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RECOGNIZING THE BURNING STATUS OF ARCHAEOLOGICAL QUARTZ PEBBLES COUPLING THERMOLUMINESCENCE AND ELECTRON PARAMAGNETIC RESONANCE SPECTROSCOPY

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ABSTRACT

Although the methodology to date pottery and pebbles by thermoluminescence (TL) is based on identical principles, the procedure followed to retrieve the accumulated dose in burnt pebbles offers extra difficulties. In addition, the distinction between burnt and unburnt pebbles is not always free from the subjective influence inherent to the visual inspection. Based on the dependencies of the 110ºC TL peak and the electron paramagnetic resonance (EPR) signal of the E'_1 center with the heating temperature of quartz, the aim of this study is to extend the method to classify burnt and unburnt pebbles independently from visual inspection. For this, several pebbles collected from the East block of the Pedra Furada rock shelter (São Raimundo Nonato, Piauí, Brazil) were used to create a burning pattern assessing TL and EPR responses as a function of the heating temperature. The results showed that the 110ºC peak was not observed in those pebbles that were heated below 400ºC and the intensity of the E'_1 signal abruptly decreased with heating above 400ºC. TL and EPR signals of specimens previously classified as "burnt" and "unburnt" appeared in good agreement with the values related to the burning pattern. This method offered the possibility to estimate the temperature in which some pebbles were heated in the past.

KEYWORDS: Quartz Pebble, Burning Pattern, TL Dating, EPR Spectroscopy, Pedra Furada

INTRODUCTION

Pottery fragments and burnt pebbles have major importance in the dating of archaeological sites where bones, charcoal fragments or any other organic material are not preserved. This is the situation of numerous sites located in the Northeast Region of Brazil due to the acidic nature of the sedimentary deposits and the severe weathering and erosion conditions (Martin, 2005).

The dating of ceramic and non-ceramic materials is based on the thermoluminescence (TL) response of quartz and feldspar minerals (McKeever, 1985; Wintle, 2008). Compared to pottery, pebbles of flint and quartz offer additional difficulties for the dating route due to the following reasons. In numerous situations it is not simple to distinguish burnt from unburnt specimens by visual inspection. Essentially, the visual inspection is based in macroscopic analysis taking into account the following features: shape; integrity; distribution of surface cracking; discoloration pattern (Parenti, 1991). Even for trained eyes, the visual inspection is not completely free from subjective arguments. The matter is more complex for quartz pebbles. Running burning experiments with quartz pieces of different sizes, Driscoll and Menuge (2011) concluded that burnt quartz cannot be easily recognizable as burnt flint if only considering macroscopic features such as fracture density, transparency, lustre and coloration.

The second difficulty comes up from the thermal history of the archaeological pebble and the low thermal conductivity of quartz itself. In order to obtain reliable TL-ages, it is essential that the firing used in the past was adequate to reset the geological signal. The distribution of temperature into the volume of a pebble will be homogeneous only if the specimen is exposed to heat during a long period of time. If the heat is not enough to keep temperature of the whole pebble above 400°C, the TL signal of quartz grains will not be completely free from the effect of the geological dose (McKeever, 1985; Duttine et al., 2005). The consequence for dating partially zeroed pebbles will be an overestimation of the accumulated dose. The difficulty of distinguishing burnt from partially unburnt pebbles was reported by Michab et al. (1998) who suggested that datable quartz pebbles are those that exhibit in natural TL curves a distinct peak near 300°C in addition to that one near 400°C.

The third difficulty is due to the hardness of the massive pebbles. In order to obtain a representative grain size for TL readings, the pebble should be severely crushed, dissipating a substantial amount of heat throughout the grains. Due to the mechanoluminescent effect, the crushing process decreases the sensitivity of the 110°C TL signal of crushed grains and causes a TL reduction in the near-surface layer of unetched grains (Toyoda et al., 2000; Takeuchi et al., 2006). Although the washing of coarse grains with hydrofluoric acid has been considered efficient to eliminate the spurious effects attributable to crushing, most dating experiments with burnt stones were carried out with flints (for e.g. see Aitken 1985; Göksu et al., 1989; Valladas 1992). There are much fewer studies dealing with the TL dating of burnt quartz pebbles (Valladas, 1981; Michab et al., 1998; Valladas et al,. 2003).

In the present study, the problem of dating burnt quartz pebbles is revisited by combining TL measurements of the 110°C peak and the electron paramagnetic resonance (EPR) signal of the E'_1 center. These features were independently used in the past to help the dating of flint (Göksu et al., 1989) and volcanic material (Falguères et al., 1994). More recently, Duttine et al. (2005) coupled the TL signal of the 380°C peak with the EPR signal of the E'_1 centre to investigate the effectiveness of annealing in dating protocols of archaeological flints. In the present study, it is shown that accurate measurements of the 110°C TL peak and the E'_1 signal in quartz grains prepared from pebbles burnt at different temperatures open the possibility to estimate the equivalent temperature in which an unidentified pebble was heated.

SITE AND SAMPLE COLLECTION

The pebbles used in this work were collected in the Pedra Furada rock shelter located in the Parque Nacional Serra da Capivara. As shown in Fig. 1, this park is located nearby São Raimundo Nonato district (Piauí, Brazil). Due to the large amount of lithic material and the numerous varieties of paintings, Pedra Furada is one of the most important sites located in South America (Santos et al., 2003).

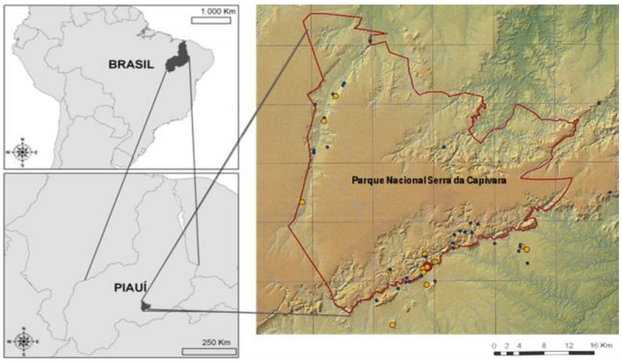

Figure 1: Localization of the Parque Nacional Serra da Capivara (FUNDHAM archives).

The Pedra Furada (PF) rock shelter was discovered in 1973 and has been excavated by several missions since 1978 (Guidon and Delibrias, 1986; Guidon and Arnoud, 1991; Parenti, 1991). This site assumed major importance in relation to the peopling of South America, since the discovery of well-structured hearths associated with lithic tools in levels dated at 25.000 years BP (Valladas et al., 2003; Santos et al., 2003). The sedimentary layers showed fifteen phases of occupation, subsequently grouped into six cultural phases: PF I, II and III (Pleistocene groups), and Serra Talhada (ST) I and II (groups between 12000 and 6000 years) and the last occupation started from 6000 years BP named Agreste (Holocene). Although the numerous studies carried out by different experts and with complementary techniques, the anthropic origin of the lithic industry and the origin of the hearths found in PF remain unclear for some part of the scientific community.

The excavations left behind a stratigraphic block (the "East block" with 12 m long, 2 m wide and 4 m high in the deepest part) that remained covered by masonry since 1988. The East block (Fig. 2) was partially excavated in 2010 when the pebbles used in this study were collected. Fourteen pebbles with length < 10 cm were collected 70 cm below the surface corresponding to the ST II phase. Based on surface signs, the pebbles used in this study were initially classified by the archaeologists in three categories: "burnt", "unburnt" and "unidentified".

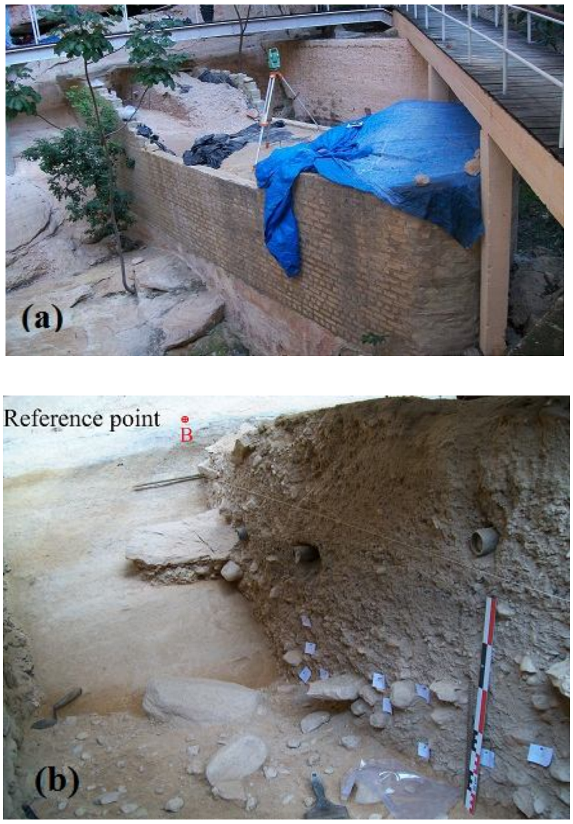

Figure 2: Overview of the remaining stratigraphic block of Pedra Furada rock shelter (a) and detail of the excavation initiated in 2010 (b).

The specimens were labelled and put inside black sacks. Some specimens are shown in Fig. 3. The pebbles classified as "unburnt" were randomly found in the sediment and did show no sign of ancient firing such as surface cracking, discoloration or reddening patterns.

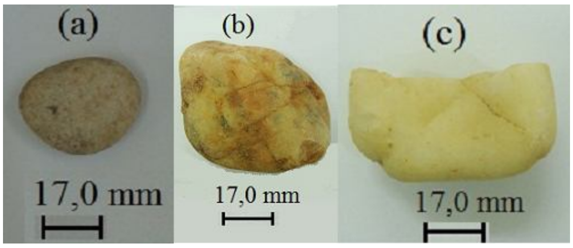

Figure 3: Pebbles collected from the East block in the Pedra Furada site initially classified as "burnt" (a), "unidentified" (b) and "unburnt"(c).

METHODOLOGY

Sample preparation

In order to eliminate the effect of sun light exposure during sample collection, a similar procedure as that introduced by Fleming (1970) to extract quartz grains from pottery fragments was adopted. According to Aitken (1985) the reduced level of TL signal in the outer surface of a pottery fragment can be avoided by removing a 2 mm layer from the surface. In this study, each pebble had 3 mm of its external part removed by grinding with sandpaper and etching with hydrofluoric (HF) acid at 40% (1:1) during 2 hours. The inner part of each pebble was crushed with impact strokes using an agate mortar. The grains were separated to 75 -150 µm using stainless steel sieves. The grains were treated with HF (45 minutes) and subsequently with hydrochloric (HCl) acid (45 minutes) for removing the superficial part of the grains that could exhibit possible spurious TL signals due to the mechanical action of the crushing (Aitken 1985; Takeuchi et al., 2006). The sample preparation was carried out in a dark room in subdue red light.

TL and EPR measurements

Three aliquots of approximately 40 mg of each sample were irradiated in a Co-60 gammacell irradiator (~3.8 Gy h^{-1}) with a dose of 10 Gy. The incidence of the radiation on the sample was from all directions since the Co-60 source has a circular geometry. To avoid the fading of the 110°C TL peak at room temperature (McKeever, 1985), the aliquots were immediately put into an ice-bath after the irradiation and were stored in a freezer at ~ -5°C. The TL glow curves were measured with a Harshaw 3500 reader with a heating rate of 4°C s^{-1} from 50 to 450°C. In total, the TL signals of six aliquots of ~ 10 mg were read. Besides the 110°C, three other peaks nearby 140, 300 and 380°C were found. Thus, the intensity of the TL emission was determined by integrating the areas of the glow curve in two separated regions as follows: from 70 to 180°C and from 250 to 425°C.

The EPR signal of the E'_1 center (an oxygen vacancy with an unpaired electron localized in one of the two non-equivalent Si atoms) was measured at room temperature with a Bruker EMX 10^+ spectrometer operating in the X-band with a high sensitive cylindrical cavity. The signals were recorded in the range from 3470 to 3500 Gauss with the parameters as follows: modulation frequency: 100 kHz; modulation amplitude: 1 Gauss; power: 0.2 mW; time constant: 81.92 ms; conversion time: 48 ms; gain: $1x10^3$; number of scans: 5. The calibration of the magnetic field and the sensitivity of the spectrometer were checked with a DPPH standard. The measurements were performed with natural (as-prepared) aliquots of 50 mg placed in a tube of vitreous silica of high purity (inner diameter: 2 mm). For each spectrum, the g-factors of the E'_1-center (Ikeya, 1993) were identified and the peak-to-peak intensity of the g_2-component was quantified.

Burning pattern

In order to create a burning pattern with the TL signal of the 110°C peak, similar to that performed by Göksu et al. (1989) with burnt flint, several pebbles initially classified as "unburnt" were heated at different temperatures from 200 to 600°C. These pebbles were heated in a muffle furnace with a rate of 10°C min^{-1}. Each specimen was kept 2 hours at the maximum temperature and then cooled slowly. The inner part of each specimen was crushed and prepared according to the procedure described before. EPR measurements, gamma irradiation with 10 Gy and subsequent TL readouts were carried out as previously described. The signal intensities were not normalized because TL and EPR measurements were carried out in the same conditions with aliquots of the same weight.

Because the actual condition of the pebbles used to create the burning pattern has major significance in this study, and since the initial classification of them was the visual inspection performed during the sample collection, the heating plateau test (Aitken, 1985) was used to check the classification of the burning condition of some specimens initially classified as "unburnt". The Fig. 4 compares the plateau curves of pebbles previously classified as "unburnt" and "burnt" with those of pebbles that were deliberately fired; as explained in the next subsection. It is observed that the TL intensity ratios of the pebbles classified as "unburnt" are higher than those calculated by the burnt pebbles. As expected, the natural TL signal should be higher for unburnt pebbles and is almost zero for those pebbles that were recently burnt in the camp-fire. Further evidence supporting the initial classification arises from the similarities between the glow curves of "unburnt" pebbles shown in Fig. 5 with the glow curves shown by Michab et al. (1998) who studied "natural" pebbles from the same archaeological site.

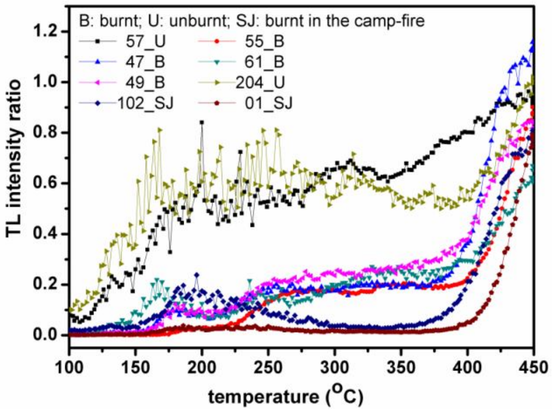

Figure 4: Plateau-test of pebbles initially classified as "unburnt" and "burnt" and for two pebbles that were burnt in a camp-fire.

Burning experiment

Two specimens were burnt in a camp-fire in order to verify the differences noticed in TL and EPR signals for quartz prepared from pebbles classified as "burnt" and "unburnt". The pebbles were placed in the inner part of the fire and were burnt during more than 10 hours until the fire naturally extinguished. After about two weeks, the quartz grains were prepared and TL and EPR measurements were conducted as previously described.

RESULTS AND DISCUSSION

TL glow curves

Typical TL glow curves of coarse quartz grains prepared from burnt and unburnt pebbles are shown in Fig. 5. The burnt pebble is one of those specimens that were heated in the camp-fire whereas the "unburnt" specimen corresponds to one of those specimens used for the heat plateau test shown in Fig. 4. Fig. 5 shows the TL glow curves obtained with quartz grains in natural (as-prepared) condition and after being irradiated with a dose of 10 Gy (natural + 10 Gy). The TL peak at 110°C is observed only for the burnt sample irradiated with 10 Gy. This peak is unstable, decreasing progressively with time at room temperature (McKeever, 1985; Khoul, 2008). For this reason, it does not occur in natural samples. However, it can be observed immediately after irradiation in laboratory or if the sample is kept at low temperature during the irradiation-reading interval. Besides the 110°C peak, the first region of the glow curves of burnt pebbles usually shows a peak nearby 140°C. In addition, Fig. 5 shows that the TL region above 250°C is different between burnt and unburnt pebbles.

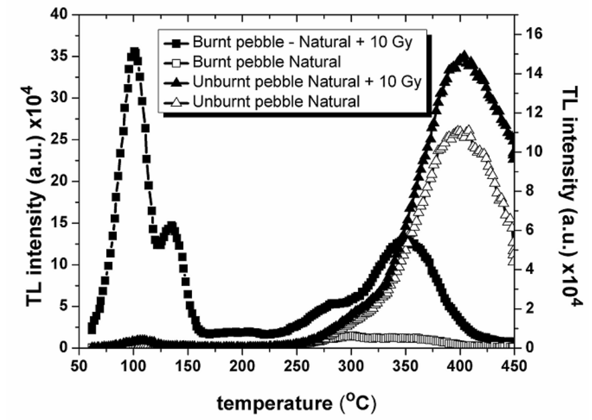

Figure 5: TL glow curves of quartz grains extracted from pebbles initially classified as "burnt" and "unburnt" in natural and irradiated (natural + 10 Gy) conditions.

The unburnt pebble shows a broad peak centred nearby 400°C related to the accumulated dose during the geological time. As it was expected, this peak increases after administering the artificial dose of 10 Gy. In case of the burnt specimen, two peaks centred near 280 and 350°C are clearly observed after the artificial irradiation with 10 Gy. The glow curves in Fig. 5 are in fair agreement with the original results published by Michab et al. (1998) that showed that artificially irradiated quartz grains from pebbles of Pedra Furada shows two glow peaks near 300 and 400°C after being heated at temperatures ≥ 500°C. The difference in peak position observed in our measurements can be explained by the heating conditions that were not the same as that used by Michab et al. (1998).

Fig. 6 shows the TL glow curves of the pebbles heated at different temperatures followed by gamma irradiation. The results show that, with the exception of the pebble burnt at 300ºC, the intensity of the glow peak at 110ºC increases with the temperature increasing. Similar results were obtained by Göksu et al. (1989) when five flint specimens from different geological origins were heated between 200 and 700ºC. The discrepancy noticed for the pebble heated at 300ºC is probably associated with the content of point defects acting as electron traps, luminescent centers or even non-luminescent centers into the quartz lattice that are competing each other during the TL read-out. As it was extensively reported, the TL response of natural quartz is very sensitive to the environmental condition in with the crystal was grown and its thermal history (Liritzis 1982; Jani et al., 1983; Guzzo et al., 2009; Sawakuchi et al., 2011). Fig. 6 shows that the peak at 140ºC also increases with the heating temperature but its intensity is much lesser than that of the 110ºC. Thus, the intensity of both peaks was considered together to the construction of the burning pattern.

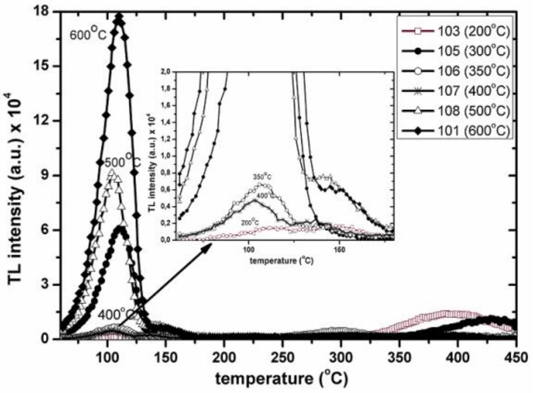

Figure 6: TL glow curves of quartz grains prepared from pebbles heat-treated at different temperatures. Test-dose: 10 Gy.

EPR signals

Fig. 7 shows typical EPR spectra of burnt and unburnt pebbles obtained in the region of the magnetic field where the *E′₁* center can be observed. This signal is not observed in the specimen that was burnt in the camp fire. The same result was obtained for those pebbles that were initially classified as "burnt". On the other hand, the *E′₁* signal was observed in all specimens that were initially classified as "unburnt". It was noticed that the intensity of this signal varied significantly from one specimen to the other. The values of the g-factor of this signal (g_1: 2.0018; g_2: 2.0006; g_3: 2.0003) are in good agreement with those from the literature (for e.g. see Ikeya, 1993; Benny et al., 2002).

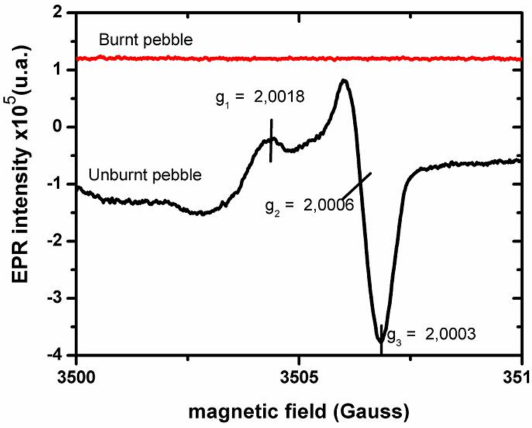

Figure 7: EPR spectra of quartz grains prepared from pebbles initially classified as "burnt" and "unburnt".

The EPR spectra of some samples used to create the burning pattern are shown in Fig. 8. It is observed that the signal of the *E′₁* center grows from 200 to 300ºC, and it decreases at 400ºC. This signal disappears for heating temperatures above 500ºC. According to Ikeya (1993), the intensity of the *E′₁* center increases above 200ºC and it disappears when quartz is annealed around 370ºC. It was also reported that its regeneration is strongly reduced by annealing at 500ºC. Falguères et al. (1994) studied the EPR signals in quartz grains extracted from volcanic materials. They did not observe the *E′₁* signal in samples that were heated in temperatures above 400ºC for at least 1 hour. Thus our results are in good agreement with those shown by previous authors.

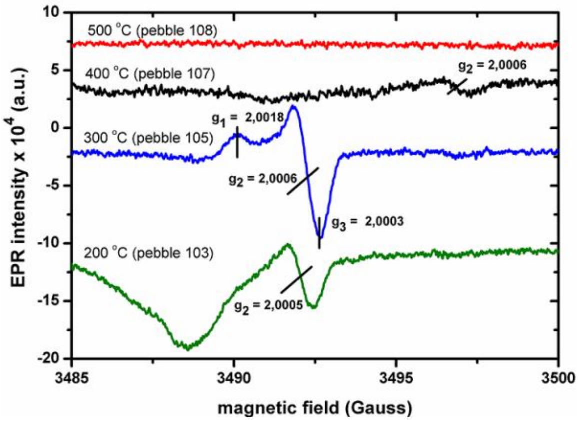

Figure 8: EPR spectra of quartz grains prepared from pebbles heat-treated and different temperatures.

Classification

Combining the intensities of the 110°C peak and that of E'_1 center of the pebbles heated in laboratory between 200 and 600°C, a burning pattern was created in order to verify the situation of the other pebbles initially classified by the visual inspection. Fig. 9 shows this pattern where it is observed the increase of the 110°C peak and the decrease of the E'_1 signal with the temperature increasing. Based on a direct comparison of both intensities, the results obtained with previously classified pebbles were joined with those of the burning pattern. Thus, it was possible to estimate the equivalent temperature of pebbles burnt in the past and also the equivalent temperature of the pebbles burnt in the camp fire (102_SJ and 01_SJ). It was also possible to check the situation to those pebbles that were initially classified as "unidentified". The results indicate that one of them (pebble 47) was burnt at an equivalent temperature around 550°C and the other (pebble 49) was probably burnt above 600°C. The combination of the intensities of the 110°C peak and E'_1 centre was essential to clarify the situation of the pebble 61. Initially, this pebble was classified as "unburnt" but the situation changed due to the occurrence of the E'_1 signal coupled with a low intensity of the 110°C peak, suggesting that this pebble could not be heated above 500°C. According to the burning pattern, it has been suggested that the equivalent temperature in which this specimen was burnt in the past might be included in 300 - 500°C interval. This result was confirmed by the methodology proposed by Michab et al. (1998). The TL curve of this pebble shows two peaks centred on 290 and 380°C excluding the possibility to this specimen to be a geological ("unburnt") pebble. However, this pebble is not datable because the equivalent temperature in which it appears in the burning pattern is not sufficient to erase the geological TL signal completely.

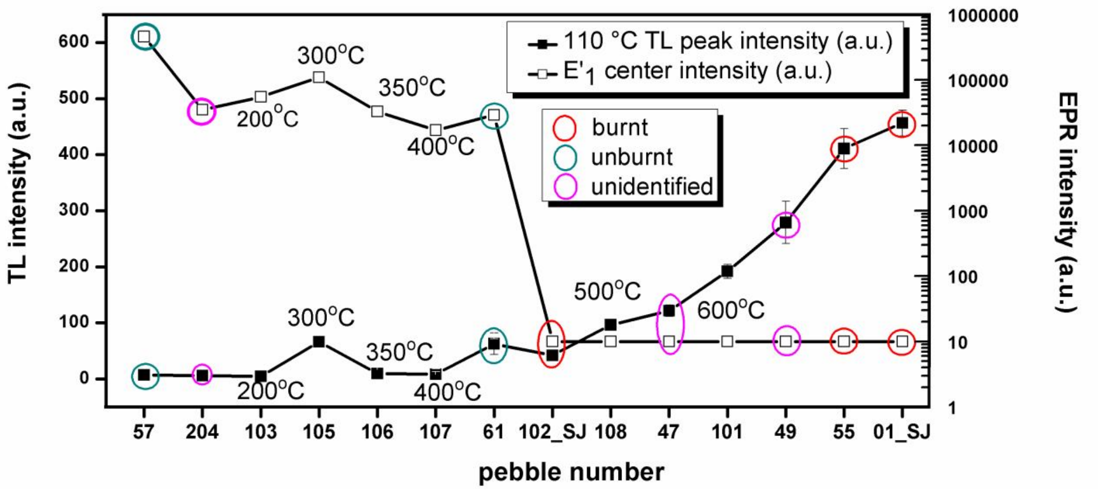

Figure 9: TL and EPR signals of the burning pattern together with signals measured in pebbles initially classified as "burnt", "unburnt" and "unidentified".

CONCLUSION

The results of this study showed that the methodology based in visual features to classify quartz pebbles in relation to their burning history can be improved by combining TL and EPR measurements. Based on the burning pattern proposed here two requirements must be satisfied simultaneously to classify a pebble as an archaeological datable specimen as follows: the absence of the EPR signal of the E'_1-center and the occurrence of the 110°C TL peak after a test-dose of few Grays. The analysis of some specimens taken from the Pedra Furada site has shown the necessity to improve the resolution of the burning pattern including additional specimens that should be heated between 400 and 500°C and also above 600°C. Nevertheless the results of this study suggests that it is possible to estimate the equivalent temperature in which the pebble was burnt in the past by coupling the EPR signal from the E'_1 center and the frozen intensity of the 110°C TL peak of quartz.

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