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WHAT THE BRICKS TELL US FROM A TEMPLE AT BURKHAN KHALDUN MOUNTAINS: CHRONOLOGICAL INSIGHTS FROM PIRIR LUMINESCENCE

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Abstract: *The Burkhan Khaldun Mountains (Mongolia) and its surrounding sacred landscape are associated with Genghis Khan's birth and burial place as described in "The Secret History of the Mongols". It was listed as a UNESCO World Heritage Site on 4 July 2015 under the title «Great Burkhan Khaldun Mountain and its surrounding sacred landscape."*

This study offered a great opportunity to apply the recently developed post-IR infrared luminescence (pIRIR) approach to feldspar using coarse and polymineral fine grain techniques and determine the manufacturing date of a brick sample associated with the ruins of the Buddhist temple at the Burkhan Khaldun Mountains. Furthermore, the mineralogical composition of different blue-grey colored bricks from various temple buildings such as the Buddhist temples in Karakorum, Dugan in Erdene Zuu and Avargyn Balgas were studied. The original place and date of manufacturing of the bricks was revealed using the $pIRIR_{180}$ and $pIRIR_{240}$ from coarse and fine grains from a heated feldspar sample and were 1280±40 AD and 1230±50 AD, correspondingly, which falls into the time period of extensive constructions in Karakorum.

Keyboards*: luminescence dating, pIRIR, RHX, compositional analysis, brick;*

INTRODUCTION

Historical building dating using luminescence methods measures the last heating of the mineral grains in fired ceramics, including bricks and tiles. Brick dating, however, in comparison to pottery or geological sediment dating is rather rarely reported in literature; Bailiff [1] used optically stimulated luminescence (OSL) [2] on quartz inclusions extracted from brick, obtaining advantages in terms of signal sensitivity compared with thermoluminescence (TL) ones [3]. The problem of re-using of Roman bricks in the construction of the churches is frequently reported [4]. However, luminescence technique is unique when no organic material for radiocarbon dating is available. In Mongolia, luminescence dating was introduced and successfully applied by Saran [5] to the brick fragments, tiles and terracotta figures in order to get information about the construction phases at the ancient resident city Karakorum (Mongolia). Fourteen down-draught kilns for burning clay bricks were excavated by the Mongolian-German Karakorum Expedition (MDKE); nine of the recovered kilns were almost completely intact [6]. According to this evidence the Orkhon kiln site and manufactures of the 13th/14th century was a demonstrable part of the "imperial manufacturing city" of Karakorum.

Extensive luminescence studies on bricks samples from Karakorum, Mongolia revealed the effect of the strong medium component on the dose evaluation [7] in heated quartz extracted from blue-grey colored bricks. In contrast, the red-colored brick showed a high OSL intensity with dominant fast component. In particular, a comparative study of quartz and feldspar doses was carried out [8] in order to get an accurate dose estimate on the chronology of the brick production [7], as well as the construction phases of the Buddhist temple complex at the Karakorum [8].

However, the application of infrared stimulated luminescence (IRSL) dating of K-feldspars has long been limited due to anomalous fading [9]. Recently developed single-aliquot post-IR IRSL protocol employs infrared stimulation of K-feldspar at elevated temperatures following the IR stimulation at 50^{o}C, which empties charge carriers in unstable traps before measuring the signal from the more stable traps without any contribution of those unstable traps.

The purpose of this work was to retain all information about the bricks associated with the ruins of the Buddhist temple "Dund Ovoo" at the Burkhan Khaldun Mountain. The study offered a great opportunity to apply the pIRIR [10] approach to fired feldspar using coarse grain and polymineral fine grain techniques. In addition, a mass-gain dating method based on the slow rehydroxylation process of clay fired in the past - the RHX dating method [11] for ceramics will be tested. Compositional analysis will be used on a set of bricks of the medieval manufacture to determine where they might have been manufactured.

Archaeological context and samples

The Burkhan Khaldun is one of the Khentii Mountains in the Khentii Province of northeastern Mongolia. The mountain and its surrounding sacred landscape are associated with Genghis Khan's birth place as described in 'The Secret History of the Mongols'. It was inscribed as a UNESCO World Heritage Site on 4 July 2015 under the title «Great Burkhan Khaldun Mountain and its surrounding sacred landscape.» The brick sample Bkh-8 associated with the remains of the Buddhist temple 'Dund Ovooni sum' located at the Burkhan Khaldun Mountain formed basis for this study.

Table 1. Sample description

Sample ID	Archaeological context	Macroscopic features
Bkh-8	Buddhist temple 'Dund ovooni takhilyn sum' at Burkhan Khaldun Mountain	Brick, blue grey colored, D=40 mm
K02/06	Erdene-Zuu wall trench on the outside of the Northern wall, 2005	Brick, blue grey colored, D=45 mm
EZ-1	Erdene-Zuu main church building 'Gol Dugan', 2010	Brick, grey, D=55 mm
KhD-1	Avargyn Balgas, Khentii province, Delgerkhaan, 2003	Brick fragment, Blue grey colored
KhD-2	Avargyn Balgas, Khentii province, Delgerkhaan, 2003	Brick fragment, Blue grey colored

The known historical facts about the temple chronology suggest that the spiritual temple was built at the beginning of 14th century, which gave an opportunity to test the luminescence method. However, the possibility of re-using ceramic building materials cannot be ruled out; it means there is a risk that the bricks are not contemporary to the construction of the structure under study.

For the compositional analysis additional samples of the medieval manufacture were introdu-ced. Sample K02/06 was collected from Erdene Zuu temple wall trench on the outside of the Northern wall, in Karakorum (MDKE, Mongolian-German Karakorum Expedition, 2005). K02/06 was dated back to the early 13th century suggesting that the walls under Erdene Zuu were constructed in early construction phase of Karakorum at 1235±40 AD. Sample EZ-1 originates from the Buddhist temple Erdene-Zuu and its main church building "Gol Dugan". The brick fragments KhD-1, KhD-2 were collected from the excavations carried at medieval Avargyn Balgas, Khentii province in 2003. For the later samples no OSL data were available. Brick samples collected for mineralogical analysis, their archaeological context and main macroscopic features of the samples are summarized in Table 1.

Experimental details

All luminescence measurements were carried out at the luminescence laboratory at the MPI, Leipzig. The outer layer of sample was removed and the obtained core of the sampled brick fragments was carefully crushed in a hydraulic press and sieved. The coarse grains (>100μm) were then treated with hydrogen peroxide (H_2O_2, 15% and 30%) to remove any organic material, followed by digestion in hydrochloric acid (HCl, 10%) to dissolve carbonates, prior to density separation using a heavy liquid (lithium heterotungstate) to obtain quartz and potassium feldspar rich fractions. No coarse grains of quartz (90-180μm) were available; therefore 2mm feldspar aliquots were prepared for luminescence measurements. Polymineral fine-grains (4-11μm) were treated with hydrogen peroxide (H2O2, 15% and 30%), hydrochloric acid (HCl, 10%), dispersal using sodium oxalate, followed by isolation via Stokes settling [12]. Single aliquot discs were prepared for fine grain measurement using a pipette with the material suspended in a water-based solution.

All luminescence measurements were done using a Risø TL-DA-20 reader [13] equipped with $^{90}Sr/^{90}Y$ beta source. The infrared stimulated (870±40nm, 130mWcm^{-2}) signal from feldspar was detected using D410 interference filter. Equivalent doses of feldspar and of polymineral fine grains were determined using the post-infrared infrared stimulated luminescence (pIRIR) [10] protocol. The luminescence signal was calculated as the integral from the initial 4 s minus the final 10s of each 100s IR_{50}, and from the initial 7.6s minus the final 20s of the 100s $pIRIR_{225}$ stimulation, respectively.

For the evaluation of alpha and beta contribution from the ceramic the external layers of the brick sample were used. The radionuclide concentrations of K, U and Th measured in powdered sample using neutron activation analysis (NAA) were converted to dose rate data using the conversion factors from [14]. The internal dose rate was calculated for feldspar assuming a potassium content equal to 12.5±0.5% [15]. The beta dose rates determined on pulverized portion of brick directly by beta counting were compared with the calculated using NAA and the ratio was 1.19 .For polymineral fine-grains, 'α-values' of 0.10±0.02 and 0.12±0.02 were used for IR_{50} and pIRIR measurements, respectively. The contribution from cosmic radiation to the dose rate was calculated following [16], assuming an uncertainty of 5%. In-situ water content measured shortly after sampling (0.22%) was taken into account.

RHX (rehydroxylation) measurements were carried out using the procedure described in [14], X-ray diffraction and infrared FT-IR measurements were carried out at the Institute of Physics and Technology. The mineral phase analysis was carried out on a Shimadzu Maxima XDR-700 diffractometer (CuKα radiation, 0.02° 2Θ step size, 5°-80°). X-ray fluorescence was carried out in Geological laboratory using a AXIOS - wave-length-dispersive X-ray fluorescence spectrometer (PANalytical) with an Rh excitation source.

Table 2. Concentration of radionuclides measured using the low-background gamma spectrometry, neutron activation analysis NAA and X-ray fluorescence analysis.

Sample	K (%)	U(ppm)	Th(ppm)
BKh-08	3.45±0.24[a]	2.48±0.13[a]	13.07±1.96[a]
BKh-08	2.80±0.168[b]	1.70±0.153[b]	8.34±0.25[b]
BKh-08	2.87[c]		13[c]

[a] Gamma spectrometry (Dresden)

[b] Neutron activation analysis NAA, (CEZA Mannheim).

[c] X-ray fluorescence analysis XRF, (Geolab, Ulanbaatar)

RESULTS AND DISCUSSION

1. XRD, XRF and RHX results

The major minerals (Fig. 1) of the Burkhan Khaldun bricks are quartz, feldspar (microcline) and plagioclase (albite). However, KA6 contained muscovite, which is thermally stable up to 900°C. The maximum thermal stability of plagioclase and potassium feldspar is at 1000–1050°C, implying the firing temperature of Bkh-8 was between 900 and 1000oC. Set of data of ceramic samples from Karakorum and set from ceramic from Avargyn Balgas were introduced to figure out where the samples might have come from. Comparing the major and trace elements of the bricks, we can determine whether the bricks came from the same sources or raw materials. Similarities in mineral composition within the groups can be recognized between the brick Bkh-8 and K2/06; later is grey colored brick collected from the wall trench on the outside of the Northern wall of Erdene Zuu.

The results obtained using the RHX dating approach on Bkh-8 yield a date 1270±60 AD.

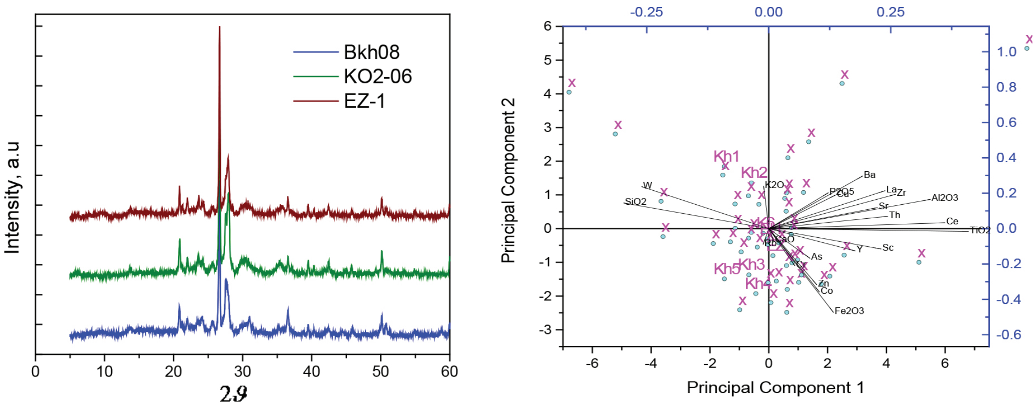

Figure 1. Left: The XRD patterns of the sample Bkh08 compared to the samples from collected Karakorum. Right: Principal component analyses

2. Luminescence results

In the following, the conventional $IRSL_{50}$ and the pIRIR protocols (Table 3) were tested on coarse grains as well as on polymineral fine grains from fired feldspar sample Bkh-8.

Table 3. Protocols used in this work – conventional IRSL and pIRIR.

Step	Protocol: IRSL	Protocol: pIRIR210
1	Give dose , Di	Give dose , Di
2	Preheat for 10 s at 210°C-290°C	Preheat for 10 s at 180°C-290°C
3	Optical stimulation with IRSL for 100 s at 50°C	Optical stimulation with IR for 100 s at 50°C
		Optical stimulation with IR for 100 s at 150°C-240°C
4	Test dose, 1 G y	Test dose, 1 G y
5	Preheat for 10 s at 210°C-290°C	Preheat for 10 s at 180°C-290°C
6	Optical stimulation with IRSL for 100 s at 50°C	Optical stimulation with IR for 100 s at 50°C
		Optical stimulation with IR for 100 s at 150°C-240°C
7	Return to step 1	Return to step 1

2.1. $IRSL_{50}$ and pIRIR on coarse grains of feldspar

The temperature dependence of $IRSL_{50}$ was studied for a preheat temperature between 210°C and 290°C and the mean De values for IRSL50 signals are plotted against the preheat temperature in Fig.2. It should be noted that the measurements were carried out using 1mm aliquots, which is nearly a single-grain technique. There is an overall De plateau between 225°C and 260°C with the mean De of 3.08±0.12 Gy; however, the $IRSL_{50}$ signals (Fig.2b) appear to decrease with the increasing preheat temperature. No fading measurements were done on this sample, but assuming g values ~2% (g-values between 0 and 5% was observed for different samples from different regions of the world); a preliminary value of 3.48Gy can be expected for Bkh-8.

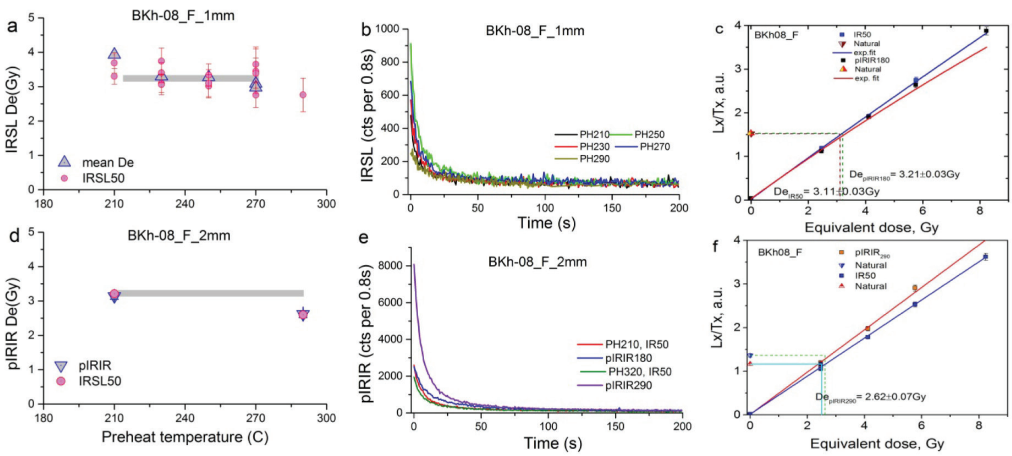

Figure 2. a) Effect of preheat temperature on IRSL De values for feldspar sample. Each data point represents the average of four 1mm aliquots. The grey bar represents the De plateau; b) IRSL decay curves; d) IR_{50} and $pIRIR_{180}$, $pIRIR_{290}$ De values for feldspar. e) IR_{50} and $pIRIR_{180}$, $pIRIR_{290}$ decay curves; c, f) The dose response curves Lx/Tx = f (De) constructed using $pIRIR_{180}$ and $pIRIR_{290}$ measurements for feldspar sample Bkh-8. De obtained using $pIRIR_{180}$ and $pIRIR_{290}$.

Subsequently, a pIRIR protocol for young samples was employed using a preheat of 210°C and stimulation temperature of 180°C; at low stimulation temperatures the residual dose is minimized [15]. Twelve 2mm aliquots of coarse feldspar were measured using $pIRIR_{180}$ protocol and yielded De_{IR50}=3.26±0.03Gy; $De_{pIRIR180}$=3.14±0.03Gy and over-dispersion OD=1.2%. Another set of aliquots were measured using the high temperature $pIRIR_{290}$ protocol which yielded De_{290} of 2.62±0.07Gy; the corresponding dose response curves Lx/Tx= f(De) for both measurements are shown in Fig.2c,f. The decrease in pIRIR De towards higher preheat temperature was observed; this is similar to the previous $IRSL_{50}$ measurements, however, this is in contrast to the increase in pIRIR De reported for unheated feldspar, e.g.[16], which is explained in terms of the hard-to-bleach signals [17]. The results based on $pIRIR_{290}$ are considered as erroneous due to the behavior of the $pIRIR_{290}$ signals; signals stimulated at higher temperature become harder to bleach resulting in considerable residual doses; therefore the pIRIR signal at lower stimulation has been suggested for young samples [16].

On the other hand, the sensitivity of pIRIR signals in unheated samples was reported to decrease with the increase of the stimulation temperature [17]. We examined the $pIRIR_{180}$ and $pIRIR_{290}$ test dose signal intensities shown in Fig.2e; when comparing the $pIRIR_{290}$ intensities with the corresponding IR_{50} intensities, the ratio increased from ($pIRIR_{180}$/IR_{50}) to ($pIRIR_{290}$/IR_{50}) from ~1.5 to ~5. Our study shows that for the well-bleached fired sample Bkh-8, the $pIRIR_{290}$ sensitivity increased towards higher preheats which can lead to the decrease of De estimates. Further investigations are needed to understand this effect.

2.2. pIRIR on polymineral fine grains

In the following, the pIRIR approach is carried out on polymineral fine grains; it is assumed that pIRIR on polymineral stimulates predominantly feldspar fine grains only [18]. The dependence of De values on the first IR stimulation temperature was investigated for fine grains for temperatures between 180°C and 270°C. Fig.3a,d show that both IR50 and pIRIR De values are decreasing towards higher stimulation temperatures. This is in agreement with the previous results obtained from IRSL and pIRIR measurements on coarse grains of feldspar. De obtained using $pIRIR_{150}$ is 6.7±0.2Gy, whereas De obtained using $pIRIR_{240}$ yielded 3.87±0.09Gy; nevertheless a preheat plateau is present between 240°C and 270°C.

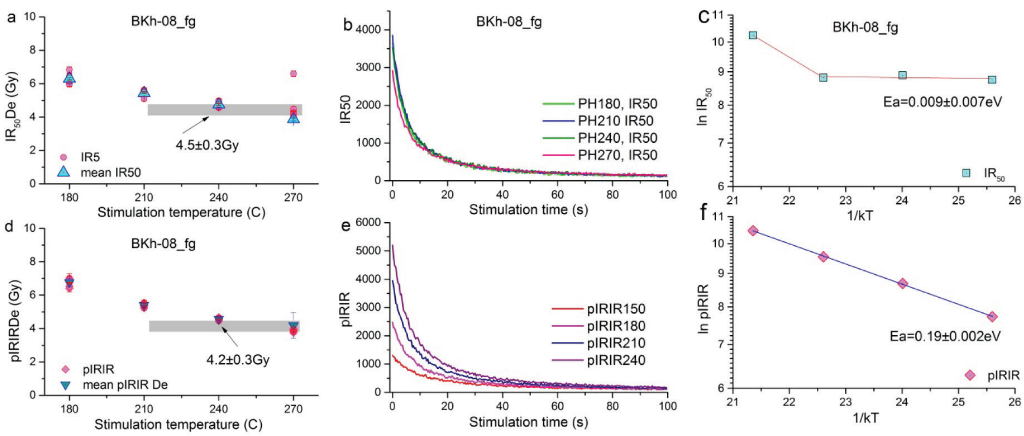

Figure 3. Dependence of De values on the first IR stimulation temperature for the polymineral fine grain sample Bkh-8 for a) IR_{50} and d) pIRIR De. Each data point represents the mean of four aliquots. b) The IR_{50} test dose signals and e) the pIRIR test dose signals; c) and f) Arrhenius plot of the IR_{50} and pIRIR signals.

The IR_{50} and pIRIR test dose signal intensities are shown in Fig.3b and Fig.3e; the pIRIR signals are increasing towards the higher preheat temperatures. For the IR_{50} there is almost no change in the intensity until the first IR stimulation temperature reached 240°C. Examination of the pIRIR/IR_{50} ratio intensities exhibited an increase from 0.5 to ~2 as the stimulation temperature increased, the pIRIR intensity was higher than the IR_{50} signal at ~240°C.

The IR_{50} and pIRIR test dose signal intensities are shown in Arrhenius plots in Fig.3c,f; the slope of the plot is interpreted as the activation energy for each signal. For the pIRIR signal the values of 0.19±0.002eV were obtained which were consistent with the value of 0.22±0.025eV reported in [19]. Interestingly, our measurements results shown in Fig.3f for IR_{50} indicate that there is no change in the intensity of IR_{50} up to temperature 240°C, but an abrupt change in the activation energy coincides with the existing preheat plateau between 240°C and 270°C. The results imply that $pIRIR_{240}$ measurements yielded the De values of 3.87±0.09 Gy which are derived from the most stable traps.

Luminescence ages

The range of doses between 2.60±0.20 Gy to 3.87±0.09 Gy based on the $IRSL_{50}$ and pIRIR signals from coarse and fine grains of feldspar were obtained in this study. From the $IRSL_{50}$ and $pIRIR_{180}$ measurements coarse feldspar doses of 3.08±0.20 Gy and 3.14±0.03 Gy were obtained, which are consistent with each other.

Dating polymineral fine grains, a higher preheat is more likely to give a stable pIRIR signal without any contribution of unstable traps. In our case, the signals in unstable traps were not emptied up to 240°C and contributed to the pIRIR signals. Therefore the De's obtained in the temperature range from 180°C and 240°C reflect the contribution of unstable traps; the doses range from 6.7±0.2Gy to 4.2±0.3 Gy. A high preheat temperature of 270°C generated a more stable $pIRIR_{240}$ signal yielding the values of 3.87±0.09 Gy.

Table 4. Luminescence results.

Sample Mineral fraction	*Method*	*Dose rate Dr (Gy/ka)*	*CAM De (Gy)*	*Age, years*	*Date, AD*
4÷11µm	*$pIRIR_{240}$*	*4.92±0.25*	*3.87±0.09*	*790±50*	*1230±50*
100÷150µm	*IRSL50*	*4.26±0.14*	*3.08±0.20*	*720±60*	*1290±60*
	$pIRIR_{180}$		*3.14±0.03*	*740±40*	*1280±40*

The final luminescence ages are derived from the ration of the absorbed radiation dose De and the dose-rate Dr; the data are summarized in Table 4. Given that luminescence dating determines the last heating event, the dates of 1290±60AD and 1230±60AD can be considered as the date of manufacturing of bricks, but not necessarily the time of temple construction. Based on the archaeological interpretations, the site is believed to have been constructed during the end of the 13th century; however, if the bricks had been reused or perhaps been relocated or reheated, a date of construction later than 1290±60 AD is expected for the temple.

CONCLUSIONS

The coarse and polymineral fine grains of feldspar have been investigated using the conventional IRSL and pIRIR approach; all IRSL, IR_{50} and pIRIR signals showed a decreasing tendency of the De plateau, in particular, the pIRIR signals do not deplete at higher preheats although the fired brick sample under study had been well bleached prior burial. The effect of residuals contained in the hard-to-bleach pIRIR signals was not observed; indeed the effect of increasing sensitivity changes was detected. For the polymineral fine grains, a higher preheat is more likely to give a stable pIRIR signal without any contribution of the unstable traps below 270°C.

The convergence of all luminescence ages (e.g. $pIRIR_{180}$ on coarse feldspar of 1280±40 AD, $pIRIR_{240}$ on fine grains of 1230±50AD) was found; in addition, a similarity of the mineral composition of the brick under study with the bricks associated with the first construction phase at the Karakorum was observed. Consequently, the original place and date of the temple's manufacture in Burkhan Khaldun is Karakorum city at 1280±40 AD. However, the possibility of re-using of building materials which falls into the time period of the extensive construction in Karakorum cannot be ruled out, and the temple construction at the later time period is expected.

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