

# 6 EQUATIONS OF RADIOACTIVE DECAY AND GROWTH

The mathematical expressions presented in this chapter are generally applicable to all those processes in which the transition of the *parent nucleus* to a *daughter nucleus*, i.e. the process of radioactive decay, is governed by statistical chance. This chance of decay is equivalent to the degree of instability of the parent nucleus. Each radioactive nuclide has its specific degree of instability which, as we will see, is going to be expressed by the half-life assigned to this nuclide.

The radioactivity of a sample is more complicated if it consists of two or more components, such as: (i) in the case of a mixture of independent activities, (ii) if one specific type of nuclide shows two modes of decay, so-called branching decay, and (iii) if we are dealing with a nuclear decay series in which also the daughter nuclides are radioactive. All these phenomena will be discussed separately.

## 6.1 LAW OF RADIOACTIVE DECAY

The fundamental law of radioactive decay is based on the fact that the decay, i.e. the transition of a parent nucleus to a daughter nucleus is a purely statistical process. The disintegration (decay) probability is a fundamental property of an atomic nucleus and remains equal in time. Mathematically this law is expressed as:

$$dN = \lambda N \cdot dt \quad (6.1)$$

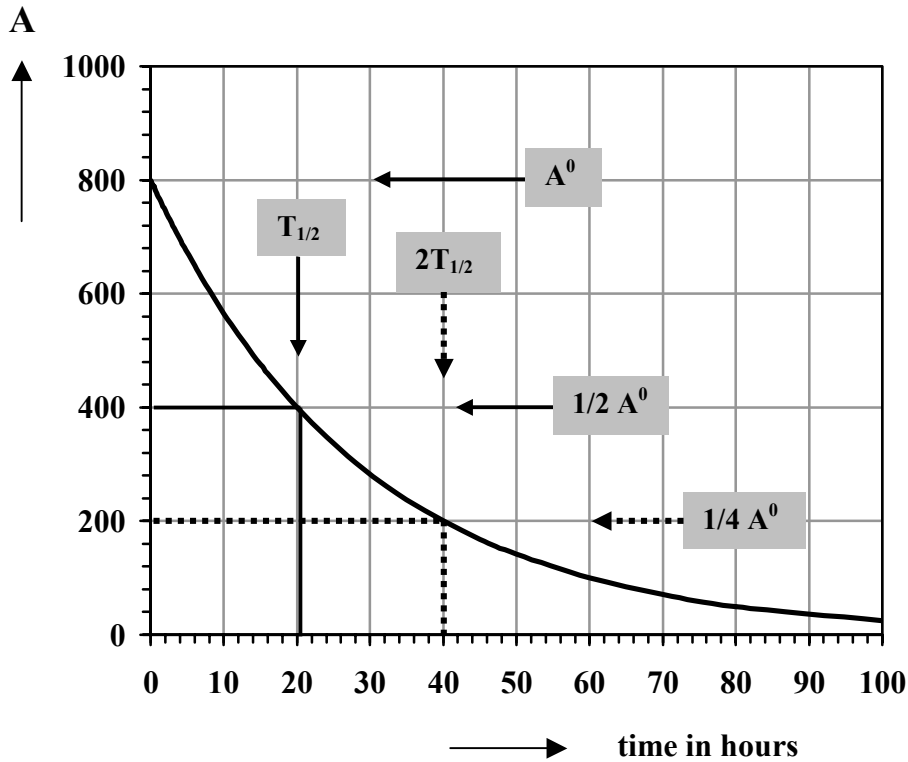
and

$$\lambda = \frac{(-dN/dt)}{N} \quad (6.2)$$

where  $N$  is the number of radioactive nuclei,  $-dN/dt$  the decrease (negative) of this number per unit of time and  $\lambda$  is thus the probability of decay per nucleus per unit of time. This *decay constant*  $\lambda$  is specific for each decay mode of each nuclide.

The *radioactivity* or *decay rate* is defined as the number of disintegrations per unit of time:

$$A = -dN / dt = \lambda N \quad (6.3)$$



**Fig.6.1** The rate of radioactive decay. After each subsequent half-life of 20 hours the number of radioactive nuclei and the original radioactivity of 800 units are divided into half.

By integration of this relation and applying the boundary conditions that at in the beginning  $t = 0$  and  $N = N^0$  we obtain:

$$\ln(N/N_0) = -\lambda t \quad (6.4)$$

and subsequently the equation of *exponential decay*:

$$N = N^0 e^{-\lambda t} \quad (6.5)$$

or using Eq.6.3:

$$A = A^0 e^{-\lambda t} \quad (6.6)$$

The time during which  $A^0$  decreased to  $A$  (= the age of the material) is:

$$T = (1/\lambda) \ln(A/A_0) \quad (6.7)$$

The relations of Eqs.6.5 and 6.6 indicate the rate at which the original number of radioactive nuclei ( $N^0$ ) and the original radioactivity ( $A^0$ ) decrease in time (Fig.6.1).

## 6.2 HALF-LIFE AND MEAN LIFE

It is a common practice to use the *half-life* ( $T_{1/2}$ ) instead of the decay constant ( $\lambda$ ) for indicating the degree of instability or the decay rate of a radioactive nuclide. This is defined as the period of time in which half of the radioactivity has disappeared (half of the nuclei have disintegrated, Fig.6.1):

$$T_{1/2} = (-1/\lambda)\ln(1/2) \quad (6.8)$$

from which:

$$\lambda = \frac{\ln 2}{T_{1/2}} = \frac{0.693}{T_{1/2}} \quad (6.9)$$

The *mean life* of a nuclide is the sum of the life times of a certain number of nuclei (before they have all disintegrated) divided by the number of nuclei. During the time interval  $dt$  a number of  $dN$  nuclei disintegrate. These "lived" during a period  $t$ , which amounts to a total life time for  $dN$  nuclei of (cf. Eq.6.3):

$$t \cdot dN = t \cdot \lambda N \cdot dt$$

Integrating over all nuclei ( $N$ ) gives the mean life (time):

$$\begin{aligned} \tau &= \frac{1}{N^0} \int_0^{\infty} t \cdot \lambda N \cdot dt = \lambda \int_0^{\infty} t \cdot e^{-\lambda t} dt = \lambda \left\{ -\frac{t}{\lambda} e^{-\lambda t} \Big|_0^{\infty} + \frac{1}{\lambda} \int_0^{\infty} e^{-\lambda t} dt \right\} \\ &= \lambda \left\{ 0 + \frac{1}{\lambda} \left( -\frac{1}{\lambda} e^{-\lambda t} \Big|_0^{\infty} \right) \right\} = \frac{1}{\lambda} \end{aligned} \quad (6.10)$$

As an example, the mean life of a  $^{14}\text{C}$  nucleus with  $T_{1/2} = 5730$  a is 8267 years. Then  $\lambda = 1/8267$ , which means that a sample activity decreases by 1‰ in about 8 years; a  $^3\text{H}$  sample activity ( $T_{1/2} = 12.43$  a) decreases by 5.6% per year.

## 6.3 ACTIVITY, SPECIFIC ACTIVITY AND RADIONUCLIDE CONCENTRATION

The *activity* of a certain sample is the number of a radioactive disintegrations per sec for the sample as a whole. The *specific activity*, on the other hand, is defined as the number of disintegrations *per unit weight or volume* of sample (see spec.act. of  $^{14}\text{C}$  and  $^3\text{H}$ , Chapter 8). The unit of radioactivity is the *Becquerel* (Bq), which is defined as a decay rate of one disintegration per second (dps) or the now obsolete *Curie* (Ci) which was defined as a decay rate of  $3.7 \times 10^{10}$  dps.

As an example of the relation between the specific activity of a sample and the actual *concentration* of the radioactive nuclide, we will calculate the specific tritium ( $^3\text{H}$ ) activity of water containing one  $^3\text{H}$  atom per  $10^{18}$  hydrogen atoms (equivalent to 1 TU = Tritium Unit (Chapter 8):

$$A_{\text{spec}} = \lambda N \text{ (per litre)}$$

where:

$$\lambda = (\ln 2)/T_{1/2} = (\ln 2)/12.43 \text{ a} \quad (1 \text{ year} = 3.16 \times 10^7 \text{ s})$$

$$N = 2 \times 10^{-18} \times (G/M) \times A \quad (G/M = \text{number of moles})$$

$$A = \text{Avogadro's number} = 6.02 \times 10^{23}/\text{mol}$$

$$M = \text{molecular weight} = 18.0$$

$$\text{pCi} = \text{pico-Curie} = 10^{-12} \text{ Curie} = 3.7 \times 10^{-2} \text{ dps} = 0.037 \text{ Bq}$$

The numerical result for water with 1 TU of tritium is:

$$A_{\text{spec}} = 0.118 \text{ Bq/L} = 3.19 \text{ pCi/L} \quad (6.11)$$

As another example we can calculate the  $^{14}\text{C}$  concentration in carbon, which has a specific  $^{14}\text{C}$  activity of 13.56 dpm per gram of carbon (in the year AD 1950) (see  $^{14}\text{C}$  standard activity, Chapter 8):

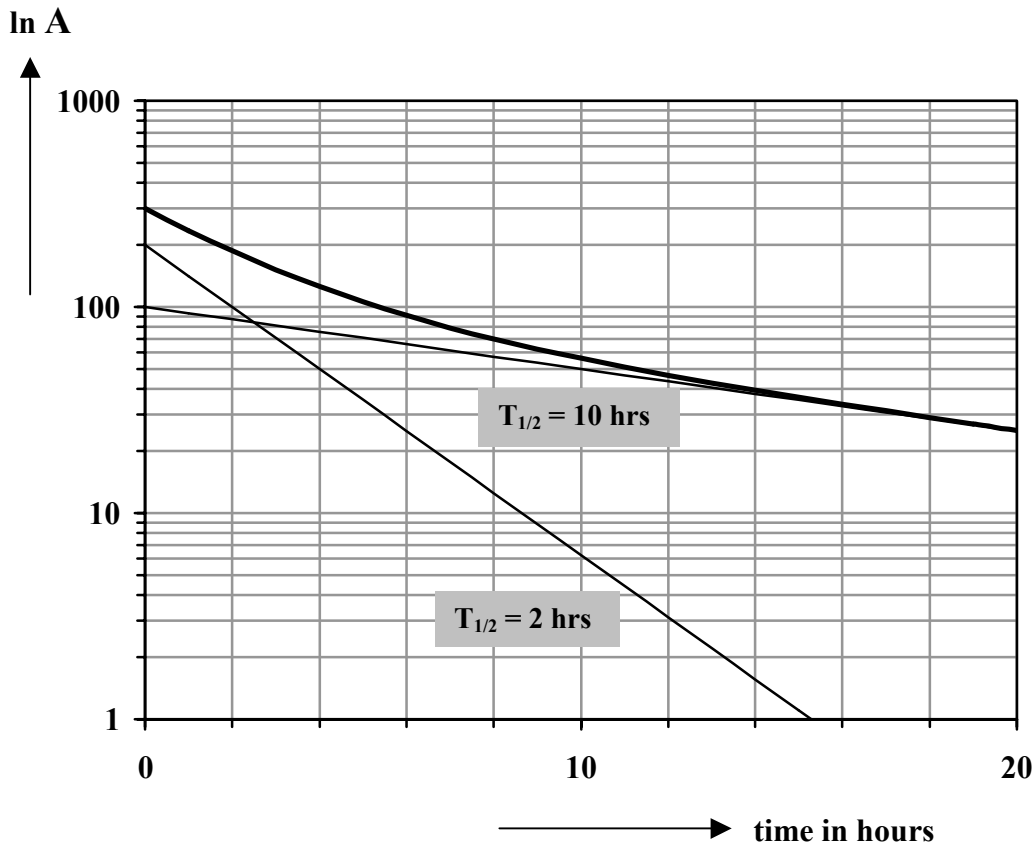
$$\frac{^{14}\text{C}}{\text{C}} = \frac{13.56 \times 5730 \times (3.16 \times 10^7) \times 12}{60 \times \ln 2 \times (6.02 \times 10^{23})} = 1.2 \times 10^{-12} \quad (6.12)$$

## 6.4 MIXTURE OF INDEPENDENT RADIOACTIVITIES

Fig.6.2 shows a semi-logarithmic decay curve of a mixture of two activities that are completely independent, a *composite decay curve*. If the half-life values are sufficiently apart, it is seen to be possible to unravel the two separate decay curves, starting at the right-hand side of the curve, where the one activity has already disappeared. Subtracting the straight semi-logarithmic curve from the composite curve gives the straight line for the shorter-lived nuclide.

## 6.5 BRANCHING DECAY

Also in nature radioactive nuclides exist that show two different modes of decay. One example is presented by  $^{40}\text{K}$  that can decay with the emission of a  $\beta^-$  or a  $\beta^+$  particle (Fig.5.1).



**Fig.6.2** Semi-logarithmic plot of a composite decay curve for a mixture of two independent radioactive compounds with half-lives of 2 and 10 hours. The longer-lived activity can be subtracted from the sum curve (heavy line) to produce the semi-logarithmic straight decay curve for the shorter-lived nuclide.

Each decay mode has its specific decay constant or half-life. The total decay is simply the sum of both single chances and is thus given by:

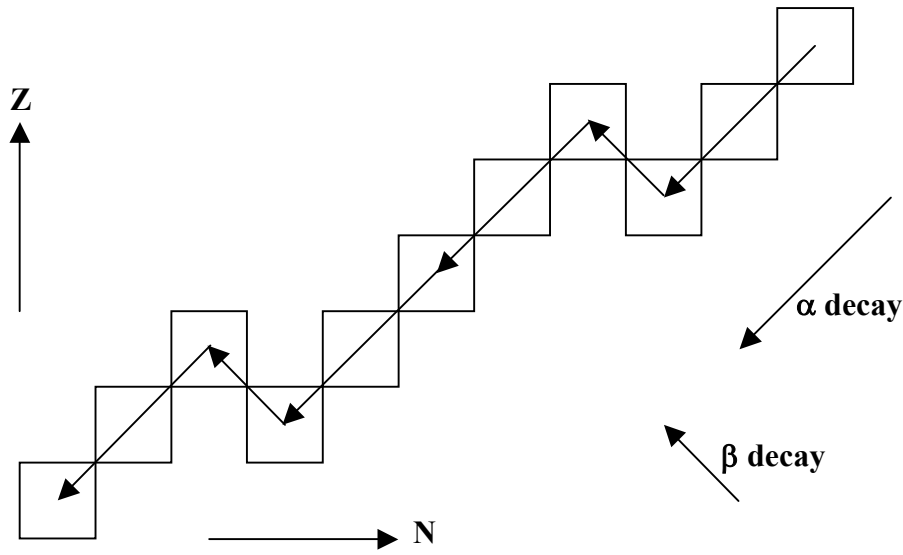
$$\lambda_{\text{total}} = \lambda_1 + \lambda_2 \quad (6.13)$$

and the total half-life consequently by:

$$(1/T_{1/2})_{\text{total}} = (1/T_{1/2})_1 + (1/T_{1/2})_2 \quad (6.14)$$

## 6.6 RADIOACTIVE DECAY SERIES

If a radioactive nuclide is situated in the Chart of Nuclides far from the stability line (for the light elements at  $Z=N$ ), the daughter nucleus after radioactive decay may be radioactive as well. In nature this occurs with the heavy nuclides in the uranium and thorium decay series (Chapter 12). Here the original decay of  $^{238}\text{U}$  or  $^{232}\text{Th}$  is followed by a series of radioactive decay products. Fig.6.3 shows schematically how the elements of such decay series are related.



**Fig.6.3** Schematic representation of a hypothetical multiple decay curve, analogous to the decay curves of the U and Th decay series (Sect.12.14-12.16).

In this context we can restrict ourselves to one element from the multiple decay chain: the relation between a parent and a daughter activity. The relations in a second and higher degree have been treated elsewhere (see textbooks by Friedlander et al. and by Faure).

The parent nucleus decays according to the equations of radioactive decay which we have treated in this section:

$$A_1 = -\frac{dN_1}{dt} = \lambda_1 N_1 \quad (6.15)$$

and

$$N_1 = N_1^0 e^{-\lambda_1 t} \quad \text{and} \quad A_1 = A_1^0 e^{-\lambda_1 t} \quad (6.16)$$

The amount of daughter nuclei is determined by two processes: (i) radioactive decay and (ii) radioactive growth by decay of the parent nuclei, respectively:

$$\frac{dN_2}{dt} = -\lambda_2 N_2 + \lambda_1 N_1 \quad (6.17)$$

The solution of this differential equation is:

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t} \quad (6.18)$$

while at zero activity at zero time ( $N_2^0 = 0$ ):

$$A_2 = \lambda_2 N_2 = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \quad (6.19)$$

The last term represents the decay of the amount of daughter nuclide that was present at the time  $t = 0$ . It is obvious that the ratio between  $\lambda_1$  and  $\lambda_2$  is the dominating factor that determines the course of the daughter activity in time. We shall now briefly mention the 3 different cases for this ratio.

### 6.6.1 SECULAR EQUILIBRIUM

This type of relation between parent and daughter activity occurs when the half-life of the parent nuclide is infinitely larger than that of the daughter nuclide. Examples are the relations between the long-living isotopes of uranium and thorium,  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$ , and their decay products (see Chapter 12):

$$\lambda_1 \ll \lambda_2$$

Eq.6.16 properly describes the parent activity in time, whereas Eq.6.19 turns into:

$$A_2 = A_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \quad (6.20)$$

or for  $\lambda_1 = 0$ ,

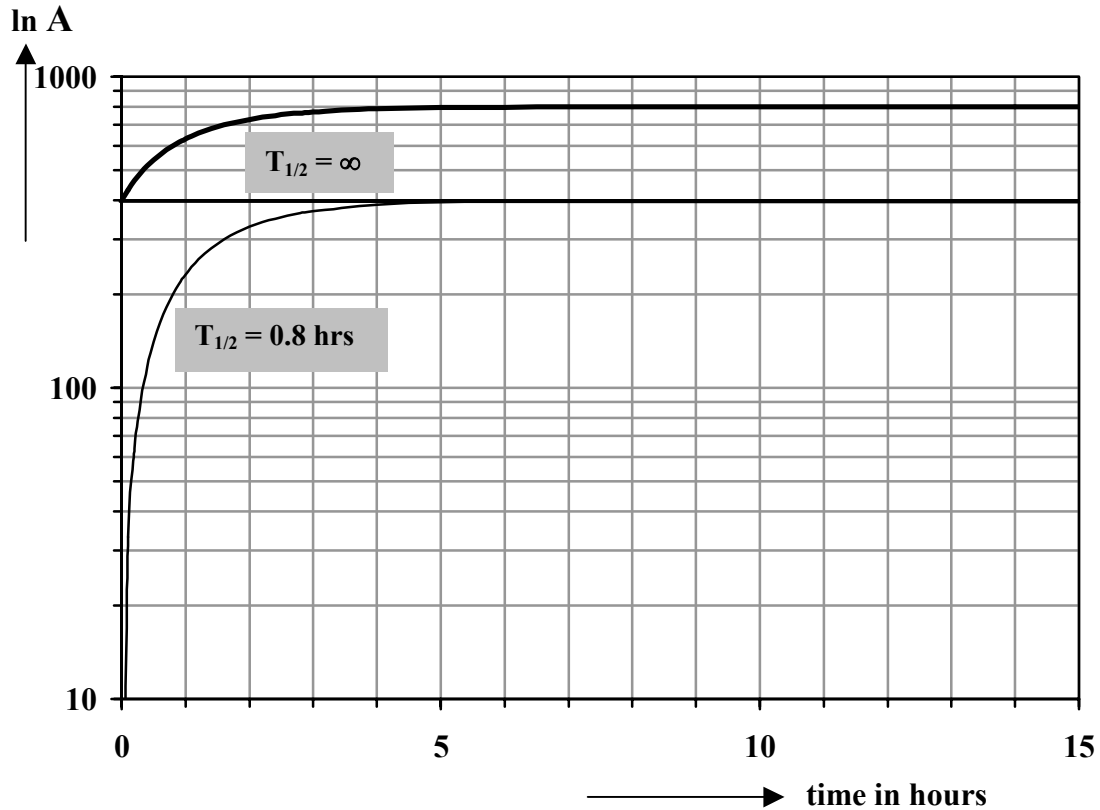
$$A_2 = A_1^0 (1 - e^{-\lambda_2 t}) \quad (6.21)$$

describing the growth of the daughter activity in time if we take  $A_2 = 0$  in the beginning. Fig.6.4 shows the courses of both activities. Finally (at  $t \rightarrow \infty$  with  $\lambda_2 t \rightarrow \infty$  in Eq.6.20) the daughter activity reaches a value of:

$$A_2 = A_1^0 e^{-\lambda_1 t} = A_1 \quad (6.22)$$

in other words, parent and daughter activity become equal.

The phenomenon that in a sample containing a long-living nuclide a short-living daughter activity may grow is applied, for instance in cases where the radioactivity measurement of the daughter nuclide is easier than that of the parent activity. Such case is the activity determination of  $^{32}\text{Si}$  with a half-life of about 140 years, which decays through a low-energetic  $\beta^-$  decay to  $^{32}\text{P}$  with a half-life of 14.3 days and a high-energetic  $\beta^-$  decay. Eq.6.21 shows that after separating chemically a pure  $^{32}\text{Si}$  activity  $^{32}\text{P}$  is growing into this sample at a



**Fig.6.4** Relation between the radioactivities of a parent (straight line) and a daughter nuclide when the parent nuclide decays infinitely slow as compared to the daughter nuclide (half-lives of  $\infty$  and 0.8 hours, respectively), i.e. the case of secular equilibrium. The upper line represents the sum of parent and daughter activity.

rate such that after one daughter half-life the daughter activity has increased to 50 % of its maximum value of  $A_1$ :

$$A_2 = 1/2 A_1 \text{ after one } T_{1/2}$$

$$A_2 = 3/4 A_1 \text{ after two } T_{1/2}$$

$$A_3 = 7/8 A_1 \text{ after three } T_{1/2}, \text{ and so on.}$$

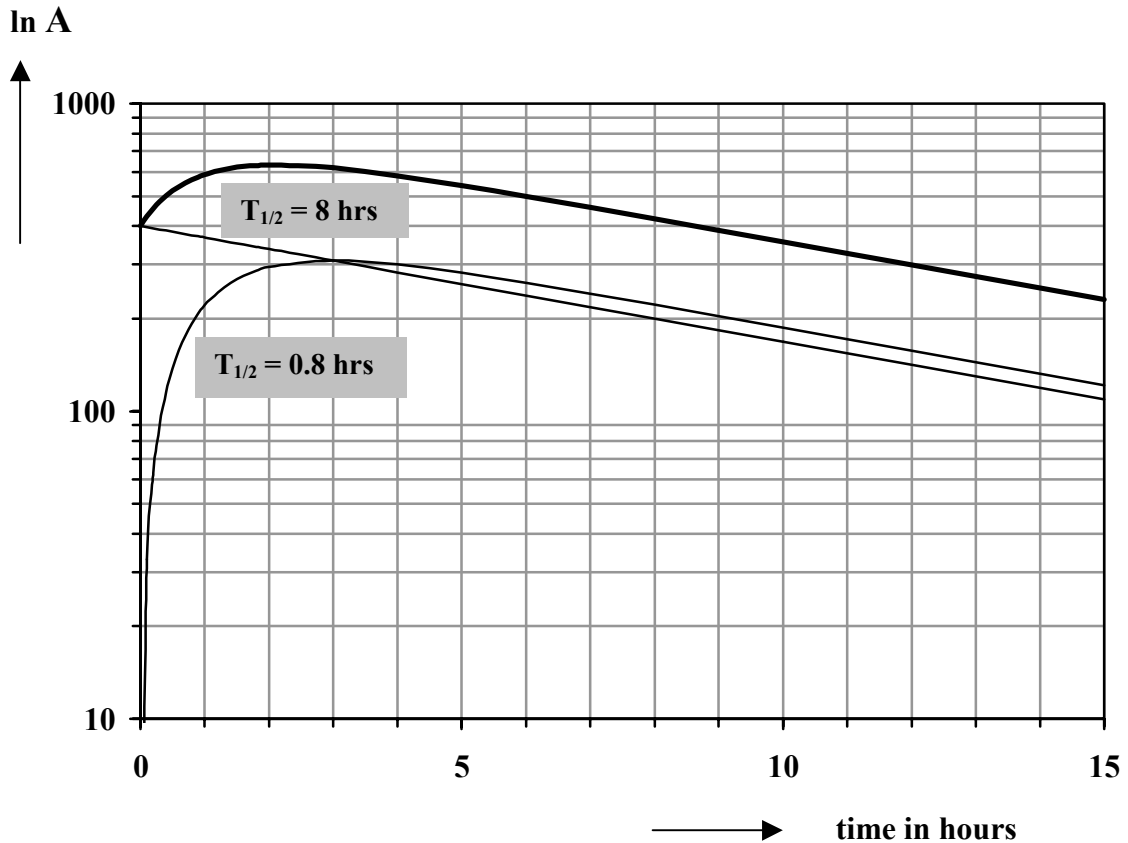
This consideration gives the time required to gain sufficient daughter activity for a proper measurement after chemical separation of the parent and daughter nuclides.

### 6.6.2 TRANSIENT EQUILIBRIUM

In this case, the half-life of the parent nuclide is still larger than that of the daughter nuclide but not infinitely:

$$\lambda_1 < \lambda_2$$





**Fig.6.5** Relation between the radioactivities of a parent and a daughter nuclide, when the half-life of the parent is larger (but not infinitely) than that of the daughter (half-lives of 8 and 0.8 hours, respectively): the case of transient equilibrium. The heavy line shows the sum activity of parent and daughter.

Growth of the daughter after zero activity at zero time now occurs according to Eq.6.19. A stationary state is reached after sufficient time in which the daughter activity is larger than the parent activity, as is to be expected. Fig.6.5 shows the course of both activities:

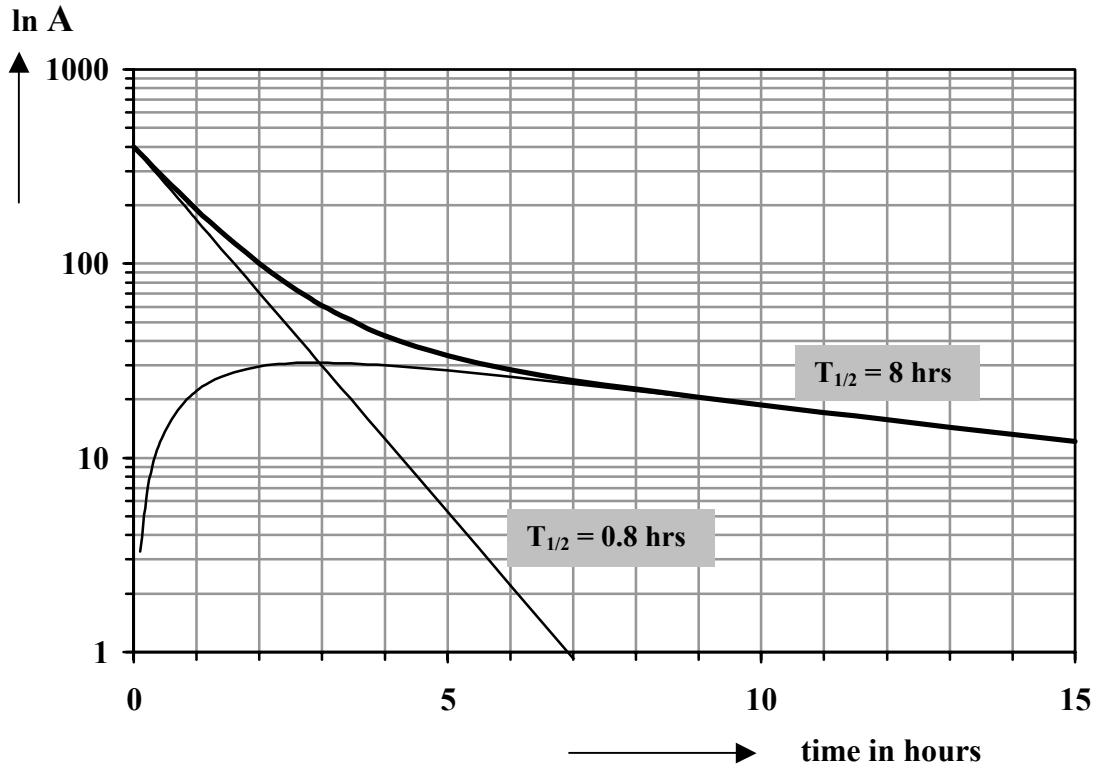
$$A_2 = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1^0 e^{-\lambda_1 t} = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1 \quad (6.23)$$

### 6.6.3 NO-EQUILIBRIUM

Here the half-life of the daughter nuclide is larger than that of the parent:

$$\lambda_1 > \lambda_2$$

The daughter activity is growing in the sample according to Eq.6.19 (Fig.6.6).



**Fig.6.6** Relation between the radioactivities of a parent and a daughter nuclide, when the half-life of the daughter exceeds that of the parent (half-lives of 8 and 0.8 hours, respectively): the case of no-equilibrium. The straight line is the semi-logarithmic plot of the parent activity. The upper curve is the total activity of the mixture.

Finally, after sufficiently long time, only daughter activity will be left, since the parent activity is disappearing at a higher rate:

$$A_2 = \frac{\lambda_2}{\lambda_1 - \lambda_2} A_1^0 e^{-\lambda_2 t} \quad (6.24)$$

After a period of  $t_{\max}$  the daughter activity reaches a maximum value for:

$$\frac{dA_2}{dt} = 0 = \frac{-\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} A_1^0 e^{-\lambda_1 t_{\max}} + \frac{\lambda_2^2}{\lambda_2 - \lambda_1} A_1^0 e^{-\lambda_2 t_{\max}}$$

or

$$\lambda_1 e^{-\lambda_1 t_{\max}} = \lambda_2 e^{-\lambda_2 t_{\max}} \quad (6.25)$$

The maximum value of the daughter activity is thus reached at:

$$t_{\max} = \frac{1}{\lambda_2 - \lambda_1} \ln \frac{\lambda_2}{\lambda_1} \quad (6.26)$$

Inserting Eq.6.25 in Eq.6.19 shows that at the time this maximum is reached, the parent and daughter activity are equal (Fig.6.6).

## 6.7 ACCUMULATION OF STABLE DAUGHTER PRODUCT

A special case of "no-equilibrium" as discussed in the preceding paragraph occurs if the daughter product is not radioactive, in other words:

$$\lambda_2 = 0$$

This can be illustrated by two examples, (i) the accumulation of  $^{40}\text{Ar}$  during the decay of  $^{40}\text{K}$  in rock (Fig.6.2) and the accumulation of  $^3\text{He}$  in water during the decay of  $^3\text{H}$ . We will take the latter process as an example to calculate the age of a sample from the remaining parent activity and the accumulated amount of daughter product.

Starting from the general relation of Eq.6.18 or 6.19 we can simply correct for  $\lambda_2 = 0$  and the absence of an original quantity of daughter (a hard condition for a successful application of the dating methods mentioned):

$$N_2 = N_1^0 (1 - e^{-\lambda_1 t}) \quad (6.27)$$

where the amount of gas accumulated ( $V$  in litres STP) is related to the number of atoms  $N_2$  by:

$$V = \frac{N_2}{6 \times 10^{23}} 22.4 \text{ L}$$

Since, instead of the original  $^3\text{H}$  activity, the activity after the unknown period of time is known (to be measured),  $N_1^0$  in Eq.6.27 has to be replaced by  $N_1$ , and subsequently  $N_1$  by the activity ( $A_1 = \lambda N_1$ ), so that:

$$\frac{6 \times 10^{23}}{22.4} V = N_1 e^{\lambda T} (1 - e^{-\lambda T}) = \frac{A_1}{\lambda} e^{+\lambda T} (1 - e^{-\lambda T}) = \frac{A_1}{\lambda} (e^{\lambda T} - 1)$$

The period of time elapsed since time zero (the "age"  $T$ ) is:

$$T = \frac{1}{\lambda} \ln \left( 2.7 \times 10^{22} \frac{\lambda}{A_1} V + 1 \right) \quad (6.28)$$

This dating method –especially applied in oceanography, but recently also in hydrology (Schlosser et al., 1998)– makes a very strong appeal to the experimental (mass spectrometric) technique, as the amount of  $^3\text{He}$  produced is extremely small. This is shown here by an example of one litres of water with a present-day  $^3\text{H}$  activity of 100 TU which over a period of 20 years has accumulated of  $5.1 \times 10^{-10}$  mL STP ( $0^\circ\text{C}$  and 1033 hPa) of  $^3\text{He}$ .

## 6.8 RADIOACTIVE GROWTH

For the sake of completeness we will mention here the production of radionuclides by nuclear reactions, because this phenomenon is, from a mathematical point of view, very similar to the case of secular equilibrium as discussed in Sect.6.6.1, if the production rate (P) is constant. The reactions may take place in a nuclear particle accelerator or in a nuclear reactor. The production rate of the radionuclide is:

$$\frac{dN}{dt} = P - \lambda N \quad (6.29)$$

where N is the number of radioactive nuclei and  $\lambda$  is the decay constant. Similar to Eq.6.21, the solution for the activity produced is:

$$\lambda N = A = P(1 - e^{-\lambda t}) \quad (6.30)$$

At time approaches infinity, a stationary is being reached in which the radionuclide production and decay are equal. Thus, at  $t = \infty$ :

$$A_{\max} = P \quad (6.31)$$

This is shown in Fig.6.7, representing the course of the radioactivity in time, comparably to Fig.6.4.

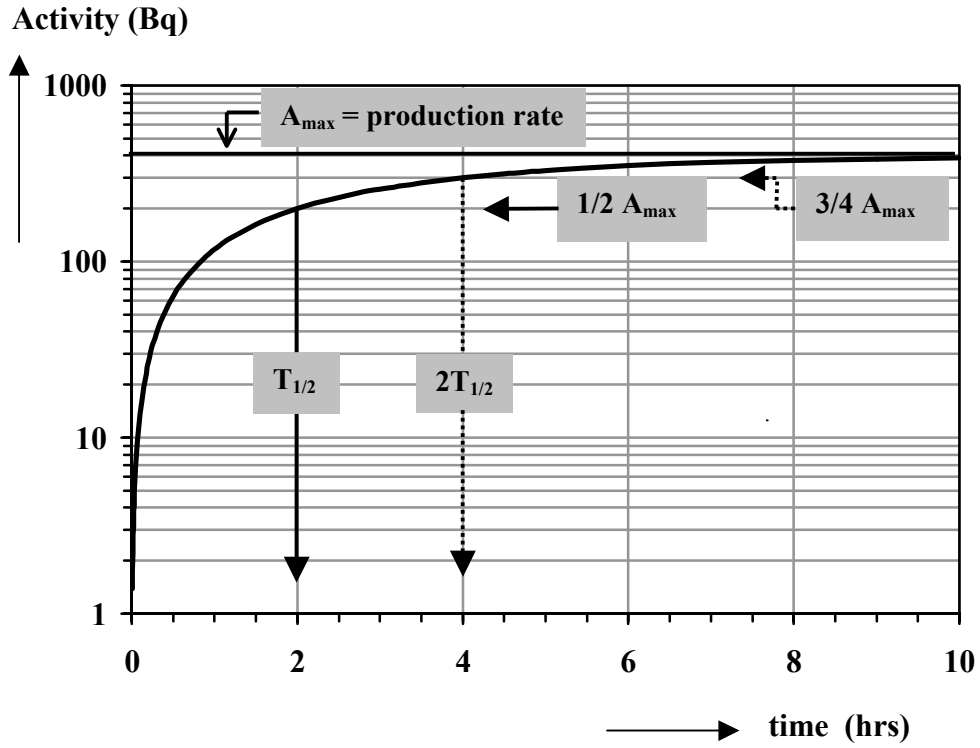
The time required to produce certain fractions of the maximum attainable activity is now:

$$A = 1/2 P = 1/2 A_{\max} \text{ after one half-life}$$

$$A = 3/4 P \text{ after a period of time} = 2T_{1/2}$$

$$A = 7/8 P \text{ after } 3T_{1/2}, \text{ and so on.}$$

This means that after 3 half-lives the maximum attainable activity has practically been reached.



**Fig.6.7** Growth of radioactivity by a constant production rate  $P$  of 400 nuclides/sec, resulting in a maximum activity of 400 Bq. The half-life of the produced nuclide is 2 hours.

