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ScienceDirect
Radiation Measurements 43 (2008) 781–785
Radiation Measurements
www.elsevier.com/locate/radmeas

Comparative study of infrared-stimulated luminescent and thermoluminescent dating of archaeological artefacts

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Abstract

In this study, thermoluminescence (TL) dating of archaeological artefacts from Geboltskirchen (Upper Austria) and Haselbach (Lower Austria) has been carried out. To receive a comparison with TL dating, all samples have been investigated additionally by means of infrared-stimulated luminescence (IRSL) analysis. The samples were prepared according to the common fine-grain technique. All important dating parameters such as potassium concentration, thorium/uranium ratio, moisture content, etc. were determined. A methodology for IRSL was developed, describing all steps of the procedure from sample preparation to measurement for any kind of ceramic artefacts. Dating results from TL and IRSL investigations of ancient pottery from Geboltskirchen and Haselbach will be discussed.

Keywords: Thermoluminescence; Infrared-stimulated luminescence; Dating

1. Introduction

Thermoluminescence (TL) can be applied as an absolute dating method, giving ages that are independent of any other chronology (Aitken, 1970). The TL method was thus used as a reference for dating of ancient pottery from Geboltskirchen (Upper Austria) and Haselbach (Lower Austria). To confirm the validity of the method, all samples were investigated additionally by means of infrared-stimulated luminescence (IRSL).

This work addresses two concrete problems regarding the age determination of different samples (brick, pottery, etc.). The first question concerns samples of the so-called *Ziegelroit*, an area close to Geboltskirchen (Upper Austria). The bricks and pottery fragments were distributed over an area of approximately $30 \times 50\,m^2$, while no traces from surfaces and edges were found that could be used to identify the form of the original bricks. Some historians and regional researchers reported in early publications remnants of a Roman signal tower. Other sources interpreted the remnants found at this burial site to stem from a gentile temple or a watch tower of the former castle of Gröbming. There are also speculations from miners that an underground fire could have burned the existing loam soil, as enormous brown coal deposits are located in the proximity. Another possibility considered by specialists of the Upper Austrian State Museum in Linz would be a field brick-kiln—a dome smeared outside with loam—to burn bricks. The distributed fragments could as well be parts of this broken-down dome. A contradiction to all these possibilities is the name *Ziegelroit* itself. The term *Roit*, referring to the time of settlement, appears shortly after 1000 AD. However, there are no analogies which give further cognitions, implying that physical dating methods are particularly suitable to get necessary information about the age and possible origin.

The second question concerns samples from Haselbach (Lower Austria). During completion of a high-speed rail line between Vienna and St. Pölten twenty archaeological sites have been discovered and investigated since the year 2003. For the archaeologists, the landscapes Perschlingtal and Southern Tullnerfeld are of greatest importance. The presence of several population groups could be confirmed from the Neolithic to the Roman Era by archaeological investigations. Since March 2004, excavations were carried out in the region of Haselbach revealing—according to archaeologists—Late Celtic pottery fragments. This assumption should be confirmed or disproved by dating of provided samples.

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doi:10.1016/j.radmeas.2008.02.005

Within these tasks, a methodology for IRSL dating should be developed which describes all steps of the procedure from sample preparation to measurement for any kind of ceramic artefacts.

2. Experimental

2.1. Sample preparation

The pottery fragments were buried by different depths of soil. Samples from Geboltskirchen were retrieved from depths of 5 cm (Cer3, Cer4) and 10 cm (Cer1, Cer2), respectively, whereas the range of depths from which the Haselbach samples were excavated is unknown to the authors. After retrieval, the samples were processed according to the well-established fine-grain technique (Zimmerman, 1971), following standard preparation procedures in subdued red light. The fine-grain technique meets two essential requirements (Aitken, 1985). First, the grains utilized are small enough to be penetrated entirely by alpha particles. Second, the grains are obtained in the form of a monolayer suitable for determination of alpha particle effectiveness. By washing the extracted sample powder in acetone and 5% acetic acid, a suspension of fine grains is obtained, an ultrasonic bath being used to disperse coagulation. The settling time is determined by diameter and—partly in combination with a centrifuge tube—permits separation of grains in the size range of 1–8 μm. The separated grains were then re-suspended with a ratio of 1.5–1.8 mg of sample to 1 ml of acetone and allowed to deposit on aluminium discs of 10 mm in diameter.

2.2. TL Measurements

The fully automated in-house developed TL reader (Berger et al., 2002) primarily consists of a Thorn EMI 9235 QA photomultiplier tube (Thorn EMI Gencon, Inc., Fairfield, NJ, USA) with an optical UV band pass filter (UG11, Schott AG, Mainz, Germany), alpha (^{241}Am) and beta sources (^{90}Sr/^{90}Y) for sample irradiations, an automated 60-sample changer and a heating system for thermal stimulation. All measurement parameters, e.g., the number of samples, the type and duration of irradiation, the heating rate, etc., can be controlled by sophisticated computer software. All TL measurements were carried out using a heating rate of $\beta = 5\,°C\,s^{-1}$ up to a maximum temperature of 500 °C in ultra-pure (5.0) dry nitrogen atmosphere. The samples were pre-heated on the same planchet at 190 °C for 1 min to remove thermally unstable traps. A subsequent background measurement was made. Artificial irradiations to produce growth curves were performed using the internal ^{90}Sr/^{90}Y beta and ^{241}Am alpha sources with dose rates of 0.580 and 3.18 Gy min^{-1}, respectively.

The TL measurement sequence used was as follows. At first, a test with four discs was carried out in order to estimate the expected equivalent dose (ED). Two discs were measured to receive the natural TL. After readout, the same discs were exposed to beta radiation from the internal source for 5 and 10 min, respectively, and re-measured to analyze the second-glow TL for determination of the intercept (I). Two more discs were beta-irradiated for 5 and 10 min to receive the TL from natural and laboratory doses. With the collected data from all these measurements, the ED can be assessed roughly. Based on this estimation, additional discs were irradiated with appropriate doses. A typical set of TL glow curves is shown in Fig. 1. The illustration shows a natural glow curve, along with several artificial glow curves from aliquots irradiated with different beta doses. According to the results of the plateau test, TL signals were integrated over a temperature range of ∼275–320 °C.

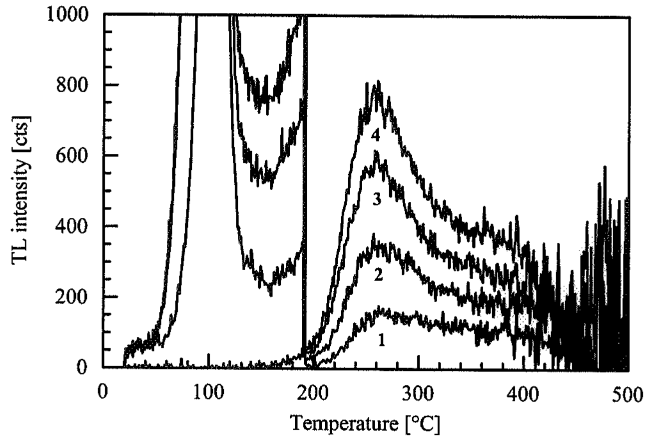

Fig. 1. Typical natural and artificial glow curves from ceramic artefacts: (1) natural TL; (2) 2.6 Gy, (3) 5.2 Gy, (4) 7.8 Gy beta irradiation.

2.3. IRSL measurements

The in-house developed IRSL reader primarily consists of a ring-shaped holder with 27 GaAlAs infrared light-emitting diodes (SFH 484, Osram GmbH, Regensburg, Germany) emitting at a wavelength $\lambda = 880 \pm 80$ nm and three photodiodes (BPW 34, Osram GmbH, Regensburg, Germany), a sample chamber and a Thorn EMI 9235 QA photomultiplier tube (Thorn EMI Gencon, Inc., Fairfield, NJ, USA) with an optical band pass filter (BG39, Schott AG, Mainz, Germany). The filter was used to discriminate between stimulation and emission light and to reduce the signal due to scattered light from the diodes. The IRSL measuring system is not automated and therefore both irradiations and pre-heating of the samples had to be carried out externally. All IRSL measurements used a stimulation time of 200 s in the $\lambda = 880 \pm 80$ nm wavelength range at room temperature of ∼25 °C. The samples were pre-heated at 220 °C for 10 min (Li, 1991) in order to eliminate thermally unstable traps before the 200 s IRSL analysis started. A subsequent background measurement was made. Artificial irradiations to produce growth curves were performed using external ^{90}Sr/^{90}Y beta and ^{241}Am alpha sources with dose rates of 3 and 4 Gy min^{-1}, respectively.

The following IRSL measurement sequence was used. First, a test with three discs was carried out in order to distinguish between the natural IRSL signal and the background. Two more discs were beta-irradiated for 5 and 10 min to receive the IRSL from natural and laboratory doses. Subsequently, the discs were pre-heated at 220 °C for 10 min and then measured. The collected data from all these measurements allow a rough assessment of the ED. Based on this estimation, additional discs were irradiated with appropriate doses. After readout, the same discs were bleached for 2–3 h under a solar simulator (LOT Oriel GmbH & Co. KG, Darmstadt, Germany) equipped with a 300 W Xe short arc lamp, followed by irradiation and pre-heating as before. Subsequent analysis revealed the second-decay IRSL—the analogue to the second-glow TL—for determination of the intercept. Fig. 2 illustrates the IRSL measurement sequence. A typical set of IRSL decay curves is shown in Fig. 3. The illustration shows a natural decay curve, along with several artificial decay curves from aliquots irradiated with different beta doses. According to the results of the plateau test, IRSL signals were integrated over a time range of ~75–175 s.

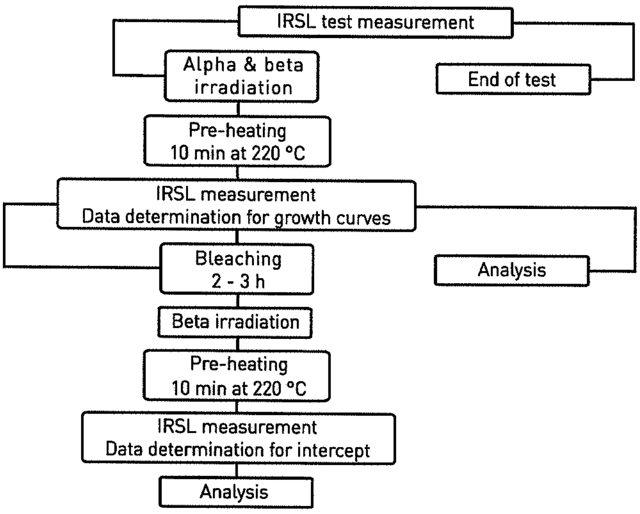

Fig. 2. Schematic representation of the IRSL measurement sequence.

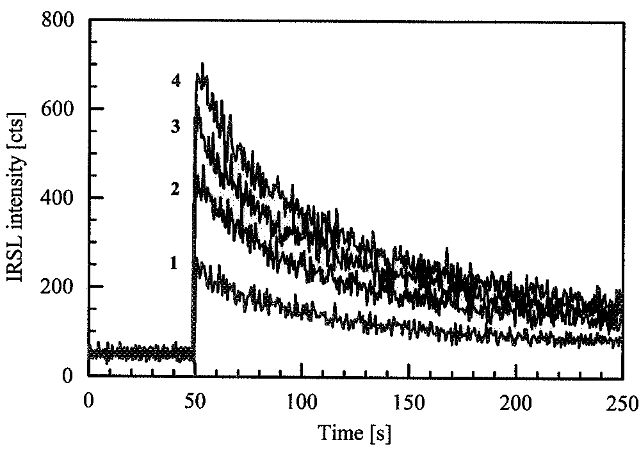

Fig. 3. Typical natural and artificial decay curves from ceramic artefacts: (1) natural IRSL; (2) 2.6 Gy, (3) 5.2 Gy, (4) 7.8 Gy beta irradiation.

2.4. Dose rate measurements

The condition of the ceramic artefacts was carefully determined, particularly regarding moisture content and saturation. Ceramic moisture contents (water/dry weight) ranged between 1% and 12% for the samples from Geboltskirchen and between 6% and 16% for the artefacts from Haselbach. A mean saturation water uptake of 28% for the samples from Geboltskirchen and 39% for the artefacts from Haselbach was observed. The dose rate from the material's potassium content was evaluated by neutron activation analysis at the Vienna TRIGA Mark-II research reactor. The dose rate from uranium and thorium was assessed by alpha counting using a Daybreak 582 alpha counter (Daybreak Nuclear and Medical Systems, Inc., Guildford, CT, USA). The environmental dose rate from the burial soil was determined from in-situ gamma dose rate measurements with a portable NaI(Tl) scintillation detector and TL dosimeters buried over sufficiently long times at the sampling points. The measured dose rates have been verified by additional gamma

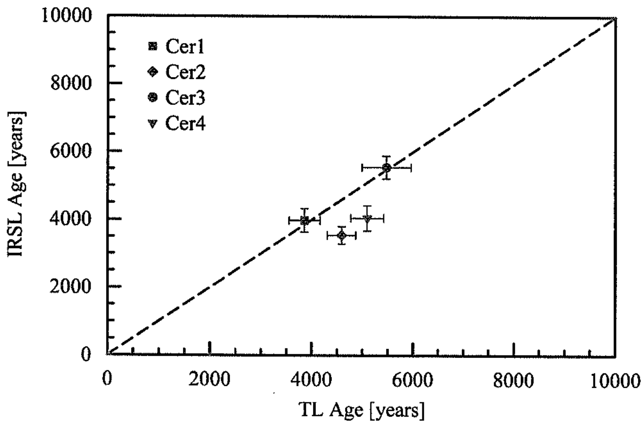

Fig. 4. Comparison of TL and IRSL ages for samples from Geboltskirchen.

Table 1
Corrected equivalent dose, D_E(=ED + I), annual dose rate, $\dot{D}$, a-value and age determined for each sample derived from Geboltskirchen, Upper Austria, using TL and IRSL methods

Sample	TL D_E (Gy)	IRSL D_E (Gy)	TL a-value	IRSL a-value	TL $\dot{D}$ (mGy/a)	IRSL $\dot{D}$ (mGy/a)	TL-Age (a)	IRSL-Age (a)
Cer1	20.45 ± 1.61	30.49 ± 1.27	0.12	0.29	5.31 ± 0.16	7.67 ± 0.27	3854 ± 308	3976 ± 342
Cer2	25.20 ± 0.78	24.07 ± 1.54	0.11	0.20	5.49 ± 0.16	6.80 ± 0.22	4588 ± 282	3539 ± 258
Cer3	25.63 ± 2.30	29.80 ± 0.69	0.13	0.19	4.68 ± 0.14	5.37 ± 0.16	5474 ± 482	5547 ± 336
Cer4	31.30 ± 1.19	42.02 ± 2.26	0.11	0.35	6.14 ± 0.19	10.39 ± 0.39	5096 ± 326	4045 ± 371

Table 2
Corrected equivalent dose, D_E(=ED + I), annual dose rate, $\dot{D}$, a-value and age determined for each sample derived from Haselbach, Lower Austria, using TL and IRSL methods

Sample	TL D_E (Gy)	IRSL D_E (Gy)	TL a-value	IRSL a-value	TL $\dot{D}$ (mGy/a)	IRSL $\dot{D}$ (mGy/a)	TL-Age (a)	IRSL-Age (a)
Has2	10.86 ± 0.71	14.06 ± 0.42	0.13	0.35	3.21 ± 0.10	5.17 ± 0.19	3382 ± 253	2721 ± 223
Has3	13.37 ± 0.40	9.31 ± 1.05	0.13	0.27	3.54 ± 0.11	4.92 ± 0.17	3778 ± 240	1895 ± 161
Has4	8.60 ± 0.71	17.42 ± 1.26	0.14	0.20	3.94 ± 0.12	4.54 ± 0.14	2181 ± 213	3831 ± 374
Has5	6.04 ± 0.63	7.19 ± 0.57	0.08	0.23	2.91 ± 0.08	4.06 ± 0.13	2076 ± 217	1770 ± 151

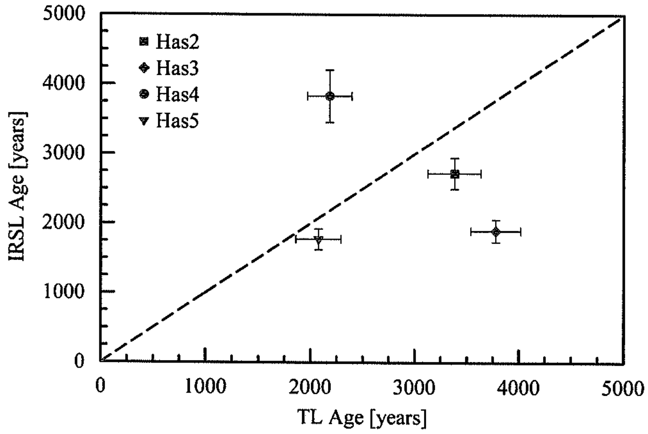

Fig. 5. Comparison of TL and IRSL ages for samples from Haselbach.

spectrometric analysis of the burial soil. The dose rate from cosmic rays were known from the Austrian Radiation Atlas (Strahlenkarte Österreichs) published by the Federal Ministry for Health and Environment. It is likely that the gamma dose rate changed throughout burial time due to changes in the thickness of the overlying layers (Aitken, 1985). This introduces a systematic uncertainty in the assessment of the external gamma dose rate. Its effect on the overall uncertainty of the dose rate is however reduced by the relative larger contributions of the well quantifiable alpha and beta dose rate so that it is not anticipated that the conclusions drawn in Sections 3 and 4 will be affected.

3. Results and discussion

3.1. Samples from Geboltskirchen

The dating results from Geboltskirchen are given in Table 1. TL age determination for the samples showed a high degree of variability between 3854 ± 308 and 5474 ± 482 years, depending on the burial depth from which the fragment has been removed. The statistical uncertainties ranged between 6% and 9%. IRSL measurements of the same pottery revealed an age in the range between 3539 ± 258 and 5547 ± 336 years with statistical uncertainties between 6% and 10%. For the majority of cases, good agreement between the TL and IRSL age was observed (Fig. 4). The achieved data suggest that the artefacts from Geboltskirchen were burnt in the Middle and Late Neolithic period.

3.2. Samples from Haselbach

The dating results from Haselbach are given in Table 2. TL ages of the Haselbach samples ranged from 2076 ± 217 to 3778 ± 240 years with statistical uncertainties between 6% and 11%. IRSL ages of the same fragments were determined to be between 1770 ± 151 and 3831 ± 374 years with uncertainties between 8% and 10%. Agreement between TL and IRSL dating is not as good as for the pottery from Geboltskirchen (Fig. 5). The achieved data suggest that the artefacts from Haselbach were fired in the Early Bronze Age and Late Iron Age.

4. Conclusions

Fragments of ancient pottery from Geboltskirchen and Haselbach, Austria, were dated by means of TL and IRSL analysis. Comparison of the TL and IRSL ages shows good agreement for the samples from Geboltskirchen, while the larger deviations found for the artefacts from Haselbach are related to significant feldspar content. The BG39 optical filter employed for IRSL measurements suppresses the potential feldspar UV emission at 280 nm which is known to be thermally unstable (Krbetschek et al., 1997). This emission is fully detected in TL but only a minor part falls into the IRSL detection window. Careful investigation of the samples may exclude anomalous fading as a source for the observed discrepancies between TL and IRSL ages. The Geboltskirchen site misses a correlation between burial depth and age of the fragments. A preliminary theory for the observed variability of ages (harmonized with geologists from the Montanuniversität Leoben) confirms the previous suggestion of large underground fires of substantial seams of lignite coal. The generated heat baked the nearby sediments to a form of natural brick. As these fires are usually burning over considerably long periods of several hundred to several thousand years, natural erosion and collapsing surfaces may lead to shifting of layers in a way that the artefacts found at lesser depths are older.

References

Aitken, M.J., 1970. Physics applied to archaeology I. Dating. Rep. Prog. Phys. 33 (3), 941–1000.

Aitken, M.J., 1985. Thermoluminescence Dating. Academic Press, London, 351pp.

Berger, T., Hajek, M., Primerano, W., Vana, N., 2002. Thermoluminescence dating of archaeological artefacts from Middle Neolithic, Bronze Age and the Roman Empire Period. Radiat. Prot. Dosim. 101 (1–4), 363–365.
Krbetschek, M.R., Götze, J., Dietrich, A., Trautmann, T., 1997. Spectral information from minerals relevant for luminescence dating. Radiat. Meas. 27 (5/6), 695–748.
Li, S.-H., 1991. Removal of the thermally unstable signal in optical dating of K-feldspar. Ancient Thermoluminescence 9 (2), 26–29.
Zimmerman, D.W., 1971. Thermoluminescent dating using fine grains from pottery. Archaeometry 13, 29–52.