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Comparison between fine-grain and ultrathin TLD in the measurement of alpha dose-rate

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Introduction

In our laboratory of TL dating, the palaeodose and the annual dose-rate are measured by fine-grain samples and ultrathin thermoluminescence dosimeters (TLD) respectively (Wang, 1983a, 1983b). The determination of internal dose-rates due to alpha and beta radiation in pottery using ultrathin TLD is quite convenient (Wang, 1983c), although TLD does not have the same geometry as the fine grains. Just as Aitken (1985) pointed out, since the thickness of the phosphor layer is about 10 μm, it might be expected that there would be significant attenuation of alpha dose-rate. Hence the question is whether the measured values of the alpha dose-rate are in good agreement with those using fine grains. If the two results are different, a correction may need to be made to the alpha dose-rate measured using ultrathin TLD. We have made a special study of this problem, the results from which are reported in this paper. Measurements of alpha and beta dose-rate using ultrathin TLD has been reported (Wang, 1983c).Here we describe the method of measurement of alpha dose-rate using fine-grain TLD.

Experimental

Sample preparation

Mix $CaSO_4$:Tm grains with the powdered sherd; their grain sizes should be the same, in the range 20-150 μm. The mixture is ground further in a ball mill, producing a limited number of fine-grains. The samples used in the study and laced in this manner are given in table 1.

Experimental Method

The laced samples and several ultrathin TLD were annealed for 60 mins at 400 °C, after which the ultrathin TLD were put into a glass container with the laced sample according to Wang's method (1983c). Hence the (beta-equivalent) annual alpha dose, D_α, and beta plus gamma annual dose, $D_{\gamma+\beta}$, are obtained by two equations,

$$D_\alpha = \frac{2\,(D_a - D_b)}{t} \qquad (1)$$

$$D_{\beta+\gamma} = \frac{D_b}{t} \qquad (2)$$

where,

D_a and D_b are respectively the average dose of type a and b dosemeters, t is the storage time in years, and γ denotes the environmental background (for further details, see Wang, 1983c) † .

After withdrawal of the ultra-thin TLD, the fine grain discs were made from the laced sample. The fine-grain samples on the discs contain both $CaSO_4$:Tm and pottery powder. However the TL contribution from the pottery powder is negligible because its sensitivity is very much lower than the $CaSO_4$:Tm. The annual equivalent beta dose of the disc sample, D_F, is given by

$$D_F = \frac{D_f}{t} \qquad (3)$$

where D_f is the equivalent beta dose for the disc sample. The (beta equivalent) annual alpha dose for the disc sample, D'_α is also obtained by the equation

$$D'_\alpha = D_F - D_{\beta+\gamma} \qquad (4)$$

In equation (4) we assume that the measurement of $D_{\beta+\gamma}$ is reliable using the ultrathin TLD method. If $D_\alpha = D'_\alpha$, we consider that the ultrathin TLD method is also reliable in measurement of alpha dose-rate. If not, a correction must be made for the ultrathin TLD method. It should be noted that all the dose-rates measured using the two methods are standardized to an identical laboratory beta source. Hence the calibration error of the beta source is not important.

Results and discussion

Tables 2.1 and 2.2 give the comparison between fine-grain and ultrathin TLD of measured of alpha dose-rate. Tables 2.1 and 2.2 show that the ultrathin TLD is 8.5 ± 4.9% less efficient in measurement of alpha dose-rate than the fine-grain TLD method when using 2 mg cm^{-2}, and is 2.8 ± 5.2% less when using 1 mg cm^{-2}. A comparison between the neutron activation analysis (NAA) and ultrathin TLD is given in tables 3 and 5 (laced samples) and tables 4 and 6 (ancient pottery and bricks).

The alpha dose-rate given represents the beta-equivalent dose-rate, and k is the ratio of alpha to beta sensitivity; k = 0.29 (±3%) for 1 mg cm^{-2} and 0.27

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†*Ed. note.*
From Wang's 1982 paper, the construction of each type of dosemeter is as follows:
Type a - phosphor deposited onto 8 mm diameter, 10 μm thick aluminium foil to a thickness of 2 mg cm^{-2}.
Type b - as type a with a layer of foil covering the phosphor surface ie completely shielded from alpha particles.

(±3%) for 2 mg cm^{-2}. Tables 3 and 5 show that as long as different values of k are used, the ultrathin TLD of different thicknesses give the same alpha dose-rate. In fact, the k value acts as a compensator for thickness. The k-value was calibrated by using New Brunswick Laboratory samples (No.109 and No.105) containing respectively, 0.01% of thorium series and 0.001% of uranium series in silica, provided by the Oxford Laboratory and calculated according to

$$k = \frac{2\,(D_a - D_b)}{(t\,D_{\alpha_o})} \qquad (5)$$

where D_{α_o} is the alpha dose-rate for NBL samples according to their specified contents of thorium and uranium.

Tables 3 and 4 show that the ultrathin TLD results are in reasonable agreement with NAA; conversely, the alpha dose-rate is obviously decreased if the k-value of 0.29 is used for a TLD thickness of 2 mg cm^{-2}. The measurement of beta dose-rate is also shown in table 4.

Even so, the ultrathin results still have a tendency to be less than the NAA results. The difference arises from two factors:

1. The NAA method involves the assumption of equilibrium of long-lived radionuclides. In fact, the equilibrium cannot be assumed for younger clay sediments. The TLD method measures the dose-rate for the state of disequilibrium under present conditions; therefore, the TLD method is more reliable than the NAA method.
2. As mentioned above, using TLD of 1 mg cm^{-2} the measured alpha dose-rate was 2.8% less than with NAA; perhaps it is one of the reasons giving rise to the differences between the two methods. Nonetheless, the experimental results show that ultrathin TLD enables measurement of alpha dose-rate, providing the thickness of phosphor on the aluminium foil is less than the least alpha particle range. The phosphor thickness for the ultrathin TLD of 1 mg cm^{-2} is about a few microns, and it can be penetrated by nearly all alpha particles. But for the ultrathin TLD of thickness 2 mg cm^{-2}, a correction is necessary; however the correction is included in the k-value.

If the aim is only to date, it is unnecessary to make excessive demands concerning the size of the phosphor grains. Because the age equals the ratio of palaeodose to annual dose (providing the size of fine grains of the archaeological sample and the phosphor are the same) the palaeodose and the annual dose suffer the same attenuation and there is no influence on the determined age.

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PR. Reviewer's comments (M. J. Aitken)

This note follows on from the pottery dating procedure described by Wang and Zhou (1983), a procedure which is evidently in routine use at the Shanghai Museum. Because the annual dose is assessed by phosphor dosimetry it has the strong advantage that the dates obtained do not depend on radioactive source calibration, at any rate to first order. The present note investigates the question of how thin the ultrathin phosphor film must be in order to avoid significant attenuation of alpha particles; it is good news that for a thickness of 1 mg cm^{-2} there is no significant attenuation (to within ± 5% experimental error).

Table 1. Composition of experimental samples

Lab. No.	Source of sherds	$CaSO_4$:Tm content, %
10	Songze site, Qingpu County, Shanghai	30
11	Fuquanshan site, Qingpu County, Shanghai	40
12	Fuquanshan site, Qingpu County, Shanghai	45
14	Tinglin site, Jinshan County, Shanghai	50

Table 2.1 Comparison of alpha dose-rate measurements obtained using fine-grain and ultrathin (2 mg cm^{-2}) TLD methods

No.	Alpha dose-rate (beta equivalent) D'_α fine-grain	Alpha dose-rate (beta equivalent) D_α ultrathin	$\frac{D'_\alpha - D_\alpha}{D'_\alpha}$
	mGy/a	mGy/a	%
10	5.44 ± 0.05	4.82 ± 0.27	+11.0 ± 4.9
	5.06 ± 0.05	4.69 ± 0.29	+7.3 ± 5.8
	5.33 ± 0.08	5.02 ± 0.20	+5.8 ± 4.0
	5.25 ± 0.06	4.77 ± 0.20	+9.1 ± 4.0
Average	5.27 ± 0.06	4.83 ± 0.24	+8.3 ± 4.7
11	5.09 ± 0.07	4.69 ± 0.18	+7.9 ± 3.8
	4.79 ± 0.09	4.48 ± 0.17	+6.5 ± 4.0
	4.96 ± 0.07	4.32 ± 0.32	+11.3 ± 6.7
	5.29 ± 0.12	4.81 ± 0.19	+9.1 ± 4.3
Average	5.03 ± 0.09	4.58 ± 0.22	+8.9 ± 4.7
12	3.99 ± 0.09	3.55 ± 0.21	+11.0 ± 5.7
	3.62 ± 0.05	3.28 ± 0.14	+9.4 ± 4.1
	4.03 ± 0.12	3.81 ± 0.23	+5.5 ± 6.5
	4.30 ± 0.08	4.02 ± 0.18	+6.5 ± 4.6
	4.10 ± 0.05		
	3.78 ± 0.03		
Average	3.97 ± 0.07	3.67 ± 0.19	+7.6 ± 5.1
14	3.97 ± 0.06	3.74 ± 0.13	+5.8 ± 3.6
	4.17 ± 0.10	3.82 ± 0.21	+8.4 ± 5.6
	3.73 ± 0.05	3.44 ± 0.17	+7.8 ± 4.8
	3.80 ± 0.06	3.29 ± 0.22	+13.0 ± 5.8
	3.68 ± 0.07		
	4.15 ± 0.09		
	4.01 ± 0.11		
Average	3.93 ± 0.08	3.57 ± 0.18	+9.2 ± 5.0
Average for 4 groups = + 8.5 ± 4.9 %			

Table 2.2 Comparison of alpha dose-rate measurements obtained using fine-grain and ultrathin (1 mg cm^{-2}) TLD methods.

No.	Alpha dose-rate (beta equivalent) D'_α fine-grain	Alpha dose-rate (beta equivalent) D_α ultrathin	$\frac{D'_\alpha - D_\alpha}{D'_\alpha}$
	mGy/a	mGy/a	%
10		4.96 ± 0.20	+5.9 ± 4.0
		5.04 ± 0.26	+4.4 ± 5.1
		5.31 ± 0.32	-0.76 ± 6.0
Average	5.27 ± 0.06	5.10 ± 0.26	+3.2 ± 5.0
11		4.93 ± 0.19	+2.0 ± 4.2
		4.61 ± 0.27	+8.3 ± 5.6
Average	5.03 ± 0.09	4.77 ± 0.23	+5.2 ± 4.9
12		3.74 ± 0.25	+5.8 ± 6.5
		4.08 ± 0.19	-2.8 ± 5.2
		3.88 ± 0.12	+2.3 ± 3.6
		4.22 ± 0.21	-6.3 ± 5.6
Average	3.97 ± 0.07	3.98 ± 0.19	-0.25 ± 5.1
14		3.77 ± 0.15	+4.1 ± 4.4
		3.82 ± 0.18	+2.9 ± 5.2
		4.10 ± 0.27	-4.3 ± 7.1
		3.53 ± 0.23	+1.0 ± 6.1
Average	3.93 ± 0.08	3.81 ± 0.21	+3.1 ± 5.8
Average for 4 groups = + 2.5 ± 5.2 %			

Table 3. Comparison of fine grain, ultrathin TLD and NAA methods for measurement of alpha dose-rate.

No.	Fine-grain k =0.29	Ultrathin TLD 1 $mg.cm^{-2}$ k=0.29	2 $mg.cm^{-2}$ k=0.27	2 $mg.cm^{-2}$ k=0.29	NAA
10	18.17 ± 0.21	17.59 ± 0.90	17.89 ± 0.89	16.66 ± 0.83	17.56 ± 0.67
11	17.34 ± 0.31	16.45 ± 0.79	16.96 ± 0.81	15.79 ± 0.76	18.71 ± 0.69
12	13.69 ± 0.24	13.72 ± 0.65	13.59 ± 0.70	12.66 ± 0.66	15.44 ± 0.56
14	13.55 ± 0.28	13.14 ± 0.72	13.22 ± 0.67	12.31 ± 0.62	14.01 ± 0.49

Alpha dose-rate, mGy a^{-1}

Table 4. Comparison; measurement of alpha dose-rate in ancient pottery and bricks using ultrathin TLD and NAA methods.

No.	Alpha dose-rate Ultrathin TLD 2 $mg.cm^{-2}$	NAA	Beta dose-rate Ultrathin TLD 2 $mg.cm^{-2}$	NAA
SB 185	22.65 ± 1.31	23.11 ± 0.79	2.20 ± 0.05	2.46 ± 0.09
SB 186	23.10 ± 1.55	24.39 ± 0.95	2.77 ± 0.11	2.51 ± 0.09
SB 191	15.98 ± 1.11	17.51 ± 0.58	2.56 ± 0.13	2.44 ± 0.09
SB 192	18.35 ± 1.10	18.10 ± 0.58	2.49 ± 0.07	2.33 ± 0.08
SB 193	22.23 ± 1.18	21.50 ± 0.75	2.89 ± 0.05	3.28 ± 0.11
SB 203	17.95 ± 1.08	18.23 ± 0.71	2.31 ± 0.04	2.51 ± 0.10
SB 204	17.68 ± 1.29	18.37 ± 0.65	2.34 ± 0.06	2.12 ± 0.07
SB 205	14.49 ± 1.88	17.68 ± 0.60	2.15 ± 0.21	2.32 ± 0.09
SB 206	27.42 ± 1.51	29.89 ± 1.02	2.32 ± 0.06	2.13 ± 0.10
SB 207	20.15 ± 0.99	19.64 ± 0.69	3.28 ± 0.08	3.06 ± 0.11

mGy a^{-1}

Table 5. The ratio of TLD/NAA for alpha dose-rate measurement

No.	Fine-grain k_{TLD}=0.29	Ultrathin TLD 1 $mg.cm^{-2}$ k_{TLD}=0.29	2 $mg.cm^{-2}$ k_{TLD}=0.27	2 $mg.cm^{-2}$ k_{TLD}=0.29
10	1.03 ± 0.04	1.00 ± 0.06	1.02 ± 0.06	0.95 ± 0.06
11	0.93 ± 0.04	0.88 ± 0.05	0.91 ± 0.05	0.84 ± 0.05
12	0.89 ± 0.04	0.89 ± 0.05	0.88 ± 0.06	0.82 ± 0.05
14	0.97 ± 0.04	0.94 ± 0.06	0.94 ± 0.06	0.88 ± 0.05
Average	0.96 ± 0.04	0.93 ± 0.06	0.94 ± 0.06	0.87 ± 0.05

Table 6. The ratio of TLD/NAA measurement of alpha and beta dose-rate in archaeological samples

No.	TLD/NAA (Ultrathin TLD: 2 mg cm^{-2}, k_{TLD} = 0.27) Alpha dose-rate	Beta dose-rate
SB 185	0.98 ± 0.07	0.89 ± 0.04
SB 186	0.95 ± 0.07	1.10 ± 0.06
SB 191	0.91 ± 0.07	1.05 ± 0.07
SB 192	1.01 ± 0.07	1.07 ± 0.05
SB 193	1.03 ± 0.07	0.88 ± 0.03
SB 203	0.98 ± 0.07	0.92 ± 0.04
SB 204	0.96 ± 0.08	1.10 ± 0.05
SB 205	0.82 ± 0.11	0.93 ± 0.10
SB 206	0.92 ± 0.06	1.09 ± 0.05
SB 207	1.03 ± 0.06	1.07 ± 0.05
Average	0.96 ± 0.07	1.01 ± 0.05