ALPHA THERMOLUMINESCENCE DOSIMETRY IN DATING OF POTTERY

Wang Weida(王维达) and Xia Junding(夏君定)

(*Shanghai Museum, Shanghai 200231*)

ABSTRACT

This article describes the measurement of internal alpha dose-rate in pottery using ultrathin $CaSO_4$: Tm thermoluminescence dosimeter. Among the advantages of the technique are not only convenience, accuracy, lowcost, but also the beta dose-rate from pottery can be obtained at the same time.

Keywords Pottery, Thermoluminescence dating, Annual alpha dose, Ultrathin thermoluminescence dosimeter

1 INTRODUCTION

The key to successful application of thermoluminescence (TL) dating is to estimate as accurately as possible the annual dose in the pottery body. The determination of internal beta dose-rate using TL dosimeters (TLD) has been reported, but the measurement of internal alpha dose-rate in pottery using TLD still is very difficult although Aitken attempted to solve this problem early in 1968[1]. This article describes the measurement of internal alpha dose-rate using ultrathin $CaSO_4$: Tm TL dosimeters and discusses the relation of K-value (effectiveness of alpha in inducing TL relative to that of beta and gamma) versus the $CaSO_4$ thickness. As a result, the alpha dosimetry procedure would be simplified, the determination errors decreased and the cost of experiment reduced as well. In addition, the beta dose-rate in the pottery can also be obtained at the same time.

2 PREPARATION OF ULTRATHIN TLD

The preparation method of ultrathin $CaSO_4$: Tm dosimeters is about the same way as the fine-grain sample of pottery[2]. $CaSO_4$: Tm phosphor is crushed by ballmill, and the grains in the size of less than 60 micron are selected by sieving. Put the selected grains into a beaker, pour distilled water into the beaker until the height is 70 mm. After stirring, the beaker is kept still for 10 min, so that the grains in the size of greater than 8 μm deposit at the bottom of the beaker. Slowly pour the suspension into other beaker. This is then kept still for 60 min; at the end of this period grains having a size of less than 3 μm are still suspension. Finally, this suspension is tipped into another beaker thereby obtaining grains in the size of 3 to 8 microns. A flat-bottomed funnel with a stopcock at exit is used for the preparation of ultrathin TLD. A copper wire ring with a handle

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is placed within the funnel at the bottom; a large piece of glass is placed on this ring with a 10 μm thick aluminium foil wrapped round this glass. The separated fine-grain $CaSO_4$: Tm in the beaker are resuspended in distilled water and then the suspension is poured into the funnel. After all the fine-grain $CaSO_4$: Tm have been deposited onto the aluminium foil the valve is opened and the water is drained off. The draining is initially rapid but gradually reduced until it is drop by drop. The dripping rate is controlled by the stopcock. When the water is drained away the glass piece is then taken out of the funnel by means of the handle on the copper ring, the $CaSO_4$: Tm aluminium foil is dried at 100°C, and at the same time a few binder is dropped onto the aluminium foil. After dry this foil is punched into various sizes required.

3 MEASUREMENT OF ANNUAL ALPHA DOSE

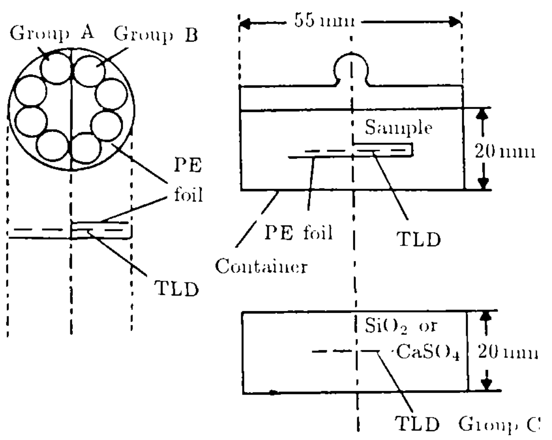

Fig.1 Irradiation configuration for ultrathin TLD determination of alpha dose rate

Twelve ultrathin TLDs of 8 mm in diameter are annealed at 400°C for 60 min then divided into three groups A, B and C. Group B dosimeters are shielded on both sides by 8 mg·cm^{-2} thick polyethylene (PE). Group A dosimeters are shielded only on aluminium foil by the same thick PE. Both groups A and B are embedded in a glass container of powdered pottery (see Fig.1). Group C dosimeters are deposited in either SiO_2 or $CaSO_4$ (both blank samples) to measure environmental radiation. The containers are sealed and stored in the dark for at least one month in order to ensure build-up of radon. At the end of the storage period, the all dosimeters are withdrawn, and TL is measured by placing the $CaSO_4$: Tm aluminium foil directly on the heater plate in the apparatus of TL measurement. The TL of group B dosimeters derives from beta and environmental radiation only while group A dosimeters produce TL resulting from an additional alpha particle dose. However, since the group A dosimeters are alpha-shielded on one side, their alpha contribution results from one-half the infinite matrix alpha dose-rate. Hence the annual alpha dose D_α and annual beta dose D_β are obtained by two equations,

$$D_\alpha = 2(D_{\mathrm{A}} - D_{\mathrm{B}})/t \cdot K \tag{1}$$

$$D_\beta = (D_{\mathrm{B}}/t) - D_o \tag{2}$$

where D_{A} and D_{B} are respectively the average dose-rate of group A and B dosimeters, t is the storage time in years, K is the ratio of alpha to beta sensitivity for the ultrathin

TL dosimeter, and D_o is the dose-rate of environmental background from the group C dosimeters. The D_o is a constant for a certain place.

The K-value is related to the process technology of the $CaSO_4$: Tm. This ultrathin $CaSO_4$:Tm TL dosimeter has a higher alpha sensitivity than the early using[3]; K is equal to 0.29 for $CaSO_4$:Tm thickness of 1 mg cm^{-2}; it is favourable for the determination of alpha dose-rate.

Table 1
The results of dose-rate measurement in ancient pottery and brick

Lab. No.	α dose-rate /$mGy \cdot a^{-1}$		β dose-rate /$mGy \cdot a^{-1}$	
	TLD (K=0.27) 2 $mg \cdot cm^{-2}$	α counting	TLD 2 $mg \cdot cm^{-2}$	α counting
SB155	17.20±1.2	14.28±0.34	2.05±0.12	1.98±0.08
SB156	15.43±0.93	17.72±0.81	2.75±0.05	2.95±0.09
SB157	16.84±1.10	16.46±0.51	2.44±0.05	2.38±0.10
SB158	15.87±1.00	16.64±0.50	2.52±0.04	2.46±0.07
SB175	12.12±0.51	13.57±0.48	1.78±0.06	1.59±0.05
SB176	12.83±0.64	14.19±0.52	2.19±0.08	2.01±0.05
SB177	16.25±1.14	18.32±0.55	2.51±0.11	2.94±0.10
SB222	9.89±0.59	10.96±0.33	1.70±0.03	1.92±0.05
SB223	25.14±1.31	28.08±0.84	3.28±0.07	3.10±0.08
SB276	20.85±1.24	21.41±0.63	2.64±0.06	2.18±0.11
SB278	23.42±1.76	21.84±0.68	3.01±0.08	2.88±0.13
SB279	11.98±1.08	13.88±0.56	1.95±0.05	2.10±0.08

The results of alpha TLD measurements on 12 samples are shown in Table 1. Also shown are the results of alpha counting on the same samples[4]. It is has been proved that the thick source alpha counting is reliable in measurement of alpha dose-rate, and with an uncertainty of about 5% for pottery samples[5], Table 1 shows that here is reasonable agreement between TLD and alpha counting in most cases.

4 CORRECTION FACTOR OF ULTRATHIN TL DOSIMETRY

In TL dating, the palaeodose and the annual dose are measured by the fine-grain samples of archaeological materials and the ultrathin TLD. As mentioned above, the determination of internal alpha and beta dose-rate in pottery using ultrathin TLD is quite convenient and low cost, although TLD does not have the same geometry as the fine-grain. Just as Aitken pointed out, since the thickness of the phosphor layer is about 10μm, it might be expected that there would be significant attenuation of alpha dose-rate[6]. Hence the question is whether the measured values of alpha dose-rate are in good agreement with those using fine-grains. If the two results are different, a correction must be made to the alpha dose-rate measured by using ultrathin TLD. We have made a special study on this problem. Since ultrathin alpha TL dosimetry has been mentioned above, now we describe the method of measurement of alpha dose-rate using fine-grain $CaSO_4$:Tm.

4.1 Sample preparation

Mix $CaSO_4$:Tm grains with pottery powder; their grain sizes should be the same, in the range of 20—150 μm. The mixture is ground further in a ballmill, producing a limited number of fine-grains. The samples used in the study and laced in this manner are given in Table 2.

4.2 Experimental

The laced samples and several ultrathin TLD are annealed for 60 min at 400 ℃, after which the ultrathin TLD are put into a glass container with the laced sample according to aforementioned method. Hence the (beta-equivalent) annual alpha dose D'_α and beta plus gamma annual dose $D_{\beta+\gamma}$ are obtained,

$$D'_\alpha = 2(D_a - D_b)/t \tag{3}$$

$$D_{\beta+\gamma} = D_b/t \tag{4}$$

where D_a and D_b are respectively the average dose of type A (group A) and B (group B) dosimeters, and γ denotes the environmental background.

Table 2
Composition of laced samples

Lab. No.	Source of pottery fragments	$CaSO_4$: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

After withdrawal of the ultrathin TLD, the fine-grain discs are made from the laced sample[2]. 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

$$D_F = D_f/t \tag{5}$$

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

$$D''_\alpha = D_F - D_{\beta+\gamma} \tag{6}$$

In equation (6) 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 ultrathin TLD method. It should be noted that all the dose-rates measured by using the two methods are standardized to an identical laboratory beta source. Hence the calibration error of the beta source is not important.

4.3 Results and discussion

Tables 3 and 4 give the comparison between the results from fine-grain or ultrathin TLD method. Table 3 and 4 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 2mg $\cdot cm^{-2}$, and is 2.5±5.2% less when using 1mg $\cdot cm^{-2}$. A comparison between the alpha counting and ultrathin TLD is given in Tables 5 and 6 (laced samples) and Tables 1 and 7 (ancient pottery and brick). The TLD method had compared with the neutron activation analysis as well as the alpha counting[7].

Table 3

Comparison of the results of α dose-rate measurements by using fine-grain or ultrathin TLD method (2 mg·cm^{-2})

Lab.No	α dose-rate/mGy·a^{-1} (beta equivalent) D''_α fine-grain	D'_α TLD	$\frac{D''_\alpha - D'_\alpha}{D''_\alpha}$ /%	Lab.No	α dose-rate/mGy·a^{-1} (beta equivalent) D''_α fine-grain	D'_α TLD	$\frac{D''_\alpha - D'_\alpha}{D''_\alpha}$ /%
10	5.44±0.05	4.82±0.27	+11.0±4.9		4.30±0.08	4.02±0.18	+6.5±4.6
	5.06±0.05	4.69±0.29	+7.3±5.8		4.10±0.05	—	—
	5.33±0.08	5.02±0.20	+5.8±4.0		3.78±0.03	—	—
	5.25±0.06	4.77±0.20	+9.1±4.0	(Average)	3.97±0.07	3.67±0.19	+7.6±5.1
(Average)	5.27±0.06	4.83±0.24	+8.3±4.7	14	3.97±0.06	3.74±0.13	+5.8±3.6
11	5.09±0.07	4.69±0.18	+7.9±3.8		3.80±0.06	3.29±0.22	+13.0±5.8
	4.79±0.09	4.48±0.17	+6.5±4.0		4.17±0.10	3.82±0.21	+8.4±5.6
	4.96±0.07	4.32±0.32	+11.3±6.7		3.73±0.05	3.44±0.17	+7.8±4.8
	5.29±0.12	4.81±0.19	+9.1±4.3		3.68±0.07	—	—
(Average)	5.03±0.09	4.58±0.22	+8.9±4.7		4.15±0.09	—	—
12	3.99±0.09	3.55±0.21	+11.0±5.7		4.01±0.11	—	—
	3.62±0.05	3.28±0.14	+9.4±4.1	(Average)	3.93±0.08	3.57±0.18	+9.2±5.0
	4.03±0.12	3.81±0.23	+5.5±6.5	$(D''_\alpha - D'_\alpha)/D''_\alpha$ average for 4 groups = +8.5±4.9%			

Table 4

Comparison of the results of α dose-rate measurements by using fine-grain or ultrathin TLD method (1 mg·cm^{-2})

Lab.No	α dose-rate/mGy·a^{-1} (beta equivalent) D''_α	D'_α	$\frac{D''_\alpha - D'_\alpha}{D''_\alpha}$ /%	Lab.No	α dose-rate/mGy·a^{-1} (beta equivalent) D''_α	D'_α	$\frac{D''_\alpha - D'_\alpha}{D''_\alpha}$ /%
10		4.96±0.20	+5.9±4.0			3.88±0.12	+2.3±3.6
		5.04±0.26	+4.4±5.1			4.22±0.21	-6.3±5.6
		5.31±0.32	-0.76±6.0	(Average)	3.97±0.07	3.98±0.19	-0.25±5.1
(Average)	5.27±0.06	5.10±0.26	+3.2±5.0	14		3.77±0.15	+4.1±4.4
11		4.93±0.19	+2.0±4.2			3.82±0.18	+2.9±5.2
		4.61±0.27	+8.3±5.6			4.10±0.27	-4.3±7.1
(Average)	5.03±0.09	4.77±0.23	+5.2±4.9			3.53±0.23	+1.0±6.1
12		3.74±0.25	+5.8±6.5	(Average)	3.93±0.08	3.81±0.21	+3.1±5.8
		4.08±0.19	-2.8±5.2	$(D''_\alpha - D'_\alpha)/D''_\alpha$ average for 4 groups = +2.5±5.2%			

The alpha dose-rate equals that the (beta-equivalent) alpha dose-rate divided by K; $K = 0.29$ (±3%) for 1 mg·cm^{-2} and 0.27(±3%) for 2mg·cm^{-2}. Tables 5 and 6 show that as long as different values of K are used, the ultrathin TLD of different thickness give the same alpha dose-rate. In fact, the K acts as a compensator for thickness. The K-value is 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

$$K = 2(D_a - D_b)/tD_{\alpha o} \qquad (7)$$

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

Table 5

Comparison of fine-grain, ultrathin TLD and α counting methods for measurement of α dose-rate

mGy·a^{-1}

Lab. No	Fine-grain	Ultrathin TLD			α counting
	K=0.29	1mg·cm^{-2} K=0.29	2mg·cm^{-2} K=0.27	2mg·cm^{-2} K=0.29	
10	18.17±0.21	17.59±0.90	17.89±0.89	16.66±0.83	18.62±0.61
11	17.34±0.31	16.45±0.79	16.96±0.81	15.79±0.76	18.10±0.54
12	13.69±0.24	13.72±0.65	13.59±0.70	12.66±0.66	15.32±0.53
14	13.55±0.28	13.14±0.72	13.22±0.67	12.31±0.62	13.22±0.42

Table 6

The ratio of TLD to α counting for α dose-rate measurement

Lab. No	Fine-grain	Ultrathin TLD		
	K=0.29	1 mg·cm^{-2} (K=0.29)	2 mg·cm^{-2} (K=0.27)	2 mg·cm^{-2} (K=0.29)
10	0.98±0.05	0.94±0.07	0.96±0.07	0.89±0.06
11	0.96±0.05	0.91±0.07	0.94±0.07	0.87±0.06
12	0.89±0.05	0.90±0.07	0.89±0.07	0.83±0.06
14	1.02±0.08	0.99±0.09	1.00±0.09	0.93±0.09
(Average)	0.96±0.06	0.94±0.08	0.95±0.08	0.88±0.07

Table 7

The ratio of TLD to α counting for α and β dose-rates of archaeological samples

Lab.No	TLD/α counting (Ultrathin TLD: 2mg·cm^{-2},K=0.27)		Lab.No	TLD/α counting (Ultrathin TLD: 2mg·cm^{-2},K=0.27)	
	α dose-rate	β dose-rate		α dose-rate	β dose-rate
SB155	1.20±0.09	1.04±0.07	SB177	0.89±0.07	0.85±0.05
SB156	0.87±0.06	0.93±0.03	SB222	0.90±0.06	0.89±0.03
SB157	1.02±0.07	1.03±0.05	SB223	0.90±0.05	1.06±0.04
SB158	0.95±0.07	1.02±0.03	SB276	0.97±0.06	1.21±0.07
SB175	0.89±0.05	1.12±0.05	SB278	1.07±0.09	1.05±0.06
SB176	0.90±0.06	1.09±0.05	SB279	0.86±0.08	0.93±0.04
			Average	0.95±0.07	1.02±0.05

Table 5 shows that the ultrathin TLD results are in reasonable agreement with alpha counting; 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}.

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

a. The alpha counting 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 alpha counting method.

b. As mentioned above, using TLD of 1 mg·cm^{-2} the measured alpha dose-rate is 2.5% less than with fine-grain; Table 8 also shows that the thickness of $CaSO_4$ thin layer must be≤0.5mg·cm^{-2}, and the thickness of 1 mg·cm^{-2} is lightly larger than that of calculation; perhaps it is one of the reasons giving rise to the differences between 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 rang. 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 it is included in the K-value.

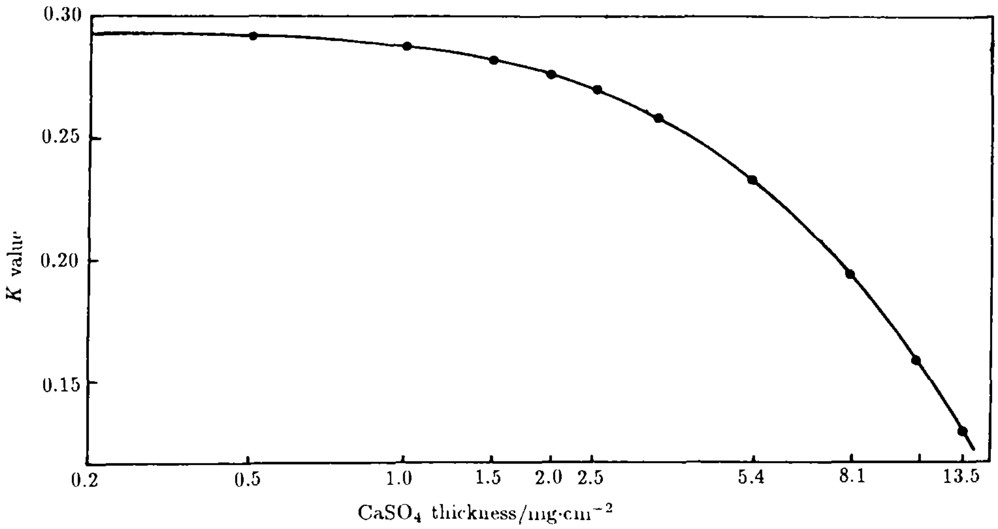

Fig.2 Variation of the K-value as a function of $CaSO_4$ thickness

4.4 Correction curve

These results can also be obtained in theory. Table 8 and Fig.2 give the variation of the K-value as a function of $CaSO_4$ thickness. The variation of K-value for various thickness is due to the alpha dose attenuation in $CaSO_4$ thin layer. The calculation of alpha dose attenuation is based on the mathematical formalism developed by Charlton and Cormark[8], which is also the bases for Bell's calculations[9].

Table 8

K-value in the different thickness of $CaSO_4$ layer

Thickness /μm	1.9	3.7	5.6	7.4	9.3	11.1	20	30	40	50
Thickness /mg·cm^{-2}	0.5	1.0	1.5	2.0	2.5	3.0	5.4	8.1	10.8	13.5
S_α	0.982	0.964	0.945	0.927	0.907	0.888	0.790	0.659	0.535	0.443
K-value	0.292	0.287	0.281	0.276	0.270	0.264	0.235	0.196	0.159	0.132

In Table 8, the S_α is absorbed dose fraction for alpha particles from the thorium and uranium series in $CaSO_4$ thin layer[8]. The functional relation of the K-value versus the $CaSO_4$ thickness is derived from the variation in mean absorbed dose for the diameter of quartz grains. Table 8 shows that the calculational K values are in agreement with the experimental one.

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 the 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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