EFFECT OF THE RATIO Th/U ON TL DATING ACCURACY

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ABSTRACT

In routine thermoluminescence (TL) dating, there is often a tendency to omit an absolute determination of the thorium and uranium content in the sample and simply measure the total alpha count rate. The annual dose is then calculated assuming equal activities for a Th/U weight ratio of about 3.17. In fact this ratio is varied significantly in pottery. Calculation of the annual dose conversion factors from data using the total alpha count rate in a selected range of Th/U weight ratios shows significant differences. The results indicate that the additional error contribution to the final TL age is in the range ±2.5% to ±4.1% for most cases where the Th/U ratio lies between 1.1 and 9.5.

Keywords TL dating, Thick source alpha counting, Th/U ratio, Error analysis

1 INTRODUCTION

For the determinating the annual alpha dose in pottery, thick source alpha counting is a simple method with a number of advantages compared with other more sophisticated methods, such as gamma spectrometry, neutron activation analysis and fission track analysis. However the method entails a complication in the conversion from α, β and γ annual dose[1]. For the annual β dose, the dose rate of unit specific activity for the uranium chain is higher than that for the thorium chain. However for the annual γ dose the reverse is true. Therefore error will be introduced in the estimated annual β and γ doses if they are calculated from the α count rate without considering the exact Th/U content ratio.

Theoretically the "slow pair" (or "fast pair") technique[2] may be used to separate the Th (or U) α count rate from the total α count rate. Unfortunately due to the very low value of the typical "pair" rate, the technique has rather low usefulness in practice. For example a typical pottery sample with a specific activity 41 Bq/kg would have a "pair" rate of only 0.2 cpks. As a result ten additional weeks of counting time is required to get a result with ±3 % statistic error.

In routine work therefore, only the total α count rate is obtained and the annual dose rate is calculated under an assumption of equal specific activity of Th and U, *i.e.*

Manuscript received date: 1993-11-25

a Th/U weight ratio of 3.17 (in fact the assumption is usually for an equal α count rate for Th and U, but this only introduces a factor difference of 1.073). In fact much pottery commonly has a weight ratio of about 3. However, were the ratio to vary over a rather larger range, the annual dose and thus the age estimate would be subjected to significant error. This paper analyses the effect of the Th/U ratio on the annual dose and hence the age estimate.

2 DOSE RATE CONVERSION FACTOR

To study the variation of dose rate under the influence of arbitrary Th/U ratios, conversion factors for annual α, β and γ dose have been calculated in terms of the total α count rate covering the entire range of possible Th/U ratios (*i.e.* $0 \leq \mathrm{Th/U} \leq \infty$).

Table 1

Annual dose conversion factor

Th/ppm	U/ppm	Ratio Th/U		$\dot{\alpha}$/cpks	Annual dose conversion factor /mGy·a^{-1}· (cpks)$^{-1}$		
		weight	activity		F_α	F_β	F_γ
0.50	0.00	∞	∞	0.2525	1.480	0.0574	0.1029
1.00	0.02	50	15.769	0.5394	1.490	0.0591	0.1008
3.00	0.10	30	9.462	1.6867	1.496	0.0601	0.0995
10.0	0.50	20	6.380	5.9081	1.502	0.0613	0.0981
10.0	1.00	10	3.154	6.7658	1.519	0.0642	0.0944
8.00	1.00	8.0	2.523	5.7557	1.527	0.0654	0.0929
4.00	0.80	5.0	1.577	3.3924	1.544	0.0684	0.0892
12.0	4.00	3.0	0.946	12.921	1.564	0.0719	0.0847
6.00	3.00	2.0	0.631	8.1761	1.581	0.0748	0.0812
2.00	2.00	1.0	0.315	4.4406	1.605	0.0789	0.0760
1.20	1.50	0.8	0.252	3.1790	1.611	0.0800	0.0746
3.00	5.00	0.6	0.189	10.091	1.618	0.0812	0.0731
1.00	2.50	0.4	0.126	4.7932	1.626	0.0825	0.0715
0.20	1.00	0.2	0.063	1.8163	1.634	0.0840	0.0696
0.30	3.00	0.1	0.032	5.2973	1.639	0.0848	0.0686
0.00	0.70	0.0	0.00	1.2007	1.644	0.0857	0.0675

This is conditioned by the long equilibrium period for the Th and U chains.

Once the Th/U content ratio is selected and combined with ppm overall weight ratios, the Th/U activity ratio can be estimated using the equivalences: 1 ppm = 13 Bq/kg for the Th chain and 1 ppm = 4.1 Bq/kg for the U chain. The total α count rate $\dot{\alpha}$ is then given by[1]

$$\begin{aligned} \dot{\alpha} = \dot{\alpha}_{\mathrm{Th}} + \dot{\alpha}_{\mathrm{U}} &= (C_{\mathrm{Th}}/1.98 + C_{\mathrm{U}}/0.583)_{\mathrm{weight}} \\ &= (0.123C_{\mathrm{Th}} + 0.132C_{\mathrm{U}})_{\mathrm{activity}} \end{aligned} \tag{1}$$

where $\dot{\alpha}_{\mathrm{Th}}$ and $\dot{\alpha}_{\mathrm{U}}$ are the respective α count rates (cpks), $(C_{\mathrm{Th}}$ and $C_{\mathrm{U}})_{\mathrm{weight}}$ represents the weight concentration (ppm) and $(C_{\mathrm{Th}}$ and $C_{\mathrm{U}})_{\mathrm{activity}}$ represents the specific parent activity (Bq/kg), in relation to the Th and U chains.

For the annual α dose of Th only or U only[1]:

$D_{\alpha Th} = 1.480\dot{\alpha}_{Th}$; $D_{\alpha U} = 1.644\dot{\alpha}_U$; with the total annual dose given by:

$D_\alpha = D_{\alpha Th} + D_{\alpha U} = 1.480\dot{\alpha}_{Th} + 1.644\dot{\alpha}_U$ (Note that $\dot{\alpha} = \dot{\alpha}_{Th} + \dot{\alpha}_U$)

A conversion factor F_α may be defined for the annual α dose

$$F_\alpha = [1.480R_{Th/U} + 1.644]/(R_{Th/U} + 1) \quad (2)$$

where $D_\alpha = F_\alpha\dot{\alpha}$; $R_{Th/U}$ is the ratio $\dot{\alpha}_{Th}/\dot{\alpha}_U$.

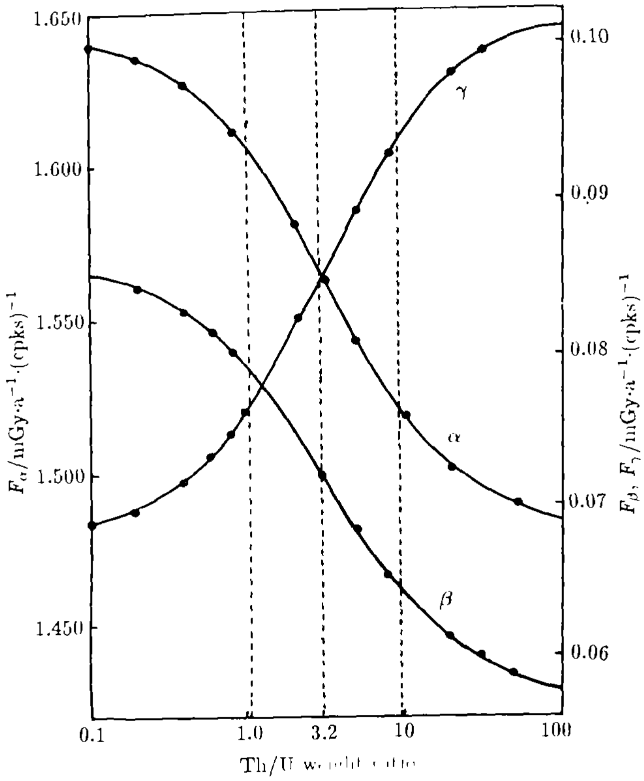

Fig.1 Variation of F_α, F_β and F_γ with Th/U weight ratio

In practice the specific activity ratio is a more useful parameter for expressing the content variation, so $R_{Th/U}$ is in fact taken to be C_{Th}/C_U, the activity content ratio (this is quite close to $\dot{\alpha}_{Th}/\dot{\alpha}_U$ with a conversion factor of about 1.073).

The same concept may be used to define conversion factors F_β and F_γ, for the annual β dose,

$$F_\beta = (0.0574R_{Th/U} + 0.0857)/(R_{Th/U} + 1) \quad \text{and} \quad D_\beta = F_\beta\dot{\alpha} \quad (3)$$

while for the annual γ dose,

$$F_\gamma = (0.1029R_{Th/U} + 0.0675)/(R_{Th/U} + 1) \quad \text{and} \quad D_\gamma = F_\gamma\dot{\alpha} \quad (4)$$

where the factor pairs 0.0574 and 0.0857, 0.1029 and 0.0675 respectively for β and γ conversions only[1].

All calculated conversion factors F_α, F_β and F_γ are shown in Table 1, where the weight content ratio of Th and U is varied from 0 to ∞.

For example taking $C_{\rm Th} = 8.00\,{\rm ppm}$, $C_{\rm U} = 1.00\,{\rm ppm}$, gives $\dot{\alpha} = (8.00/1.98) + (1.00/0.583) = 5.756$ (cpks) and

$$F_\alpha = \frac{1.480 \times (8 \times 4.1/13) + 1.644}{(8 \times 4.1/13) + 1} = 1.527\,({\rm mGy \cdot a^{-1}/cpks})$$

$$F_\beta = 0.0654\,({\rm mGy \cdot a^{-1}/cpks}) \qquad F_\gamma = 0.0929\,({\rm mGy \cdot a^{-1}/cpks})$$

Fig.1 shows the variation of the three conversion factors F_α, F_β and F_γ for a weight ratio range from 0.1 to 100. Outside this range all three curves appear to reach limiting values. The central value of the Th/U weight ratio is taken as 3.2 (*e.g.* 1 ppm activity ratio 13/4.1) which is the value normally chosen in pottery dating.

3 UNCERTAINTY OF CONVERSION FACTOR

At the central value (Th/U weight ratio 3.2) the total annual dose in terms of the total α count rate only is given by $D_\alpha = F_\alpha \dot{\alpha}$

where $F_\alpha = (1.48 + 1.644)/2 = 1.562\,({\rm mGy \cdot a^{-1}/cpks})$

the standard variation of F_α should be

$$\sigma_\alpha = \sqrt{\frac{\sum_{i=1}^{n}(x_i - \bar{x})^2}{n(n-1)}} = \sqrt{\frac{(1.480 - 1.562)^2 + (1.644 - 1.562)^2}{2}} = 0.0820$$

then $D_\alpha = 1.562\dot{\alpha} \pm 0.0820\dot{\alpha}(\pm 5.2\%)$ (mGy/a).

The same procedure at the central value for the annual β and γ dose gives:

$$\left.\begin{aligned} F_\beta &= (0.0574 + 0.0857)/2 = 0.0716\ ({\rm mGy \cdot a^{-1}/cpks}) \\ \sigma_\beta &= \sqrt{\frac{(0.074 - 0.0716)^2 + (0.0857 - 0.0716)^2}{2}} = 0.0142 \\ D_\beta &= 0.0716\dot{\alpha} \pm 0.0142\dot{\alpha}(\pm 19.8\%)\ ({\rm mGy/a}) \end{aligned}\right\} \tag{6}$$

$$\left.\begin{aligned} F_\gamma &= (0.1029 + 0.0675)/2 = 0.0852\ ({\rm mGy \cdot a^{-1}/cpks}) \\ \sigma_\gamma &= \sqrt{\frac{(0.1029 - 0.0852)^2 + (0.067 - 0.0852)^2}{2}} = 0.0177 \\ D_\gamma &= 0.0852\dot{\alpha} \pm 0.0177\dot{\alpha}\,(\pm 20.8\%)\ ({\rm mGy/a}) \end{aligned}\right\} \tag{7}$$

It can be seen that the maximum errors of the annual α, β and γ dose are $\pm 5.2\%$, $\pm 19.8\%$ and $\pm 20.8\%$ respectively. Now this refers to two extreme cases (pottery containing Th without U or U without Th) which never in fact occur.

Nevertheless, while it would be more accurate to be able to select appropriate conversion factors for the actually occuring Th/U ratios, in practice it might be more convenient to estimate the relative standard error when selecting the central conversion factor instead of measuring that actually occuring in the archaeological pottery samples. Although it would then be unrealistic to consider the extreme limit case given above. a reasonable range might be taken as a half that of the extreme limiting case. The six conversion factors respectively:

$$\left.\begin{aligned}
F_{\alpha\min} &= (1.562 + 1.480)/2 = 1.521\,(\mathrm{mGy\cdot a^{-1}/cpks}) \\
F_{\alpha\max} &= (1.562 + 1.644)/2 = 1.603\,(\mathrm{mGy\cdot a^{-1}/cpks}) \\
F_{\beta\min} &= 0.0645\,(\mathrm{mGy\cdot a^{-1}/cpks}) \\
F_{\beta\max} &= 0.0787\,(\mathrm{mGy\cdot a^{-1}/cpks}) \\
F_{\gamma\min} &= 0.0764\,(\mathrm{mGy\cdot a^{-1}/cpks}) \\
F_{\gamma\max} &= 0.0941\,(\mathrm{mGy\cdot a^{-1}/cpks})
\end{aligned}\right\} \tag{8}$$

The corresponding Th/U weight ratio range is now 1.1 to 9.5.

Then assuming a weight ratio of 3.2, the calculated annual dose obtained from the total α count rate and the maximum associated error is given by:

$$D_\alpha = 1.562\dot{\alpha} \pm 0.041\dot{\alpha}\,(\pm 2.6\,\%)\ (\mathrm{mGy/a}) \tag{9}$$

$$D_\beta = 0.0716\dot{\alpha} \pm 0.0071\dot{\alpha}(\pm 9.9\,\%)\ (\mathrm{mGy/a}) \tag{10}$$

$$D_\gamma = 0.0852\dot{\alpha} \pm 0.0085\dot{\alpha}(\pm 10\,\%)\ (\mathrm{mGy/a}) \tag{11}$$

The uncertainties are larger than those given by Aitken *et al*[3], which are

$$\left.\begin{aligned}
D_\alpha &= 146.0C_\alpha \pm 0.7\,\%\ (\mathrm{mrad/a}) \\
D_\beta &= 6.2C_\alpha \pm 3.9\,\%\ (\mathrm{mrad/a}) \\
D_\gamma &= 8.8C_\alpha \pm 6.8\,\%\ (\mathrm{mrad/a})
\end{aligned}\right\} \tag{12}$$

where C_α is the total α count rate of the thick sample (cpks) with a sample diameter of ϕ 42 mm and a count efficiency of 85 %.

They are however rather closer to those given by R. Sasidheran *et al*[4]:

$$\left.\begin{aligned}
D_\alpha &= 500.6C'_\alpha \pm 1.7\,\%\,(\mathrm{mrad/a}) \\
D_\beta &= 23.0C'_\alpha \pm 9.2\,\%\,(\mathrm{mrad/a}) \\
D_\gamma &= 27.5C'_\alpha \pm 11.3\,\%\,(\mathrm{mrad/a})
\end{aligned}\right\} \tag{13}$$

where C'_α is the total count rate per hour per cm^2.

In practice most pottery and soil samples will have Th/U ratios within 1.1 to 9.5 range, so that equations (9)–(11) can give satisfactory error estimates.

4 UNCERTAINTY IN TL DATING

Having obtained equations (9)–(11) to estimate the annual dose and associated uncertainty, it is now possible to inquire how much uncertainty is introduced into the age obtained from TL dating. The fine-grain technique and the quartz inclusion technique are analysed respectively.

4.1 Fine-grain technique

In the fine-grain technique, the effective annual α dose from the Th chain represents 21 % of the total annual dose, and that from the U chain represents 24 % (supposing α relative effectiveness K=0.15)[2], giving a combined 45 %. The annual β dose from the Th chain provides 6 % of total with 8 % coming from the U chain (14 % combined). The annual γ dose from Th chain contributes 10 % to the total, with 7 % coming from the U chain (17 % combined)[2].

From equations (9)–(11), the total uncertainty of the annual dose (used in the relationship: age = equivalent dose/annual dose) is:

$$E_D = [(0.026 \times 0.45)^2 + (0.099 \times 0.14)^2 + (0.10 \times 0.17)^2]^{1/2} = 2.5\,\%$$

If the entire Th/U ratio range (0 to ∞) is considered, E_D becomes about 5.1 %. Comparing either of these values to the uncertainty normally associated with the equivalent dose of around 10 % shows their effect on the age determination to be small.

4.2 Quartz inclusion technique

In quartz inclusion technique, the annual β dose from the Th chain contributes 10 % of the total dose, with 16 % coming from the U chain. The annual γ dose from Th chain contibutes 19 % of the total, with 12% coming from the U chain[2].

Again the total uncertainty in the annual dose will be:

$$E'_D = [(0.099 \times 0.26)^2 + (0.10 \times 0.31)^2]^{1/2} = 4.1\%$$

for the limited Th/U ratio range (8.3% for the 0 to ∞ range).

5 SUMMARY

Generally, during TL dating of pottery and soil samples, the total α count rate is measured rather than the "pair rate", and a "central value" of 3.2 is assumed for the Th/U weight ratio in order to calculate the annual α, β and γ dose. In this case a maximum possible error of ±5.1 % is introduced for the fine grain technique and ±8.3 % for the quartz inclusion technique ($0 \leq$ Th/U $\leq \infty$). In fact in most practical situations the Th/U ratio lies in the range $1.1 \leq$ Th/U ≤ 9.5, so that the maximum possible errors introduced by the "central value" assumption become only ±2.5% and ±4.1% respectively.

ACKNOWLEDGEMENT

The work was financially supported by City Polytechnic of Hong Kong through a Strategic Research Grant.

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