\ p \neq j}}^n (k_p - k_j)} + \frac{S_i(1 - e^{-k_j t})}{k_j \prod_{\substack{p=i \\ p \neq j}}^n (k_p - k_j)} \right) \right] \quad (2.1)$$

for the particular case (of most interest) one is interested in the decay chain starting from a single parent nuclide with no source term S . In this case the above relation reduces to:

$$N_n(t) = \left(\prod_{j=1}^{j=n-1} k_{j,j+1} \right) \times \sum_{j=i}^{j=n} \frac{N_i(0)e^{-k_j t}}{\prod_{\substack{p=i \\ p \neq j}}^n (k_p - k_j)} \quad (2.2)$$

It is of interest to construct the first few terms, i. e.

$$\begin{aligned}N_1 &= N_1(0)e^{-k_1 t} \\ N_2 &= k_{1,2} \left\{ \frac{N_1(0)e^{-k_1 t}}{k_2 - k_1} + \frac{N_1(0)e^{-k_2 t}}{k_1 - k_2} \right\} \\ N_3 &= k_{1,2} k_{2,3} \left\{ \frac{N_1(0)e^{-k_1 t}}{(k_2 - k_1)(k_3 - k_1)} + \frac{N_1(0)e^{-k_2 t}}{(k_1 - k_2)(k_3 - k_2)} + \frac{N_1(0)e^{-k_3 t}}{(k_1 - k_3)(k_2 - k_3)} \right\}\end{aligned} \quad (2.3)$$

These relations allow one to update the numbers of atoms from time $t=0$ to time t . It is also of interest to calculate the numbers at various times in the range $0, t$ (for example, for plotting purposes). This can be done by specifying the total time t over which the calculation is to be made, and the number of time-steps L to reach t . The time interval for each calculation is then $\Delta t = t/L$. For