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AN INTERNATIONAL COMPARISON STUDY ON OSL DATING

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OSL TARİHLENDİRME ÜZERİNE ULUSLARARASI BİR KARŞILAŞTIRMA ÇALIŞMASI

Abstract:

International inter-laboratory comparison tests are an important need to ensure the accuracy of measurements in experimental laboratories, on Optical Stimulated Luminescence (OSL) dating technique, these tests have unfortunately been organized very limited in number so far. The latest inter-laboratory comparison study on this subject was started by the Nordic Luminescence Laboratory in Denmark in 2006. After the results from 30 different laboratories were collected in a period of 6 years, the study ended in 2012. The results of the study were published in 2015. In this paper, within the scope of the aforementioned comparison study, the result of OSL dating of quartz-rich aeolian and/or coastal marine sand sample as inter-comparison one was presented. Equivalent dose (De) of the sample was determined using Single Aliquot Regenerative Dose (SAR) protocol. For the calculation of annual dose rate (Da), concentrations of radioactive isotopes (^{238}U, ^{232}Th, and ^{40}K) were measured by using a high-purity germanium detector. OSL age was determined as 4530 ± 41 years for the sample. Consequently, within the error limits, the age of the sample was determined close to the expected age. By participating in this comparison test, we had the opportunity to compare our results with other luminescence dating laboratories and to ensure the accuracy of our results. Meanwhile, we had the chance to interpret, separately, each step of dating measurements (preparation of pure quartz minerals, determination of equivalent dose and annual dose etc.) performed in TL/OSL Dosimetry Laboratory of Turkish Atomic Energy Authority (TAEA) on an international scale.

Özet:

Uluslararası laboratuvarlar arası karşılaştırma testleri, deneysel laboratuvarlarda yapılan ölçümlerin doğruluğunu sağlamak için önemli bir ihtiyaçtır. Ancak şimdiye kadar Optik Uyarmalı Lüminesans (OSL) tekniği ile tarihlendirme konusunda uluslararası karşılaştırma testleri veya ölçümlerinin yapıldığı organizasyonlar maalesef oldukça az sayıda düzenlenmiştir. Bu konuda en son laboratuvarlar arası karşılaştırma çalışması Danimarka Risø da bulunan Nordic Lüminesans Laboratuvarı tarafından 2006 yılında başlatılmıştır. Farklı 30 laboratuvarın sonuçları 6 yıl içinde toplandıktan sonra, çalışma 2012 yılında sona ermiştir. Çalışmanın sonuçları 2015 yılında yayınlanmıştır. Bu makalede, yukarıda bahsedilen karşılaştırma çalışması kapsamında kuvars bakımından zengin rüzgârla oluşan ve/veya kıyısal deniz kumu numunesinin OSL tekniği ile tarihlendirilmesi sonrasında elde edilen sonuçlar karşılaştırmalı olarak sunulmuştur. Numunenin Eşdeğer dozu (D_e), Tek tablet Yineleme Doz (SAR) yöntemi kullanılarak ölçülmüştür. Örnekler için yıllık çevresel radyasyon dozunun (D_a) hesaplanması için gereken uzun yarı ömürlü radyoaktif izotopların miktarlarını belirlemek için yüksek

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saflıkta germanyum dedektör kullanılmıştır. Örneğin OSL yaşı 4530 ± 41yıl olarak hesaplanmıştır. Sonuç olarak, hata sınırları içinde, numunenin yaşı beklenen yaşa oldukça yakın olarak belirlenmiştir. Bu karşılaştırma testine katılarak, sonuçlarımızı dünyadaki diğer lüminesans tarihlendirme laboratuvarları ile karşılaştırma, sonuçlarımızın doğruluğunu test etme ve aynı zamanda, Türkiye Atom Enerjisi Kurumu, TL/OSL Laboratuvarı tarafından yapılan OSL tarihlendirme ölçümlerinin her aşamasını (saf kuvars minerallerinin hazırlanması, eşdeğer dozun ve yıllık dozun belirlenmesi vb.) uluslararası ölçekte yorumlama fırsatımız olmuştur.

Keywords: OSL dating, inter-laboratory comparison, equivalent dose, quartz, SAR.

Anahtar Kelimeler: OSL tarihlendirme, laboratuvarlar arası karşılaştırma, eşdeğer doz, kuvars, SAR.

1. Introduction

In OSL dating, the time elapsed from the last daylight exposure of the sample is determined. Since the samples are exposed to daylight just before the burial, it is the widely used method to dating of the time of deposition of sediments which are the subject of this study (Huntley, Godfrey-Smith & Thewalt, 1985; Huntley, Hutton & Prescott, 1993; Huntley and Lamothe, 2001; Murray and Funder 2003; Vandenberghe, 2004; Madsen, Murray & Andersen, 2007; Madsen and Murray, 2008; Murray, Buylaert & Thiel, 2015). Natural mineral grains of quartz and/or feldspar are used in OSL dating of sediments.

International inter-laboratory comparison tests is an important need to ensure the accuracy of measurements in experimental laboratories , on OSL dating, these tests have unfortunately been organized very limited in number so far. In this context, the most recent inter-laboratory comparison was initiated by the Nordic Luminescence Laboratory in Risø, Denmark in 2006. 30 different laboratories from 17 different countries participated to this organization which only TAEA- TL/OSL Dosimetry Laboratory joined from Turkey. The results obtained by participating laboratories were interpreted comparatively and were presented as a statistical distribution by giving only laboratory codes without laboratory names. (Murray, Buylaert & Thiel, 2015). The code of our laboratory was AF.

Within the scope of the aforementioned comparison study, aeolian and/or coastal marine sand sample ridge sample was dated by Optically Stimulated Luminescence (OSL) technique. In the article of Murray, Buylaert & Thiel 2015, obtained results getting from 30 different laboratories (equivalent dose, annual dose, determined age, etc.) by using the inter-comparison sample were given, however details of the each laboratory studies were not presented. In the current study, we present the details of our OSL dating study performed in the framework of this inter-comparison test having laboratory code of AF.

2. Material and Method

2.1. Methodology

In OSL dating studies, samples are taken generally from a certain depth below ground level (except that buildings, caves etc.). At this depth, the sample is exposed to natural ionizing radiation (mostly from ^{238}U, ^{232}Th, ^{40}K radioactive elements and cosmic rays) during the burial time. This radiation releases some electrons in natural quartz and feldspar minerals. Some of these free electrons are stored in traps formed by crystal structure defects exist in minerals. When minerals are exposed to daylight or heat, the trapped electrons release and combine with

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luminescence centers. A luminescence light is emitted at the end of this event. The emitted luminescence can be measured by heat or light; if by heat, the luminescence emitted from the mineral is called Thermoluminescence (TL), and if released by light, it is called Optically Stimulated Luminescence (OSL). Thus, TL and OSL dating provides an estimate of the time elapsed since the sample was last heated and exposed to daylight, respectively.

In TL and OSL dating applications, it is assumed that the dose in and/or around the sample during elapsed time comes at a constant speed to the sample. The absorbed dose by the sample from the past to the present will increase in proportion to the natural radiation. These approaches are expressed as follows with the age equation (Aitken, 1985).

Age= D_e (Gy) / D_a (Gy/ka)

The amount of the absorbed dose by the sample during the burial time is called the paleodose or equivalent dose (D_e). Annual dose rate (D_a) is absorbed radiation dose by the sample per year from natural radionuclides and cosmic rays. The ages so far determined by luminescence methods range from decades to millions of years (Aitken, 1985; Aitken 1998; McKeever, 2001; Bøtter-Jensen, McKeever Buylaert & Thiel Wintle, 2003). Details of the physical basis of luminescence dating can be found elsewhere (Aitken, 1985; Aitken, 1998).

2.2. Sampling

In this study, aeolian and/or marine ridge/swale sand sample was taken from Skagen peninsula region, about 2 km south of the town of jerup, in Denmark (latitude: 57° 31′, longitude: 10° 24 ′) with an expected age of approximately 5000 years was dated using OSL technique. All the details related to the collection of the samples from the sampling site and their preparation for the inter-comparison study were described detail in the work of Murray, Buylaert & Thiel, 2015. All procedures such as preparing samples in moisture-proof packages of 250 grams and labeling them with a special laboratory sample code and sending them to the relevant participating laboratories were performed by Nordic Luminescence Laboratory in Risø (Denmark).

2.3. Sample preparation

In luminescence dating applications, the samples are brought to a very small size so that the luminescence emitted from the sample can reach the photo tube more easily, and more efficient luminescence is obtained. Sample preparation technique varies according to the kind of mineral (quartz, feldspar, calcite or polymineral) and the size of the mineral (coarse grain or fine grain). In this study, coarse grain quartz minerals (180-250 μm) were prepared by using conventional sample preparation technique, except that the heavy liquid separation stage.

The conventional sample preparation procedure was performed as follows:

- Wet sieving in the range 180-250 μm.
- 1 h %10 HCl (Hydrochloric acid) to dissolve any carbonates.
- 8 h concentrated H_2O_2 (Hydrogen peroxide) to remove organic matters.
- 1 h %40 HF (Hydrofluoric acid) to remove feldspar minerals and etching the surface of the quartz grains.
- 1 h %10 HCl again to remove any remaining fluorides.

The sample was washed 3 times with distilled water after each chemical treatment. The extracted quartz minerals were stored at a temperature of 50 ° C for 1 night. Finally, the minerals were sieved again to interval 180-250 μm and were checked for residual feldspar grains using an Infrared (IR) test. IR test results showed that the ratio of infrared stimulated luminescence

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to blue light stimulated luminescence is less than %1 and all subsequent OSL measurements were performed using these highly pure quartz minerals. All samples were prepared under subdued orange light at a wavelength of 590 nm. The obtained pure quartz minerals were mounted on stainless-steel discs using silicone spray. These minerals were used for equivalent dose measurements. After the first wet sieving the samples not in the range of 180-250 μm were dried again and used to determine the annual dose rate.

3. Measurement

The equivalent dose measurements were performed using Risø TL/OSL reader (Model TL/OSL-DA-20) equipped with light-emitting diodes (870 nm for infrared and 470 nm for blue LEDs) and $^{90}Sr/^{90}Y$ beta source with a calibrated dose rate of 0.140 Gy/s for coarse grains. All continuous wave optically stimulated luminescence (CW- OSL) decay curves were detected through a 7.5mm thick Hoya U-340 filter. The luminescence signal was measured during 40 s and luminescence intensity was calculated as the first 0.8 s of the resulting decay curve minus the background, which was averaged over the last 4 s. All OSL measurements were performed at temperature of 125 °C for 5 s. All heating's were performed in a nitrogen atmosphere with constant heating rate of 5 °C/s. All measurements were carried out in subdued red light.

In this study, the typical standard Single Aliquot Regeneration (SAR) protocol (with 260ºC, preheat for 10s, and 220ºC cut heat) was used to convert the measured luminescence intensity to the equivalent dose (Murray and Wintle, 2000). In this protocol, firstly, one or more aliquots are prepared and the intensity of natural OSL is measured. Then the same aliquots are compared with the intensity of OSL measured in the laboratory by dosing with a known radiation source (usually Sr-90 beta source) in the laboratory. This procedure is repeated several times at varying laboratory doses and the dose response curve (growth curve) is plotted to show the change in OSL signal intensity depending on the beta radiation dose. The equivalent dose (D_e) is obtained by determining the dose corresponding to the natural luminescence count on the growth curve (Murray and Wintle 2000; Wintle and Murray, 2006.)

The concentrations of uranium (^{238}U), thorium (^{232}Th) and potassium (^{40}K) radioactive elements in the sediment were measured in Turkish Atomic Energy Agency (TAEA) - Gamma Spectrometry Laboratory by using the Canberra a high purity germanium gamma detector.

4. Results and discussion

All luminescence signals were measured during 40 s and luminescence intensity was calculated as the first 0.8 s of the resulting decay curve minus the background, which was averaged over the last 4 s. Preheat temperature was 260ºC for 10s, 1.4 Gy test beta dose was given the aliquots with cut heat 220ºC. The mean dose recovery ratio (measured/given dose) for six aliquots was calculated as 1.00 ± 0.47. This calculated value shows the reliability of OSL dating method.

The D_e value was obtained from the interpolating method by using 26 aliquots. The best correlation between beta doses and OSL signal intensities was obtained for the linear function (y=ax+n, a=1.06 E-001±2.12E-003, n=0). In this mathematical function, y is the measured OSL signal intensity, x is the dose of the artificial beta ray, n is the OSL signal intensity at zero applied beta dose mean that the relative amount of OSL signal intensity of un-irradiated sample and a is the rate of defect production and/or radiation yield upon irradiation at room temperature. Recycling ratio and recuperation were 1.00 ± 0.02 and 1.11 ± 0.15 %, respectively. And the growth curves pass very close to the origin for almost all aliquots. These data show that SAR protocol worked correctly in the studied sample. Average equivalent dose value for

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studied sediment sample, based on accepted 17 aliquots, was determined as De=4.76 ± 0.29 Gy by using OSL technique.

The measured concentrations of ^{238}U, ^{232}Th and ^{40}K in the sediment were given in Table 1. These elemental concentrations were converted into dose rates using conversion factors in the literature (Guerin et al., 2011). In this study, a beta-ray attenuation-correction factor was used following Mejdahl (1979). The fraction of the water uptake during burial (F) was considered as 0.6. The saturated water content of the sample (W) was measured as 21.40 using PresicaXM60 gravimetric moisture analyzer. The contribution of the cosmic dose to the annual dose rate was considered to be 0.17±0.01 Gy/ka by assuming the altitude, geomagnetic latitude, longitude and depth from the ground level (0 km, 50°N, 10°E and 150 cm) of the sampling site. For coarse grain dating, both external and internal alpha dose rate contributions (are) eliminated (Aitken, 1998). Therefore, the annual dose rate was calculated by taking into account beta dose rate plus gamma dose rate within the sediment and contribution from the cosmic rays. When the size range is around 100 µm, the quartz grains are free of from internal radioactivity (Aitken 1998). However, for quartz particles over 100 µm, the internal dose rate resulting from ^{238}U and ^{232}Th in (of) grains becomes important. For this reason, internal dose rate value was assumed as 0.06 ± 0.03 Gy/ka (Mejdahl, 1987). Considering all these conditions, the total beta and gamma dose rate values were calculated as 1.05 ± 0.06 Gy/ka. For sediment sample, radionuclide concentrations and calculated annual dose rates of beta, gamma and cosmic rays are given in Table 1. Beta and gamma dose rate values were corrected for average particle size of 215 µm.

Table 1. Annual dose rate components of the sediment sample.

Radionuclide concentrations (Bq/kg)			**Dose rate (Gy/ka)**			
^{238}U	^{232}Th	^{40}K	Beta (assumed)	Gamma (measured)	Cosmic (calculated)	Total
2.86 ± 1.24	3.70 ± 0.32	264 ± 22	0.06 ±0.03	0.82 ±0.06	0.17±0.01	1.05±0.06

OSL age of the above mentioned geological sample was found as 4.53±0.41 ka. When we compare our results with other laboratories, the following descriptions were obtained:

- The average of the equivalent dose (D_e) results obtained from the all laboratories was 4.48 ± 0.12 Gy. The equivalent dose value (4.76 ± 0.29 Gy) determined in our laboratory was almost the same with the average value within the experimental error limits. It was also showed that we were able to isolate almost pure quartz minerals from the sample without damaging the luminescence signal in the sample, and so determine accurately the equivalent dose within the error limits.
- The average saturated water content of the sediment samples were measured as 19±2% by all other laboratories. The same quantity was measured as 21.40± 0.685% in our laboratory. Therefore, we can say that this quantity, which is important in OSL age calculations, can also be measured with big accuracy in our laboratory.

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- The annual dose rate results, more than half of the participants, including us showed that, the dose rate measured lower than the average. The average dose rate of all laboratories was declared as 1.15 ± 0.02 Gy/a, similarly we determined this value as 1.05 ± 0.06 Gy/a. Guerin et al. 2015 showed that in well sorted sediments , differences in particle size, variation in potassium feldspar amounts, and the presence of radioactive minerals such as zircon may lead to different dose rate measurements. Therefore, the variations in dose ratios are expected situation for a quartz-rich beach sand intercomparison sample used in this study.
- Since the dose rate we measured was slightly lower than the average, the OSL age of the sample was determined as slightly higher than the mean. Nevertheless, we found the average OSL age of sample within the error limits. The mean OSL age value was calculated as 4.53 ± 0.41 ka by us, while the average of all other laboratories was 3.99 ± 0.14 ka.

5. Conclusion

In conclusion, the calculated OSL age of the geological sample is an agreement with the expected age value within the error limits. By participating in this comparison test, we have gained many points of view; firstly; to compare our results with other luminescence dating laboratories results which are world's leading dating laboratories , secondly to demonstrate the accuracy of our results and lastly; to consolidate separately each step of dating measurements (preparation of pure quartz minerals, determination of equivalent dose, determination of saturated water content, and calculation of annual dose etc.) performed in our laboratory on an international scale.

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