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Original Article

Thermoluminescence and optically stimulated luminescence dating of bricks from the Thung Tuk archaeological site, Southern Thailand

Santi Pailoplee[1]*, Krit Won-In[2], Boonyarit Chaisuwan[3], and Punya Charusiri[1]

[1] *Morphology of Earth Surface and Advanced Geohazards in Southeast Asia Research Unit (MESA RU), Department of Geology, Faculty of Science, Chulalongkorn University, Pathum Wan, Bangkok, 10330 Thailand.*

[2] *Department of Earth Sciences, Faculty of Graduate School, Kasetsart University, Chatuchak, Bangkok, 10900 Thailand.*

[3] *The 15th Regional Office of Fine Arts, Phuket. Srisoonthon, Thalang, Phuket, 83110 Thailand.*

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Abstract

Thermoluminescence (TL) and optically stimulated luminescence (OSL) based dating were applied to ancient fired bricks to derive the chronology of the Thung Tuk archaeological site (TT), southern Thailand. In order to test the feasibility of brick dating, the inside and outside portions of the brick mass were dated separately. From the obtained results, the outside portion of the brick mass was found to be more suitable for luminescence dating than the inside portion since the inside might be incompletely fired during the production process. Among the brick ages obtained using the outside portion, both the TL and OSL (with a minimum model) analyses were all in agreement with the known ages of the TT, except for one sample that appeared to be much younger. This likely represents a subsequent renovation brick. Thus, the assessment of renovation and other imports into sites should be carefully considered in future luminescence dating.

Kywords: thermoluminescence, optically stimulated luminescence, brick, Thung Tuk archaeological site, Southern Thailand

1. Introduction

Because of the limited dating range (< 45,000 y) of radiocarbon dating (Roberts *et al.*, 1994) and the lack of organic materials exposed in excavation sites, luminescence dating by thermoluminescence (TL; Aitken, 1985) and optically stimulated luminescence (OSL; Murray and Wintle, 2002) have become common current alternative methods for assessing the chronological information of archaeological sites. Using residual ceramics (e.g., Robertson and Prescott, 1987, 1988), or more recently bricks (e.g., Changkian and Kaewtubtim, 1999; Won-in *et al.*, 2008) from ancient remain within the site, both TL and OSL can date the time since these materials were burnt (fired) and from this the potential age of the ancient community can be implied.

Among various archaeological sites discovered in Thailand, the Thung Tuk archaeological site (TT), which is located in the Ko Khao Island (Figure 1a), Southern Thailand, is archeologically significant. The site contains plenty of artifact assemblages, such as chinaware, local earthenware, color beads and so on (Figure 1b), and was likely to represent a significant trading community along the Andaman Coast and Strait of Malacca (Srisuchart, 1986).

After excavation and renovation, at least eight ancient remains look similar to the basements of a building exposed in this area (Figure 1c). From a chronological point of view, Srisuchart (1986) defined the age of TT as being around 1,000-1,300 y old, based on typological comparisons of the

* Corresponding author.
Email address: pailoplee.s@gmail.com

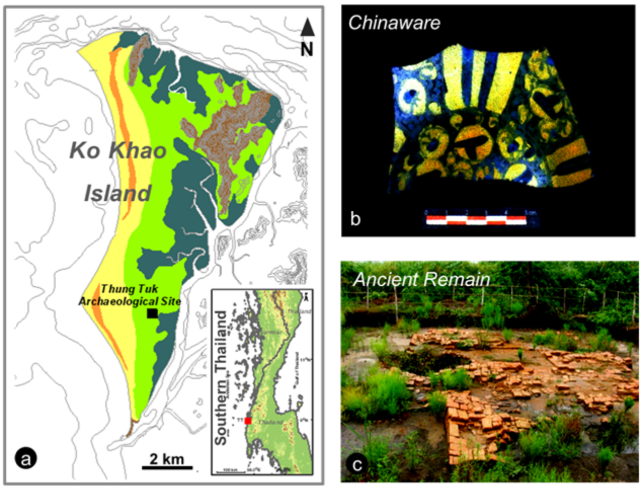

Figure 1. (a) Map of Ko Khao Island showing the location of the Thung Tuk archaeological site (TT; black square). Examples of the (b) antiques and (c) ancient building remains found in the vicinity of the TT (Chaisuwan and Naiyawatt, 2002).

artifacts found during excavation of the site. Meanwhile, Chaisuwan and Naiyawatt (2002) reported the radiocarbon dates of organic fragments from the TT as being 1,070-1,310 y old, in good agreement. Thereafter, Pailoplee *et al.* (2010) preliminary dated five basement bricks from the TT by TL dating to obtain a broader estimated age of 840-1,500 y that, however, still broadly agrees with the two previous assigned dates. However, some brick samples showed much older dates (2,800±270 y), leading to the assumption of an incompletely burnt (fired) brick during the production process (Pailoplee *et al.*, 2010). Therefore, the main aim of this study was to constrain the TT's age using both TL and OSL dating techniques. In addition, the feasibility of using luminescence dating with incompletely fired bricks (or similar) materials was clarified.

2. Sample Collection and Preparation

The elapsed time since the brick production (firing) and the present was determined from the luminescence dating using Equation 1 (Aitken, 1985);

$$\text{Luminescence date} = \frac{\text{Equivalent dose (ED)}}{\text{Annual dose (AD)}}, \qquad (1)$$

where the ED (Gy) is measured from the luminescence emitted during heating (TL) or optical stimulation (OSL) of the sample. Meanwhile, the AD (Gy/y) was evaluated from the concentration of three abundant natural radioisotopes, uranium (U), thorium (Th) and potassium (K), in the environment surrounding the sample.

In order to date the TT, six samples of bricks were collected, one from each of six different ancient remains. In order to avoid sunlight bleaching and soil contamination during the sampling process, the outer 3 mm of the brick surface was removed and discarded (Bailiff and Holland, 2000) and then half of the remaining brick mass was crushed and sieved. A 300-g portion of the obtained grains (diameter <90 mm) was dried and then packed in a plastic vessel ready for the AD evaluation. The water content, as the absorbing part of the radiation, was ascertained during this process.

After cutting across the brick mass, most of them revealed contrasting colors between the inside and outside portions of the brick mass (Figure 2). Based on literature reviews, some research works that have done previously mentioned that the different color mentioned above implies variously the physical and chemical properties of the brick including the technique of brick production. For instance, Karaman *et al.* (2006) conducted the relationship between the color and compressive strength of brick, which significant economically for defining the quality of of brick. Regarding to the chemical composition, Gredmaier *et al.* (2011) suggested that the inner portion of bricks consists normally of the calcium sulphate due to the combing between sulphur and calcium during the firing process. In addition, according to Maritan *et al.* (2006), the different technique of brick firing causes the different color within the brick mass. Pit firing technique, defined by a high heating rate and short time span produced the homogeneous color within the brick mass. Meanwhile, the technique of kiln firing, with a low heating rate and long time span generated the different color between the core and margin of brick.

As a result, in order to investigate further the relationship between the color within the brick and the luminescence signal including the luminescence date, we assumed that the color changes in the bricks (Figure 2) might be due to the incomplete heating (firing) of the brick mass during their manufacture (Pailoplee *et al.*, 2010). In order to demonstrate

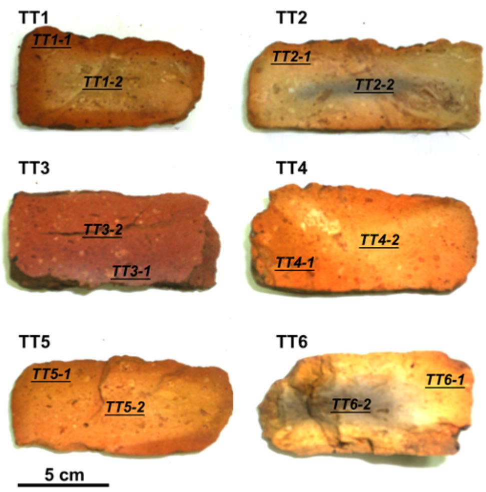

Figure 2. Photographs of the six bricks from TT used in this study (TT1-6), showing the contrasting colors in the brick mass from the outside to the inside. The labeled number is equivalent to the sample number used in the luminescence dating.

the non-resetting luminescence signal and the potential of TL and OSL dating of such incompletely fired bricks, the ED was evaluated by analyzing the outer and inner parts of the brick mass separately. The sampling location of each brick for evaluation of the ED is shown in Figure 2 along with the designated sample code. Accordingly from the six collected bricks, a total of 12 samples were dated using both the TL and OSL approaches.

For sample treatment, the enrichment of the quartz inclusion fraction was performed using the previously reported separation procedures (Takashima and Honda, 1989) under dim red light (wavelength > 600 nm). Each sample was crushed into a size range of 74-250 µm and then washed in 10% (w/w) hydrochloric acid. In order to eliminate the residual feldspar, each sample was etched using 40% (w/w) hydrofluoric acid for 45 min. Thereafter, the ferro-minerals were eliminated using an iso-dynamic magnetic separator. From the X-ray diffraction analysis (XRD), all the treated samples were found to contain least 95% (atom/atom) quartz, which is the main mineral used for the ED evaluation.

3. Measurements

3.1 Radiometric measurements

In order to determine the AD, the concentration of U, Th and K were measured. To satisfy this, about 300 g of each sample was contained in a plastic vessel and kept at room temperature for 1 month. Thereafter, in the radioactive concentration assessments, each sample was measured by thallium-doped sodium iodide gamma-ray spectrometry for 48 hrs. Based on the obtained concentration of these three radionuclides plus the water content (obtained as in section 2), the AD values of each sample were evaluated according to the standard table proposed by Bell (1979). The degree of radiation attenuation with respect to the grain-size distribution and water content were also recognized in this AD determination (Aitken, 1985).

3.2 TL measurements

The TL was measured using an automated TL reader (Risø TL/OSL-DA-15) with a mounted Schott BG12 filter and a built in ^{90}Sre^{90}Y beta source. The ED was evaluated according to the multi-aliquot regenerative technique (MAR; Veronese *et al*., 2006). Each sample was divided into 12 aliquots and then allocated into four groups (three aliquots per group). The first group was used to measure the natural TL signal (N), while the other three groups were heated and exposed to an additional single (R1), double (R2) or triple (R3) beta dose, respectively. The TL glow curves of sample TT 1-1 are shown as a representative example in Figure 3a. To select a suitable stable sensitivity, a plateau test procedure (Aitken, 1985) was employed. Normally, the 300°C TL-peak was selected for the representative dosimetry, growth curve contribution (Figure 3b), and ED determination (Table 1).

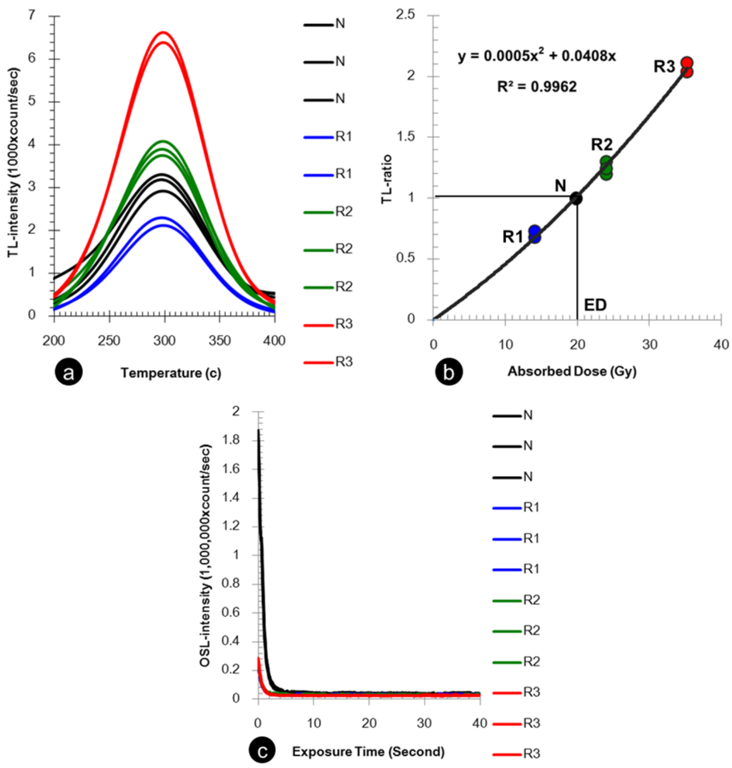

Figure 3. Representative (a) TL glow curves, (b) TL growth curve, and (c) OSL decay curves of brick sample TT1-1 where N and R1-3 represent the natural and single, double and triple regenerative-doses, respectively.

Table 1. TL and OSL dating results of brick samples, Thung Tuk archaeological site, southern Thailand.

Sample	U (ppm)	Th (ppm)	K (%)	W (%)	AD (Gy/Ka)	TL		OSL (Mean ED)		OSL (Min ED)	
						ED (Gy)	Dates (Yr)	ED (Gy)	Dates (Yr)	ED (Gy)	Dates (Yr)
TT1-1	9.43±0.23	61.85±2.2	2.95±0.1	3.52	9.99±7.68	19.84±0.61	**1,980±140**	24.17±2.06	**2,410±170**	16.14±1.47	**1,610±110**
TT1-2	9.43±0.23	61.85±2.2	2.95±0.1	3.52	9.99±7.68	20.35±0.63	**2,030±140**	33.78±15.32	**3,380±240**	21.40±1.82	**2,140±150**
TT2-1	11.66±0.22	64.41±1.99	3.00±0.09	4.57	10.71±7.24	7.56±1.65	**700±50**	16.58±0.58	**1,540±100**	12.82±0.56	**1,190±70**
TT2-2	11.66±0.22	64.41±1.99	3.00±0.09	4.57	10.71±7.24	10.46±1.15	**970±60**	21.6±1.34	**2,010±130**	16.12±0.31	**1,500±100**
TT3-1	19.3±0.27	77.2±2.16	4.07±0.1	0.62	14.85±10.03	0.77±0.01	**51±2**	0.41±0.10	**27±10**	0.28±0.04	**18±9**
TT3-2	19.3±0.27	77.2±2.16	4.07±0.1	0.62	14.85±10.03	0.13±0.00	**8±0**	1.04±0.29	**70±10**	0.28±0.02	**18±3**
TT4-1	4.94±0.19	29.96±1.5	2.14±0.09	5.38	5.70±2.57	7.77±2.23	**1,360±170**	10.55±0.28	**1,850±220**	9.38±0.37	**1,640±200**
TT4-2	4.94±0.19	29.96±1.5	2.14±0.09	5.38	5.70±2.57	10.91±1.70	**1,910±230**	11.98±2.42	**2,100±260**	9.45±0.68	**1,650±200**
TT5-1	9.98±0.18	59.37±1.93	3.07±0.08	9.38	9.78±6.16	16.41±0.91	**1,670±110**	18.09±0.72	**1,850±130**	16.20±0.58	**1,650±110**
TT5-2	9.98±0.18	59.37±1.93	3.07±0.08	9.38	9.78±6.16	16.42±1.33	**1,670±110**	27.78±9.06	**2,840±200**	17.74±8.43	**1,810±130**
TT6-1	6.27±0.16	33.32±1.26	2.39±0.08	4.70	6.54±2.44	8.28±1.16	**1,260±130**	7.54±0.25	**1,150±120**	7.12±0.21	**1,080±110**
TT6-2	6.27±0.16	33.32±1.26	2.39±0.08	4.70	6.54±2.44	7.21±1.96	**1,100±120**	12.7±0.73	**1,940±210**	11.17±0.45	**1,700±180**

3.3 OSL measurements

For measurement of the OSL, the samples were optically stimulated using the blue light emitted from a diode array (470±30 nm) with two filters, HA-3 for heat rejection and U-340 for detection. The ED was measured using the single-aliquot regenerative technique as developed by Murray *et al.* (1998). Therefore, the result of one disc represents a single-aliquot ED. In this study, 24 discs of aliquots per sample were measured. The fixed test dose (usually 10-20% of the natural dose) was applied to correct for any sensitivity changes. In order to remove any unstable TL peaks, samples were preheated at 220°C while measuring the natural or regeneration doses, and at 160°C while measuring the test dose, with a read temperature of 125°C for 40 s. The OSL decay curves are shown in Figure 3c, where the black lines correspond to the natural dose (N) and the other color lines correspond to the additional doses (R1-3). In order to obtain the OSL growth curve and ED estimation, the OSL signals were calculated by integration of the first 2 s of the data and subtracting the background (40-50 s). The series of obtained ED results are shown as histograms in Figure 4. According to Duller *et al.* (2008), four different age models that can be applied in the analysis of ED datasets are the (i) central or average age model, (ii) common age model, (iii) minimum age model and (iv) finite mixture model. It is noted that each model depends on its own specific assumption that has a valid, but different value and unique meaning. For instance, the central model is commonly used when the population of the ED distribution exists. Meanwhile the minimum age model assumes that only part of the sample was bleached or heated. Based on the distribution of ED (Figure 4), the central and minimum age models were used in this study and the results are shown in Table 1.

4. Results and Conclusions

Conceptually, brick dating is meaningful because the brick firing date represents the date of brick production, which in general is relatively closely related to the construction and age of the ancient community (Vieillevigne *et al.*, 2000; Kresten *et al.*, 2003). In order to date the TT archaeological site and demonstrate the routine procedure for brick dating, both TL and OSL analyses were performed (Table 1). The ED values and ages obtained by TL were broadly similar to those obtained by OSL with the minimum age model, with the ED_{OSL}/ ED_{TL} ratios being between 0.8 and 1.5, illustrating the broad agreement of the TL and OSL (min) results and level of authenticity. However, there was a small tendency for the OSL with the central age model to overestimate the age compared to that for both the TL and OSL (min) analyses.

Comparing the dates obtained from the outside and inside portions of the bricks, both the TL- and OSL-derived dates for the inside portion were much older than those for the corresponding outside portion (Figure 5a), which suggests that the inner portion of the brick mass may be

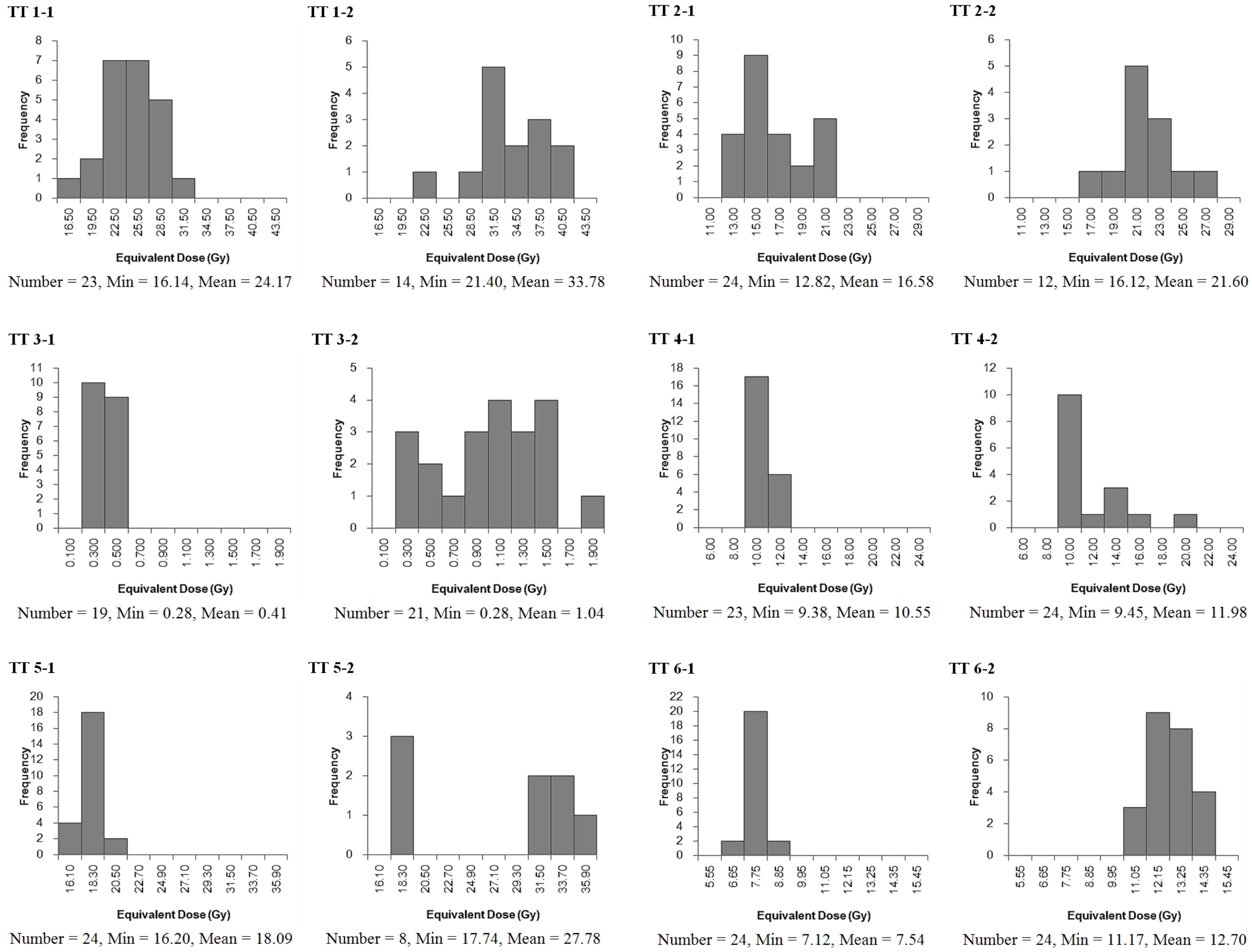

Figure 4. Histograms showing the equivalent dose distributions of the 12 brick samples (outer (-1) and inner (-2) samples from each of six different bricks (TT1-6), see Figure 2).

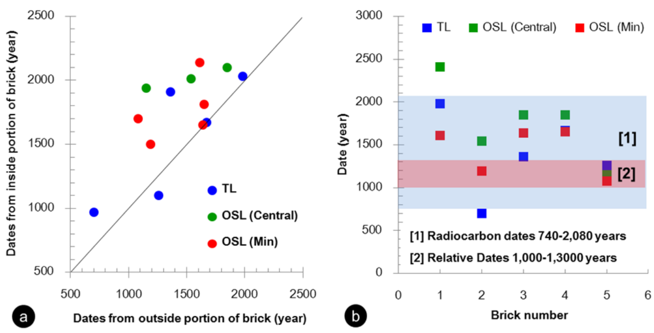

Figure 5. (a) Calibration of the obtained luminescence ages obtained from the inside and outside portions of the five old bricks (TT3 is excluded as a likely introduced artifact). (b) Comparison of the obtained luminescence ages with the reference radiocarbon ages.

significantly incompletely fired. This was supported by the observed variation in the ED values obtained from the inner brick portions, while the ED values from the outer portions showed a lower variation and greater level of consistency (Figure 4).

Recognizing only the ages obtained from the outside portion, most bricks yielded ages in the range of 1,000-2,000 y, with the exception of one peculiar brick (sample TT3-1) that gave an age of 10-50 y (Table 1). The former ages were in line with the reference age (Figure 5b), whereas the latter was not. Regarding to the prominent young age, not only do weak TL or OSL emissions lead to an underestimation of the dates, but also the estimation of the dose rate should be recognized, as mentioned by Roberts (1997). In the TT area, there is no evidence to support the existence of radon loss (Meakins *et al.*, 1979) or uranium enrichment (Guibert *et al.*, 1994) conditions. In addition, there is no archaeological evidence to support the occurrence of the latter communities after 1,000-2,000 y ago, let alone such recent ones as 10-50 y ago. Therefore, the specific physical features of the brick color (sample no. TT3 in Figure 2) and the prominent young age (sample TT3-1 in Table 1) allowed us to conclude this was a recently introduced brick, and probably used for renovation work during the excavation time span. Overall, both the TL and OSL dating techniques appear to be effective and viable methods to authenticate the age of ancient fired materials, or at least specifically for bricks.

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