

Dating Ancient Remains by Thermoluminescence: Implications of Incompletely Burnt Bricks

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Received 24 February 2010 Accepted 27 April 2010

Abstract

In this study, the feasibility of thermoluminescence dating for ancient burnt bricks related to archaeometry are investigated at the Thung Tuk archaeological site. All samples were prepared by the quartz inclusion treating technique and evaluate equivalent dose by using the regeneration technique. The natural radionuclide concentration of uranium (U), thorium (Th) and potassium (K) were determined by using gamma ray spectrometry for natural dose rate assessment. The TL-dating results of brick pieces indicated two different age ranges –one around 840-1,500 years BP and another at about 2,800 years BP. The younger date range corresponded quite well to the previous chronological results of the site (1,000-1,300 year BP from antique typology and 1,070-1,310 years BP from conventional radiocarbon dating) whereas the contrasting old date of brick sample had been subjected to incomplete burning of brick during the production process. In final, we concluded that the age of the Thung Tuk archaeological site is about 840-1,500 years BP based on TL dating results of bricks.

Keywords: Thermoluminescence dating, TL, brick, Thung Tuk, archaeological site, southern Thailand

1. Introduction

Up to the present, the possibility of applying TL dating to different fields and materials has been explored. In particular, good results have been obtained in dating burnt flints (Martini et al., 2001; Mercier and Valladas, 2003), metallurgical slag (Krbetschek et al., 1997; Haustein et al., 2001), clay-cores of bronzes (Marco and Emanuela, 2001), volcanic lava (Takashima and Watanabe, 1994; Bonde et al., 2001), stalagmitic calcite (Franklin et al., 1990; Roque et al., 2001), archaeological pottery (Zimmerman, 1971; Abdel-Wahab et al., 1996; Yanchou et al., 1998; Yaroslav et al., 2001), ceramics (Guibert et al., 1994; Bougrov et al.,

1995; Robertson and Prescott, 1998; Zacharias et al., 2005), fault related materials (Hutton et al., 1994; Toyoda et al., 2000) and also geological sediment (Aitken, 1985 ; Taylor and Aitken, 1997).

As well as for these TL-dating applications, TL dating of burnt bricks from ancient remains is also of great interest, particularly in archaeological building sites whose ages are from prehistoric times (with no written records to document the story of the past), in the absence of antique typology and beyond the upper limit of 40,000 years that is known as the radiocarbon barrier (Roberts et al., 1994). Brick dating is meaningful because the date of brick production implies the date of

the structure built out of burnt bricks. Therefore, the absolute age of brick can be a representative image of the chronology of ancient community construction (Vieilleveigne et al., 2000). Though the TL concept is analogous to the dating of ceramics, archaeological potteries or sediment etc., the TL dating of bricks is still rarely discussed and is less widespread (Cechak et al., 2000). Particularly in Thailand, a brick date has only been reported by Changkian and Kaewtubtim (1999). They applied TL dating with an ancient brick from Yarang historical site, Pattani province, southern Thailand. The TL age of a brick piece was 538 ± 15 years BP which was in line with the 445 ± 85 years BP conventional radiocarbon date determined by Reotrit (1987). Due to the current scarcity of research into TL dating of brick, the prime objective of this study is to prove and ascertain the reliability of TL dating within the framework of ancient remains dating from the representative brick samples at the TT.

2. The site and significance

The Thung Tuk (TT) archaeological site is located in southern Thailand, between latitude $8^{\circ}49'N$ and longitude $98^{\circ}16'E$ (Figure 1). The site is at Ko Khao Island between the ends of Muang Thong Canal and Thung Tuk Canal which is the mouth of the Takua Pa River area. The significance of the site rests on the fact that it contains a rich collection of artifact assemblage representative of ancient community cultures and was well known for a long time as one of the most significant communities and trading points along the Andaman coast and Strait of Malacca (Wales, 1947; Srisuchart, 1986).

The majority of archaeological experts have resolved that the TT was the location of an ancient seaport town with which Indians, Arabians, and Malaysians were so well acquainted due to its being an important spice market situated on the west bank of the Malayan Peninsula (Srisuchart, 1986). The TT is located in a suitable locality having appropriate the natural circumstances to anchor in safety from big storms. In addition it has perfect natural resources since it is situated near to the deep open sea where big vessels can gain access conveniently and yet is located at the mouth of the Takua Pa River which is also the center of water communication.

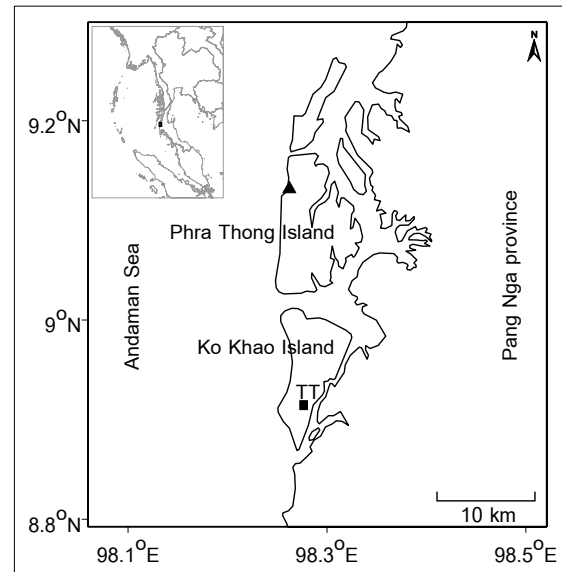


Figure 1. Map of southern Thailand showing the location of Ko Khao Island. Thung Tuk archaeological site is located in the southern part of the island (black square).

Previous research at the TT was commenced by Wales (1947) who explored the Thung Tuk area and discovered historical remains constructed from brick and vestigial remains, such as pottery from China, glassware from Persia, local earthenware and a large number of colored beads (Figure 2).

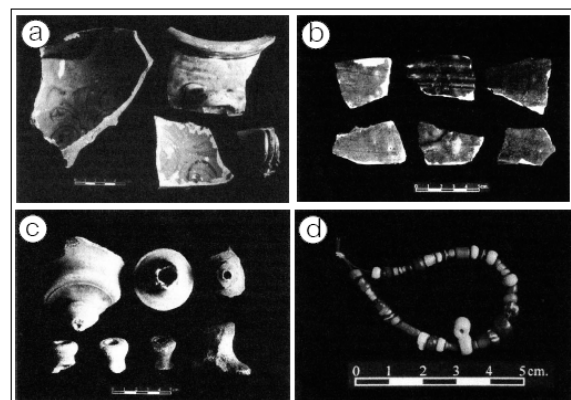


Figure 2. Some of the ancient antiques that were discovered at the Thung Tuk archaeological site referred relative age; (a) Chinaware, (b) Persia glassware, (c) Local earthenware, and (d) Bead.

After that, Srisuchat et al. (1986) excavated and identified the age of the TT by artifact typology. They summarized that the artifact assemblage was indicative of the Tang's dynasty (12-15 Buddhist periods). Then, between 1988 and 1989, Chaisuwan and Naiyawatt (2002) reviewed the literatures and renovated the ancient remains at the site. They also proposed the first scientific date information of about 1,070-1,310 years BP using the conventional radiocarbon dating method.

According to current field investigation, the boundary of the Thung Tuk town plan is mapped as shown in Figure 3. Geologically, the area is a flat plain covered mainly with sand, trees and small bushes, some spots are barren areas with sporadic artificial remains. At least eight archaeological remains look similar to the basements of a building or sanctuary exposed in this area. In addition, pieces of chinaware, local earthenware, colored glass containers as well as various types and colors of beads and Indian coins have been found scattered on the ground of these archaeological remains.

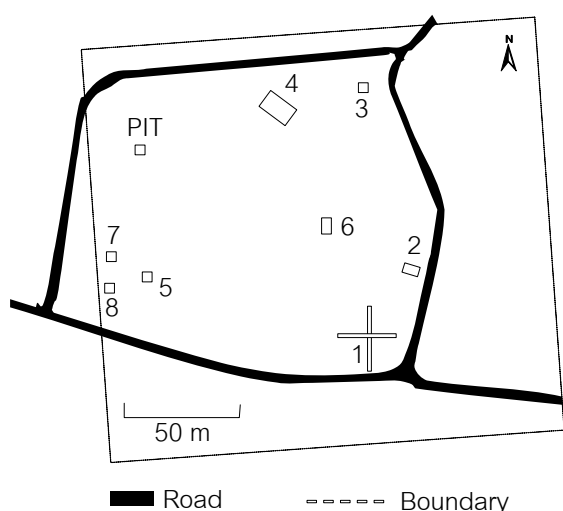


Figure 3. Town plan of TT showing location of 8 ancient remains (modified from Chaisuwan and Naiyawatt, 2002).

3. Methodology

3.1 Sample collection

Due to the specific TL characteristics of bricks, it has become necessary to analyze brick samples from many locations in order to obtain a relevant date for the building of a site (or, more precisely, the firing of its bricks). Consequently, a total of eight brick samples from the basement of

ancient remains were collected from five selected locations namely 1, 3, 4, 6, and 8 (see Figure 3). The lists of sample collection are described in Table 1.

3.2 Sample preparation and measurement

All samples taken from the site as a lump (with the dirt around them) were placed in dark plastic bags to prevent light exposure. The samples were transported and opened in the laboratory under red light. Brick preparation started with removing a 2 mm layer from the outermost surface of the brick mass, as suggested by (Baillif and Holland, 2000). Its outer layer should discard because it may reduce the luminescence level due to the effect of sunlight effect and soil contamination on the brick surface. Through cutting across the mass of brick, we found that some brick samples showed contrasting colors in brick mass (Figure 4).

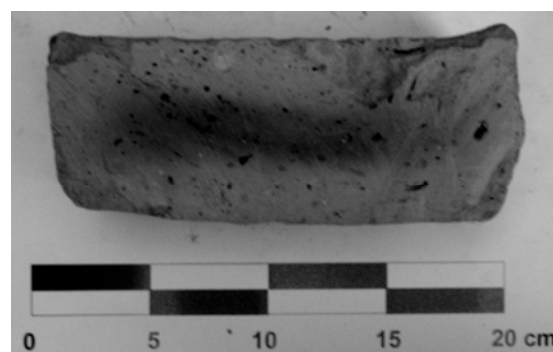


Figure 4. Photograph of a brick sample showing contrasting colors in brick mass after firing.

We assumed that the color change was due to incomplete heating of the brick mass in the production process. This also affected the non-resetting TL signal in the inner part of brick. Therefore, we decided to evaluate equivalent dose by using both of the specific outer part of brick (sample TT1-1, TT3-3, TT3-4, TT4-1, TT4-3, TT6-1, and TT8-1) and also the mixed mass of contrasting color brick (sample TT3-1) to prove our theories.

After the brick pre-treatment procedure was complete, the brick fragments were subjected to the same process using the method described by Takashima and Watanabe

(1994). First of all, they were mechanically crushed with a rubber hammer. Part of the crushed samples was sieved into two portions – one having a grain size that would pass through a 20 mesh ($<841\mu\text{m}$) for annual dose assessment. Then the remainder was obtained by re-grinding the fragments in an agate mortar and sieving into a grain size range of 74-250 μm for equivalent dose evaluation (Fleming, 1970).

In the case of the equivalent dose evaluation procedure, quartz mineral was extracted to represent the TL-sensitivities in our TL dating study following the method described by Bailiff and Holland (2000). HF etching (to eliminate feldspar composition) followed by washing in HCl (to eliminate organic composition) and then re-washing in distilled water of the sample continuously. After drying the samples using 40-50 °C heating, the dark minerals (e.g. zircon, garnet, and metallic minerals) were removed by using a magnetic separator (Frantz's isodynamic magnetometer). The contaminated mineral of the final treated quartz was checked by using X-ray diffraction analysis (XRD). As the XRD traces, the main minerals observed after chemical treatment should at least 95% quartz.

In TL-sensitivity measurement step, the regeneration technique described by Takashima and Honda (1998) was applied for this TL dating approach.

Luminescence measurements were performed with 0.1 mg individual aliquots of treated pure quartz, deposited as a monolayer onto stainless steel discs. All aliquots were measured by TL performed with the standard commercial Risø (Denmark) and a high-sensitivity home-made apparatus from Akita University, Japan, developed by Takashima and Honda (1989).

In radioactive concentration assessments, the prepared 300-g samples were contained in a plastic vessel. Chemical analyses were performed by gamma ray spectrometry at Akita University, Japan. The annual dose values computed by transform the radioactive (K, U and Th) concentrations with the standard table from Bell (1979).

4. Results and discussions

Based on the equivalent dose evaluation process, the general TL characteristics of samples were particularly suitable for dating. The TL glow curves typical of the samples discussed herein are briefly shown in Figure 5a. To select the suitable stable sensitivity, a plateau test procedure (Aitken, 1985) was employed in a temperature range of 280-320 °C. Normally, the 320°C TL peak was selected for the representative dosimetry (Figure 5b).

For the TL dating of bricks, from the date of construction would be the date of the last firing (Kresten et al., 2003), therefore TL dates and radiocarbon dates would be theoretically equivalent.

Most bricks yielded ages with the range of 840-1,500 years BP, with the exception of one peculiar brick (sample TT3-1) which gave a date of 2,800 years BP (Figure 6). The former dates were in line with the reference age whereas the later was not. Archaeologically, there is no evidence to support the occurrence of earlier communities. Therefore, the specific physical features and the old date results of brick (sample TT3-1) allowed us to “re-think” the traditional brick production procedure. We therefore consider that the apparent age difference can readily be explained as a difference in firing temperature. There is no need to invoke arguments, such as “an older culture” as opposed to “a younger culture”. It is definitely a difference in firing temperature and the older brick date seems to indicate an erroneous result and has become chronologically meaningless.

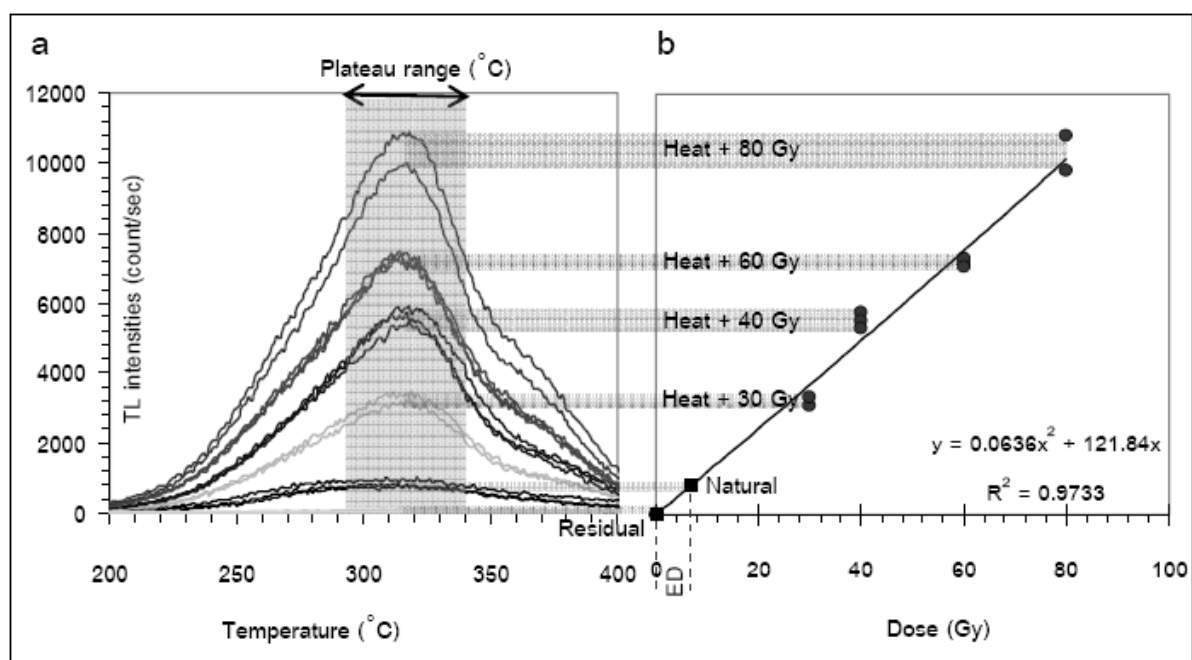


Figure 5. (a) An example of the TL glow curve behavior of brick sample TT1-1 showing TL intensities relates to natural and additional artificial irradiation. (b) Fitted TL dose-growth curves for sample TT1-1 at various irradiation levels.

Table 1. TL dating results of sediment and brick sample from ancient remains, Thung Tuk archaeological site. Wt is water content, AD is annual dose, and ED is equivalent dose.

Sample no.	U ppm	Th ppm	K ₂ O (%)	Wt (%)	AD (Gy/ka)	ED (Gy)	TL date (Year)
TT1-1	10.49	58.78	1.88	9.69	5.83	6.66	1,140 ± 120
TT3-1	9.83	46.58	2.33	14.00	7.27	207	2,800 ± 270
TT3-3	22.35	62.57	1.93	1.55	9.26	11.73	1,270 ± 370
TT3-4	22.35	62.57	1.93	1.55	9.26	14.30	1,500 ± 660
TT4-1	9.18	44.06	2.32	14.00	6.96	7.52	1,100 ± 270
TT4-3	6.99	40.50	1.93	10.77	5.34	5.49	1,000 ± 340
TT6-1	20.85	72.74	2.13	0.28	10.187	11.22	1,100 ± 440
TT8-1	19.58	85.44	2.51	1.09	11.28	9.45	840 ± 300

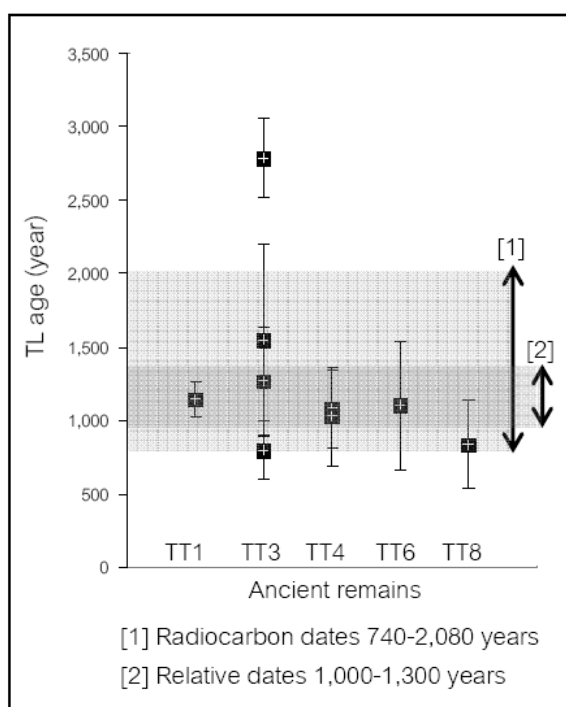


Figure 6. Comparison of TL dates from brick samples with radiocarbon and relative dates.

5. Conclusion and recommendation

From what has been presented in the preceding sections, a concentrated conclusion can be drawn regarding the possible application of TL dating with archaeological materials particularly with ancient burnt bricks. In this TL dating approach, we conclude that TL properties (i.e. glow curve shapes, peak temperatures, sensitivities and sensitivity changes) can be successfully considered as the characterization parameters of ancient burnt bricks. Calibration of interpreted well dates with brick dates and the previous reference dates (radiocarbon and relative dates) from the site were carried on successfully. Most of the brick dating results are in good agreement with those of the chronological evidence. However, the strange old dating results associated with the heterogeneous mass of brick sample reveal that ancient brick has the possibility of being incompletely heated at the time of brick production, particularly, ancient Thai brick. This brick produces a dangerous date when use the whole brick mass to evaluate equivalent dose. The TL dating of incompletely heated brick is therefore indirect.

The age is not obtained from the brick production itself, and caution is needed in the interpretation and presentation of the result.

To approach a high accuracy of TL dating with brick, we propose a new brick treatment trick for TL dating. After eliminating a few millimeters of the outer rim to avoid exposed grain along the travel time from the site to laboratory (Bailiff and Holland, 2000), a few of the outmost portions should be used for equivalent dose evaluation. Then, the remaining portions can be used for annual dose assessment through determination of radioactive element concentration (U, Th, and K).

Finally, we conclude that TL dating in ancient burnt brick appears more likely to produce high precision, as with other archaeological objects, such as pottery. Moreover, TL dating with brick is more powerful than the other materials in the sense that the mass of brick is sufficient for the TL dating required. The providing of sufficient samples taken from a homogenous brick and sufficient thickness can minimize radiation non-equilibrium caused by the proximity of the external radiation environment to the brick sample.

6. Acknowledgements

Thermoluminescence dating has been financially supported by The 15th Regional Office of Fine Arts, Phuket which is most gratefully acknowledged. We also thank the EATGRU research centers, Chulalongkorn University for providing the TL facilities. Thanks also extended to the Commission of Higher Education and Ratchadapiseksomphot Endowment Fund (CC508B), the Faculty of Science, Chulalongkorn University and the Thai Government Stimulus Package 2 (TKK2555:PERFECTA) for the research funds to S.P. We are grateful for the comments from many anonymous referees.

7. References

- Abdel-Wahab, M. S., El-Fiki, S. A., El-Fiki, M. A., Gomaa, M., Abdel-Kariem, S., El-Faramawy, N., 1996. Annual dose measurement and TL dating of ancient

- Egyptian pottery. *Radiation Physics and Chemistry*; 47 (5), 697-700.
- Aitken, M.J., 1985. Thermoluminescence dating. London: Academic Press.
- Aitken, M. J., 1990. Science-based dating in archaeology. London: Longman.
- Bailiff, I. K., Holland, N., 2000. Dating bricks of the last two millennia from Newcastle upon Tyne: a preliminary study. *Radiation Measurements*; 32 (5-6), 615-619.
- Bell, W.T., 1979. Attenuation factors for the absorbed radiation dose in quartz inclusions for thermoluminescence dating. *Ancient TL*; 8, 2-13.
- Bevilacqua, F., Chiavari, C., Di Francesco, C., Migliorini, E., Martini, M., Sibilina, E., 1999. Thermoluminescence analysis in historical architecture: application to the ornamentations "in cotto" of the atrium of the Abbey of Pomposa. Sixth International Conference on Non Destructive Testing and Microanalysis for the Diagnostics and Conservation of the Cultural and Environmental Heritage, Roma, 17–20 May 1999.
- Bonde, A., Murray, A., Friedrich, W. L., 2001. Santorini: Luminescence dating of a volcanic province using quartz. *Quaternary Science Reviews*; 20 (5-9), 789-793.
- Bougrov, N. G., Vlasov, V. K., Kiryukhin, O. V., Fatkulbayanova, N. L., 1995. Thermoluminescence measurements of ceramic samples from accidentally polluted territory of southern Urals. *Radiation Measurements*; 24 (4), 493-498.
- Cechak, T., Dynybyl, V., Gerndt, J., Kanaval, J., Kubelik, M., Musilek, L., Pavlik, M., Zyma, J., 2000. Thermoluminescence-Reader for Dating of Brick Structures, CTU Prague, Workshop 2000, Paper JAD/18.
- Cechak, T., Gerndt, J., Hirsl, P., Jirousek, P., Kanaval, J., Kubelik, M., Musilek, L., 2001. Automation of TL brick dating by ADAM-1. *Radiation Physics and Chemistry*; 61 (3-6), 729-731.
- Chaisuwan, B., Naiyawatt, R., 2002. Thung Tuk ancient seaport. Phuket. Phuket: 15th Regional Office of Fine Arts.
- Chankian, S., Kaewtubtim, P. 1999. TL dating of ancient pottery of the Yarang historical site, Amphur Yarang, Pattani Province. *Songklanakarin Journal of Science and Technology*; 21, 347-353.
- Colman, S. M., Pierce, K. L., 2000. Classifications of quaternary geochronologic methods. In J. S. Noller, J. M. Sowers, and W. R. Lettis (eds.), *Quaternary Geochronology: Methods and Applications*, pp2-5. Washington D.C.: American Geophysical Union.
- Colman, S. M., Pierce, K. L., Birkeland, P. W. 1987. Suggested terminology for quaternary dating methods. *Quaternary Research*; 28, 314-319.
- Duller, G.A.T., Botter-Jensen, L., Murray, A.S., 2000. Optical dating of single sand-sized grains of quartz: sources of variability, *Radiation Measurements*; 32, 453–457.
- Feathers, J. K. 2003. Use of luminescence dating in archaeology. *Measurement Science and Technology*; 14, 1493-1509.
- Franklin, A. D., Hornyak, W. F., Pagonis, V., Kristianpoller, N., 1990. Thermoluminescence study of annealing a geological calcite. *International Journal of Radiation Applications and Instrumentation. Part D. Nuclear Tracks and Radiation Measurements*; 17 (4), 517-523.
- Guibert, P., Schvoerer, M., Etcheverry, M. P., Szepertyski, B., Ney, C., 1994. IXth millenium B.C. ceramics from Niger: detection of a U-series disequilibrium and TL dating. *Quaternary Science Reviews*; 13 (5-7), 555-561.
- Haustein, M., Krbetschek, M. R., Trautmann, T., Roewer, G., Stolz, W., 2001. A luminescence study for dating archaeometallurgical slag. *Quaternary Science Reviews*; 20 (5-9), 981-985.
- Hutton, J. T., Prescott, J. R., Bowman, J. R., Dunham, M. N. E., Crone, A. J., Machette, M. N. Twidale, C. R., 1994. Thermoluminescence dating of Australian palaeo-earthquakes. *Quaternary Science Reviews*; 13 (2), 143-147.
- Kalchgruber, R., Göksu, H. Y., Hochhäuser, E., Wagner, G. A., 2002. Monitoring environmental dose rate using Risø

- TL/OSL readers with built-in sources: recommendations for users. *Radiation Measurements*; 35, 585-590.
- Krbetschek, M. R., Götze, J., Dietrich, A., Trautmann, T., 1997. Spectral information from minerals relevant for luminescence dating. *Radiation Measurements*; 27 (5-6), 695-748.
- Kresten, P., Goedicke, C., Manzano, A. 2003. TL-dating of vitrified material. *Geochronometria*; 22, 9-14.
- Martini, M., Sibilìa, E., 2001. Radiation in archaeometry: archaeological dating. *Radiation Physics and Chemistry*; 61 (3-6), 241-246.
- Martini, M., Sibilìa, E., Croci, S., Cremaschi, M., 2001. Thermoluminescence (TL) dating of burnt flints: problems, perspectives and some examples of application. *Journal of Cultural Heritage*; 2 (3), pp 179-190.
- Mercier, N. Valladas, H., 2003. Reassessment of TL age estimates of burnt flints from the Palaeolithic site of Tabun Cave, Israel. *Journal of Human Evolution*; 45 (5), 401-409.
- Murray, A. S., Mejdahl, V., 1999. Comparison of regenerative-dose single-aliquot and multiple-aliquot (SARA) protocols using heated quartz from archaeological sites. *Quaternary Geochronology*, 18, 223-229.
- Reotrit, S., 1987. Archaeological exploration in the Yarang historical site, Amphur Yarang, Pattani Province. Pattani: Center of Southern Thailand Study, Songklanakarin University, Pattani.
- Roberts, R. G., 1997. Luminescence dating in archaeology. *Radiation Measurements*; 27, 819-892.
- Roberts, R. G., Jones, R., Smith, M. A. 1994. Beyond the radiocarbon barrier in Australian prehistory. *Antiquity*; 68, 611-616.
- Robertson, G. B., Prescott, J. R., 1988. The THAI Ceramics Archaeological Project: TL characteristics of the artifacts. *International Journal of Radiation Applications and Instrumentation (Part D). Nuclear Tracks and Radiation Measurements*; 14 (1-2), 299-305.
- Roque, C., Guibert, P., Vartanian, E., Bechtel, F., Schvoerer, M., 2001. Thermoluminescence-dating of calcite: study of heated limestone fragments from Upper Palaeolithic layers at Combe Sauniere, Dordogne, France. *Quaternary Science Reviews*; 20 (5-9), 935-938.
- Singhvi, A. K., Wagner, G. A., 1986. Dating Young Sediments. In A.J. Hurford, E. Jager, and I. A. M. Tencate (eds.), pp. 159-197. Bangkok.
- Srisuchart, T., 1986. Ta Kua Pa: Ancient town. Southern culture Encyclopedia 3. Center of southern culture study. Srinakharinwirot University, Song Khla province. p. 1217.
- Takashima, I., Honda, S. 1989. Comparison between K-Ar and TL dating results of pyroclastic flow deposits in the Aizutajima area, Northeast Japan. *Journal of Geological Society*; 95, 807-816.
- Takashima, I., Watanabe, K. 1994. Thermoluminescence age determination of lava flows/ domes and collapsed materials at Unzen volcano, SW Japan. *Bulletin of the Volcanological Society of Japan*; 39, 1-12.
- Taylor, R.E., Aitken, M.J., 1997. *Chronometric Dating in Archaeology*, Plenum Press, London.
- Toyoda, S., Rink, W. J., Schwarcz, H. P., Rees-Jones, J., 2000. Crushing effects on TL and OSL on quartz: relevance to fault dating. *Radiation Measurements*; 32 (5-6), 667-672.
- Vieillellevigne, E., Guibert, P., Bechtel, F., 2000. Exploration of the possibilities of dating mediaeval construction's building by optically stimulated luminescence: the case of the bricks citadel of Termez (Ouzbekistan). Institut de Recherche sur les Archéomatériaux, UMR 5060 CNRS - Université Bordeaux 3. Centre de Recherche en Physique Appliquée à l'Archéologie (CRP2A), Maison de l'Archéologie, 33607 Pessac cedex, France. 2 p.
- Wales, D. C., Wales, H. G. Q., 1947. Further work on Indian sites in Malaya. *JMBRAS*; 5, 1-2.
- Yanchou, L., Jingzhao, Z., Xueli, X. J., 1988. TL dating of pottery sherds and baked soil from the Xian Terracotta Army Site, Shaanxi Province, China. *International*

- Journal of Radiation Applications and Instrumentation* (Part D) Nuclear Tracks and Radiation Measurements; 14 (1-2), 283-286.
- Yaroslav, V. K., Hall, S., Tite, M. S., Bailey, R., O'Malley, J. M., Medvedev, V. E., 2001. Radiocarbon and thermoluminescence dating of the pottery from the early Neolithic site of Gasya (Russian Far East): initial results. *Quaternary Science Reviews*; 20 (5-9), 945-948.
- Zacharias, N., Buxeda i Garrigós, J., Mommsen, H., Schwedt, A., Kilikoglou, V. 2005. Implications of burial alterations on luminescence dating of archaeological ceramics. *Journal of Archaeological Science*; 32 (1), 49-57.
- Zimmerman, D. W. 1971. Thermoluminescence dating using fine grains from pottery. *Archaeometry*; 13 (1), 29-56.

Received 24 February 2010

Accepted 27 April 2010