# New Concept For Calculating The Age Of A Fossil Bone By Uranium-Series Method

Samer Farkh, Abdallah Zaiour, Jalal Abdallah, Samir Farkh, Hanadi Ismail, Ahmad elMoll, Mirvat Ballout, Nafez Harb, Katia Haddad, Zaher Zeaiter

**Abstract:** The age of a fossil bone can be determined based on the chain of radioactive uranium, as far as it behaves like a closed system. Uranium is incorporated into the bone after its death and disintegrates into thorium. The calculation of age is based on the assumption that living tissue does not contain uranium and at the entrance of uranium in the dead tissue, it did not contain thorium. Uranium is soluble in water and easily enters the bone while the thorium adsorbs on the clay soil particles since it is insoluble in water. In this work, we determined the equations needed to calculate the age of a bone sample. We have shown that age cannot be determined from the activity ratio between uranium-234 and its source uranium-238 because ratio had to be 1 from 3.5 million years, before they both entered the bone. Thus, we used the activity ratio of thorium-230 and its source uranium-234 which allowed us to go backward up to 350 000 years. In the case where $^{234}U/^{238}U$ is greater than 1, an amount of exogenous $^{234}U$ should be deducted.

**Keywords**: bone, age, uranium, thorium, equilibrium, radioactive chain

## INTRODUCTION

A bone is composed of phosphates forming hdroxyapatite crystals $(Ca_5PO_4)_3OH$ and of calcium carbonate $CaCO_3$. This mineral matrix is intimately linked with an organic matrix named ossein formed mainly by collagen fibers. These two matrices give bone its rigidity and resistance to physical and chemical factors that can alter it. The dating of fossil bones started by Szabo et al. (1969), they are often the only materials available for chronological reconstruction of important archaeological sites. They also exhibit advantage over speleothems, they contain a high uranium content up to 1000 ppm (Szabo, 1980). However, since the bone can be an open system for uranium, it cannot be dated directly and it is therefore necessary to develop models for the incorporation of uranium. To calculate the correct age, a fossil bone must be well preserved. In other words, all of the uranium entered into the bone just after burial and in a very short time interval relative to the age of the bone. This time interval corresponds to that of the breakdown of the organic matter, estimated by 2700 years. This is the early incorporation model (EU) (Szabo, 1980). Indeed, the breakdown of the organic material allows the passage of uranium, under a soluble uranyl form, to its final position in the crystal matrix of hydroxyapatite. The application of this model on the dating of early modern humans Skhul, allowed a better estimate of the age between 100 and 135 kyears (Grün et al., 2005). This result is consistent with the TL ages obtained on flint burned 119 +/- 18 kyears (Mercier et al., 1993). However, another model called linear (LU) can affect bone where the entrance of uranium could be continuous and steady over time (Labeyrie and Lalou, 1981). This model has the effect of rejuvenating the age of the samples (Masaoudi et al., 1994).


- *Samer Farkh, Abdallah Zaiour, Hanadi Ismail, Mirvat Ballout, Zaher Zeaiter. Lebanese University (I), Beirut, Lebanon. E-mail: zeaiterzaher@yahoo.com*
- *Samir Farkh, Ahmad elMoll, Nafez Harb, Katia Haddad. Lebanese University (III), Tripoli, Lebanon.*
- *Jalal Abdallah. Academia SINICA, Institute of Physics, CERN, Switzerland.*


It is the same for the model (RU) statemed by Blackwell et al. (1992) where an amount of late uranium is added to the sample. However, in most cases, the incorporation of the uranium is between the two models (EU) and (LU). But since there are many exponential forms between these two models and because of differences found between age calculated by the U-Th method (Uranium Series) and by the ESR method (electronic spin resonance) on the same sample, Grün et al. (1988) have developed a general model that calculates an incorporation factor p of uranium by combining the U-Th and ESR data. This is the model series of uranium (US) but corrected by the ESR ages obtained on the same sample, named US-ESR. Indeed, the application of the combined US-ESR model, is an estimate of the minimum age (Michel et al., 2008). This model US-ESR has been successfully applied on a human mandible in Mala Balanica cave and was used to estimate his age between 397 and 525 kyears (Rink et al., 2013). Recently, a new radial diffusion model (RDA) uses the cylindrical geometry to describe the incorporation of uranium in fossil bones. Because bone is cylindrical, this model examines the incorporation of uranium around this cylinder. It showed that the spatial distribution of uranium in bone is not the same (Cid et al., 2014). It gives, therefore, a better understanding of poorly preserved fossil bones.

## Decay of uranium in the sample

The chain of radioactive decay of the uranium is represented as follows:

$$^{238}U \rightarrow {}^{234}Th \rightarrow {}^{238}Pa \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra \rightarrow {}^{222}Rn \rightarrow {}^{218}Po \rightarrow {}^{214}Pb \rightarrow \ldots {}^{206}Pb$$

Uranium-238, head of the chain, decays slowly (with a half-life of 4.47 billion years) to be transformed into uranium-234 (half-life of 248,000 years), which then decays into thorium -230 (half-life of 75,200 years). Unlike uranium, thorium is insoluble in water (Kaufman, 1969; Gascoyne, 1992). Living bone contains a negligible amount of uranium (Yokoyama et al, 1981; O'Meara et al., 1997). In contact with water, uranium is incorporated into the bone in a uranyl form $UO_2^{(2+)}$, then the uranium $U^{(4+)}$ is substituted for the calcium $Ca^{(2+)}$, because they have the same ionic radius 0.09 nm (Rae and Ivanovich, 1986). Similarly, stalactites and stalagmites are crystallized or even floors stalagmite formed by infiltration of water on the walls of the cave and in the archaeological layers. As uranium-

234 decays, thorium-230 accumulates and its content increases toward its secular equilibrium following an exponential law. Thus, the isotopic ratio of thorium over uranium provides a measurement of the elapsed time. Previously, scientists used the ratio $^{230}Th/^{234}U$ to calculate the age of a sample. Then it was reported in the literature that uranium-238 and uranium-234 are not in equilibrium when they enter the sample and the $^{234}U/^{238}U$ ratio is greater than 1 (Cherdyntsev et al. 1955; Thurber, 1962). Therefore, the ratio of $^{230}Th/^{234}U$ is currently is used for age calculation taking in account the excess of $^{234}U$ originated from the ratio of ($^{234}U/^{238}U$ >1). In fact, this excess should be large while the equilibrium is established between $^{234}U$ and $^{230}Th$, around 350,000 years, especially that $^{234}U$ has a period of 248,000 years which is very close to this equilibrium. We will first seek to calculate the amount of time needed for the equilibrium between $^{234}U$ and $^{238}U$ to be established, before they got incorporated into the system to be dated.

### Time of the equilibrium between $^{238}U$ and $^{234}U$ before they got incorporated into the sample

For Uranium $^{238}U$, head of the chain, and according to the decay law, we can state:

$$dN_{238} = -\lambda_{238}N_{238}dt$$

In fact, we note that the number of atoms which gives rise to radioactive phenomena (dN) is proportional to the time of observation (dt) and to the total number of radioactive atoms (N). The disintegration constant (λ) control the kinetics of the radioactive phenomenon for a given type of isotope. The (-) sign indicates that the number (dN) decreases from the total number (N). We have:

$$\mathbf{dN}/\mathbf{N_{238}} = -\lambda_{238}\mathbf{dt} \Rightarrow \int_{N_0}^{N} \mathbf{dN}/\mathbf{N_{238}} = -\int_{t_0}^{t} \lambda_{238}\mathbf{dt}$$

$$\Rightarrow \mathbf{Ln}\left(\mathbf{N_{238}}/\mathbf{N_{0238}}\right) = -\lambda_{238}\mathbf{t} \Rightarrow \mathbf{N_{238}}/\mathbf{N_{0238}} = \mathbf{e}^{-\lambda_{238}\mathbf{t}}$$

$$\Rightarrow \boxed{\mathbf{N_{238}} = \mathbf{N_{0238}}\mathbf{e}^{-\lambda_{238}\mathbf{t}}}$$

This is an exponential formula which provides that for an initial number $N_0$ of radioactive nuclei (or atoms), it remains N after a time t. The exponent of e (Napierian number ≈ 2.72) is negative, so this exponential law is decreasing. For Uranium $^{234}U$, descendant of uranium $^{238}U$, the decay equation is as follows:

$$dN_{234} = -\lambda_{234}N_{234}dt + \lambda_{238}N_{238}dt$$

or: $dN_{234} + \lambda_{234}N_{234}dt = \lambda_{238}N_{238}dt$ **(1)**

To solve the equation (1), we must first solve the equation without a second member:

$$dN_{234} + \lambda_{234}N_{234}dt = 0 \Rightarrow dN_{234} = -\lambda_{234}N_{234}dt$$

$$\Rightarrow \mathbf{N}_{234} = \mathbf{N_0}_{234}\, e^{-\lambda_{234}t}$$

Then it must be assumed that $\mathbf{N_0}_{234}$ is a function of **t**:

$N_{234} = X(t)e^{-\lambda_{234}t} \Rightarrow dN_{234} = dX(t)e^{-\lambda_{234}t} - \lambda_{234}e^{-\lambda_{234}t}X(t)dt$ **(2)**

$$\Rightarrow dN_{234} = dX(t)e^{-\lambda_{234}t} - \lambda_{234}N_{234}dt$$

$$\Rightarrow dN_{234} + \lambda_{234}N_{234}dt = dX(t)e^{-\lambda_{234}t}$$

When compared with (1), it could be written:

$$dX(t)e^{-\lambda_{234}t} = \lambda_{238}N_{238}dt = \lambda_{238}\mathbf{N_0}_{238}e^{-\lambda_{238}t}dt$$

This implies that:

$$dX(t) = \lambda_{238}\mathbf{N_0}_{238}e^{-\lambda_{238}t}e^{\lambda_{234}t}dt = \lambda_{238}\mathbf{N_0}_{238}e^{-(\lambda_{238}t-\lambda_{234}t)}dt$$

By integration:

$$\int_{X_0}^{X} \mathbf{dX(t)} = \int_{t_0}^{t} \lambda_{238}\mathbf{N_{0238}}\mathbf{e}^{-(\lambda_{238}-\lambda_{234})\mathbf{t}}\mathbf{dt}$$

So:

$$\mathbf{X} - \mathbf{X_0} = \frac{-\lambda_{238}\mathbf{N_{0238}}}{\lambda_{238}-\lambda_{234}}\mathbf{e}^{-(\lambda_{238}-\lambda_{234})\mathbf{t}} + \frac{\lambda_{238}\mathbf{N_{0238}}}{\lambda_{238}-\lambda_{234}}$$

So X =

$$\mathbf{X} = \frac{-\lambda_{238}\mathbf{N_{0238}}}{\lambda_{238}-\lambda_{234}}\left[\mathbf{e}^{-(\lambda_{238}-\lambda_{234})\mathbf{t}} - \mathbf{1}\right] + \mathbf{X_0}$$

$$= \frac{\lambda_{238}\mathbf{N_{0238}}}{\lambda_{238}-\lambda_{234}}\left[\mathbf{1} - \mathbf{e}^{-(\lambda_{238}-\lambda_{234})\mathbf{t}}\right] + \mathbf{X_0}$$

By replacing X by its value in (2) we obtain:

$$N_{234} = \frac{\lambda_{238}N_{0238}}{\lambda_{238}-\lambda_{234}}\left[1 - e^{-(\lambda_{238}-\lambda_{234})t}\right]e^{-\lambda_{234}t} + X_0e^{-\lambda_{234}t}$$

But at t = 0, $N_{234} = 0 = X_0$ so:

$$N_{234} = \frac{\lambda_{238}N_{0238}}{\lambda_{238}-\lambda_{234}}\left[1 - e^{-(\lambda_{238}-\lambda_{234})t}\right]e^{-\lambda_{234}t}$$

$$N_{234} = \frac{\lambda_{238}N_{0238}}{\lambda_{238}-\lambda_{234}}e^{-\lambda_{234}t} - \frac{\lambda_{238}N_{0238}}{\lambda_{238}-\lambda_{234}}e^{-\lambda_{238}t}$$

$$\boxed{N_{234} = \frac{\lambda_{238}N_{0238}}{\lambda_{238}-\lambda_{234}}\left(e^{-\lambda_{234}t} - e^{-\lambda_{238}t}\right)}$$

At the radioactive equilibrium, the activity of $^{238}U$ is equal to that of $^{234}U$: $\lambda_{238}N_{238} = \lambda_{234}N_{234}$

$$\Rightarrow \lambda_{238}N_{238} = \lambda_{234}\frac{\lambda_{238}N_{0238}}{\lambda_{238}-\lambda_{234}}\left(e^{-\lambda_{234}t} - e^{-\lambda_{238}t}\right)$$

$$\Rightarrow \lambda_{238}N_{0238}e^{-\lambda_{238}t} =$$

$$\lambda_{234}\frac{\lambda_{238}N_{0238}}{\lambda_{238}-\lambda_{234}}e^{-\lambda_{234}t}-\lambda_{234}\frac{\lambda_{238}N_{0238}}{\lambda_{238}-\lambda_{234}}e^{-\lambda_{238}t}$$

$$\Rightarrow e^{-\lambda_{238}t}=\lambda_{234}\frac{e^{-\lambda_{234}t}}{\lambda_{238}-\lambda_{234}}-\lambda_{234}\frac{e^{-\lambda_{238}t}}{\lambda_{238}-\lambda_{234}}$$

$$\Rightarrow e^{-\lambda_{238}t}+\lambda_{234}\frac{e^{-\lambda_{238}t}}{\lambda_{238}-\lambda_{234}}=\lambda_{234}\frac{e^{-\lambda_{234}t}}{\lambda_{238}-\lambda_{234}}$$

$$\Rightarrow\frac{\lambda_{238}e^{-\lambda_{238}t}-\lambda_{234}e^{-\lambda_{238}t}+\lambda_{234}e^{-\lambda_{238}t}}{\lambda_{238}-\lambda_{234}}=\lambda_{234}\frac{e^{-\lambda_{234}t}}{\lambda_{238}-\lambda_{234}}$$

$$\Rightarrow\lambda_{238}e^{-\lambda_{238}t}=\lambda_{234}e^{-\lambda_{234}t}$$

$$\Rightarrow e^{-(\lambda_{238}-\lambda_{234})t}=\frac{\lambda_{234}}{\lambda_{238}}$$

$$\Rightarrow-(\lambda_{238}-\lambda_{234})t=\mathrm{Ln}\left(\frac{\lambda_{234}}{\lambda_{238}}\right)\Rightarrow t=\frac{\mathrm{Ln}\left(\frac{\lambda_{234}}{\lambda_{238}}\right)}{-(\lambda_{238}-\lambda_{234})}$$

$$\lambda_{238}=\frac{\mathrm{Ln}2}{T_{238}}\quad\text{and}\quad\lambda_{234}=\frac{\mathrm{Ln}2}{T_{234}}$$

But:

So:

$$\boxed{t=\frac{\mathrm{Ln}\left(\frac{T_{238}}{T_{234}}\right)}{-\left(\frac{\mathrm{Ln}2}{T_{238}}-\frac{\mathrm{Ln}2}{T_{234}}\right)}}$$

We have $T_{238}$ = 4.47 $10^9$ years and $T_{234}$ = 248000 years. These 2 elements have already been in equilibrium for a long time, which could be calculated from the equation **t ≈ 3.500.000 years**. We can determine the activity ratio at the equilibrium time:

The activity of $^{234}$U at a time t = 3.500.000 years.

$$^{234}U=\lambda_{234}N_{234}=\lambda_{234}\frac{\lambda_{238}N_{0238}}{\lambda_{238}-\lambda_{234}}\left(e^{-\lambda_{234}t}-e^{-\lambda_{238}t}\right)$$

The activity of $^{238}$U at a time t = 3.500.000 years.

$$^{238}U=\lambda_{238}N_{238}=\lambda_{238}N_{0238}e^{-\lambda_{238}t}$$

$$R=\frac{^{234}U}{^{238}U}=\frac{\lambda_{234}\frac{\lambda_{238}N_{0238}}{\lambda_{238}-\lambda_{234}}\left(e^{-\lambda_{234}t}-e^{-\lambda_{238}t}\right)}{\lambda_{238}N_{0238}e^{-\lambda_{238}t}}$$

$$=\frac{\lambda_{234}}{\lambda_{238}-\lambda_{234}}\frac{\left(e^{-\lambda_{234}t}-e^{-\lambda_{238}t}\right)}{e^{-\lambda_{238}t}}=1$$

So the activity ratio between $^{234}$U and $^{238}$U should be 1 before they enter into the bone. Therefore, the age cannot be calculated from the ratio between the two. However, in nature this report is generally greater than 1. Several authors explained the reason for the imbalance between $^{234}$U and $^{238}$U. Cherdyntsev et al. (1961) considered that the alpha emissions from $^{238}$U bring the $^{234}$U out from its stable position in the rocks. Uranium-234 is easily carried away by runoff waters that reach the buried bones. This alpha recoil hypothesis was confirmed by Rasilainen et al. (2005). Rosholt et al. (1963) considered that $^{234}$U is more soluble in water than $^{238}$U. The latter hypothesis seems more likely. Indeed, if the alpha particles emitted by the $^{238}$U move the $^{234}$U from its position, then the alpha particles emitted by the $^{234}$U will also move the $^{230}$Th, which will distort calculated ages by the U-Th method, i.e. it casts doubt on the principle of the method.

It should be noted here:
i) in nature, this ratio can be greater than 1, it is necessary to take account of excess uranium-234 derived from uranium-238 in the sample.
ii) when this ratio is greater than 1, the balance is restored after some time in the sample.

**Restoring equilibrium between $^{234}$U and $^{238}$U**

$$\frac{^{234}U}{^{238}U}=\frac{^{238}U_0-{}^{238}U_0e^{-\lambda_{234}t}+{}^{234}U_0e^{-\lambda_{234}t}}{^{238}U_0e^{-\lambda_{238}t}}$$

$$\Rightarrow R=\frac{^{238}U_0\left(1-e^{-\lambda_{234}t}+e^{-\lambda_{234}t}.\,{}^{234}U_0\big/{}^{238}U_0\right)}{^{238}U_0e^{-\lambda_{238}t}}$$

$$=\frac{\left(1-e^{-\lambda_{234}t}+e^{-\lambda_{234}t}.\,{}^{234}U_0\big/{}^{238}U_0\right)}{e^{-\lambda_{238}t}}$$

$$R=\frac{1-e^{-\lambda_{234}t}}{e^{-\lambda_{238}t}}+R_0\frac{e^{-\lambda_{234}t}}{e^{-\lambda_{238}t}}$$

So:

But in approximation $e^{-\lambda_{238}t}=1$, so: $\mathbf{R = 1 - e^{-\lambda_{234}t} + R_0e^{-\lambda_{234}t}}$

So: $\mathbf{R-1 = (R_0-1)\,e^{-\lambda_{234}t}}$ **(A)**

**R** is the activity ratio at time **t** between $^{234}$U and $^{238}$U and $\mathbf{R_0}$ is the activity ratio, initially >1, between $^{234}$U and $^{238}$U. In a closed system, if we had initially a ratio $R_0$ = $(^{234}U/^{238}U)_0$ >1, and as time elapses, this ratio tends toward 1.

**Calculation of the activity of $^{234}$U in the sample**

As already stated above: $dN_{238}=-\lambda_{238}N_{238}dt$

$$dN\big/N_{238}=-\lambda_{238}dt\Rightarrow\int_{N_0}^{N}dN\big/N_{238}$$

$$= -\int^{t} \lambda_{238} dt \Rightarrow Ln\left( {N_{238}}/{N_{0238}} \right) = -\lambda_{238} t$$

$$\Rightarrow {N_{238}}/{N_{0238}} = e^{-\lambda_{238} t} \Rightarrow N_{238} = N_{0238} e^{-\lambda_{238} t}$$

$dN_{234} = -\lambda_{234} N_{234} dt + \lambda_{238} N_{238} dt$

or: $$dN_{234} + \lambda_{234} N_{234} dt = \lambda_{238} N_{238} dt \quad (1)$$

To solve the equation (1), we must first solve the equation without a second member.

$$dN_{234} + \lambda_{234} N_{234} dt = 0$$

$$\Rightarrow dN_{234} = -\lambda_{234} N_{234} dt \Rightarrow \boxed{N_{234} = N_{0234} e^{-\lambda_{234} t}}$$

$$N_{234} = X(t) e^{-\lambda_{234} t} \quad 2)$$

$$\Rightarrow dN_{234} = dX(t) e^{-\lambda_{234} t} - \lambda_{234} e^{-\lambda_{234} t} X(t) dt$$

$$\Rightarrow dN_{234} = dX(t) e^{-\lambda_{234} t} - \lambda_{234} N_{234} dt$$

$$\Rightarrow dN_{234} + \lambda_{234} N_{234} dt = dX(t) e^{-\lambda_{234} t}$$

When compared to (1) we can write:

$$dX(t) e^{-\lambda_{234} t} = \lambda_{238} N_{238} dt = \lambda_{238} N_{0238} e^{-\lambda_{238} t} dt$$

This implies that:

$$dX(t) = \lambda_{238} N_{0238} e^{-\lambda_{238} t} e^{\lambda_{234} t} dt$$
$$= \lambda_{238} N_{0238} e^{-(\lambda_{238} t - \lambda_{234} t)} dt$$

By integration:

$$\int_{X_0}^{X} dX(t) = \int_{t_0}^{t} \lambda_{238} N_{0238} e^{-(\lambda_{238} - \lambda_{234}) t} dt$$

So:

$$X - X_0 = \frac{-\lambda_{238} N_{0238}}{\lambda_{238} - \lambda_{234}} e^{-(\lambda_{238} - \lambda_{234}) t} + \frac{\lambda_{238} N_{0238}}{\lambda_{238} - \lambda_{234}}$$

Thus:

$$X = \frac{-\lambda_{238} N_{0238}}{\lambda_{238} - \lambda_{234}} \left[ e^{-(\lambda_{238} - \lambda_{234}) t} - 1 \right] + X_0$$
$$= \frac{\lambda_{238} N_{0238}}{\lambda_{238} - \lambda_{234}} \left[ 1 - e^{-(\lambda_{238} - \lambda_{234}) t} \right] + X_0$$

When replacing $X$ by its value in (2) we get:

$$N_{234} = \frac{\lambda_{238} N_{0238}}{\lambda_{238} - \lambda_{234}} \left[ 1 - e^{-(\lambda_{238} - \lambda_{234}) t} \right] e^{-\lambda_{234} t} + X_0 e^{-\lambda_{234} t}$$

But at t = 0, $N_{234} = X_0 e^{-\lambda_{234} t}$

So: $X_0 = N_{0234}$.

Thus:

$$N_{234} = \frac{\lambda_{238} N_{0238}}{\lambda_{238} - \lambda_{234}} \left[ 1 - e^{-(\lambda_{238} - \lambda_{234}) t} \right] e^{-\lambda_{234} t} + N_{0234} e^{-\lambda_{234} t}$$

$$N_{234} = \frac{\lambda_{238} N_{0238}}{\lambda_{238} - \lambda_{234}} e^{-\lambda_{234} t} - \frac{\lambda_{238} N_{0238}}{\lambda_{238} - \lambda_{234}} e^{-\lambda_{238} t} + N_{0234} e^{-\lambda_{234} t}$$

$$N_{234} = \frac{\lambda_{238} N_{0238}}{\lambda_{238} - \lambda_{234}} \left( e^{-\lambda_{234} t} - e^{-\lambda_{238} t} \right) + N_{0234} e^{-\lambda_{234} t}$$

But:

$$T_{238} \gg T_{234} \Rightarrow \frac{Ln2}{T_{238}} \ll \frac{Ln2}{T_{234}} \Rightarrow \lambda_{238} \ll \lambda_{234},$$

in approximation $e^{-\lambda_{238} t} = 1$

Thus:

$$N_{234} = \frac{\lambda_{238} N_{0238}}{\lambda_{234}} \left( 1 - e^{-\lambda_{234} t} \right) + N_{0234} e^{-\lambda_{234} t} \quad (3)$$

To obtain the activity of $^{234}U$ we multiply by $\lambda_{234}$. So :

$$\lambda_{234} N_{234} = \lambda_{238} N_{0238} \left( 1 - e^{-\lambda_{234} t} \right) + \lambda_{234} N_{0234} e^{-\lambda_{234} t}$$

$$\Rightarrow {}^{234}U = {}^{238}U_0 \left( 1 - e^{-\lambda_{234} t} \right) + {}^{234}U_0 e^{-\lambda_{234} t}$$

$$\Rightarrow {}^{234}U = {}^{238}U_0 - {}^{238}U_0 e^{-\lambda_{234} t} + {}^{234}U_0 e^{-\lambda_{234} t}$$

**Calculating the activity of $^{230}$Th in the sample**

At first we calculate the number of atoms of $^{230}$Th, $N_{230}$:

Or: $$dN_{230} = -\lambda_{230} N_{230} dt + \lambda_{234} N_{234} dt$$

$$dN_{230} + \lambda_{230} N_{230} dt = \lambda_{234} N_{234} dt \quad (4)$$

First, the solution for the equation

$$dN_{230} + \lambda_{230} N_{230} dt = 0 \text{ is } N_{230} = N_0 e^{-\lambda_{230} t}.$$

Second, we assume that $N_0$ is a function of *t*:

$$N_{230} = Y(t) e^{-\lambda_{230} t} \quad \textbf{(5)}$$

$$\Rightarrow dN_{230} = dY(t) e^{-\lambda_{230} t} - \lambda_{230} e^{-\lambda_{230} t} Y(t) dt$$

$$\Rightarrow dN_{230} = dY(t) e^{-\lambda_{230} t} - \lambda_{230} N_{230} dt$$

$$\Rightarrow dN_{230} + \lambda_{230} N_{230} dt = dY(t) e^{-\lambda_{230} t}$$

When compared with (4), we can write:

$$dY(t) e^{-\lambda_{230} t} = \lambda_{234} N_{234} dt \quad (6)$$

We replace $N_{230}$ in (3) by its value in (6):

$$dY(t) e^{-\lambda_{230} t} = \lambda_{234} \left[ \frac{\lambda_{238} N_{0238}}{\lambda_{234}} \left( 1 - e^{-\lambda_{234} t} \right) + N_{0234} e^{-\lambda_{234} t} \right] dt$$
$$= \left[ \lambda_{238} N_{0238} \left( 1 - e^{-\lambda_{234} t} \right) + \lambda_{234} N_{0234} e^{-\lambda_{234} t} \right] dt$$

So: **dY(t)** =

$$\left[\frac{\lambda_{238}N_{0238}\left(1-e^{-\lambda_{234}t}\right)}{e^{-\lambda_{230}t}}+\frac{\lambda_{234}N_{0234}e^{-\lambda_{234}t}}{e^{-\lambda_{230}t}}\right]dt$$

$$=\left[\lambda_{238}N_{0238}\left(e^{\lambda_{230}t}-e^{-(\lambda_{234}-\lambda_{230})t}\right)+\lambda_{234}N_{0234}e^{-(\lambda_{234}-\lambda_{230})t}\right]dt$$

By integration:

$$\int_{Y_0}^{Y} dY(t) =$$

$$\int_{t_0}^{t}\left[\lambda_{238}N_{0238}\left(e^{\lambda_{230}t}-e^{-(\lambda_{234}-\lambda_{230})t}\right)dt+\lambda_{234}N_{0234}e^{-(\lambda_{234}-\lambda_{230})t}dt\right]$$

So: **Y-Y$_0$** =

$$(\lambda_{238}N_{0238})\left(\frac{e^{\lambda_{230}t}}{\lambda_{230}}-\frac{e^{-(\lambda_{234}-\lambda_{230})t}}{\lambda_{230}-\lambda_{234}}\right)+\lambda_{234}N_{0234}\frac{e^{-(\lambda_{234}-\lambda_{230})t}}{\lambda_{230}-\lambda_{234}}$$

$$-\left[\lambda_{238}N_{0238}\left(\frac{1}{\lambda_{230}}-\frac{1}{\lambda_{230}-\lambda_{234}}\right)+\frac{\lambda_{234}N_{0234}}{\lambda_{230}-\lambda_{234}}\right]$$

But at t = 0, $^{230}N_0 = 0$ so $Y_0 = 0$.

By replacing $Y$ by its value in (5) we get:

$$N_{230}=e^{-\lambda_{230}t}\lambda_{238}N_{0238}\left(\frac{e^{\lambda_{230}t}}{\lambda_{230}}-\frac{e^{-(\lambda_{234}-\lambda_{230})t}}{\lambda_{230}-\lambda_{234}}\right)+e^{-\lambda_{230}t}\lambda_{234}N_{0234}\frac{e^{-(\lambda_{234}-\lambda_{230})t}}{\lambda_{230}-\lambda_{234}}$$

$$-e^{-\lambda_{230}t}\left[\lambda_{238}N_{0238}\left(\frac{1}{\lambda_{230}}-\frac{1}{\lambda_{230}-\lambda_{234}}\right)+\frac{\lambda_{234}N_{0234}}{\lambda_{230}-\lambda_{234}}\right]$$

$$N_{230}=\lambda_{238}N_{0238}\left(\frac{1}{\lambda_{230}}-\frac{e^{-\lambda_{234}t}}{\lambda_{230}-\lambda_{234}}\right)+\lambda_{234}N_{0234}\frac{e^{-\lambda_{234}t}}{\lambda_{230}-\lambda_{234}}$$

$$-\lambda_{238}N_{0238}\left(\frac{e^{-\lambda_{230}t}}{\lambda_{230}}-\frac{e^{-\lambda_{230}t}}{\lambda_{230}-\lambda_{234}}\right)-\lambda_{234}N_{0234}\frac{e^{-\lambda_{230}t}}{\lambda_{230}-\lambda_{234}}$$

To obtain the activity of $^{230}$Th we multiply by $\lambda_{230}$:

$$\lambda_{230}N_{230}=$$

$$\lambda_{238}N_{0238}\left(1-\lambda_{230}\frac{e^{-\lambda_{234}t}}{\lambda_{230}-\lambda_{234}}\right)+\lambda_{234}N_{0234}\lambda_{230}\frac{e^{-\lambda_{234}t}}{\lambda_{230}-\lambda_{234}}$$

$$-\lambda_{238}N_{0238}\left(e^{-\lambda_{230}t}-\lambda_{230}\frac{e^{-\lambda_{230}t}}{\lambda_{230}-\lambda_{234}}\right)-\lambda_{234}N_{0234}\lambda_{230}\frac{e^{-\lambda_{230}t}}{\lambda_{230}-\lambda_{234}}$$

**$^{230}$Th =**

$$^{238}U_0\left(1-e^{-\lambda_{234}t}\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\right)+{}^{234}U_0e^{-\lambda_{234}t}\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}$$

$$-{}^{238}U_0\left(e^{-\lambda_{230}t}-e^{-\lambda_{230}t}\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\right)-$$

$$^{234}U_0e^{-\lambda_{230}t}\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}$$

$^{230}$Th is the activity of $^{230}$Th at time *t*, $^{238}U_0$ is the initial activity of $^{238}$U, $^{234}U_0$ is the initial activity of $^{234}$U.

**$^{230}$Th =**

$$^{238}U_0-{}^{238}U_0e^{-\lambda_{234}t}\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}+{}^{234}U_0e^{-\lambda_{234}t}\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}$$

$$-{}^{238}U_0e^{-\lambda_{230}t}+{}^{238}U_0e^{-\lambda_{230}t}\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}-{}^{234}U_0e^{-\lambda_{230}t}\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}$$

**$^{230}$Th =**

$$\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\left({}^{234}U_0e^{-\lambda_{234}t}-{}^{238}U_0e^{-\lambda_{234}t}+{}^{238}U_0e^{-\lambda_{230}t}-{}^{234}U_0e^{-\lambda_{230}t}\right)$$

$$+{}^{238}U_0\left(1-e^{-\lambda_{230}t}\right)$$

The equation of $^{230}$Th is composed of 2 parts, let us process each one separately:

**First**: $$^{238}U_0\left(1-e^{-\lambda_{230}t}\right)\quad(7)$$

We already demonstrated that:

$$N_{238}=N_{0238}e^{-\lambda_{238}t}\Rightarrow\lambda_{238}N_{238}=\lambda_{238}N_{0238}e^{-\lambda_{238}t}$$

$$\Rightarrow{}^{238}U={}^{238}U_0e^{-\lambda_{238}t}$$

But in approximation $e^{-\lambda_{238}t}=1$ so $^{238}U={}^{238}U_0$.
We replace $^{238}U_0$ by $^{23}$[illegible] $^{234}$U, we get:

$$^{238}U.\frac{^{234}U}{^{234}U}\left(1-e^{-\lambda_{230}t}\right)=\frac{^{234}U\left(1-e^{-\lambda_{230}t}\right)}{^{234}U/_{238U}}$$

**Second**:

$$\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\left({}^{234}U_0e^{-\lambda_{234}t}-{}^{238}U_0e^{-\lambda_{234}t}+{}^{238}U_0e^{-\lambda_{230}t}-{}^{234}U_0e^{-\lambda_{230}t}\right)\quad(8)$$

We already demonstrated that:

$$^{234}U={}^{238}U_0-{}^{238}U_0e^{-\lambda_{234}t}+{}^{234}U_0e^{-\lambda_{234}t}\qquad(9)$$

$$\Rightarrow{}^{238}U_0-{}^{234}U={}^{238}U_0e^{-\lambda_{234}t}-{}^{234}U_0e^{-\lambda_{234}t}$$

$$=\left({}^{238}U_0-{}^{234}U_0\right)e^{-\lambda_{234}t}$$

But with respect to (8):

$$\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\left({}^{234}U_0e^{-\lambda_{234}t}-{}^{238}U_0e^{-\lambda_{234}t}+{}^{238}U_0e^{-\lambda_{230}t}-{}^{234}U_0e^{-\lambda_{230}t}\right)$$

$$= \frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\left[{}^{234}U - {}^{238}U_0 + \left({}^{238}U_0 - {}^{234}U_0\right)e^{-\lambda_{230}t}\frac{e^{-\lambda_{234}t}}{e^{-\lambda_{234}t}}\right]$$

$$= \frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\left[{}^{234}U - {}^{238}U_0 + \left({}^{238}U_0 - {}^{234}U_0\right)e^{-\lambda_{234}t}e^{-(\lambda_{230}t-\lambda_{234}t)}\right]$$

$$= \frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\left[{}^{234}U - {}^{238}U_0 + \left({}^{238}U_0 - {}^{234}U\right)e^{-(\lambda_{230}t-\lambda_{234}t)}\right]$$

$$= \frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\left({}^{234}U - {}^{238}U_0\right)\left(1 - e^{-(\lambda_{230}t-\lambda_{234}t)}\right)$$

$$= \frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\,{}^{234}U\left(1 - \frac{{}^{238}U_0}{{}^{234}U}\right)\left(1 - e^{-(\lambda_{230}t-\lambda_{234}t)}\right)$$

$$= \frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\,{}^{234}U\left(1 - \frac{1}{{}^{234}U/{}^{238}U}\right)\left(1 - e^{-(\lambda_{230}t-\lambda_{234}t)}\right)$$

$$= {}^{234}U\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\left(1 - \frac{1}{{}^{234}U/{}^{238}U}\right)\left(1 - e^{-(\lambda_{230}t-\lambda_{234}t)}\right)$$

**Age equation**

$$^{230}Th = {}^{234}U\frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\left(1 - \frac{1}{{}^{234}U/{}^{238}U}\right)\left(1 - e^{-(\lambda_{230}t-\lambda_{234}t)}\right) + \frac{{}^{234}U\left(1 - e^{-\lambda_{230}t}\right)}{{}^{234}U/{}^{238}U}$$

Or:

$$^{230}Th/{}^{234}U = \frac{\lambda_{230}}{\lambda_{230}-\lambda_{234}}\left(1 - \frac{1}{{}^{234}U/{}^{238}U}\right)\left(1 - e^{-(\lambda_{230}t-\lambda_{234}t)}\right) + \frac{\left(1 - e^{-\lambda_{230}t}\right)}{{}^{234}U/{}^{238}U} \quad (B)$$

This equation is the activity ratio of $^{230}Th/^{234}U$ depending on the activity ratio of $^{234}U/^{238}U$, (this ratio is initially >1).
$\lambda_{230}$, $\lambda_{234}$ are radioactive constants or probabilities of disintegration of $^{230}Th$ and $^{234}U$, respectively.
Both ratios are determined by the alpha detector, time t which is the age of the sample is calculated.

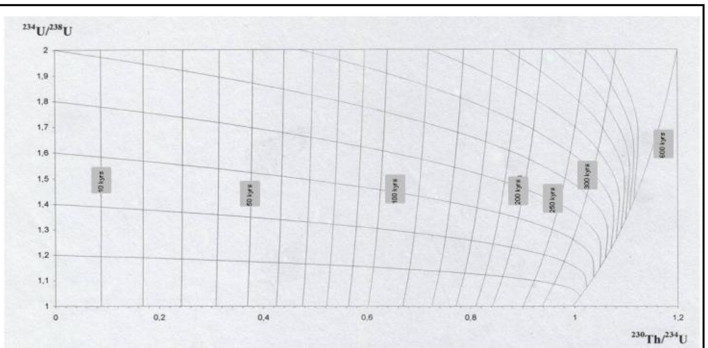


***Fig 1.*** *Resolution of equations (A) & (B). Variation of the $^{230}Th/^{234}U$ in function of that of $^{234}U/^{238}U$ represented by the vertical lines. We note here that the $^{230}Th/^{234}U$ ratio is independent of that of $^{234}U/^{238}$ up to 30 kyrs.*

If the ratio $^{234}U/^{238}U$ is = 1 (which is not the case), we should have calculated the age by a simplified equation derived from (B), and it will then be:

$$^{230}Th/{}^{234}U = \left(1 - e^{-\lambda_{230}t}\right)$$

## CONCLUSION

The uranium-series (US) method is based on a well-known chemical principle. The age of the sample is determined through mathematically confirmed calculation steps. The fossil bones, although they may behave as an open system, but they are valuable materials for dating. Indeed, their high uranium content reduces the error of chemical handling and increases the detecting of alpha emissions by spectrometry. When $^{234}U$ and $^{238}U$ enters the bone in a variable ratio, we are then forced to move from a simple equation to a complicated one to calculate the age. Beside, our understanding of the geological history of the bone burying becomes more difficult especially when uranium enters into the bone in a continuous manner. Here, the choice of the uranium incorporation model is important and it is advisable to make a US-ESR combination for the resulting age.

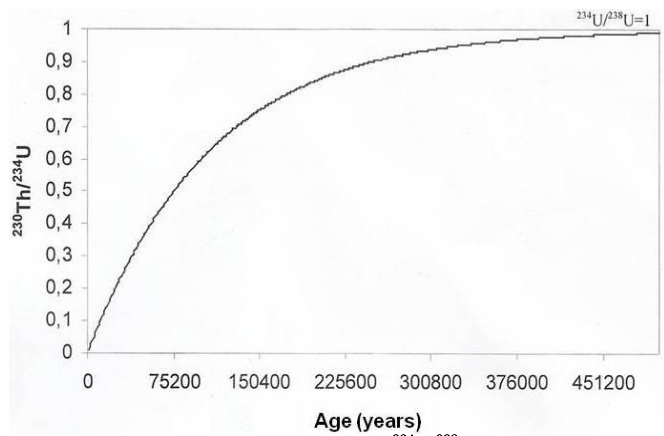


***Fig 2.*** *Resolution of equation (C), when $^{234}U/^{238}U$ = 1. Variation of the $^{234}U/^{238}U$ ratio over time, i.e. the restoration of balance between $^{234}U$ and $^{238}U$ represented by horizontal curves (see equation (A)).*

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