Applied Radiation and Isotopes 89 (2014) 58–64

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Uncertainty propagation in nuclear forensics

S. Pommé [a,*], S.M. Jerome [b], C. Venchiarutti [a]

[a] European Commission, Joint Research Centre, Institute for Reference Materials and Measurements, Retieseweg 111, B-2440 Geel, Belgium
[b] National Physical Laboratory, Teddington, Middlesex TW11 OLW, UK

HIGHLIGHTS

- Uncertainty propagation formulae for age dating with nuclear chronometers.
- Applied to parent–daughter pairs used in nuclear forensics.
- Investigated need for better half-life data.

ARTICLE INFO

Article history:
Received 29 November 2013
Received in revised form
31 January 2014
Accepted 6 February 2014
Available online 14 February 2014

Keywords:
Nuclear forensics
Uncertainty
Half-life
Decay data
Accuracy
Non-proliferation

ABSTRACT

Uncertainty propagation formulae are presented for age dating in support of nuclear forensics. The age of radioactive material in this context refers to the time elapsed since a particular radionuclide was chemically separated from its decay product(s). The decay of the parent radionuclide and ingrowth of the daughter nuclide are governed by statistical decay laws. Mathematical equations allow calculation of the age of specific nuclear material through the atom ratio between parent and daughter nuclides, or through the activity ratio provided that the daughter nuclide is also unstable. The derivation of the uncertainty formulae of the age may present some difficulty to the user community and so the exact solutions, some approximations, a graphical representation and their interpretation are presented in this work. Typical nuclides of interest are actinides in the context of non-proliferation commitments. The uncertainty analysis is applied to a set of important parent–daughter pairs and the need for more precise half-life data is examined.

1. Introduction

Age dating of radioactive material by means of radiometric or mass spectrometry measurements is a long-established technique in geological and archaeological sciences (Hamilton, 1965; Magill and Galy, 2005). It uses statistical decay laws to provide the link between the activity or atom concentration of radionuclides and their daughter nuclide(s) and the time elapsed since certain initial conditions. Nuclear forensics is a relatively young scientific discipline in which age dating is performed using the same principles. Here, the age refers to the time elapsed since a radionuclide of interest, usually an actinide isotope, was chemically separated from its decay products (Mayer et al., 2007; 2013). Such information taken with other evidence is crucial for identification of the sampled material (see e.g. Schwantes et al. (2009)). The precision by which the age of the material can be determined not only depends on the precision of the analytical measurement, but is also limited by the state of knowledge of the half-lives involved.

In this paper, exact mathematical formulae are presented of the uncertainty propagation factors involved with age dating in nuclear forensics, based on the measurement of either atom ratios or activity ratios. From the exact equations, approximate formulae are derived which are applicable under specified conditions. Graphs of the uncertainty propagation as a function of time for two hypothetical cases are presented and interpreted. The equations are used specifically to examine the constraints imposed on the attainable precision of the age due the current uncertainties on the nuclear half-lives of the parent–daughter pairs involved.

2. Age dating by atom ratio measurements

2.1. Exact formulae

Age dating is applied to a material containing a radionuclide and its decay products, assuming that no physicochemical processes other than radioactive decay have altered their relative

* Corresponding author. Tel.: +32 14 571 289; fax: +32 14 571 864.
E-mail address: stefaan.pomme@ec.europa.eu (S. Pommé).

http://dx.doi.org/10.1016/j.apradiso.2014.02.005

concentrations over time. The number of atoms of the parent and daughter nuclides, $P(t)$ and $D(t)$ respectively, obey statistical decay rules as a function of time:

$$P(t) = P(0)e^{-\lambda_P t} \tag{1}$$

$$D(t) = D(0)e^{-\lambda_D t} + P(0)\frac{\lambda_P}{\lambda_D - \lambda_P}(e^{-\lambda_P t} - e^{-\lambda_D t}) \tag{2}$$

in which the decay constants λ_P and λ_D are inversely proportional to the parent and daughter half-lives, $T_P = \ln(2)/\lambda_P$ and $T_D = \ln(2)/\lambda_D$, respectively.

The ratio of Eq. (2) and Eq. (1) gives the equation for the atom ratio of daughter and parent atoms at any time $t > 0$:

$$R(t) = \frac{D(t)}{P(t)} = R(0)e^{-(\lambda_D - \lambda_P)t} + \frac{\lambda_P}{\lambda_D - \lambda_P}(1 - e^{-(\lambda_D - \lambda_P)t}) \tag{3}$$

It is assumed that at time $t = 0$, the parent nuclei were separated completely from the daughter nuclei such that $R(0) = 0$, so that $D(t)$ can be fully ascribed to ingrowth after $t = 0$. Determining the 'age' of the material consists of calculating the most likely amount of time elapsed since separation, using measured values of the atom ratio, $\hat{R}(t)$, and literature values of the decay constants, $\hat{\lambda}_P$ and $\hat{\lambda}_D$:

$$\text{Age} = \frac{1}{\hat{\lambda}_P - \hat{\lambda}_D}\ln\left(1 - \hat{R}(t)\frac{\hat{\lambda}_D - \hat{\lambda}_P}{\hat{\lambda}_P}\right) \tag{4}$$

Linear propagation of uncertainty on the atom ratio $R(t)$ and the decay constants λ_P and λ_D results to:

$$\begin{aligned}\left(\frac{\sigma(t)}{t}\right)^2 = &\left(\frac{\lambda_D}{\lambda_P - \lambda_D}\left(\frac{T}{t} - \frac{\lambda_P}{\lambda_D}\right)\right)^2\left(\frac{\sigma(\lambda_P)}{\lambda_P}\right)^2 \\ &+ \left(\frac{-\lambda_D}{\lambda_P - \lambda_D}\left(\frac{T}{t} - 1\right)\right)^2\left(\frac{\sigma(\lambda_D)}{\lambda_D}\right)^2 \\ &+ \left(\frac{T}{t}\right)^2\left(\frac{\sigma(R)}{R}\right)^2\end{aligned} \tag{5}$$

in which the variable T is defined as

$$\frac{T}{t} = \frac{e^{(\lambda_D - \lambda_P)t} - 1}{(\lambda_D - \lambda_P)t} \tag{6}$$

and the relative uncertainties on the decay constants are equal to (minus) the relative uncertainties on the half-lives, i.e. $\sigma(\lambda)/\lambda = -\sigma(T_{1/2})/T_{1/2}$.

The uncertainty on R arises mainly from the uncertainty of the analytical measurement result, which may be expected to be smallest around $R = 1$ and to increase in approximate proportion to $|\log(R)|$ when parent and daughter concentrations differ by orders of magnitude. Another uncertainty component that should be taken into account is the possibility of an incomplete chemical separation at time $t = 0$ (Williams and Gaffney, 2011; Eppich et al., 2013). This would increase $R(t)$ by a relative amount $R(0)/R(t)e^{-(\lambda_D - \lambda_P)t} \neq 0$ (Eq. (3)), which also propagates by a factor T/t to the relative uncertainty of the age estimate (Eq. (5)).

2.2. Approximate equations

2.2.1. Atom ratio

Serial expansion of Eq. (3) shows that $R(t) \approx \lambda_P t$ if the age of the material is low compared to the half-lives of the nuclides involved ($|\lambda_D - \lambda_P|t < 1$). This means that the initial ingrowth of the daughter happens at the rate of the decay of the parent and is, to a first order approximation, independent of the half-life of the daughter. Further changes to $R(t)$ with time depend on which nuclide has the longest half-life. In the case of a *long-lived parent*, where $\lambda_P \lll \lambda_D$, the atom ratio tends towards the secular equilibrium value $R(t) \approx \lambda_P/\lambda_D$ for $\lambda_D t > 1$ and reaches about half that value after one half-life of the daughter nuclide, $R(T_D) \approx \lambda_P/2\lambda_D$. As a result, the expectation value of the daughter concentration remains smaller than that of the parent concentration. In the case of a *long-lived daughter*, where $\lambda_D \lll \lambda_P$, the atom ratio reaches unity after about one half-life of the parent, $R(T_P) \approx 1$, and then increases exponentially by $R(t) \approx e^{\lambda_P t}$ for $\lambda_P t > 1$.

2.2.2. Uncertainty propagation

The uncertainty propagation formulae in Eq. (5) can be stated as a series expansion, resulting in approximate formulae valid for $|\lambda_D - \lambda_P|t < 1$:

$$\begin{aligned}\left(\frac{\sigma(t)}{t}\right)^2 \approx &\left(-1 - \frac{\lambda_D t}{2} - \frac{\lambda_P(\lambda_D - \lambda_P)t^2}{6}\right)^2\left(\frac{\sigma(\lambda_P)}{\lambda_P}\right)^2 \\ &+ \left(\frac{\lambda_D t}{2} + \frac{\lambda_P(\lambda_D - \lambda_P)t^2}{6}\right)^2\left(\frac{\sigma(\lambda_D)}{\lambda_D}\right)^2 \\ &+ \left(1 + \frac{(\lambda_D - \lambda_P)t}{2} + \frac{(\lambda_D - \lambda_P)^2 t^2}{6}\right)^2\left(\frac{\sigma(R)}{R}\right)^2\end{aligned} \tag{7}$$

In Table 1, approximate equations are summarised for the propagation factors under boundary conditions. For material with a relatively young age compared to the half-lives involved, $\lambda_P t \lll 1$ and $\lambda_D t \lll 1$, the propagation factors for λ_P and R are unity, while the factor for λ_D is insignificantly small. Consequently, age determinations based on *atom ratios* are more sensitive to the *parent half-life* then to the daughter half-life. This is compatible with the fact that $R(t) \approx \lambda_P t$ for small values of t, i.e. linear with λ_P and independent of λ_D.

Only for old material, $|\lambda_D - \lambda_P|t \ggg 1$, does the uncertainty propagation of the daughter half-life become important, but under these conditions the over-all accuracy of the method is relatively poor, as the propagation factors increase almost exponentially with $\lambda_D t$.

2.3. Graphical representation

The propagation factors are presented for two hypothetical cases: a long-lived parent nuclide ($\lambda_P \lll \lambda_D$) in Fig. 1 and a long-lived daughter nuclide ($\lambda_D \lll \lambda_P$) in Fig. 2.

2.3.1. Long-lived parent

In the top graph of Fig. 1, there is no visible difference between the dominant propagation factors of λ_P and R. They remain close to unity as long as $\lambda_D t < 1$. For larger values of t, the propagation factors increase exponentially with $\lambda_D t$. The age dating method

Table 1
Exact and approximate equations for uncertainty propagation factors for age dating via atom ratio measurements in specific conditions. The variable T has been defined in Eq. (6). Exact equations are also presented as a function of the half-lives T_P and T_D instead of the decay factors λ_P and λ_D.

Condition	Age from atom ratio, using Eq. (3) Propagation factor		
	$\left\lvert\left(\frac{\partial t}{\partial \lambda_P}\right)\frac{\lambda_P}{t}\right\rvert$	$\left\lvert\left(\frac{\partial t}{\partial \lambda_D}\right)\frac{\lambda_D}{t}\right\rvert$	$\left\lvert\left(\frac{\partial t}{\partial R}\right)\frac{R}{t}\right\rvert$
Always valid	$\left\lvert\frac{\lambda_D}{\lambda_P - \lambda_D}\left(\frac{T}{t} - \frac{\lambda_P}{\lambda_D}\right)\right\rvert$	$\left\lvert\frac{-\lambda_D}{\lambda_P - \lambda_D}\left(\frac{T}{t} - 1\right)\right\rvert$	$\left\lvert\frac{T}{t}\right\rvert$
Always valid	$\left\lvert\frac{T_P}{T_D - T_P}\left(\frac{T}{t} - \frac{T_D}{T_P}\right)\right\rvert$	$\left\lvert\frac{-T_P}{T_D - T_P}\left(\frac{T}{t} - 1\right)\right\rvert$	$\left\lvert\frac{T}{t}\right\rvert$
$\underline{\lambda_P \ll \lambda_D}$	Long-lived parent		
$\lambda_D t \ll 1$	1	$\frac{\lambda_D t}{2}$	1
$\lambda_D t \gg 1$	$\frac{e^{\lambda_D t}}{\lambda_D t}$	$\frac{e^{\lambda_D t}}{\lambda_D t}$	$\frac{e^{\lambda_D t}}{\lambda_D t}$
$\underline{\lambda_P \gg \lambda_D}$	Long-lived daughter		
$\lambda_P t \ll 1$	1	$\frac{\lambda_D t}{2}$	1
$\lambda_P t \gg 1$	1	$\frac{\lambda_D}{\lambda_P}$	$\frac{1}{\lambda_P t}$