Thermoluminescence dating analysis at the site of an ancient brick structure at Pengkalan Bujang, Malaysia

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HIGHLIGHTS

• Thermoluminescence analysis of shards sample from Pengkalan Bujang.
• Fine grain technique and additive dose method were applied to determine paleodose.
• Measuring concentration of U, Th and K in the samples and their surroundings.
• TL ages obtained indicative of the last firing of the shards.

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ABSTRACT

Bujang Valley is a well-known historical complex found in the north-west of peninsular Malaysia; more than 50 ancient monuments and hundreds of artefacts have been discovered throughout the area. The discovery of these suggests Bujang Valley to have been an important South East Asian trading centre over the period from the 10th to 14th centuries. Present work concerns thermoluminescence (TL) dating analysis of shards collected from a historic monument located at Pengkalan Bujang in Bujang Valley. All the shards were prepared using the fine grain technique and the additive dose method was applied in determining the paleodose of each shard. The annual dose rate was obtained by measuring the concentration of naturally occurring radionuclides (U, Th and K) in the samples and their surroundings. The TL ages of the shards were found to range between 330±21 years and 920±69 years, indicative of the last firing of the bricks and tiles from which the shards originated, some dating back to the period during which the historical complex remained active.

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1. Introduction

The archaeological structures that are to be found in Bujang Valley, located in the north west of peninsula Malaysia, have been the subject of study for more than one hundred and fifty years, Colonel James Low discovering the first such monument in the 1840s (Shuhaimi and Mohd Yatim, 1990). Low suggested it to form part of an ancient Hindu colony (Wales, 1940). To-date, more than 80 heritage sites have been discovered in the Bujang Valley, a number of these being found at Pengkalan Bujang. Archaeological investigation in the area of Pengkalan Bujang began in 1959 with the work of Alastair Lamb, providing stratigraphic information and cultural material, supporting the hypothesis of Lamb that Pengkalan Bujang had in fact been a major entrepot trading centre (Shuhaimi and Mohd Yatim, 1990). Prominent among the archaeological sites discovered in the Pengkalan Bujang area are Sites 19, 20, 21, 22 and 23. Here it should be noted that the giving of site numbers in the Bujang Valley was introduced by Quaritch–Wales in 1940, the dense clustering of structures creating difficulties in providing a name to each individual site (Shuhaimi and Mohd Yatim, 1990).

Although archaeological activities in Pengkalan Bujang area began many decades ago, nevertheless the chronology of sites in Pengkalan Bujang remains poorly understood, particularly in regard to Site 23. This particular site, situated on the sandbanks of the Bujang River, contains the ruins of an ancient monument of...
brick-construction, first identified by Quaritch–Wales in his 1936–1937 survey. Subsequent excavation began in 1985, remaining active to present date. The paucity of archaeological data, including lack of archaeometric information for the site has represented a major limitation in seeking absolute dating of the structures.

Various Malaysian workers have made attempts to date items from the Pengkalan Bujang sites, with both unpublished work (Jasman and Mahat, 2009; Rashid et al., 2009) as well as published records existing (Mahat et al., 2010), use being made of the thermoluminescence (TL) dating method. The results show clear disparity between the age obtained by the TL method and the age estimated by archaeologists. In this study, we have conducted further dating at Site 23, measurements being carried out on shards using thick alpha source counting and the TL dosimeter CaF$_2$:Dy (commonly referred to by the product label TLD 200), as well as through atomic emission spectroscopy (the latter not being discussed herein).

2. Archaeological site

2.1. Bujang Valley

Bujang Valley, one of the most important archaeological sites in Peninsular Malaysia, is a large area in Merbok, Kedah, lying between latitude 5° 44′ N and longitude 100° 24′ E. Bujang Valley descends from the Jerai Mountain and stretches southward towards the Muda River and Cherok Tokun in Seberang Perai, meeting with the Straits of Melaka. Geomorphologically, Bujang Valley is made up of mountains, denudational hills and fluvial and marine deposits (Shuhaimi and Mohd Yatim, 1990).

Evidence for old Kedah suggests this ancient kingdom to have begun its development in the Bujang Valley. Given the existence of a number of written records, including those from Chinese and Arabian seafarers and also literature from India, the importance of the Bujang Valley sites in development of old Kedah is well-established. In an item of Tamil literature, the Bujang Valley has been referred to as Kataha and Kadaram, also being written as Kidaram. Scholars have come to the conclusion that these Indian toponyms refer to Bujang Valley. Additionally, the written record of I-Ching, a famous Chinese pilgrim, describes his voyage in 671 A.D., mentioning the location of Chieh-Ch’a (Kedah) in relation to Sri-vijaya and Malayu (written now as Melayu). From the record of I-Ching, Chieh-Ch’a was an entrepot in Kedah, archaeological evidence identifying the entrepot as the Bujang River town with its anchorage in Kuala Merbok (Braddell, 1980).

2.2. Pengkalan Bujang site

Definitely, Pengkalan Bujang is located at Kampung Pengkalan Bujang in the Kuala Muda District of present-day Kedah; it is situated along the banks of the middle reaches of the Bujang River (Fig. 1), river in Bahasa Malaysia being referred to as sungai. It is believed that in ancient times the sites were situated rather closer to the river and the sea (Shuhaimi and Mohd Yatim, 1990).

According to archaeological studies, Pengkalan Bujang was a major entrepot trading centre over the period from the 11th to 14th centuries (Lamb, 1961). Thousands of fragments of Chinese porcelain from the period of the Sung and Yuan dynasty (960–1368) have been discovered in Pengkalan Bujang, as have at least one hundred small bottles, items once widely imported from the Middle East, and numerous beads of glass, agate and terra cotta, some of which seem to have originated from India as well as perhaps Java (Lamb, 1980). All of these findings are supportive of Pengkalan Bujang being an important trading centre in the Malay region.

2.3. Site 23

In the TL analysis of present study, only one site was chosen from the Pengkalan Bujang area, namely Site 23. The site was
discovered by Quaritch-Wales in 1936. The brick structure at Site 23 appears to have been based on a square plan 2.13 mx2.13 m (Shuhaimi and Mohd Yatim, 1990).

In addition to the paucity of data on its chronology, an argument has arisen among Malaysian archaeologists as to the role of this site in ancient times. Some believe the structure to have been built as a Hindu temple, acting as a Vihara (a monastery) (Sabtu, 2014). Based on the structure of the site, others believe the structure to have been a mosque, a part of the structure being suggested to be a minbar (a place where the imam stands to deliver the khutbah or sermon), it also being directed to the kiblah, i.e. towards Makkah (Jaafar, 2013). Archaeological research on this site continues to address such interests (Figs. 2 and 3).

3. Thermoluminescence dating

The phenomenon of TL has been found useful in dating fired ceramics, including bricks, pottery and tiles. The technique, initially introduced by Aitken and his group in a paper in Nature (Aitken et al., 1964), has now been popularly applied for in excess of fifty years, remaining highly relevant in determining the age of such anthropomorphic artefacts. In Malaysia, while use of the TL dating is still very much in a nascent stage of development, several previous attempts have been made to use the technique in dating of Bujang Valley items, the TL data showing a range of ages from 248 ± 65 to 464 ± 155 years (Rashid et al., 2009; Mahat et al., 2010).

The TL dating technique is based on measurement of the intensity of light released as a result of heating of the sample, the thermal treatment releasing electrons trapped at defect centres within the medium. The firing of the pottery or tile from which each shard will have originated will have released all previously trapped electrons, resetting the chronological clock. Subsequent to firing, growth of the TL signal will be proportional to the age of the sample, a reflection that the TL signal depends on the natural radiation exposure of the sample (Aitken, 1998). The greater the duration of sample exposure, the greater the TL signal. The fundamental age equation is given by Aitken (1999), as follows:

$$\text{Age} = \frac{\text{Paleodose} \text{ (Gy)}}{\text{Annual dose} \text{ (Gy/year)}} = \frac{\text{NTL}}{sD}$$

(1)

where NTL is the natural thermoluminescence from the sample, s is the TL sensitivity of the sample and D is total dose-rate. It is to be noted however that while Eq. (1) is a simplified equation that outlines the basic TL dating approach, in practise it cannot be used directly as it does not reflect the different TL sensitivities towards the various types of radiation (alpha, beta and gamma). The detailed equation used in age determination is described in Section 4.3.

4. Experimental methods

4.1. Sampling

The shards studied herein were retrieved from a depth within the presently prevailing level of earth of more than 30 cm. In all but two cases (samples PB23BBL1 and PB23AGF1) the sampling procedures included the wrapping or covering of samples by opaque black plastic, avoiding light exposure that could bleach a fraction of the stored signal, as outlined by Aitken (1974). The consequences of leaving the two particular samples exposed to light will be addressed in the findings, presented below.

4.2. Determination of paleodose ($D_e$)

Details of the shards used for analysis are provided in Table 1. The shards are illustrated in Fig. 4. Each shard was prepared using the fine grain technique for TL analysis (Aitken, 1974). In this, the shard is gently crushed, grains of the size of 1–8 µm being obtained by settling the grains in acetone. Selected grains were then deposited on aluminium discs of 10 mm diameter and thickness 0.5 mm, each disc containing a few milligrams of grain sample. The paleodose, $D_e$ of samples was measured using the additive dose method. This method is a direct comparison of natural TL signal with induced TL signal, the latter obtained by irradiating the samples to a known irradiation dose. Seventeen sample aliquots were prepared; five aliquots were used for determination of natural irradiation of the samples post the firing of the shards, also allowing determination of the supralinearity correction. A further

![Fig. 2. The ruined structure of Site 23 (photo by S.N Sabtu).](image)

![Fig. 3. The part of the structure claimed to be a minbar, directed to the kiblah (photo by S.N Sabtu).](image)

### Table 1

<table>
<thead>
<tr>
<th>Sample no.</th>
<th>Sample type</th>
</tr>
</thead>
<tbody>
<tr>
<td>PB23BBF1</td>
<td>Clay brick</td>
</tr>
<tr>
<td>PB23BBL1</td>
<td>Clay brick</td>
</tr>
<tr>
<td>PB23BB1</td>
<td>Clay brick</td>
</tr>
<tr>
<td>PB23BB2</td>
<td>Clay brick</td>
</tr>
<tr>
<td>PB23AGF1</td>
<td>Clay tile</td>
</tr>
<tr>
<td>PB23AGL1</td>
<td>Clay tile</td>
</tr>
</tbody>
</table>
seven aliquots were used in the preparation of an additive beta irradiation TL growth curve. The remaining five aliquots were used in the construction of an additive alpha TL growth curve to determine the ratio of the effectiveness of alpha particles to that of beta particles in inducing TL, referred to as the $k$ value (Aitken, 1974). The paleodose $D_e$ was determined by extrapolation of TL response against irradiation dose. The TL output from each sample was measured by heating the sample at high temperature at a heating rate of 20 °C/s in a high-purity nitrogen atmosphere, the emitted TL being recorded using an EMI 9635QB photomultiplier fitted with a Corning 7–59 transmission filter and a Chance Pilkington heat filter on the X-Y recorder available at the University of Wollongong. All samples were analysed at a temperature of 350 °C.

4.3. Determination of annual dose

The annual dose received by a sample is due to irradiation by natural radionuclides within the sample and immediate surroundings. The main sources are nuclides from the uranium and thorium decay series, and potassium-40, with further small contributions from rubidium-87 and cosmic rays (Musilek and Kubelik, 2000). The alpha, beta and gamma dose-rates were taken into account in determining the annual dose-rate. The annual dose rate can be determined using equation below:

$$D = s_A D_A + s_B D_B + s_D D_D$$  \hspace{1cm} (2)

where $s_A$, $s_B$, and $s_D$ are TL sensitivity for alpha, beta, gamma and cosmic radiations respectively and $D_A$, $D_B$, $D_D$, and $D_C$ are dose-rate due to alpha, beta, gamma and cosmic radiations respectively. The TL sensitivity of beta, gamma and cosmic radiations are the same but for the alpha radiation the sensitivity is substantially less (by factor of between 3 and 15, depending on mineral concerned) than the other types of radiations (Aitken, 1991). Hence the Eq. (2) becomes:

$$D = s_A D_A + s_B D_B + s_D (D_D + D_C)$$  \hspace{1cm} (3)

Considering the various radiation components, the age equation (Eq. (1)) can be express to:

$$Age = \frac{NTL}{s_A D_A + s_B (D_B + D_D) + s_D (D_D + D_C)}$$  \hspace{1cm} (4)

Defining a factor and an archaeological dose $D_e = NTL / s_P$. Eq. (4) can be rewritten as

$$Age = \frac{D_e}{kD_A + D_B + D_D + D_C}$$  \hspace{1cm} (5)

The internal dose-rates due to alpha and beta dose-rates from the natural radionuclides of uranium, thorium and potassium in the shards were calculated using concentration values of those radionuclides evaluated herein by thick source alpha counting (TSAC) and atomic emission spectroscopy (Aitken, 1985; Shepherd and Price, 1990). The concentrations of uranium and thorium decay chains were determined by using calibrated TSAC over a 42 mm scintillation area. The sample material was dried, finely crushed and sealed in an alpha counting cell and left for 21 days to allow the uranium and thorium decay chains to achieve secular equilibrium. A minimum of 1000 alpha counts were accumulated for each sample, providing a counting uncertainty of 3%. The potassium content was determined by means of atomic emission spectroscopy. A value of $10 \pm 3\%$ was assigned to the average moisture content, and this correction was applied for all samples using the formula of Zimmerman (1971).

The external dose-rate due to the contribution of gamma and cosmic rays were determined using in situ measurements. High-sensitivity CaF2: Dy dosimeters (TLD-200) phosphors were used to determine the combined gamma and cosmic dose rate at the site. The phosphor dosimeters were placed in small tubes and buried at the site for 6 months. Subsequently the irradiation energy absorbed by the dosimeters from its surrounding was analysed using a standard Harshaw 3500 TLD reader.

5. Results and discussions

The internal annual doses from alpha and beta contribution were determined from the concentrations of U, Th and K present in the samples, the values being obtained at secular equilibrium for both the U and Th decay chains. The concentrations were converted to dose-rate by using conversion factors data from Adamiec and Aitken (1998). The dose-rate obtained was corrected using the $k$-value, the ratio of the effectiveness of alpha particles in inducing TL to that of beta particles and it was determined from growth curves of beta and alpha irradiation (Aitken, 1985). The $k$ values for each sample are tabulated in Table 2. The dose-rate also was calculated taking into account the moisture content and the contribution of cosmic ray (Zimmerman, 1971; Aitken, 1985).

Additionally, a rubidium-87 content of 100 ppm was assumed for all samples (Vora et al., 2002). The internal dose-rate for all of the analysed samples proved to be relatively high, predominately...
as a result of the uranium, thorium and potassium present within the samples. In line with expectation, the content of rubidium-87 and cosmic radiation was found to be relatively minor. Using TL dosimetry (CaF₂: Dy, TLD 200), the average of the gamma and cosmic dose rates from the sampling site was found to be 1.48 ± 0.09 mGy/year. The totals of annual dose (internal and external) for all shards are shown in Table 2. Equation 5 has been used to determine the age of each shard (Table 2). The uncertainties in the TL age analysis based on the assessment of systematic errors and random errors as outlined by Aitken and All-

dred (1972). All uncertainties are quadratically summed to obtain the overall uncertainty for each shard.

It is perhaps of no surprise, given the period over which the site was active, that the majority of the age estimates from this TL analysis are less than that estimated by archaeologists for the initial establishment of the area (the so-called target event). It is to be noted that brick and tile production was almost certainly on-

ternal establishment of the area (the so-called target event). It is to

The values shown assume secular equilibrium for both U and Th decay chains. The uncertainty levels indicated represent one standard deviation.

### Table 2
Results of TL analysis.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Plateau region (°C)</th>
<th>Paleodose (Gy)</th>
<th>Potassium content (%)</th>
<th>Specific activity (U + Th) (Bg/kg)</th>
<th>k value (α/β efficiency)</th>
<th>Annual dose rate (mGy)</th>
<th>Age (Year)</th>
<th>Date (Century)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PB23BBF1</td>
<td>300–475</td>
<td>4.01 ± 0.14</td>
<td>1.430 ± 0.05</td>
<td>167 ± 4</td>
<td>0.138 ± 0.010</td>
<td>6.929 ± 0.233</td>
<td>580 ± 40</td>
<td>15th</td>
</tr>
<tr>
<td>PB23BBL1</td>
<td>275–450</td>
<td>3.25 ± 0.23</td>
<td>1.420 ± 0.05</td>
<td>191 ± 3</td>
<td>0.121 ± 0.010</td>
<td>7.083 ± 0.199</td>
<td>460 ± 46</td>
<td>16th</td>
</tr>
<tr>
<td>PB23BB1</td>
<td>275–450</td>
<td>3.96 ± 0.24</td>
<td>1.530 ± 0.05</td>
<td>148 ± 3</td>
<td>0.132 ± 0.010</td>
<td>6.360 ± 0.211</td>
<td>622 ± 58</td>
<td>14th</td>
</tr>
<tr>
<td>PB23BB2</td>
<td>250–475</td>
<td>5.43 ± 0.22</td>
<td>1.450 ± 0.05</td>
<td>159 ± 3</td>
<td>0.110 ± 0.010</td>
<td>5.903 ± 0.201</td>
<td>920 ± 69</td>
<td>11th</td>
</tr>
<tr>
<td>PB23AGF1</td>
<td>Not valid</td>
<td>2.57 ± 0.09</td>
<td>0.845 ± 0.05</td>
<td>229 ± 4</td>
<td>0.128 ± 0.010</td>
<td>7.796 ± 0.228</td>
<td>330 ± 21</td>
<td>17th</td>
</tr>
<tr>
<td>PB23AGL1</td>
<td>275–375</td>
<td>4.15 ± 0.62</td>
<td>0.720 ± 0.05</td>
<td>226 ± 4</td>
<td>0.056 ± 0.010</td>
<td>5.356 ± 0.187</td>
<td>775 ± 143</td>
<td>13th</td>
</tr>
</tbody>
</table>

One other factor that may have given rise to less than anticipated ages would be past human activity. If the structure had been reused or the material from which the shards were obtained had perhaps been relocated or reheated, then these activities may have caused post target date construction modification of the materials around the brick structure at Site 23. Based on various archaeological interpretations, the structure is believed to have been constructed around 9th to 12th centuries while the surrounding area was in use up to about the 15th century (Shuhaimi and Mohd Yatim, 1990). From the TL ages obtained, samples PB23BBF1, PB23BBL1, PB23BB2 and PB23AGL1 are indicative of 11th to 15th century activity, a range that is in good agreement with other archaeological interpretations.

### 6. Conclusion

The shards from Site 23, located at Pengkalan Bujang, Bujang Valley, Kedah, Malaysia, were analysed by TL dating. The fine-grain technique was used to evaluate the paleodose. The annual dose was measured by using thick source alpha counting (TSAC) and atomic emission spectroscopy to determine the concentrations of uranium, thorium and also potassium that contribute to the internal dose rate. The gamma and cosmic dose rate was measured by means of the direct method using TL dosimetry, burying TL phosphors at the sampling site for 6 months.

Present TL analysis has confronted the prevailing paucity of information about the site. The dates tabulated in Table 3 show a clear disparity between the age obtained by previous researchers and the age obtained in this work. The dates obtained by previous researchers were outside the age range which has been estimated by archaeologists. In this work, the TL age range of the shards is indicative of activities at the site that span the 11th to 17th centuries. The upper range is in some doubt given a failure to protect two samples from light and heat exposure post-sampling.

### Table 3
Comparison of date from various literatures.

<table>
<thead>
<tr>
<th>Author</th>
<th>Dating method and protocol</th>
<th>Date (century)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shuhaimi and Mohd Yatim (1990)</td>
<td>TL dating using dosimeter LiF:Mg,Ti (TLD 100) and TL dating using dosimeter LiF:Mg,Ti (TLD 100)</td>
<td>9th to 15th</td>
</tr>
<tr>
<td>Mahat et al. (2010)</td>
<td>TL dating using dosimeter LiF:Mg,Ti (TLD 100)</td>
<td>16th to 18th</td>
</tr>
<tr>
<td>Rashid et al. (unpublished)</td>
<td>TL dating using dosimeter LiF:Mg,Ti (TLD 100)</td>
<td>17th</td>
</tr>
<tr>
<td>This work</td>
<td>TL dating using dosimeter CaF₂:Dy:TSAC and AES analysis.</td>
<td>11th to 15th (good sampling conditions) to 16th to 17th (poor sampling conditions)</td>
</tr>
</tbody>
</table>
While the ages obtained in this research span the date of suggested construction of the structure, in some cases the age represents the last firing of the material from which the shards were obtained. Thus said, the ages obtained for some samples (PB23BBF1, PB23BB1, PB23BB2 and PB23AGL1) are in good agreement with archaeological interpretations concerning the age of the site. Further study is required in obtaining a more accurate chronology of the ancient structure at Site 23. The TL dating technique has been used to good effect in obtaining present analysis.

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