Chapter 8
The Statistics of Radioactivity

The laws of Statistics find applications in the phenomenon of radioactivity. The disintegration of a nucleus is a random event which is not affected by the history of the nucleus or the conditions external to the nucleus. In this chapter we will evaluate the probabilities that govern radioactive decay. The results we will derive will be of great importance to experimental practice.

8.1 The Behavior of Large Samples. The Law of Radioactivity

Soon after the discovery of radioactivity, it was found experimentally that the *activity* (rate of decay) of a sample decreases exponentially with time [1]. In 1905, E. von Schweidler proved theoretically the law of radioactivity, considering the process of nuclear decay as a purely statistical effect. The basic assumption he made is that the probability ΔP for a certain nucleus to decay during a sufficiently small interval of time Δt is proportional to this interval,

$$\Delta P = \lambda \Delta t, \tag{8.1}$$

with the coefficient of proportionality λ being characteristic of a certain kind of nucleus and mode of decay, independent of the nucleus' history or of any other influence from neighboring nuclei or the environment. The probability for a nucleus not to decay during a certain time interval $0 \le t < \Delta t$ is

$$1 - \Delta P = 1 - \lambda \Delta t. \tag{8.2}$$

The probability the nucleus will not decay during the interval $\Delta t \le t < 2\Delta t$ is exactly the same. The combined probability that the nucleus will not decay during the time interval $0 \le t < 2\Delta t$ is, therefore,

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DOI 10.1007/978-3-319-53345-2_8

$$(1 - \Delta P)^2 = (1 - \lambda \Delta t)^2. \quad (8.3)$$

In general, the probability a nucleus will not decay during the time interval between $t = 0$ and $t = n\,\Delta t$ is

$$(1 - \Delta P)^n = (1 - \lambda \Delta t)^n = (1 - \lambda t/n)^n, \quad (8.4)$$

which, in the limit $\Delta t \to 0, \quad n \to \infty$, tends to $e^{-\lambda t}$. Thus, if initially ($t = 0$) there were N_0 nuclei of the particular isotope, at time t the surviving nuclei will be

$$N(t) = N_0 e^{-\lambda t}. \quad (8.5)$$

This is the law of radioactivity.

Alternatively, if at time t there exist $N(t)$ nuclei, in a time interval dt there will be $\lambda N(t)\,\mathrm{d}t$ decays of nuclei (we assume that the number of nuclei is large enough so that the function $N(t)$ may be considered to be continuous, as was done here, and dt is small compared to the duration of our measurement but large enough so that the difference between the real number of decays during dt from the theoretically expected would be negligible). Thus,

$$\mathrm{d}N = -\lambda N\,\mathrm{d}t \quad (8.6)$$

the solution of which is Eq. (8.5).

The constant λ is characteristic of the isotope for the particular mode of decay (if there are more than one) and is called *decay constant*. It is proved that the *mean lifetime* of the nuclei is equal to

$$\tau = \frac{1}{\lambda}. \quad (8.7)$$

The time needed for half of the initial nuclei to decay is equal to

$$\tau_{1/2} = \frac{\ln 2}{\lambda}. \quad (8.8)$$

and is called *half-life*.

The variation of the number of surviving nuclei with time is shown in Fig. 8.1.

The *activity* of a sample (number of decays per unit time), which is the magnitude measured directly, is

$$A \equiv -\frac{\mathrm{d}N}{\mathrm{d}t} = \lambda N(t) = \lambda N_0 e^{-\lambda t} \quad (8.9)$$

and is seen to decrease exponentially with time, in agreement with experiment.

Figure 8.2 shows, as a function of time, the rate $R(t)$ at which the disintegrations of the nuclei of a radioactive sample are counted, by an experimental arrangement

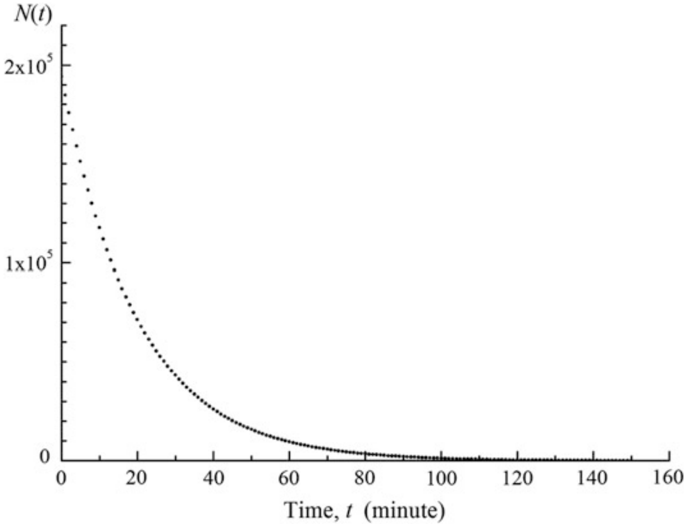


Fig. 8.1 The decay with time of the number of the nuclei of a radioactive sample. The points are at a time distance of one minute from each other. For this particular isotope it is $\tau = 20$ min

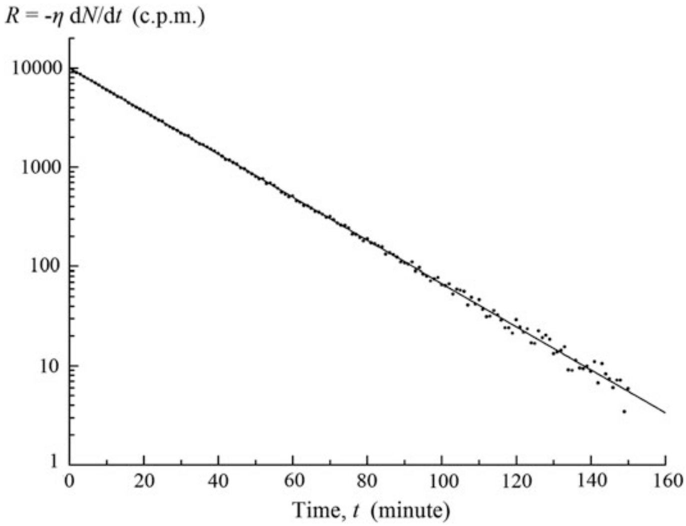


Fig. 8.2 Plot, as a function of time, of the rate of counting $R(t)$ of the disintegrations of a radioactive sample, with an experimental arrangement capable of counting only 10% of the decays $(\eta = 0.1)$. The scale of $R(t)$ is logarithmic. The continuous line shows the theoretically predicted rate, according to Eqs. (8.9) and (8.10)

which counts only 10% of the disintegrations (*efficiency* $\eta = 0.1$). Each measurement is, therefore, equal to

$$R(t) = -\eta \frac{\mathrm{d}N}{\mathrm{d}t} = \eta A \tag{8.10}$$

The units of $R(t)$ are c.p.m. (counts per minute) while those of the activity $A = -\mathrm{d}N/\mathrm{d}t$ are, usually, d.p.s (disintegrations per second). The scale of $R(t)$ in Fig. 8.2 is logarithmic. Because it is

$$\ln R(t) = \ln(\lambda N_0) - \lambda t, \tag{8.11}$$

the relationship between $\ln R(t)$ and t is linear. The same is true for the activity of the sample. The method used for the determination of the decay constant is based on plotting $\log R(t)$ as a function of time (from the slope of the straight line).

The statistical nature of the process of radioactivity must be stressed. This is something that was also highlighted theoretically by the successful interpretation that Quantum Mechanics gave to the phenomenon of α decay via the tunnel effect. It is this statistical nature of the phenomenon we will try to describe below in this chapter. At present we simply mention that, according to the relation (8.6), the average expected number of decays in a relatively small interval of time, Δt, is

$$\Delta N \approx \lambda N(t)\,\Delta t \approx \lambda N_0 \mathrm{e}^{-\lambda t} \Delta t. \tag{8.12}$$

Due to the nature of radioactive decay, the number which will be measured in practice will have fluctuations about this value. It will be proved that the fractional fluctuation is greater for small values of ΔN. This is obvious in Fig. 8.2, where, for large values of t, when the rate of disintegration is small, the differences between the measured and the theoretically predicted rate which has no fluctuations (continuous line) are proportionally large. It must be kept in mind that the scale for the rate is logarithmic and this brings out the *fractional* variations in this magnitude.

8.2 Nuclear Disintegrations and the Binomial Distribution

Assume that, initially ($t = 0$), we have in a sample N_0 radioactive nuclei and that we wish to know what is the probability that in a time t we will have exactly x decays. We may consider that we observe N_0 objects, each one of which has a probability $p(t)$ to suffer something (decay) and a probability $q(t) = 1 - p(t)$ that it will remain unchanged in the time interval between $t = 0$ and t. From the law of radioactivity, we know that it is

$$q(t) = \frac{N(t)}{N_0} = \mathrm{e}^{-\lambda t} \tag{8.13}$$

and, therefore, also

$$p(t) = 1 - \mathrm{e}^{-\lambda t}. \tag{8.14}$$

According to the binomial distribution, therefore, the probability exactly x of the N_0 nuclei to disintegrate in the time interval between $t = 0$ and t is

$$P_{N_0}(x) = \frac{N_0!}{(N_0 - x)!\,x!} p^x (1-p)^{N_0 - x}. \tag{8.15}$$

Substituting for $p(t)$ from Eq. (8.14), we have

$$P_{N_0}(x) = \frac{N_0!}{(N_0 - x)!\,x!} (1 - \mathrm{e}^{-\lambda t})^x (\mathrm{e}^{-\lambda t})^{N_0 - x}. \tag{8.16}$$

This is the exact relation for the probabilities, independently of any restrictions on the values of N_0, x and λ.

From the properties of the binomial distribution (Subsection 7.2.2), we know that the expected or mean value of the number of disintegrations x in the time interval from $t = 0$ to t is

$$\bar{x} = N_0 p = N_0 (1 - \mathrm{e}^{-\lambda t}) \tag{8.17}$$

and its standard deviation

$$\sigma_{\bar{x}} = \sqrt{N_0 pq} = \sqrt{N_0 (1 - \mathrm{e}^{-\lambda t}) \mathrm{e}^{-\lambda t}}. \tag{8.18}$$

We note that it is

$$\sigma_{\bar{x}} = \sqrt{\bar{x} \mathrm{e}^{-\lambda t}}. \tag{8.19}$$

For small values of t, i.e. for $t \ll \tau$ $(= 1/\lambda)$, the mean or expected value of the number of disintegrations in the time interval from $t = 0$ to t is equal to

$$\bar{x} = \lambda N_0 t \tag{8.20}$$

and the standard deviation of the mean $\bar{x}$ is equal to

$$\sigma_{\bar{x}} = \sqrt{\bar{x}}. \tag{8.21}$$

This is a very important characteristic of the phenomenon of radioactivity. The measurement of a number of events (disintegrations) also gives the standard deviation of this number (equal to its square root).

Example 8.1

A sample of radioactive material initially contains $N_0 = 10^9$ nuclei, whose decay constant is $\lambda = 10^{-6}\ \text{s}^{-1}$. What is the expected number of disintegrations in the time interval between $t = 0$ and $t = 10$ s ?

The mean lifetime of these radioactive nuclei is $\tau = 1/\lambda = 10^6$ s, which is much larger than the duration of the measurement, $t = 10$ s. We may, therefore, use Eqs. (8.20) and (8.21). We find $\bar{x} = \lambda N_0 t = 10^{-6} \times 10^9 \times 10 = 10\,000$ disintegrations and $\sigma_{\bar{x}} = \sqrt{\bar{x}} = 100$ disintegrations.

The expected number of disintegrations is $\bar{x} = 10\,000 \pm 100$.

The mean activity of the sample will therefore be $\bar{A} = -\overline{\text{d}N/\text{d}t} = \bar{x}/t = 1000 \pm 10$ d.p.s. (disintegrations per second). It therefore follows that measurements lasting for $t = 10$ s will give us the activity of this sample with an error of 1%.

If we perform many measurements of duration $t = 10$ s each, while the number N of the nuclei has not changed appreciably from the initial, N_0, the distribution of the results is expected to be given by the Gaussian distribution function

$$f(x) = \frac{1}{\sqrt{2\pi}\ 100} \text{e}^{-(x-10000)^2/2\times 10000},$$

where x is the total number of disintegrations measured in 10 s.

Example 8.2 [E]

The number of nuclei of isotope 1 varies with time according to the relation $N_1 = N_{10}\text{e}^{-t/\tau_1}$. The nuclei of the isotope 2 produced also decays with a mean life of τ_2. Plot the concentration $N_2(t)$ of the daughter isotope as a function of time. Given: $N_{10} = 10^6$, $\tau_1 = 10$ min, $N_{20} = 0$, $\tau_2 = 5$ min.

In a time interval dt, the number of nuclei of the daughter isotopes produced is $(\text{d}N_1/\text{d}t)\text{d}t$, while that of the nuclei decaying is $(N_2/\tau_2)\text{d}t$. The net change of the nuclei of isotope 2 is:

$$\text{d}N_2 = \frac{N_{10}}{\tau_1}\text{e}^{-t/\tau_1}\text{d}t - \frac{N_2}{\tau_2}\text{d}t \quad \text{or} \quad \frac{\text{d}N_2}{\text{d}t} + \frac{N_2}{\tau_2} = \frac{N_{10}}{\tau_1}\text{e}^{-t/\tau_1}$$

The solution of this differential equation with $N_{20} = 0$ is

$$N_2(t) = \frac{\tau_2}{\tau_1 - \tau_2} N_{10}\left(\text{e}^{-t/\tau_1} - \text{e}^{-t/\tau_2}\right).$$

Substituting in this equation

$$N_2(t) = 10^6\left(e^{-t/10} - e^{-t/5}\right).$$

with t in min.

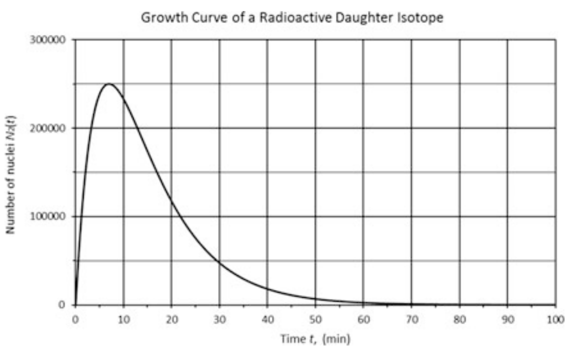
We will plot this function for $0 \le t \le 100$ min, with t increasing in steps of 0.1 min.

We place the t values in column A by the following procedure. Highlight cell A1 by left-clicking on it. Type **0** in the cell. Press **ENTER** and in cell A2 type **= A1 + 0.1**. **Fill Down** to cell A1001.

Column B will contain the values of $N_2(t)$. Highlight cell B1 by left-clicking on it. Type in this cell: **= 10^6*(exp(-A1/10)- exp(-A1/5))**. Then **Fill Down** to cell B1001.

Highlight columns A and B by left-clicking on the label A and then, holding the Shift or Control key down, left-clicking on label B. From **Insert > Recommended Charts** we select the **line chart**. The plot of $N_2(t)$ appears. We will format this graph:

1. In the chart title box type **Growth Curve of a Radioactive Daughter Isotope**.
2. Right-click on the curve. In the window that opens select **color black**. This changes the color of the curve to black. Set the thickness of the curve to 1.25 pts.
3. We left-click on a number of the Y-scale and open the **Format Axis, Axis Options** window. We set **Bounds Minimum** 0 and **Maximum** 300000, **Units Major** 100000 and **Minor** 50000. **Tick Marks**, **Major Type Outside** and **Minor Type Outside**. For the X-scale we select **Bounds Minimum** 0 and **Maximum** 100, **Units Major** 10 and **Minor** 5. **Tick Marks**, **Major Type Outside** and **Minor Type Outside**.
4. Pressing the [+] key opens the **Chart Elements** dialog box. We choose **Axis Titles**. For X-Axis we write **Time, *t* (min)**. For Y-Axis we write **Number of nuclei, $N_2(t)$**.



5. Pressing the [+] key to open the **Chart Elements** dialog box, we choose for the X-axis major gridlines to be visible. For the Y-axis, we choose both major and minor gridlines to appear.

Example 8.3 [O]

The number of nuclei of isotope 1 varies with time according to the relation $N_1 = N_{10}e^{-t/\tau_1}$. The nuclei of the isotope 2 produced also decays with a mean life of τ_2. Plot the concentration $N_2(t)$ of the daughter isotope as a function of time. Given: $N_{10} = 10^6$, $\tau_1 = 10$ min, $N_{20} = 0$, $\tau_2 = 5$ min.

In a time interval dt, the number of nuclei of the daughter isotopes produced is $(\mathrm{d}N_1/\mathrm{d}t)\mathrm{d}t$, while that of the nuclei decaying is $(N_2/\tau_2)\mathrm{d}t$. The net change of the nuclei of isotope 2 is:

$$\mathrm{d}N_2 = \frac{N_{10}}{\tau_1}e^{-t/\tau_1}\mathrm{d}t - \frac{N_2}{\tau_2}\mathrm{d}t \quad \text{or} \quad \frac{\mathrm{d}N_2}{\mathrm{d}t} + \frac{N_2}{\tau_2} = \frac{N_{10}}{\tau_1}e^{-t/\tau_1}$$

The solution of this differential equation with $N_{20} = 0$ is

$$N_2(t) = \frac{\tau_2}{\tau_1 - \tau_2}N_{10}\left(e^{-t/\tau_1} - e^{-t/\tau_2}\right).$$

Substituting in this equation

$$N_2(t) = 10^6\left(e^{-t/10} - e^{-t/5}\right)$$

with t in min.

We will plot this function for $0 \le t \le 100$min, with t increasing in steps of 0.1 min.

We place the t values in column A by the following procedure. Highlight column A by left-clicking on its label, A. Then

Column > Set Column Values

typing **(i-1)/10**, with i from 1 to 1001, in the window that opens. Then press **OK**.

Column B will contain the values of $N_2(t)$. Highlight column B by left-clicking on its label, B. Then

Column > Set Column Values

typing **10^6^*(exp(col(A)/10)-exp(col(A)/5))**, with i from 1 to 1001, in the window that opens. Then press **OK**.

Highlight columns A and B by left-clicking on the label A and then, holding the Shift or Control key down, left-clicking on label B. Then

Plot > Line > Line

The plot of $N_2(t)$ appears. We will format this graph.

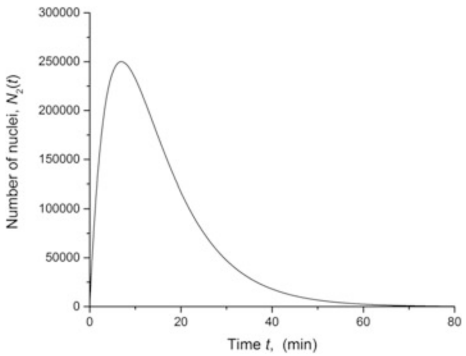
1. Delete the text box.
2. Double-click on the line and set

Line: Connect Straight, Style Solid, Width 1, Color Black Press **OK**.

3. Double-click on one of the axes and set
Scale Horizontal, from 0 to 80, Type linear, Major Ticks: Type By Increments, Value 20, Minor Ticks: Type By Counts, Count 1
Vertical from 0 to 300000, Type linear, Major Ticks: Type By Increments, Value 50000, Minor Ticks: Type By Counts, Count 1

4. We change the labels of the axes:
Double-click on the X label and write **Time *t* (min)**. Double-click on the Y label and write **Number of nuclei, *N*₂(*t*)**.
5. Save the project. Export graph as jpg (say).

The final graph is shown here.



Example 8.4 [P]

The number of nuclei of isotope 1 varies with time according to the relation $N_1 = N_{10}\mathrm{e}^{-t/\tau_1}$. The nuclei of the isotope 2 produced also decays with a mean life of τ_2. Plot the concentration $N_2(t)$ of the daughter isotope as a function of time. Given: $N_{10} = 10^6$, $\tau_1 = 10$ min, $N_{20} = 0$, $\tau_2 = 5$ min.

In a time interval dt, the number of nuclei of the daughter isotopes produced is $(\mathrm{d}N_1/\mathrm{d}t)\mathrm{d}t$, while that of the nuclei decaying is $(N_2/\tau_2)\mathrm{d}t$. The net change of the nuclei of isotope 2 is:

$$\mathrm{d}N_2 = \frac{N_{10}}{\tau_1}\mathrm{e}^{-t/\tau_1}\mathrm{d}t - \frac{N_2}{\tau_2}\mathrm{d}t \quad \text{or} \quad \frac{\mathrm{d}N_2}{\mathrm{d}t} + \frac{N_2}{\tau_2} = \frac{N_{10}}{\tau_1}\mathrm{e}^{-t/\tau_1}$$

The solution of this differential equation with $N_{20} = 0$ is

$$N_2(t) = \frac{\tau_2}{\tau_1 - \tau_2}N_{10}\left(\mathrm{e}^{-t/\tau_1} - \mathrm{e}^{-t/\tau_2}\right).$$

Substituting in this equation

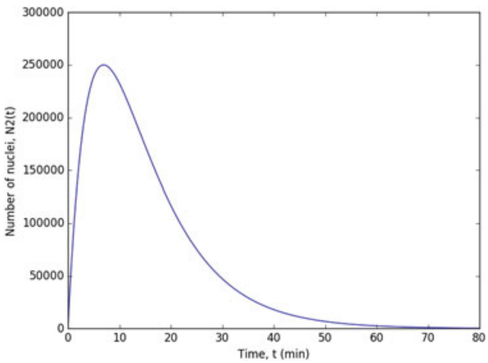
$$N_2(t) = 10^6\left(e^{-t/10} - e^{-t/5}\right)$$

with t in min.

Using matplotlib, we will plot this function for $0 \leq t \leq 100$ min, with t increasing in steps of 0.1 min.

```
from __future__ import division
import numpy as np
import matplotlib.pyplot as plt
t = np.linspace(0.0, 100.0, 1000)
N2 = 10**6 * (np.exp(-t/10.0)-np.exp(-t/5.0))
plt.plot(t, N2, '-')
plt.xlim(0, 80)
plt.ylim(0, 300000)
plt.ylabel("Number of nuclei, N2(t)")
plt.show()
```

The resulting curve is shown here.



Example 8.5 [R]

The number of nuclei of isotope 1 varies with time according to the relation $N_1 = N_{10}e^{-t/\tau_1}$. The nuclei of the isotope 2 produced also decays with a mean life of τ_2. Plot the concentration $N_2(t)$ of the daughter isotope as a function of time. Given: $N_{10} = 10^6$, $\tau_1 = 10$ min, $N_{20} = 0$, $\tau_2 = 5$ min.

In a time interval dt, the number of nuclei of the daughter isotopes produced is $(\mathrm{d}N_1/\mathrm{d}t)\mathrm{d}t$, while that of the nuclei decaying is $(N_2/\tau_2)\mathrm{d}t$. The net change of the nuclei of isotope 2 is:

$$dN_2 = \frac{N_{10}}{\tau_1} e^{-t/\tau_1} dt - \frac{N_2}{\tau_2} dt \quad \text{or} \quad \frac{dN_2}{dt} + \frac{N_2}{\tau_2} = \frac{N_{10}}{\tau_1} e^{-t/\tau_1}$$

The solution of this differential equation with $N_{20} = 0$ is

$$N_2(t) = \frac{\tau_2}{\tau_1 - \tau_2} N_{10} \left(e^{-t/\tau_1} - e^{-t/\tau_2} \right).$$

Substituting in this equation

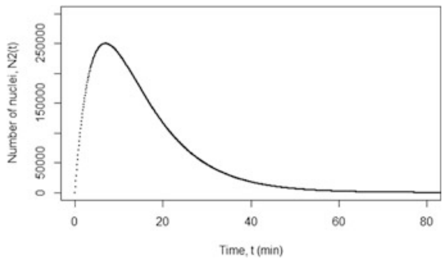
$$N_2(t) = 10^6 \left(e^{-t/10} - e^{-t/5} \right).$$

with t in min.

Just as we did above, we will plot this function for $0 \leq t \leq 100$ min, with t increasing in steps of 0.1 min.

```
> t<- seq(0, 100, by = 0.1)
> N2 <- 10^6*(exp(-t/10)-exp(-t/5))
>
> plot(t, N2, pch=20, cex=0.5, xlim=c(0, 80), ylim=c(0, 300000),
  xlab="Time, t (min)", ylab="Number of nuclei, N2(t)")
```

The graph shown in the figure appears.



Example 8.6 [E]

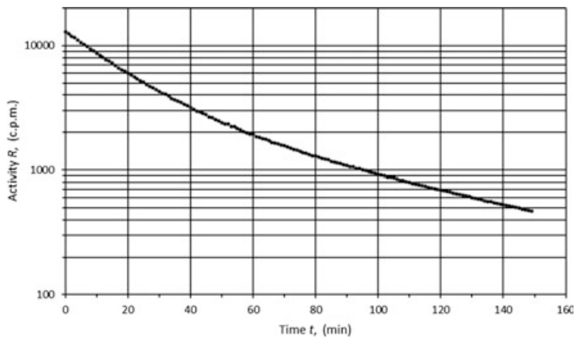
Measurements of the activity of a radioactive sample, R, are given every minute for $0 \leq t \leq 150$ min:

```
12993, 12414, 11882, 11566, 11023, 10623, 10207, 9813, 9428, 9026, 8639,
8353, 8058, 7709, 7517, 7218, 6904, 6637.86466, 6406, 6198, 5995, 5820,
5579, 5393, 5196, 5098, 4841, 4689, 4564, 4424, 4246, 4135, 4072, 3912,
3759, 3648, 3594, 3480, 3380, 3287, 3187, 3085, 2969, 2925, 2843, 2778,
2669, 2624, 2542, 1823, 1774, 1753, 1714, 1670, 1647, 1616, 1578, 1566,
1527, 1491, 1463, 1446, 1417, 1370, 1353, 1325, 1297, 1291, 1261, 1244,
```

1222, 1206, 1168, 1159, 1141, 1122, 1096, 1081, 1067, 1059, 1027, 1023, 998, 983, 964, 956, 948, 924, 913, 905, 880, 876, 865, 853, 828, 826, 810, 805, 786, 771, 762, 757, 745, 732, 719, 710, 563, 559, 550, 543, 537, 530, 525, 515, 512, 504, 497, 489, 484, 478, 469

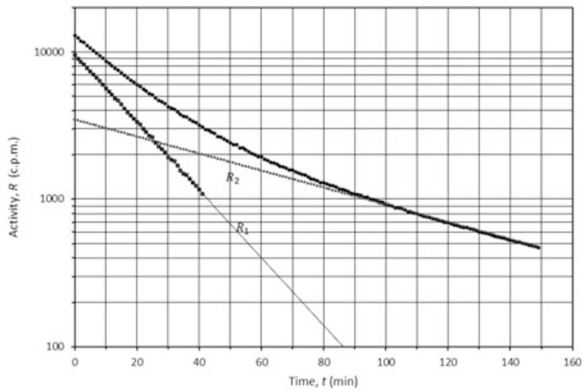
Plot log*R*(*t*) and verify that the activity seems to be due to two isotopes with different decay constants. Analyze the curve *R*(*t*) into two decay curves and find the two decay constants.

The values of *t* are entered in column A and those of *R* in column B. We plot log*R*(*t*): Highlight columns A and B by left-clicking on label A and, holding the Shift or Control key down, left-click on label B. Then, from **Insert**, **Recommended Charts**, we choose the **Scatter** plot. After some basic formatting, the graph shown in the figure below is produced.



We assume that the activity in the interval $120 \le t \le 150$ min is due almost entirely to isotope 2. From columns A and B we copy the data for $120 \le t \le 150$ min and paste them in columns C and D. We plot these points. Pressing the $\boxed{+}$ key to open the **Chart Elements** dialog box, we choose **Trendline** and an **Exponential** fit. The equation given is y = 3456.9 $e^{-0.0133x}$, which corresponds to the activity of isotope 2 being given by $R_2(t) = R_{20}e^{-t/\tau_2}$, where $R_{20} = 3457$ c.p.m. and $\tau_2 = 1/0.0133 = 75$ min.

In cell E1 we type **= 3456.9*EXP(-0.0133*A1)**. We press **ENTER** and **Fill Down** to E150. Column E now contains the values of $R_2(t)$. In cell F1 we type **= B1-E1**. We press **ENTER** and **Fill Down** to F150. Column F now contains $R_1(t)$. We fit an exponential to these data, using only those values that are greater than 1000, as these have small proportional errors. The equation given is y = 9594.2$e^{-0.053x}$, which corresponds to the activity of isotope 1 is given by $R_1(t) = R_{10}e^{-t/\tau_1}$, where $R_{10} = 9594$ c.p.m. and $\tau_1 = 1/0.053 = 18.9\,\text{min}$. In a graph we plot R_1, R_2 and $R_1 + R_2$. This is shown in the figure below.



Example 8.7 [O]

Measurements of the activity of a radioactive sample, R, are given for $0 \leq t \leq 150$ min (see Example 8.6 [E]). Plot log$R(t)$ and verify that the activity seems to be due to two isotopes with different decay constants. Analyze the curve $R(t)$ into two decay curves and find the two decay constants.

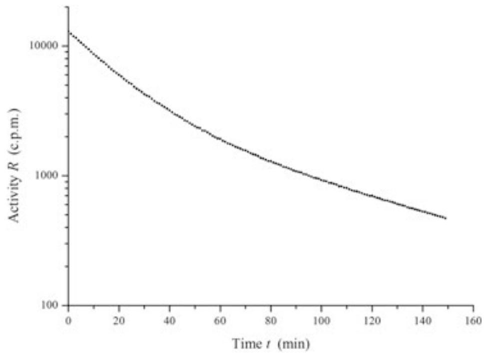
The values of t are entered in column A and those of R in column B. We plot log$R(t)$: Highlight columns A and B by left-clicking on label A and, holding the Shift or Control key down, left-click on label B. Then

Plot > Symbol > Scatter

A plot of $R(t)$ is produced. We will modify the plot to suit our requirements:

1. Delete the text box in the plot.
2. Double-click on a point and in **Plot Details > Symbol** change the 9 pt. squares to 3 pt. circles.
3. We change the labels of the axes:
   Double-click on the X label and write **Time *t* (min)**. Double-click on the Y label and write **Activity *R* (c.p.m)**.
4. 4. Double click on the t-axis. In the **Scale** window that opens, select, for Horizontal **From 0 to 160** and for Vertical **From 100 to 20 000** and **Type Log10**.

The graph produced is shown below.



We assume that the activity in the interval $120 \leq t \leq 150$ min is due almost entirely to isotope 2. From columns A and B we copy the data for $120 \leq t \leq 150$ min and paste them in columns C and D. We highlight column C by left-clicking on its label. We then set column C as an X axis by selecting **Column** and then **Set as X**. We plot a graph of these points exactly as above.

We will plot a best fit exponential curve between these points. While in the graph, we select

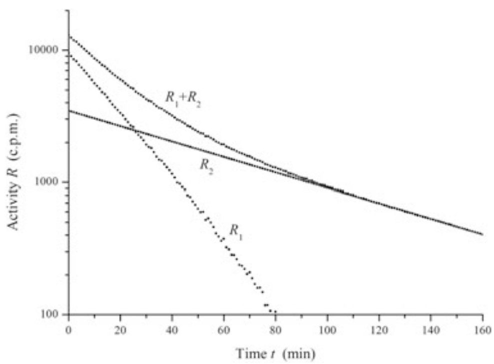
Analysis > Fitting > Exponential Fit

In the window that opens

Settings > Function Selection > Category, Exponential > Function, ExpDec1

We go to **Parameters**. We tick **Fixed** for **y0** and set its value to zero. Press **Fit**. The best fit for these points is given as y = A1*exp(-x/t1), or $R_2 = R_{20}\mathrm{e}^{-t/\tau_2}$, where $R_{20} = 3472.8 \pm 3.9$ c.p.m. and $\tau_2 = 74.52 \pm 0.47$ min.

For $0 \leq t \leq 150$, we enter $R_2 = R_{20}\mathrm{e}^{-t/\tau_2}$ in column E. The difference of col(B) – col(E) is evaluated in column F. This is the activity of the first isotope, $R_1 = R_{10}\mathrm{e}^{-t/\tau_1}$. A best fit performed on R_1 as above gives $R_{10} = 9580 \pm 14$ c.p.m. and $\tau_1 = 18.82 \pm 0.04$ min. In a graph we plot R_1, R_2 and $R_1 + R_2$. This is shown in the figure below.



Example 8.8 [P]

Measurements of the activity of a radioactive sample, R, are given for $0 \le t \le 150$ min (see Example 8.6 [E]). Plot log$R(t)$ and verify that the activity seems to be due to two isotopes with different decay constants. Analyze the curve $R(t)$ into two decay curves and find the two decay constants.

```
import numpy as np
import matplotlib.pyplot as plt
from scipy.optimize import curve_fit

t = np.arange(0, 150)
R = np.array([12993, 12414, 11882, 11566, 11023, 10623, 10207, 9813, 9428, 9026,
8639, 8353, 8058, 7709, 7517, 7218, 6904, 6637.86466, 6406, 6198, 5995, 5820, 5579,
5393, 5196, 5098, 4841, 4689, 4564, 4424, 4246, 4135, 4072, 3912, 3759, 3648, 3594,
3480, 3380, 3287, 3187, 3085, 2969, 2925, 2843, 2778, 2669, 2624, 2542, 2477, 2408,
2348, 2327, 2226, 2218, 2157, 2098, 2028, 1985, 1935, 1928, 1855, 1823, 1774, 1753,
1714, 1670, 1647, 1616, 1578, 1566, 1527, 1491, 1463, 1446, 1417, 1370, 1353, 1325,
1297, 1291, 1261, 1244, 1222, 1206, 1168, 1159, 1141, 1122, 1096, 1081, 1067, 1059,
1027, 1023, 998, 983, 964, 956, 948, 924, 913, 905, 880, 876, 865, 853, 828, 826,
810, 805, 786, 771, 762, 757, 745, 732, 719, 710, 699, 698, 685, 674, 668, 653,
645, 643, 632, 626, 616, 603, 597, 590, 584, 570, 563, 559, 550, 543, 537, 530,
525, 515, 512, 504, 497, 489, 484, 478, 469])

# We define the function R(t) = R_10 e^(-t/τ_1) + R_20 e^(-t/τ_2) in Python as follows:
def R_func(t, A1, t1, A2, t2):
  return A1*np.exp(-t/t1) + A2*np.exp(-t/t2)

# We then use the curve_fit function of the scipy.optimize sub-package to
# perform non-linear least squares fitting to the data:
popt, pcov = curve_fit(R_func, t, R, p0 = (12000, 20, 3000, 100))

# By examining the popt array,
Popt
array([ 9949.25831076, 19.8784205, 3036.47784508, 79.38078197])

# we obtain the results of the fitting:
R_10 = 9949.258 c.p.m, R_20 = 3036.478 c.p.m, τ_1 = 19.878, τ_2 = 79.381
```

Example 8.9 [R]

Measurements of the activity of a radioactive sample, R, are given for $0 \le t \le 150$ min. Plot log$R(t)$ and verify that the activity seems to be due to two isotopes with different decay constants. Analyze the curve $R(t)$ into two decay curves and find the two decay constants.

We will use non-linear least squares in order to fit a curve of the form $R(t) = R_{10}\mathrm{e}^{-t/\tau_1} + R_{20}\mathrm{e}^{-t/\tau_2}$ to the values of $R(t)$.

```
> t<- seq(0, 149, by=1)
>
> R<- c(12993, 12414, 11882, 11566, 11023, 10623, 10207, 9813, 9428, 9026, 8639,
8353, 8058, 7709, 7517, 7218, 6904, 6637.86466, 6406, 6198, 5995, 5820, 5579, 5393,
5196, 5098, 4841, 4689, 4564, 4424, 4246, 4135, 4072, 3912, 3759, 3648, 3594, 3480,
3380, 3287, 3187, 3085, 2969, 2925, 2843, 2778, 2669, 2624, 2542, 2477, 2408, 2348,
2327, 2226, 2218, 2157, 2098, 2028, 1985, 1935, 1928, 1855, 1823, 1774, 1753, 1714,
1670, 1647, 1616, 1578, 1566, 1527, 1491, 1463, 1446, 1417, 1370, 1353, 1325, 1297,
1291, 1261, 1244, 1222, 1206, 1168, 1159, 1141, 1122, 1096, 1081, 1067, 1059, 1027,
1023, 998, 983, 964, 956, 948, 924, 913, 905, 880, 876, 865, 853, 828, 826, 810,
805, 786, 771, 762, 757, 745, 732, 719, 710, 699, 698, 685, 674, 668, 653, 645,
643, 632, 626, 616, 603, 597, 590, 584, 570, 563, 559, 550, 543, 537, 530, 525,
515, 512, 504, 497, 489, 484, 478, 469)
>
> fm1 < - nls(R ~ A1*exp(-t/t1) + A2*exp(-t/t2), start = list
(A1 = 12000, t1 = 20, A2 = 3000, t2 = 100))
> fm1
  Nonlinear regression model
  model: R  ~  A1 * exp(-t/t1)  + A2 * exp(-t/t2)
  data: parent.frame()
  A1     t1     A2     t2
  9949.26  19.88 3036.48  79.38
  residual sum-of-squares: 56850
  Number of iterations to convergence: 3
  Achieved convergence tolerance: 5.324e-06
>
```

The results of the fitting are:

$$R_{10} = 9949 \text{ c.p.m.}, R_{20} = 3036 \text{ c.p.m.}, \tau_1 = 19.88\,\text{min and } \tau_2 = 79.38\,\text{min}.$$

```
> R1<-9949*exp(-t/19.88)
> R2<-3036*exp(-t/79.38)
>
```

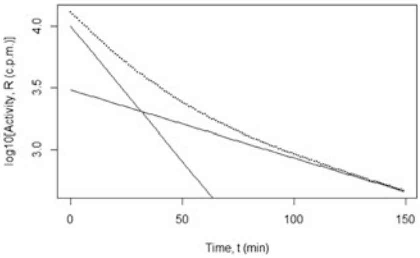
We plot $R(t)$

```
> plot(t,log10(R), pch=20, cex=0.5, xlab="Time, t (min)", ylab="log10
[Activity, R (c.p.m.)]")
>
```

and then add the curves (straight lines) for R_1 and R_2:

```
> lines(t, log10(R1))
> lines(t, log10(R2))
```

The graph shown in the figure is produced.



8.3 Radioactivity and the Poisson Distribution

Assume that, in a radioactive sample, the probability for a disintegration (i.e. an event) to occur in the small time interval dt is $\Lambda \mathrm{d}t$. The probability of having 0 events (i.e. no disintegration) in the time integral dt is, therefore, equal to $1 - \Lambda \mathrm{d}t$. The parameter Λ, which expresses a probability per unit time, is considered to remain constant during the experiment. According to Sect. 8.1, if the sample consists of N nuclei, it will be $\Lambda = \lambda N$. For Λ to remain constant, the number N of the nuclei must not change significantly during the whole of the experiment.

We will denote by $P_r(t)$ the probability that r disintegrations will occur in the time interval (0, t). (We point out that the notation for the probabilities in this section is different from that of Chap. 7).

Let us first evaluate the probability $P_0(t)$ of no disintegration in the time interval (0, t). For no disintegration to happen in the time interval (0, $t + \mathrm{d}t$), no disintegrations must happen either in the interval (0, t) or the interval (t, $t + \mathrm{d}t$). Since the probabilities are independent of each other, the probability that no disintegration will occur in the time interval (0, $t + \mathrm{d}t$) will be equal to the product of the probabilities that no disintegrations should happen either in the interval (0, t) or the interval (t, $t + \mathrm{d}t$):

$$P_0(t + \mathrm{d}t) = P_0(t)\,(1 - \Lambda \mathrm{d}t). \tag{8.22}$$

Therefore,

$$\frac{P_0(t+\mathrm{d}t) - P_0(t)}{\mathrm{d}t} = -\Lambda P_0(t) \tag{8.23}$$

Or

$$\frac{\mathrm{d}P_0(t)}{\mathrm{d}t} = -\Lambda P_0(t). \tag{8.24}$$

The solution of this differential equation is:

$$P_0(t) = A\,\mathrm{e}^{-\Lambda t} \tag{8.25}$$

where A is a constant. Given that it is certain that no disintegration will happen in the interval (0, 0), it is $P_0(0) = 1$. Therefore, $A = 1$ and

$$P_0(t) = \mathrm{e}^{-\Lambda t}. \tag{8.26}$$

We will evaluate the probability $P_1(t)$ of exactly one disintegration to occur in the time interval (0, t). There are two ways in which exactly one disintegration will occur in the interval (0, $t + \mathrm{d}t$). The first is to have one disintegration occurring in the interval (0, t) and none in (t, $t + \mathrm{d}t$). The second is to have no disintegration occurring in the interval (0, t) and one in the interval (t, $t + \mathrm{d}t$). The total probability is, therefore,

$$P_1(t+\mathrm{d}t) = P_1(t)\,(1 - \Lambda \mathrm{d}t) + P_0(t)\Lambda \mathrm{d}t \tag{8.27}$$

or

$$\frac{P_1(t+\mathrm{d}t) - P_1(t)}{\mathrm{d}t} = \frac{\mathrm{d}P_1(t)}{\mathrm{d}t} = -\Lambda P_1(t) + \Lambda P_0(t) \tag{8.28}$$

and the differential equation for $P_1(t)$ is

$$\frac{\mathrm{d}P_1(t)}{\mathrm{d}t} = -\Lambda P_1(t) + \Lambda \mathrm{e}^{-\Lambda t}. \tag{8.29}$$

whose solution, satisfying the condition $P_1(0) = 0$, is

$$P_1(t) = \Lambda t \mathrm{e}^{-\Lambda t}. \tag{8.30}$$

Generalizing, we will now evaluate the probability $P_r(t)$ for exactly r disintegrations to occur in the time interval (0, t). There are two ways in which exactly r disintegrations occur in the time interval (t, $t + \mathrm{d}t$). The first is that r disintegrations occur in the interval (0, t) and none in the interval (t, $t + \mathrm{d}t$). The second is that

$r-1$ disintegrations occur in (0, t) and one in (t, $t+\mathrm{d}t$). The total probability is, therefore,

$$P_r(t+\mathrm{d}t) = P_r(t)\,(1-\Lambda \mathrm{d}t) + P_{r-1}(t)\Lambda \mathrm{d}t \tag{8.31}$$

from which we have the recurrence differential equation

$$\frac{\mathrm{d}P_r(t)}{\mathrm{d}t} = -\Lambda P_r(t) + \Lambda P_{r-1}(t) \tag{8.32}$$

for $P_r(t)$. It is easily verified, by substitution, that the function

$$P_r(t) = \frac{(\Lambda t)^r}{r!}\mathrm{e}^{-\Lambda t} \tag{8.33}$$

is a solution of the recurrence differential equation, which gives the already known results for $P_0(t)$ and $P_1(t)$.

From the known facts for the phenomenon of radioactivity, when the sample consists of N_0 nuclei whose decay constant is λ, it will be $\Lambda = \lambda N_0$. It has therefore been found that, for a given time interval t, the probabilities that $r = 0, 1, 2, \ldots$ disintegrations will occur, are given by the Poisson distribution

$$P_r(t) = \frac{(\lambda N_0 t)^r}{r!}\mathrm{e}^{-\lambda N_0 t}. \tag{8.34}$$

The mean or expected number of disintegrations in the time interval (0, t) will be

$$\mu = \lambda N_0 t \quad (t \ll \tau) \tag{8.35}$$

Returning to the notation of Chap. 7, the probability for x disintegrations occurring in the time interval (0, t), for which the expected number of disintegrations is $\mu = \lambda N_0 t$, is given by the relation

$$P_\mu(x) = \frac{\mu^x}{x!}\mathrm{e}^{-\mu}. \tag{8.36}$$

As it is known, for the Poisson distribution the mean or expected value of x is $\bar{x} = \mu$ and its standard deviation is $\sigma_{\bar{x}} = \sqrt{\mu} = \sqrt{\bar{x}}$. These are in agreement with what has been said in Sect. 8.2.

8.4 The Counting Rate of Nuclear Disintegrations and Its Error

In an experimental arrangement for the measurement of radioactivity, only a fraction of the total number of disintegrations is measured. This fraction is called *efficiency* η of the arrangement. Thus, if the activity of the sample being measured is $A = -\mathrm{d}N/\mathrm{d}t$, the counting rate of the disintegrations will be

$$R = \eta\,(-\mathrm{d}N/\mathrm{d}t) = \eta A \tag{8.37}$$

The problem we will now examine is this: If in a time interval t, the number of nuclei disintegrating is x, then ηx of them will be counted by our experimental arrangement. The counting rate will be $R = \eta x/t$. What is the expected or mean value $\bar{R}$ of R and which is its standard deviation $\sigma_{\bar{R}}$?

For the solution of the problem, we return to the beginning of the extraction of the relation for the probabilities for x disintegrations to occur in the time interval (0, t). Since the probability of a disintegration occurring in the time interval dt is $\Lambda \mathrm{d}t$, the probability of a disintegration being counted in the time interval dt is $\eta\Lambda dt$. Substituting $\eta\Lambda$ in place of Λ in Eq. (8.33) and since it is $\Lambda = \lambda N_0$, we find

$$P_x(t) = \frac{(\eta\lambda N_0 t)^x}{x!}\mathrm{e}^{-\eta\lambda N_0 t} \tag{8.38}$$

as the probability of $x = 0, 1, 2, \ldots$ disintegrations being counted in the time interval (0, t). The counting rate of the disintegrations in the time interval (0, t) is $R = x/t$. The possible values of the counting rate are

$$R_0 = 0, \quad R_1 = \frac{1}{t}, \quad R_2 = \frac{2}{t}, \ldots, \quad R_x = \frac{x}{t}, \ldots$$

and, therefore, the mean of the counting rate is

$$\bar{R} = \sum_{x=0}^{\infty}\frac{x}{t}P_x(t) = \frac{1}{t}\sum_{x=0}^{\infty} x\,P_x(t) = \frac{\bar{x}}{t} = \frac{\eta\lambda N_0 t}{t} = \eta\lambda N_0. \tag{8.39}$$

The standard deviation or the standard error of the mean $\bar{R}$ is

$$\sigma_{\bar{R}} = \sqrt{\sum_{x=0}^{\infty}(R-\bar{R})^2 P_x(t)} = \sqrt{\sum_{x=0}^{\infty}\left(\frac{x}{t}-\frac{\bar{x}}{t}\right)^2 P_x(t)} = \frac{1}{t}\sqrt{\sum_{x=0}^{\infty}(x-\bar{x})^2 P_x(t)} = \frac{\sigma_{\bar{x}}}{t} = \frac{\sqrt{\eta\lambda N_0 t}}{t} \tag{8.40}$$

$$\sigma_{\bar{R}} = \frac{\sigma_{\bar{x}}}{t} = \sqrt{\frac{\bar{R}}{t}} = \frac{\sqrt{\bar{x}}}{t}. \tag{8.41}$$

Summarizing, if in a time interval equal to t we count M nuclear disintegrations, then we conclude that, for measurements lasting for time t, the expected or mean value of the number of the disintegrations counted, x, is

$$\bar{x} = M \pm \sqrt{M} \tag{8.42}$$

and, therefore, the expected or mean rate of counting is

$$\bar{R} = \frac{M}{t} \pm \frac{\sqrt{M}}{t}. \tag{8.43}$$

From the relation $\sigma_{\bar{R}} = \sqrt{\frac{\bar{R}}{t}}$ we see that, for a given counting rate R, in order to reduce the error in the measured rate $\bar{R}$ by a factor of 2 we must quadruple the duration of the measurement.

To avoid mistakes, it must be noted that the equation $\bar{x} = M \pm \sqrt{M}$ makes sense only if the magnitude M is a pure (i.e. dimensionless) number. Otherwise the equation would be dimensionally wrong, since the dimensions (and the units) of M and $\sqrt{M}$ would not be the same (see Appendix 2). It would be wrong, for example, to evaluate first the value of the rate $\bar{R}$ and then take the square root of this quantity as being its standard deviation!

Example 8.10

In a measurement that lasted for 8 min, 1685 nuclear disintegrations were counted in a radioactive sample. What is the counting rate and its standard deviation? If the efficiency of the measuring arrangement is equal to 10%, with negligible error, what is the estimate for the activity A of the sample?

It is $t = 8$ min and $M = 1685$. The mean value of the number of counts in an 8-minute measurement is, therefore, $\bar{x} = 1685 \pm \sqrt{1685} = 1685 \pm 40$ counts.

The counting rate is equal to $R = \frac{1685}{8} \pm \frac{40}{8} = 211 \pm 5$ c.p.m.

Given that $\eta = 0.1$ with good accuracy, the activity of the sample is $A = \frac{R}{\eta} = \frac{211 \pm 5}{0.1} = 2110 \pm 50$ d.p.m. (disintegrations per minute) or $A = \frac{2110 \pm 50}{60} = 35 \pm 1$ d.p.s. (disintegrations per second).

Example 8.11

The measurement of a radioactive sample for 100 s resulted in the recording of 635 counts. Taking the sample away and counting for 30 s, resulted in 98 counts (these counts are due to the radioactivity of the environment and is called *background*). Find the clear counting rate which is due to the sample alone, as well as its standard deviation.

The background counting rate is: $R_{\mathrm{B}} = \frac{98 \pm \sqrt{98}}{30} = 3.27 \pm 0.33$ c.p.s.

The total counting rate of source and background is: $R_T = \frac{635 \pm \sqrt{635}}{100} = 6.35 \pm 0.25$ c.p.s.

The clear counting rate of the source alone is given by the difference

$$R_S = R_T - R_B = 6.35 - 3.27 = 3.08\,\text{c.p.s.}$$

which has a standard deviation $\sigma_S = \sqrt{\sigma_T^2 + \sigma_B^2} = \sqrt{0.25^2 + 0.33^2} = 0.41$ c.p.s.

Therefore, $R_S = 3.1 \pm 0.4$ c.p.s.

Example 8.12

If R_T is the total counting rate for source and background and R_B is the counting rate for the background alone, which must the division of the available time be among the two measurements in order to obtain the best accuracy in the measurement of the rate $R_S = R_T - R_B$ of the source?

If the available time is equal to t and a time t_T is used for the measurement of R_T and time $t_B = t - t_T$ for the measurement of R_B, we will have a total of $M_T = R_T t_T$ counts when we measure the source plus background and $M_B = R_B t_B$ counts when we measure the background alone.

The standard deviation σ_S of R_S is given by the relation $\sigma_S^2 = \sigma_T^2 + \sigma_B^2$, where $\sigma_T^2 = R_T/t_T$ and $\sigma_B^2 = R_B/t_B$.

Therefore, it is $\sigma_S^2 = \frac{R_T}{t_T} + \frac{R_B}{t - t_T}$. The value of σ_S, which is always positive, has a minimum when σ_S^2 has a minimum.

This happens when it is $\frac{d\sigma_B^2}{dt_T} = \frac{d}{dt_T}\left(\frac{R_T}{t_T} + \frac{R_B}{t - t_T}\right) = 0$ or $-\frac{R_T}{t_T^2} + \frac{R_B}{(t - t_T)^2} = 0$.

It follows that, for the smallest possible error σ_S in R_S, the time must be divided according to the relation $\frac{t_T}{t_B} = \sqrt{\frac{R_T}{R_B}}$.

Substituting, we find that, in this case, the value of the minimum standard deviation of R_S is:

$$\sigma_{S,\min} = \sqrt{\frac{R_T}{t}} + \sqrt{\frac{R_B}{t}}.$$

Example 8.13

Apply the conclusions of Example 8.12 to the measurements of Example 8.11.

For $R_T = 6.35$ c.p.s. and $R_B = 3.27$ c.p.s., the best division of the total time for the measurement will be $\frac{t_T}{t_B} = \sqrt{\frac{R_T}{R_B}} = \sqrt{\frac{6.35}{3.27}} = 1.39$.

Since it is $\frac{t_T + t_B}{t_B} = 2.39$, it follows that $t_B = \frac{t}{2.39} = 0.42\,t$ and $t_T = 0.58\,t$.

The best division of time would be 42% for the measurement of the background and 58% for the measurement of source plus background. If this division had been done in Example 8.9, in which it was $t = 130$ s, we would have $\sigma_{S,\min} = 0.38$ d.p.s. instead of 0.41 d.p.s. The difference is small.

Example 8.14

The counting rate for a radioactive source was measured to be $R = 160 \pm 4$ c.p.m. What was the approximate duration t of the measurement? The background may be considered to be negligible.

If the measurement lasted for time t, the number of counts recorded was $M = Rt$, with a standard deviation $\sqrt{M} = \sqrt{Rt}$. By dividing by t, it was found that

$$\frac{M \pm \sqrt{M}}{t} = \frac{Rt \pm \sqrt{Rt}}{t} \quad \text{or} \quad R \pm \sqrt{\frac{R}{t}} = 160 \pm 4 \text{ c.p.m.}$$

$$\Rightarrow \quad R = 160, \qquad \sqrt{\frac{R}{t}} = \sqrt{\frac{160}{t}} = 4, \quad \Rightarrow \quad t = 10\,\text{minutes.}$$

Alternatively, the fractional standard deviation of the rate is $\frac{4}{160} = 0.025$. If M counts were counted in total, it will be $\frac{\sqrt{M}}{M} = 0.025$ or $M = 40^2 = 1600$.

Since it is $R = 160$ c.p.m., it follows that $t = \frac{1600}{160} = 10$ min.

Problems

8.1 Two identical samples of a long-lived radioisotope are prepared at the same time. The first sample is monitored for 5 min and is found to give a total of $N_1 = 493$ counts. The second sample gives $N_2 = 1935$ counts in 20 min. (a) What are the counting rates R_1 and R_2 and their errors for the two samples? (b) By what factor is the fractional error in the counting rate decreased in increasing the counting time from 5 to 20 min?

8.2 A sample of a long-lived radioisotope emits an average of 10 α particles per hour.
   (a) What is the expected number of particles to be emitted in 10 min?
   (b) What is the probability that no particle will be emitted in a time interval of 10 min?

8.3 In an activation experiment, a sample is bombarded with neutrons. Immediately after, the activity of the sample is measured. During the first minute $n_1 = 256$ counts are recorded and during the second minute, $n_2 = 49$. Assuming that the counts are due to only one radioisotope and neglecting the background, find the decay constant λ of the isotope and its error, $\delta\lambda$.

8.4 Preliminary measurements of radioactivity gave for the background a counting rate approximately equal to $R_B = 1.0$ c.p.s. and for the radioactive source plus background a counting rate of $R_T = 4.0$ c.p.s. If we have at our disposal 9 min in which to measure these two magnitudes, (a) how must the time be divided between the two measurements for best results in the evaluation the net counting rate from the source, R_S? (b) approximately what will the standard deviation of R_S be?

8.5 In an experiment similar to that performed by C.S. Wu et al. in 1957 in order to prove the violation of parity in the decay of ^{60}Co, the spins of the nuclei of the radioisotope are aligned in a magnetic field, at low temperatures. Over a period of time, the number of photons emitted in the direction of the nuclear spins was measured to be $N_{+} = 363$ counts and the number of photons emitted in the opposite direction was measured to be $N_{-} = 561$ counts. What is the value of the polarization ratio $r = N_{-}/N_{+}$ and what is its error, δr ?

8.6 A radioactive sample contains three radioisotopes, A, B and C. The initial numbers of nuclei and the mean lifetimes of these isotopes are $N_{\mathrm{A}} = 1\,000\,000$, $N_{\mathrm{B}} = 600\,000$, $N_{\mathrm{C}} = 270\,000$ and $\tau_{\mathrm{A}} = 100$ s, $\tau_{\mathrm{B}} = 300$ s, $\tau_{\mathrm{C}} = 900$ s, respectively. Find the counting rates $R_{\mathrm{A}}(t)$, $R_{\mathrm{B}}(t)$ and $R_{\mathrm{C}}(t)$ due to the three isotopes, assuming that all the decays are counted. Plot, as a function of time, using a logarithmic R-scale, these counting rates and the total counting rate of the sample, $R(t) = R_{\mathrm{A}}(t) + R_{\mathrm{B}}(t) + R_{\mathrm{C}}(t)$.

Reference

1. See, for example, G. Friedlander, J.W. Kennedy, E.S. Macias and J.M. Miller, *Nuclear and Radiochemistry* (J. Wiley and Sons, New York, 3rd ed., 1981). Ch. 9