

THE DETERMINATION OF EQUIVALENT AND ANNUAL DOSE IN ARCHAEOLOGICAL SHERDS

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ABSTRACT

Atoms and molecules receiving energy from surroundings will be unstable. When these atoms and molecules are stimulated, they return to the ground state. During this process light is emitted from them. This phenomenon is called luminescence. Luminescence resulted from light stimulation is called optically stimulated luminescence (OSL).

Energy is absorbed by a sample from flux of radiation is proportional to luminescence quantity occurring result of stimulation. The absorbed radiation energy is called equivalent dose. The quantity of luminescence due to equivalent dose is known as natural OSL.

There are several techniques to evaluate the equivalent dose. Principally, in evaluating the equivalent dose, the natural OSL from sample is compared with OSL of same or similar sample that is irradiated with a calibrated radioisotope source.

The equivalent dose may be used in dating in archaeological and geological materials. It can be determined that age of historical ruins or occurrence time of still appearing trace of important natural events by dating. In this dating method, our aim is to determine the last time when the material was exposed to light. Optically stimulated luminescence methods have used for dating in archeological and geological samples since 1980's. The basic process is measurement of natural (absorbed dose in buried duration) and annual (absorbed dose in buried in a year) dose of sample for dating. That is, annual dose represents yearly dose rate is absorbed from the flux of nuclear radiation provided thorium-232, uranium-238 and potassium-40 in material as well as by cosmic rays. Besides annual dose is assumed to be constant. It can be determined by measuring amount of radioactive elements in sample. The age can be calculated from the ratio of natural dose to annual dose.

In this study, the potential of OSL for determination of equivalent dose in archaeological sherds from Turkey (Aksaray, Eskişehir) was investigated.

Key Words: OSL, archeology, Annual Dose, Equivalent Dose, Infrared Stimulated Luminescence Dating, Aksaray.

1. INTRODUCTION

Optically Stimulated Luminescence (OSL) dating is a relatively new alternative approach to chronological problems. It is caused by the stimulation of trapped electrons from metastable energy levels, which are related to their subsequent recombination under photon-emission in the crystal lattice. After the archaeological sherds are buried and shielded from further light exposure ionizing radiation from the decay of naturally occurring radioisotopes of U, Th, and K produces free electrons which are subsequently trapped in crystallographic charge defects in silicate minerals. Since these charges accumulate with time, their amount and thus the intensity of the luminescence signal can be used for dating (1,2). The OSL clock, like in TL dating is reset by exposure to sunlight prior to deposition. The luminescence signal is dependent on several factors, which include mineralogy, radioactivity, temperature, grain size, preheat etc. In OSL studies, different preheats, which are necessary to eliminate the unstable OSL components in measurements of luminescence counts (3).

The first work of Huntley *et al.* (1985) showed the potential of OSL in dating applications. The OSL properties of quartz have been used in dating applications and dosimetry studies intensively afterward (2). Hütt, Jeak and Tchonka (1988) firstly showed that it was possible to stimulate a luminescence signal from feldspars using optical stimulation in the near infrared. Both quartz, feldspar and polymineral sediments, which are undatable by conventional radiocarbon methods, can be absolutely dated ($\pm \sim 10\%$) within a range of 100 to 200,000 years.

Luminescence dating was carried out using single-aliquot regeneration (SAR) and multiple-aliquot additive dose (MAAD) procedures on polymineral archaeological samples. The site, located in Aksaray/Turkey,

2. EXPERIMENTAL PROCEDURES

The apparatus (Optical Dating System 9010 Reader) used has been developed by Spooner, Aitken, Smith, Franks and McElroy (1990) (11). The basic luminescence reader incorporates an IR light-emitting diode (LED) module based on the design described by Spooner et al. (1990) (11). All data were collected using an IRSL add-on unit for the 9010 automated reader, which uses TEMT 484 IR diodes run at 40 mA giving a power of about 30 mW/cm² at the sample. Luminescence was detected using a Thorn EMI 9235 QA photomultiplier tube (5).

2.2 SAMPLE PREPARATION

Firstly, the sample was scraped by a riffler to remove 3 mm layer on material (4). Then the material was crushed and sieved to obtain the size 90–125 μm for coarse grain and the size $<20 \mu\text{m}$ for fine grain. This fraction was then washed in 10% HCl and %35 H₂O₂ to remove carbonates and organic materials respectively. The sample was washed in distilled water then in acetone and a suspension of grains was obtained. Then, the grains were resuspended in acetone and deposited on aluminum discs of 10 mm diameter and 0.5 mm thickness. Aluminum discs were put in the bottom of small glass tubes, the suspension was put in this tube and with the evaporation of acetone a thin layer of sample was produced. The using sedimentation procedures are based on Stokes' Law. These discs were then placed on sample trays of the OSL dating system. All the experiments were carried out under the red light.

3. APPLIED METHODOLOGY

There are currently two major analytical approaches; The first method is the single aliquot regeneration method (SAR), which determines an age for each aliquot on discs by matching a regenerated signal from the light reset level to the original natural level, while compensating for apparent sensitivity changes (6,7,8,9). The five discs were prepared. The preheat temperature is 205 °C for 3 min in all steps. The steps of the procedure are as follows: 1) The first aliquot was preheated and the complete IRSL signal (110 s) was measured to define the initial signal level $L(N_0)$. 2) The aliquot was irradiated for D_1 (3 Gy) and preheated. 3) The complete IRSL signal (200 s) was measured to define the first regeneration signal level, $L(N_1)$. 4) The aliquot was irradiated for D_2 (6 Gy) and preheated. 5) The complete IRSL signal was measured to define the second regeneration signal level, $L(N_2)$. 6) The aliquot was irradiated for D_3 (12 Gy) and preheated. 7) The complete IRSL signal was measured to define the third regeneration signal level, $L(N_3)$. 8) The aliquot was irradiated for D_4 (24 Gy) and preheated. 9) The complete IRSL signal was measured to define the fourth regeneration signal level, $L(N_4)$. 10) The aliquot was irradiated for D_5 (60 Gy) and preheated. 11) The complete IRSL signal was measured to define the fifth regeneration signal level, $L(N_5)$. After each irradiation, the aliquot was left for 24 h. The line for this aliquot was plotted against the laboratory dose-luminescence signal. All procedure was repeated for each aliquot. However the natural luminescence (N_0) appeared in background level.

The other multiple aliquot additive dose method (MAAD), is used principally on the fine-grained polymineral. This method applies additional doses (beta or gamma) to the natural luminescence on separate aliquots of the sample effectively building a dose response curve, simulating future dose and interpolating an paleodose dose to the solar reset level (9). The steps of the procedure are as follows: Another archaeological sherd taken from same site were scraped, crushed and sieved. In this methodology fine grain is used. 1) Again, 24 discs were prepared from samples and split into six groups with four discs each. 2) These samples were preheated for 3 min at 205C to remove unstable components and left for 24 h. 3) Samples were OSL-dated using the additive dose method. The growth curve was built from six points: First group natural (N_0), second one $N_0+0.5$ Gy, third one $N_0+2.5$ Gy, fourth one $N_0+4.5$ Gy, fifth $N_0+6.5$ Gy and last one $N_0+8.5$ Gy. Each data point is the mean of the measurements for four discs. A ⁹⁰Sr - ⁹⁰Yb radioactive source was used for irradiation. The typical strength of a 73.5 mCi SIF 1177 Sr-90 β source installed in the system is 0.0322 Gy per second on aluminum. All the IRSL signals were counted for 110 s (3). The graph of changing in luminescence versus to additive dose is drawn and then interpolated. However the natural luminescence (N_0) appeared in background level.

3.2. MEASUREMENTS OF DOSE RATE

The total dose rates quoted in Table 2 include the alpha dose rate, beta dose rate, gamma dose rate and cosmic ray component (Aitken, 1985). The alpha, beta and gamma dose rates were obtained by determining the

concentrations of the major radioactive isotopes of the uranium and thorium series and of potassium within the surrounding materials using X - ray fluorescence. Since the water content of the sample plays an important role on the absorption of the radiation, the saturation water content measurements were performed. The cosmic ray contribution, water uptake during burial and a alpha dose attenuation factor were assumed (1)

4. RESULTS AND DISCUSSION

The measurement of preheating, bleaching, and irradiation should not alter the luminescence sensitivity of the sample (10). The equivalent dose values using multiple-aliquot protocol and single-aliquot protocol were found as background level. Therefore different method were performed in addition above both methods. The 24 aliquots were exposed to sun light for bleaching. Then these aliquots groups were irradiated for 0.05 Gy, 0.1 Gy, 0.2 Gy, 0.4 Gy, 0.8 Gy and 1.6 Gy. That is, the first group was irradiated for 0.05 Gy and so on. Only the last group (1.6 Gy) represented decay signal of luminescence above background level. Otherwise by means of MAAD the signal were observed above $N_0+2.5$ Gy on third group at the first time, but not observed for $N_0+0.5$ Gy. Unfortunately the photomultiplier tube cannot perceive luminescence signal of equivalent dose smaller than 1.6 Gy for this material. Since we couldn't observe OSL signal from $N_0+0.5$, paleodose of the sample must be smaller than 1.1 Gy. It means that the sherd is not archaeological. The average ED (N_0) was assumed as 1 Gy. Annual dose consisting of external and internal dose components are given in table 1 and table 2.

Table 1. For typical pottery the external gamma dose mGy per year (1)

U(ppm)	3	0.344
Th(ppm)	10	0.514
K ₂ O(%)	1.2	0.24
	Total	1.098

Table 2. Equivalent (ED) and total annual dose rate

ED(Gy)	U(ppm)*	Th(ppm)	K ₂ O(%)	Cosmic(mGy/a)	W	F	Annual Dose(mGy/a)	Age(a)=ED/Annual dose
1±0.05	17.8	11.3	1.74	0.07	0.37	0.6	13.28	75±9

*since this value is unexpected high, it may be measured by a different apparatus later.

5. CONCLUSION

This study is successful for very young materials to date by OSL methods. SAR and MAAD methods were used and these are compatible each other. Dating study of very young materials should be resumed.

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