Thermoluminescence dating of pottery from Sringaverapura—A Ramayana site

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Abstract. The first thermoluminescence (TL) dates of pottery from a *Ramayana* associated site are reported. The TL dates for pre-NBPW Black-Slipped Ware levels are 730 and 765 B.C., while radiocarbon date takes the earliest Black Slipped Ware level (for which no TL dates are available) to 905 B.C. (uncorrected). For the OCW level, three TL dates are available which range from 1035 B.C. to 875 B.C. The early NBPW period believed to be associated with the *Ramayana* episode will thus be post-750 B.C.

Keywords. Thermoluminescence dates; fine grain technique; *Ramayana*; Sringaverapura; archaeology.

1. Introduction

An attempt was made by Lal (1979) to find a common archaeological denominator for the *Ramayana* associated sites so that the material culture contemporary to the epic may be identified and dated. Though about fifteen hundred archaeological ^{14}C dates are available today (Agrawal *et al* 1975), many archaeological sites still remain undated primarily due to paucity of datable organic material. We have now installed a thermoluminescence (TL) laboratory and have taken up a systematic study of the important sites where radiocarbon method could not be applied so far.

Sringaverapura is a site associated with the *Ramayana* and has been excavated recently. All the *Ramayana* associated sites, viz. Ayodhya, Bharadwaja-Ashrama and Nandigrama, have yielded an early Northern Black Polished Ware (NBPW) level. This level is now thought to be coeval with the *Ramayana* period (Lal 1981). At Sringaverapura, however, the NBPW level is preceded by a Black-Slipped Ware level ·which in turn, is preceded by that of Ochre Colour Ware (OCW).

The OCW culture is a controversial one and there is as yet no unanimity about its authorship (Agrawal 1969; Lal 1972). There are till today no radiocarbon dates available from unambiguous OCW levels. TL dates of some other debated OCW sites are available (Huxtable *et al* 1972) (table 1), however, these indicate a large scatter from *c.* 2600 to 1100 B.C. and therefore cannot be used for pinpointing a date bracket for the OCW culture. It should, however, be noted that these TL measurements were made in 1969 and no sherds were

tested for anomalous fading at that time.   Also most of the sherds emanated radon so that the dose would have been over-estimated, because the full implication of this had not been realized then. The dates should therefore be regarded as minimum ages for the culture (J Huxtable, personal communication).

2. Sample collection and site details

The site [Sringaverapura mound] is situated 35 km upstream from Allahabad. The mound is situated on the bank of River Ganga. A detailed excavation

Table 1. TL dates from various OCW sites as reported by Huxtable *et al* 1972.

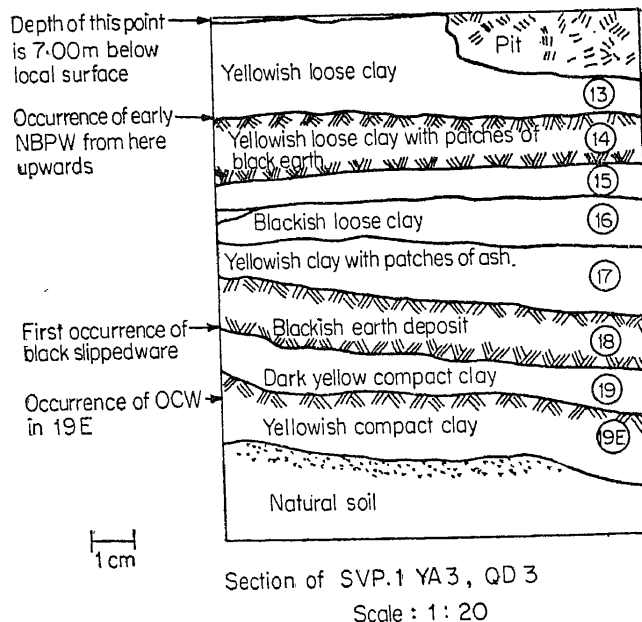| Site	Sherd No.	TL ages (years B. C.)	Mean TL age (years B. C.)
Atranjikhera	11164	1610	
	11165	1170	
	C1	2280	1690
	C2	1250	
	C3	2130	
Lal Qila	11291	1730	
	92	2030	1880
Jhinjhana	11392	1990	
	B1	1570	2070
	B2	2650	
Nasirpur	11491	1500	
	92	1180	1340

Figure 1. Section diagram along with cultural association of trench SVP I, at Sringaverapura.

yielded archaeologic material belonging to seven different cultural periods in a total deposit of nearly 9 m below the top surface (figure 1).

The pre-NBPW level was spread over a 1·65 m thick deposit of which OCW and Black-Slipped Ware were 0·45 and 1·2 m respectively. The pre-NBPW deposits gave red ware and black-slipped ware, the OCW deposit yielded a red ware of OCW culture. A reliable sampling and site details are crucial for TL dating and, with this in view, the samples were collected at the site by the authors (B B Lal and A K Singhvi) during the excavations at Sringaverapura. All the samples were carefully identified and were immediately sealed with adhering soil in double plastic bags. This method enabled effective retention of moisture which was subsequently determined in the laboratory.

3. Thermoluminescence analysis

The basic principles of TL dating and its application to archaeology have been described in detail by Singhvi and Nambi (1979). We briefly describe here the various parameters used in TL age determination.

3.1 *The age equation*

The minerals responsible for the TL of the samples lose their geologic TL during the firing of pottery. A fresh acquisition of TL restarts due to the exposure of the sample to radiation arising from natural radioactivity (uranium and thorium series and potassium) present in the sample matrix and the burial media. The TL measured in the laboratory is simply related to the time elapsed since firing of the samples as the annual dose rate is practically constant.

The age determination thus is based on: (a) the measurement of the total radiation dose *via* TL and (b) the measurement of radiation dose rate. The age equation can be written as:

$$\text{Age} = \frac{\text{Natural TL (i.e. total radiation dose)}}{\text{TL per unit radiation dose} \times \text{dose per year}}$$

which can be expressed as:

$$\text{Age} = \frac{(ED)_\beta + 1}{aD_\alpha + D_\beta + D_\gamma + D_c}, \tag{1}$$

where $(ED)_\beta = \dfrac{\text{Natural TL}}{(\text{TL per unit radiation dose})_\beta}$,

I = supralinearity correction,

$a = (\text{TL per rad})_\alpha \, / \, (\text{TL per rad})_\beta$.

D's represent dose rates due to various radiation components arising from the decay of natural U-238, Th-232 series and K-40 present in varying degrees
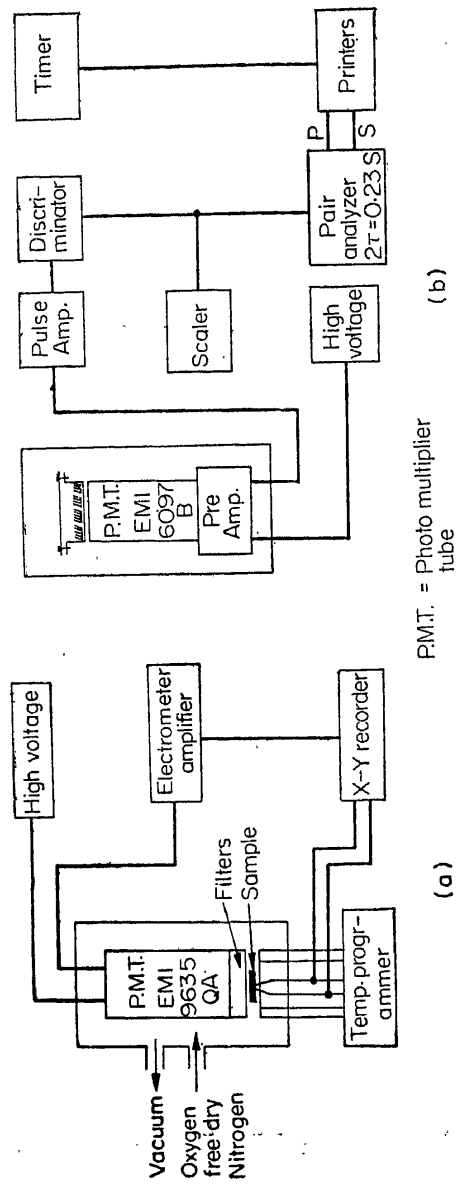
Figure 2. Block diagrams of the thermoluminescence and alpha counting systems.

P.M.T. = Photo multiplier tube

in pottery (P) and soil (S). The various contributions to these dose rates are:

$$D_\alpha = D_\alpha^p(Th, U),$$

$$D_\beta^p = D_\beta^p (Th, U) + D_\beta^p (K),$$

$$D_\gamma = D_\gamma^s (Th, U) + D_\gamma^s (K),$$

$$D_c = D_c \ (\text{cosmic rays}).$$

Here, typically D_β^p (K) is the contribution to beta dose from the beta decay of K-40 present in the pottery matrix. The scaling factor *a* described above takes into account the lower TL induction efficiency of alphas compared to that of betas.

3·2 *Experimental details*

Low light levels encountered in archaeologic ceramics put exacting requirements on the sample preparation and measurement techniques. A considerable effort was therefore put into the fabrication of TL vacuum glow oven and the electronics to ensure reliable TL measurement with negligible interference from spurious TL, etc. In what follows we describe the various procedures as also the intercalibration exercises carried out in our laboratory to ensure a reliable TL date. A schematic diagram of the TL apparatus and the alpha counting system is given in figure 2. Typical glow curves are indicated in figure 3.
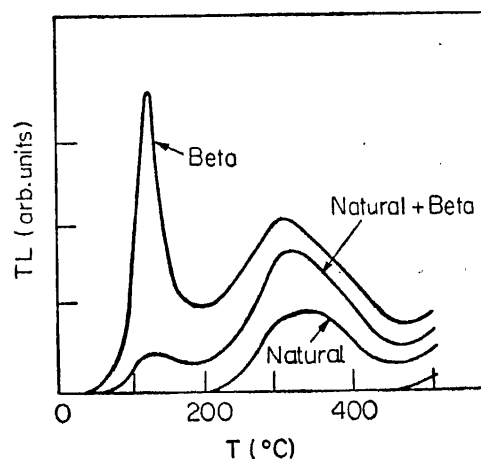

Figure 3. Typical TL glow curve of fine grain extracts from potsherds excavated from Sringaverapura.

3·2 a. TL reader

The TL reader comprised of a vacuum glow oven fabricated by us. The heater was a 0·25 mm thick kanthal strip with a flexible power supply contact to avoid any possible warp due to extension of the plate during heating. The solid angle of the sample subtended at the photocathode was frequently monitored using a CaF_2: natural phosphor disc. All the measurements were made after evacuating the oven chamber twice and flushing with ultra high purity nitrogen gas (IOLAR-Grade II supplied by the Indian Oxygen). Nitrogen flow rate was kept at about 3-3·5 l/min since lower flow rates occasionally failed to quench the spurious TL. The temperature was controlled using an automatic linear temperature programmer (Devgan *et al* 1980). The reproducibility and linearity of heating rate was checked frequently during the measurements using a Hewlett Packard time base AM 171008. Typical heating rates were usually 6-7°C/sec and the reproducibility was better than 1% throughout the measurement schedule. The optical detection system comprised of an EMI 9635QA (selected for high blue to low red sensitivity) coupled to Corning 7-59 and 5-60 filters along with two Chance Pilkington HA3 filters. This arrangement gave a good discrimination against black-body radiation upto 500°C. The PMT output was amplified through a PRL fabricated electrometer amplifier using a CK5886 electrometer tube. The stability of the optics channel was continuously monitored using a radiocarbon activated plastic phosphor (Facey 1966) and was better than $\pm$ 1·5%.

3.2 b. Radiation sources

Artificial beta irradiations were carried out using a 40 mCi $^{90}Sr/^{90}Y$ beta plaque source (type SIP). The beta irradiations were carried out 'off' plate and the irradiation facility consisted of a 1·52 cm high aluminium irradiation mount on a perspex base. The primary calibration of the beta source was done using irradiated CaF_2: nat disc obtained from Research Laboratory for Archaeology, Oxford. This was recalibrated at the Bhabha Atomic Research Centre, Bombay, where the values agreed to within 4%. The dose rate to CaF_2: nat was found to be 121·3 rads/min. The dose to pottery was taken to be 1·05 times the dose to CaF_2: nat (Wintle and Murray 1977).

Artificial alpha irradiations were carried out in six-seater vacuum alpha irradiation facility (Singhvi and Aitken 1978). The calibration of alpha source was done at Oxford *via* alpha spectrometry and by intercomparing irradiated discs of CaF_2:nat. The average source strength as seen by the sample was $S = 0·080 \ \mu m^{-2} \ min^{-1}$.

3·2 c. Sample radioactivity

The natural radioactivity *i. e.* U-238 and Th-232 was estimated using thick source ZnS (Ag) alpha counting technique (Turner *et al* 1958). Plastic screens impregnated with ZnS (Ag) were used as scintillators. A fixed 42 mm dia counting area was defined by a steel ring of appropriate dimensions. An EMI

6097B photomultiplier tube coupled to a single channel spectrometer was used as counting system. The system also had a pair counting facility with resolving time of 0·23 sec. This enabled check on abnormal U/Th ratios by measuring ^{216}Po alpha activity (half life 0·15 sec). A serial printer printed out the data every twenty minutes and this ensured a check against any electrical disturbance or on any count rate change due to system malfunctioning. The spectrometer was calibrated using two Canadian certified reference material standards BL-3 (1% U) and DL-1 (41 ppm U, 83 ppm Th). The 83% discrimination level was adjusted using BL-3. Typical background count rates were 0·1-0·2 counts/ksec. cm^2 and the pottery count rates were approximately 15 counts/ ksec. cm^2. In all the samples both unsealed (α_0) and sealed (α_1) counts were done to check the loss of radioactivity due to radon escape. As in all the cases α_0 and α_1 were almost identical within 7%, we used α_0 for estimating U and Th concentration. Equal alpha activity was assumed in estimating the concentrations of U and Th. Pairs rate was only used as a monitor for abnormal U/Th ratios. The potassium analyses were carried out using a Perkin-Elmer 305A atomic absorption spectrophotometer. The sample was dissolved by a sequential treatment of HF, $HClO_4$, HNO_3, HCl. The potassium content of soil was determined by γ-ray spectrometry using a well type 7.5×7.5 cm NaI (Tl) scintillation system. A considerable variation in soil potassium content was observed within each stratum. Therefore an average value has been used for all the samples.

The estimation of dose rates from measured alpha count rates and potassium concentration was made using conversion factors given by Aitken (1979). Appropriate wetness corrections were also applied as described later.

4. Measurements

4·1 *Sample preparation*

The measurements reported here were carried out on the fine grain extract (1-8μm) from the pottery (Zimmerman 1971). A small portion of the sherd was gently crushed in a stainless steel V-trough, and the fraction less than 100μm was sieved out. 1-8μm grains were extracted from this fraction by their known sedimentation rates (2-20 min.) in a 6 cm analar grade acetone column. These grains were resuspended in acetone and equal volume of the suspension was pipetted on to clean aluminium discs (1 cm dia.) having 1 cm column of acetone above them. For each sample, 24 discs were prepared and all the sample preparations were carried out in lights filtered with three amber solar screens. The disc-to-disc variation of TL signal was typically 3-4%.

4·2. *Estimation of ED_β, I and a*

The equivalent beta dose was measured using additive beta dose procedures. In this, four discs were used for estimation of natural TL level, three each for natural plus beta-1 and natural plus beta-2. A growth curve of TL *vs* applied dose was constructed and a linear back extrapolation of this growth

curve yielded the equivalent beta dose. The value of the supralinearity intercept was also determined using the same procedure. Care was, however, taken to ensure that only samples with exactly similar radiation and thermal histoιy were used (to avoid complications due to possible predose effect and transparency changes). The same procedure was used to construct alpha growth curve, yielding equivalent alpha exposure. The a value was calculated using the relation given by Aitken and Bowman (1975).

$$a = \frac{(ED)_\beta}{1300 \times S \times Y} , \qquad (2)$$

where S is the source strength and Y is the equivalent alpha exposure in minutes.

4.3 *Fading tests*

Plateau test for stability of TL signal in all the samples was carried out and the plateau R varied from $\pm 3\%$ to $\pm 7\%$ in samples over a temperature range of 75-125° C, beginning usually at around 275-300° C. In addition, all the samples were measured for short-term anomalous fading (Wintle 1973) and the results indicated in table 2.

4·4 *Age calculations*

The dry dose were then converted to wet dose rate using scaling relations given by Zimmerman (1971) giving true *in situ* dose rate. *In situ* water fraction was taken to be the realistic water fraction as the general stratigraphy, the depth of the samples (9 m) and the proximity of a river did indicate that water fraction did not vary appreciably during burial history of the sample. Typical saturation water content was 10% and thus a $\frac{1}{2} + \frac{1}{3}$ water content assumption would at best affect the age estimate only by 1-2%. The dose rates with alpha components appropriately scaled down were added to obtain the total annual dose. In this calculation the cosmic ray contribution was taken to be 15 mr/yr. The total archaeological dose $(ED_\beta + I)$ when divided by this dose rate gives the age.

4.5 *Estimation of errors*

The errors were assessed by a rigorous use of the procedures suggested by Aitken (1976) and Aitken and Alldred (1972). Typical measurement errors *i. e.* $(\delta Q/Q)$, $(\delta I/Q + I)$, $(\delta a/a)$ have been taken to be 5%. The random errors in alpha counting and potassium determination were taken to be 5%. The alpha and beta source calibration errors have also been taken as $\pm 5\%$. Other errors due to variation in Th/U ratio and wetness correction have been taken into account. Since a conservative estimate of errors due to all these parameters has been made, the quoted errors provide a maximal variation in the age.

Table 2. TL measurements and radioactivity data on the samples

Sample No.	TL analysis			Sherd wt. (wet/dry)	Soil wt. (wet/dry)	Radioactivity analysis (m.rads/yr.)							Age in yrs. B.P.	Anomalous fading	
	AD rads	Y min	a			D_α^p (Th,U)	D_β^p (Th,U)	D_β^p (K)	D_γ^s (Th,U)	$D_\gamma^s K$	D_c	D_t			
TL-21	2187	99.5	0.152	1.11	1.15	355	110	184	93	65	15	822	2660	?	7% in 6 weeks
TL-24 (1)	2794	105	0.162	1.03	1.08	456	130	233	105	70	15	1009	2769	?	10% in 1 week
TL-24 (2)	2725	104.3	0.168	1.02	1.08	475	125	223	105	70	15	1013	2690	<	3% in 1 week
TL-29	2864	38.5	0.17	1.05	1.06	442	121	298	107	71	15	1054	2743	<	5% 1 week
TL-33 (1)	3082	68.5	0.161	1.06	1.09	463	134	258	125	68	15	1063	2900	<	3% in 1 week
TL-33 (2)	3227	167.7	0.170	1.05	1.09	541	149	228	128	69	15	1130	2855	<	3% in 1 week
TL-38 (1)	3378	126.3	0.22	1.15	1.15	559	119	254	109	65	15	1121	3015	<	3% in 1 week

AD is the total archaeological dose ($ED_\beta + 1$); Y is the equivalent alpha exposure in minutes; a is the alpha efficiency factor; wt. (wet/dry) is the ratio of the weights of samples as retrieved and after 24 hours drying; Typically D_α (Th,U) is alpha dose rate due to radioactivity (Th,U) within the pottery; D_c cosmic rays; and D_t is total annual radiation dose.

Table 3. TL dates of the Pre-NBPW and OCW level samples from Sringaverapura Trench SVP1, Square Y A3, Quadrant 3. Radiocarbon dates are based on $\tau_{\frac{1}{2}} = 5730$ years and 1950 as reference year, while TL dates have the base year 1980. Thus there is a systematic difference of 30 years between TL and ^{14}C dates.

Sample No.	Layer	Culture	TL date (B.P. years)	Archl. estimate (B.P. years)	Radiocarbon date (B.P. years)
PRL: TL: 21	17	Red Ware-Pre-NBW	2660 ± 280	2700-3000	
PRL: TL: 24 (1)	17	Black and Red Ware Pre-NBW	2769 ± 400	2700-3000	
PRL: TL: 24 (2)	17	Black and Red Ware Pre-NBW	2690 ± 280	2700-3000	
PRL: TL 29	18	Black Slipped Ware Pre-Pre-NBW	2743 ± 300	2700-3000	2700 ± 130 (PRL-669)
—	19	Black Slipped Ware			2855 ± 200 (PRL-670)
PRL: TL: 33 (1)	19E	Ochre Colour Pottery-OCW	2900 ± 380	Pre-3000	
PRL: TL: 33 (2)	19E	Ochre Colour Pottery-OCW	2855 ± 270	Pre-3000	
PRL: TL: 38 (1)	19E	Ochre Colour Pottery-OCW	3015 ± 280	Pre-3000	

5. Results and discussion

Table 2 summarizes the sample data on TL and radioactivity measurements. Table 3 gives the sample parameters and TL dates. Archaeological and radio-carbon dates on samples from the same stratum are also included for comparison. All these dates show a good agreement, within the experimental errors. The TL dates, PRL: TL: 29, 763± 300 B.C. and radiocarbon date, 750 ± 130 B. C. for the same level agree very well. The TL dates also show a stratigraphic sequence with depth. The errors quoted are conservative estimates. Anomalous fading data are concordant with glow curve shapes (glow peaks at 110°C and 375°C) and it appears that most of the TL is contributed by quartz (Singhvi and Zim-merman 1979). It is, however, difficult to reconcile this with the high values of a (table 2) (Singhvi 1981).

Archaeologically, in the upper Ganga valley the Ochre colour ware has been thought to belong to the second half of the second millennium B. C. Thus, TL dates for OCW in the middle Ganga valley towards the end of that millennium may not be far out, particularly because there is considerable margin of error in the TL dates themselves. The date for the Black-Slipped occupation at Sringa-verapura is c. 760 B. C. These too seem to be fairly in order, allowing the same margin of error as in the case of OCW level which is datable to c. 950 B. C. Since the NBPW overlapped with the black-slipped ware at Sringaverapura, the beginning of the NBPW may well go back to the 7th-8th century B. C. As already discussed earlier, the lowest common culture at the Ramayana sites so far excavated is that of the early NBPW. It would thus follow that the Ramayana episode is unlikely to be earlier than the middle of the eighth century B. C. This problem is discussed elsewhere in detail (Lal 1980).

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