Journal of Radiation and Nuclear Applications
An International Journal

http://dx.doi.org/10.18576/jrna/040202

Luminescence Dating and Dose Estimation of Pottery from Egyptian Archaeological Sites

I. El-Mesady[1], *R. Bergmann*[2], *A. Brania*[3] *and Y. S. Rammah*[1*]

[1]Physics Department, Faculty of Science, Menoufia University, Egypt.
[2]Institute of Atomic and Subatomic Physics, Vienna University of Technology, Stadionallee 2, AT-1020 Vienna, Austria.
[3]Faculty of Archaeology, Department of Conservation, Cairo University, Egypt

Received: 13 Mar. 2018, Revised: 15 Apr. 2019, Accepted: 23 Apr. 2019.
Published online: 1 May. 2019.

Abstract: In the course of thermoluminescence (TL) dating of ancient Egyptian pottery, two pottery sherds were collected from Saqqara, Giza zone (Egypt). The common fine-grain technique was used in the preparation. Quartz (Qz), plagioclase (Plg) and k-feldspars were observed in the samples through the X-Ray Diffraction (XRD) spectra and thin section investigation. The annual dose for each sample was measured, in which an alpha counter was used for uranium and thorium contribution and low-level γ-spectrometer for the potassium contents. Alpha count rates for these samples are found to be 5.12 and 5.14 counts/ksec, in the same order, potassium contents were evaluated and found to be 1.68 and 1.97%. TL ages of sample with low and that with high K concentration are 2542±198 and 1686±194 years respectively, which are verified by historical knowledge. The results were found to be in good agreement with that estimated by archaeologists.

Keywords: Thermoluminescence dating; Quartz and feldspars; X-Ray Diffraction; Egyptian pottery.

1 Introduction

For some types and origins of archaeological materials, artifacts and sediments, luminescence dating is applied [1-9]. Luminescence dating includes both thermoluminescence (TL; stimulated by heat) and optically stimulated luminescence (OSL; stimulated by light [10-17]). In which, solution of the equation; Age $= D_e / \dot{D}$ is required, where D_e and $\dot{D}$ refer to the equivalent and the total dose rates respectively [10].
In principle, thermoluminescence can be used with any material that contains quartz or feldspar. In spite of quartz is pure SiO_2 at the most, it forms in the mineral in several geological settings, i.e. under different conditions of pressure and temperature. Because of the different defects in quartz that are either intrinsic (e.g., Si and O vacancies) or extrinsic formed by impurity atoms (e.g., Al or Ti) and dependent either on the conditions of formation of mineral or subsequent variation, its luminescent emission assorted [18]. Feldspar is describing a group of alumino-silicate minerals including $KAlSi_3O_8$ (K-feldspar) and plagioclase feldspars ($NaAlSi_3O_8$ (Ab - albite) and $CaAl_2Si_2O_8$ (An- anorthite)). Emission from feldspars mostly has been applied in accident dosimetry for a comparatively short period of time aside from dating of the archaeological and geological settings.
Thermoluminescence (TL) dating of pottery has been investigated in different laboratories over the world [19-21] and can be utilized as an absolute dating method [20]. However, very limited attention was given to this application for dating in Egypt in anticipation of high uncertainty.
The aim of the present study was to use a fully automated in-house developed TL reader to date some samples selected from Saqqara, Egypt with uncertainty between 7.7-11.5%. Low level γ spectroscopy was used to evaluate the dose rate from potassium content in the material.

2 Historical Sites

Egypt boasts a long tradition of pottery production that extends since ancient past passing by Graeco-Roman period to the present times. For approximately seven thousand years, Egyptian potters have produced a broad assortment

Corresponding author e-mail: dr_yasser1974@yahoo.com.

of vessels using raw materials, methods, and technologies [22].

Potsherds are found at archaeological sites. Sherds are important sources of information because of the rapidly change of pottery styles with time. It can give conception about occupation sites, human activities and landscape changes [23].

Pottery samples were selected from Saqqara, Egypt. Saqqara is located near the south of Cairo, on the west bank of the river Nile [24]. Its first burials, were built on the ridge of the desert plateau with the starting of the 1st Dynasty.

3 Materials and Methods

Two pottery samples were selected from Saqqara, Egypt. The first sample is red gray in color and the second is red black with thick layer texture. The fine-grain technique was used in the preparation [20], following standard processing procedures in subdued red light. It meets two essential requirements. First, size of grains obtained must be small enough to be easy to penetrate entirely by the short range alpha particles. Second, the grains are formed in a monolayer shape to be convenient for determination of alpha particle effectiveness.

The mineral composition of samples (1 and 2) was analyzed by X-ray diffraction technique. A Philips PW 1840 with CuKα radiation was used. Patterns were obtained by step scanning from 5 to 60 °2θ with generator tension 40 kV and current 25 mA. The technique was used to describe the structure of the powder crystals, in which, data obtained from diffraction are compared by a database maintained by the international center for diffraction data. Thin section which permit a very detailed description of pottery ware, was studied under a transmission light microscope. The slide is viewed in cross-polarized light known as "crossed Nicols".

The fully automated in-house developed TL reader [20, 25] with a heating rate = 5 $^{\circ}Cs^{-1}$ up to 500°C in (5.0) ultra-pure dry nitrogen atmosphere were used for all the TL measurements. Samples pre-heating were done on the same planchet at 190°C for 1 min to remove traps which thermally unstable. Measurement of background was performed in subsequent cycles. An internal $^{90}Sr/^{90}Y$ beta and ^{241}Am alpha sources with dose rates of 0.580 and 3.18 Gy min^{-1} respectively were used to irradiate the samples for producing the growth curves [21]. The TL measurement cycle was as follows. Firstly, in order to calculate the equivalent dose (D_e), a test was performed with four discs. However, two discs were measured for receiving the natural TL. After readout, the same discs were exposed to beta radiation from the internal source for 5 and 10 min, respectively, and re-measured to analyze the second-glow TL for determination of the intercept (I). Two discs more were irradiated with beta for 5 and 10 min to determine the natural and laboratory TL-doses. In this case, the growth is linear with dose, and the equivalent dose D_e can be easily found at the interception point of this line with the added radiation dose artificially. Based on this estimation, another discs were irradiated with suitable doses. The plateau test can be performed plotting the total absorbed dose obtained in different temperature intervals. A clear difference exists between the natural luminescence signal and the luminescence signal by a laboratory irradiation. There is almost no glow at temperature below 200 °C in natural sample whereas the artificially irradiation sample may show considerable glow in this region. The glow curve of the natural thermoluminescence is compared with one of the thermoluminescence by artificial irradiation. And, the division of the glow curve from natural + artificial irradiation sample by the natural glow curve gives a constant region, where the signal does not change in this region, which is called plateau region. In the plateau test, the equivalent dose is calculated in dependence on the glow curve temperature region. The additive growth curves were determined within the plateau areas of the glow curves.

In dating of geological or archeological materials, the important factor is the estimation of the amount of ionizing radiation, to which sample has been exposed since its initial formation. This quantity is called the equivalent dose, D_e. Measurement of D_e is based on the comparison of the natural luminescence signal of the samples with the signal caused from a known quantity of artificial radiation like ^{90}Sr / ^{90}Y for beta and ^{241}Am for alpha irradiation.

Daybreak 582 alpha counting system was used in dose rate measurements. The powdered sample (0.1 mm layer and approximately 0.2 mg weight) was spread on a zinc sulphide scintillation screen that has a 42 mm diameter. Then, both were positioned in a sealed Plexiglas holder. In order to eliminate the radon effect, the Plexiglas holder can be sealed off with an O-ring. The alpha counter gave results in Counts/ksec. The alpha efficiency was determined by comparing the alpha-induced luminescence from the ^{241} Am source with the beta-induced luminescence from the ^{90} $Sr/^{90}$ Y source.

4 Results and Discussion

XRD analysis and thin section microscopy determined both quartz and potassium aluminum silicate with monoclinic system in samples 1 and 2. Fig. 1 (a and b) shows the XRD spectra of both samples 1 and 2, however Figs. 2 and 3 displayed the thin section microscopy illustration of crossed Nicols and plane polarized light.

The materials are composed of quartz (Qz), plagioclase (Plg) and k-feldspars which observed through the XRD spectra and thin section investigation. Phenocrysts are in reddish brown to brown fine groundmass. Orientations of the grains can be recognized most probably due to manufacturing. The grains occupy about 15-25% of our materials. Some vagues can be seen within the materials. Feldspar (plagioclase and k-feldspars) constitutes little amount of grains not exceeding 20%, while quartz about 80% of the grains.

Figs (4-7) exhibit the natural glow curves from samples 1 and 2 along with the artificial measurements after exposure to alpha radiation from the internal source at 15.9, 31.8, and 47.7 Gy and beta doses 2.9, 5.8, and 8.7 Gy. The plateau

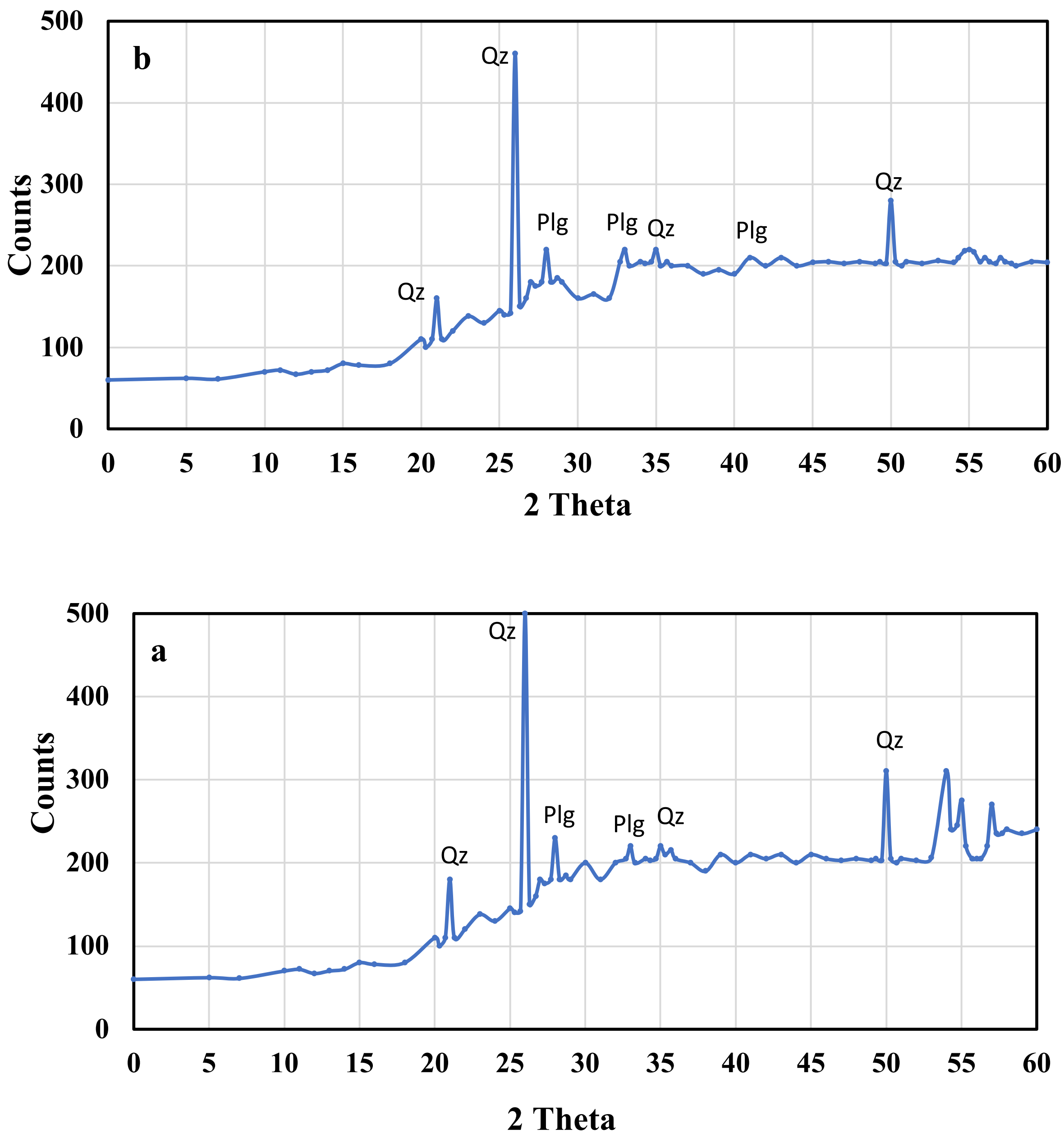


Fig.1: XRD pattern for (a) sample 1 and (b) sample 2. Peaks of quartz (Qz) and plagioclase (Plg) were defined on the spectra.

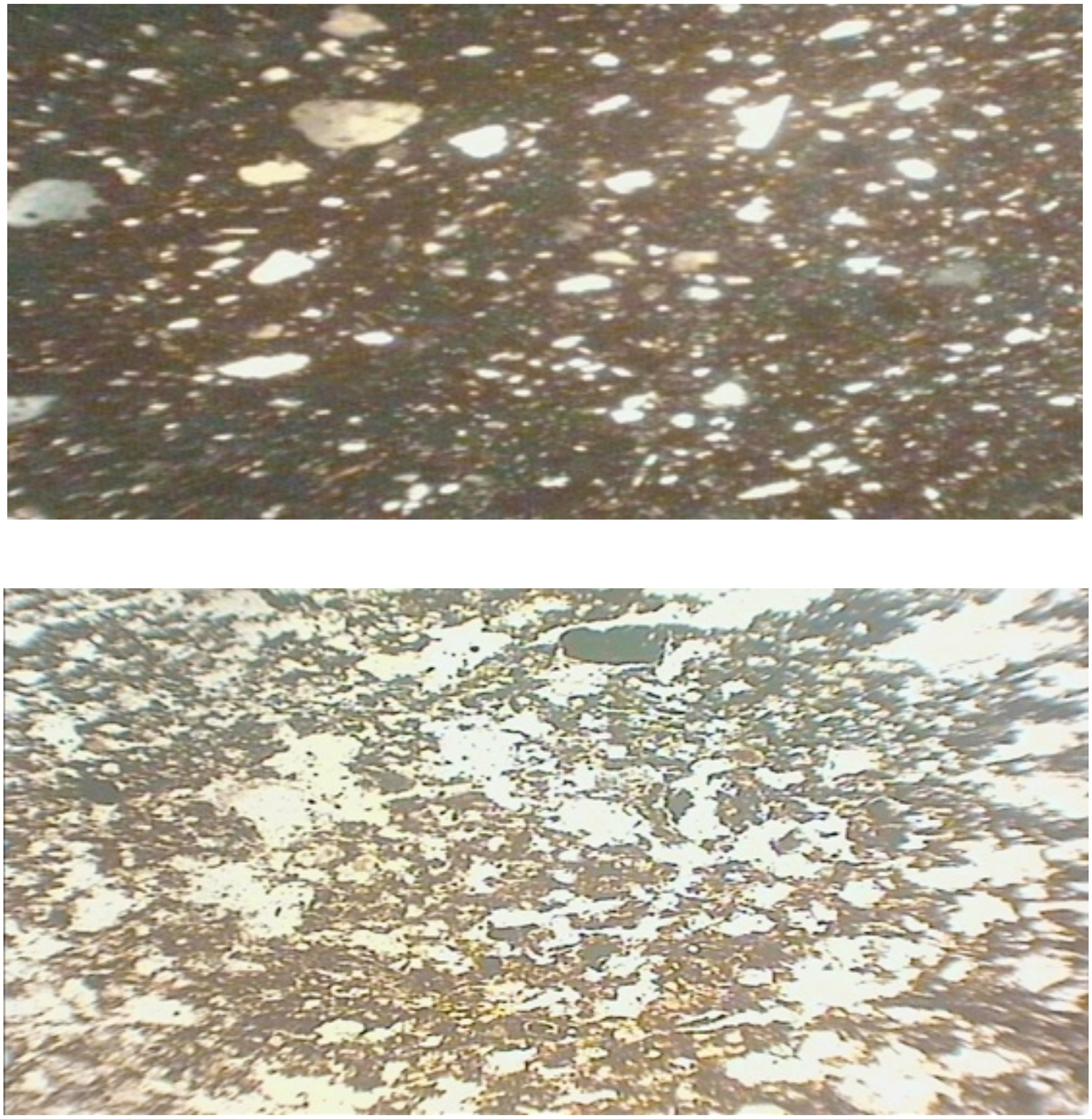


Fig.2: Thin section investigation of sample 1, light spots are mainly feldspars, dark gray crystals are quartz, large quartz grains contain feldspar inclusions: (above) if the slide is viewed in cross-polarized light (Crossed Nicols) and (down) if the polarizer only is inserted (plane polarized light).

region was determined in the temperature range from 277-312 oC, where a constant ratio can be gotten.

Comparison of the natural luminescence signal of the samples with the signal caused from artificial radiation of 90Sr / 90Y beta source and 241Am alpha source were displayed in Fig.8. The luminescence signal increases as a function of artificial and the total dose of the sample summing up its natural and artificial dose as presented in Fig.8, the growth is linear.

Fig. 9 illustrates fading of some prepared discs stored at room temperature and measured at different times, the dosimetric peak was defined at 300°C. The results demonstrated a very small decrease of the TL response during the elapsed period of time from 1 up to 75 days.The dating results were calculated for pottery samples at saturation weight increase 0.2, saturation fraction 0.8 and cosmic dose rate 0.177 mGy/year. The intercept I, alpha irradiation induced equivalent dose Q_{α}, beta irradiation induced equivalent dose Q_{β}, potassium content ^{40}K, the total dose rate and α count rate were presented for both samples 1 and 2 in Table 1. Moreover, the ages were determined depending on the measurement of dose rate from the potassium content in the sample which evaluated by low level γ spectroscopy. The TL ages of Saqqara samples 1 and 2 are 2542 and 1686 year respectively with uncertainty between 7.7-11.5% which found to be consistent with the historical knowledge about the investigated place [26].

Table 1: Specifications of samples 1 and 2.

Specifications	**Sample 1**	**Sample 2**
I (Gy)	0.37±0.21	0.24±0.43
Q_α (Gy)	74.29±3.8	23.7±1.64
Q_β (Gy)	5.38±0.23	5.15±0.45
^{41}K (%)	1.68	1.97
Total dose rate (mGy/year)	2.26	3.2
A count rate (Counts/ksec)	5.12	5.14
Age (years)	2542±198	1686±194

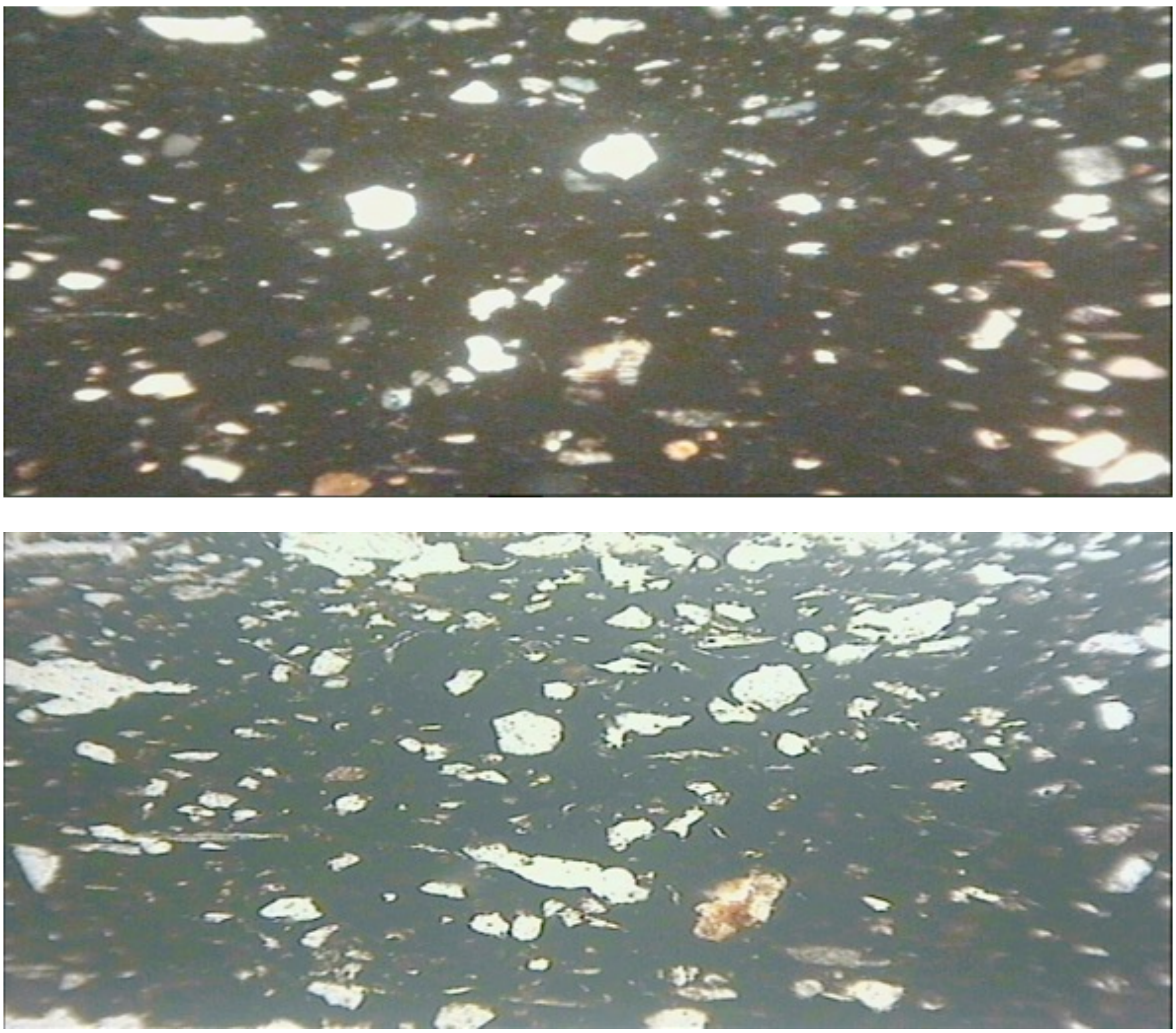


Fig.3: Thin section investigation of sample 2, light spots are mainly feldspars, dark gray crystals are quartz, large quartz grains contain feldspar inclusions: (above) if the slide is viewed in cross-polarized light (Crossed Nicols) and (down) if the polarizer only is inserted (plane polarized light).

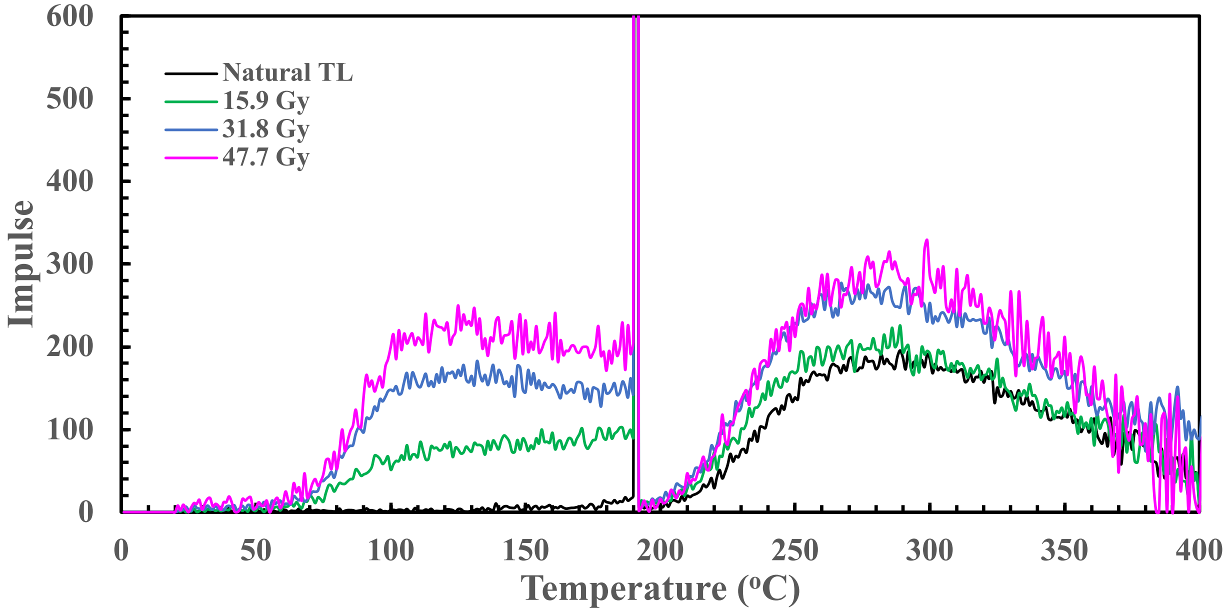


Fig. 4: The average of natural and artificial TL measurements from sample 1 (natural TL, 15.9, 31.8, and 47.7 Gy) alpha irradiation.

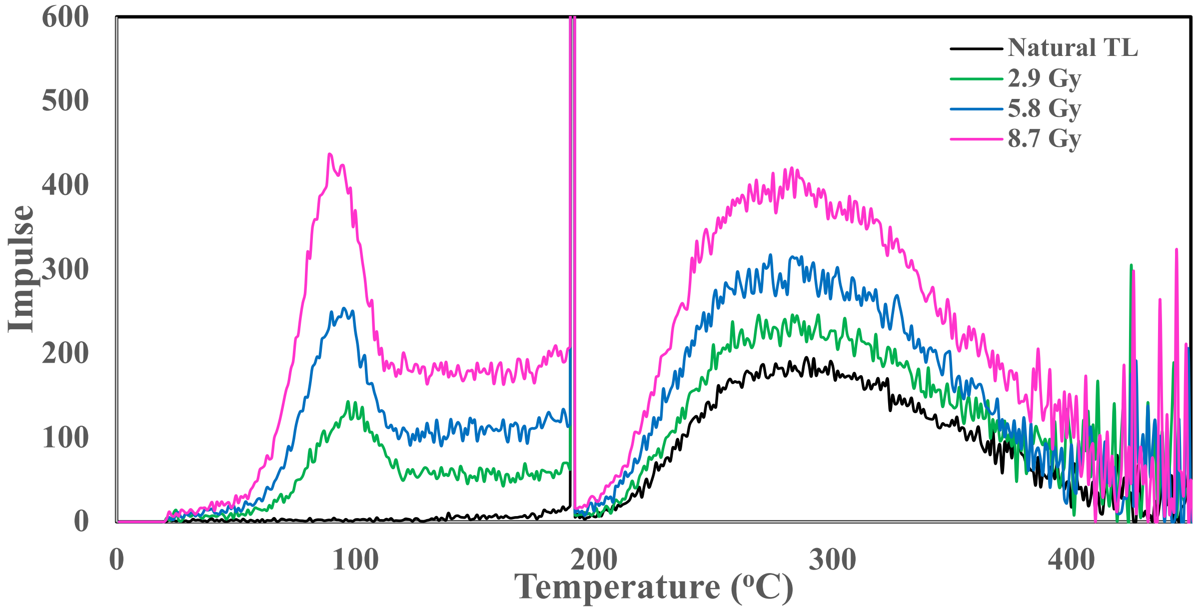


Fig. 5: The average of natural and artificial TL measurements from sample 1 (natural TL, 2.9, 5.8, and 8.7 Gy) beta irradiation.

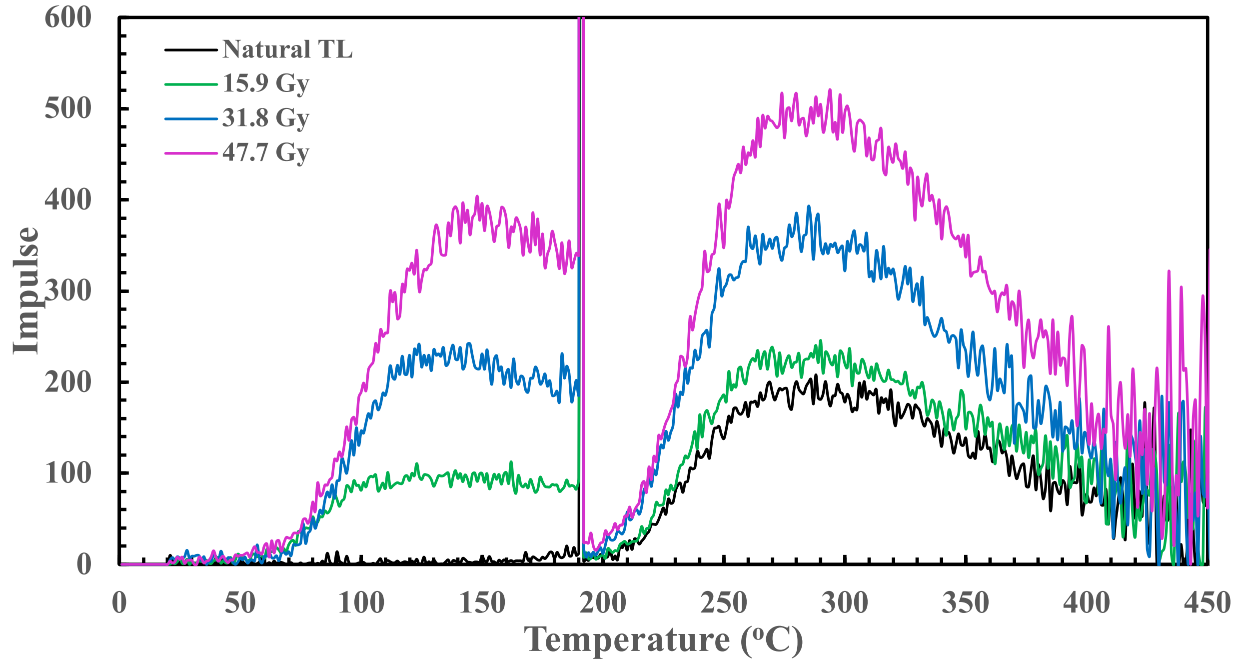


Fig. 6: The average of natural and artificial TL measurements from sample 2 (natural TL, 15.9, 31.8, and 47.7 Gy) alpha irradiation.

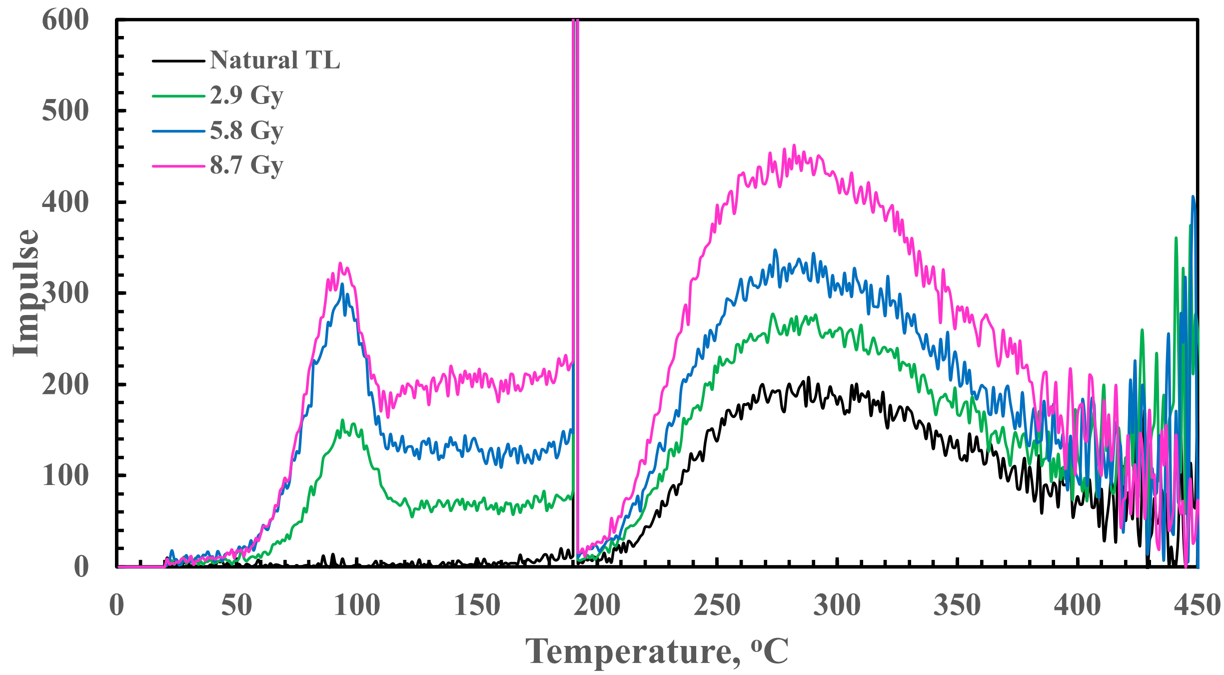
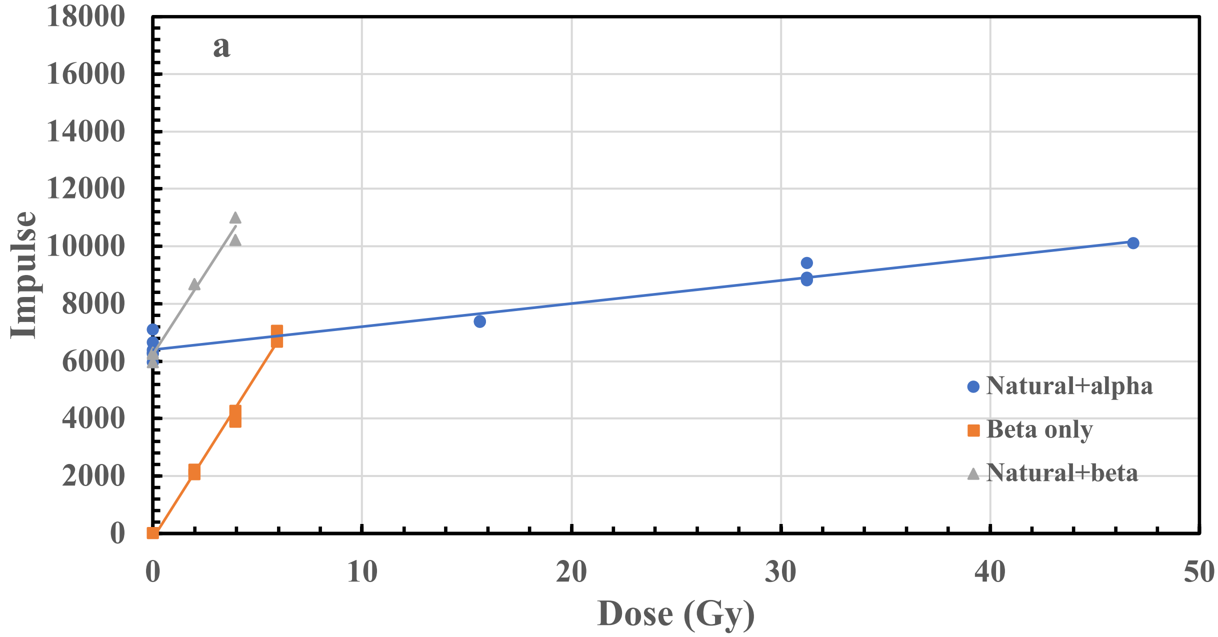


Fig.7: The average of natural and artificial TL measurements from sample 2 (natural TL, 2.9, 5.8 and 8.7 Gy) beta irradiation.



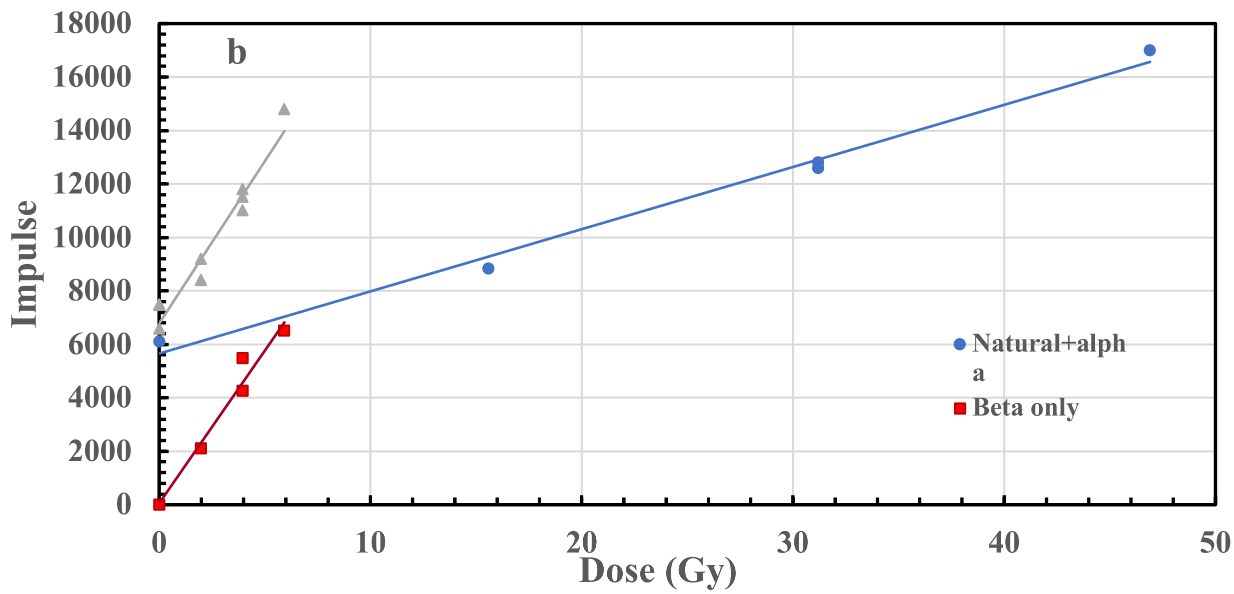


Fig.8: The natural and artificial TL signal from (a) sample 1 and (b) sample 2 as a function of dose. Green line refers to Natural + artificial alpha irradiation (N+α), blue line refers to Natural + artificial beta irradiation (N+β) and red line refers to only artificial beta irradiation.

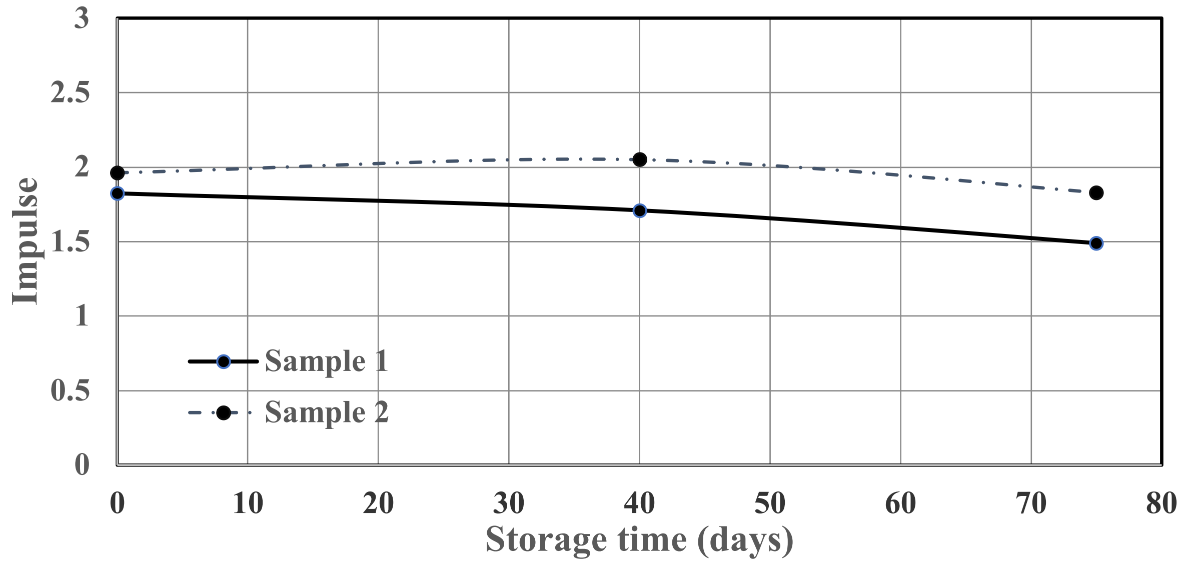


Fig.9: TL-signal versus the storage time.

5 Conclusions

In this study, pottery sherds from Saqqara archaeological site, Giza, Egypt were dated. XRD analysis and thin section microscopy identified in samples 1 and 2 both quartz and potassium aluminum silicate. They are composed of quartz, plagioclase and k-feldspars phenocrysts in reddish brown to brown fine groundmass. The grains are found to occupy about 15-25% of them. Feldspar (plagioclase and k-feldspars) constitutes little amount of grains not exceeding 20%, while quartz about 80% of the grains. The annual dose ($\dot{D}$) was calculated from the concentration of radioactive isotopes of (U, Th and ^{40}K). Alpha count rates for these samples are found to be 5.12 and 5.14 counts / ksec and ^{40}K to be 1.68 and 1.97% for sample 1 and 2 respectively. The TL ages of Saqqara samples 1 and 2 are 2542 and 1686 year respectively with uncertainty between 7.7-11.5%. These ages are compatible with the historical knowledge about the investigated place. The agreement between archaeological studies and TL dating results make a potential for TL dating to contributed significantly for establishing the chronology of Saqqara site.

Acknowledgements: Authors would like to acknowledge Prof. Dr. Saleh Ismail, Institute of Atomic and Subatomic Physics, Vienna University of Technology, Vienna, Austria, Prof. Dr. Abdel Azim Hussein, Faculty of Science, Physics Department, and Prof. Dr. Maher Dawoud, Faculty of Science, Geology Department, MenoufiaUniversity,

Referance

[1] L.T. Filho, G.M. Ferraz and S. Watanabe, Nucl Instrum. Method Phys. Res. B., **229**, 253, 2005.

[2] A.L. Yusoff, R.P. Hugtenburg and D.A. Bradley, Radiat. Phys. Chem., **74**, 459, 2005.

[3] E.M. Yoshimura and E.G. Yukihara, Nucl. Instrum. Method Phys. Res. B., **250**, 337, 2006.

[4] B. Engin, Food Control., 18, 243, 2007.

[5] J.S. Sanchez, D.F. Mosquera and J.L.M. Fenollós, Geochronometria., **31**, 21, 2008.

[6] M.A. Atlıhan, E. Sahiner and F.S. Alanyal, Radiat. Phys. Chem., **81**, 594, 2012.

[7] A. Chruścińska, A. Cicha, N. Kijek, P. Palczewski, K.R. Przegiętka and K.S. Tuszynska, Geochronometria., **41**, 352, 2014.

[8] N. Klasen, P. Fischer, F. Lehmkuhl, A. Hilgers, Geochronometria., **42**, 67, 2015.

[9] A.O. Sawakuchi, V.R. Mendes, F.N. Pupim, T.D. Mineli, L.M. Ribeiro, A. Zular, C.F. Guedes, P.C. Giannini, L. Nogueira, W.S. Filho and M.L. Assine, Brazilian journal of Geology, **46**, 209, 2016.

[10] N. Zacharias, A. Schwedt, J.B. Garrigos, C.T. Michael, H. Mommsen and V. Kilikoglou, J. Archaeol. Sci., **34**, 1804, 2007.

[11] F. Daniels, C. Boyd and D. Saunders, Science., **117**, 343, 1953.

[12] H. McDougall, Thermoluminescence of geological materials, Academic Press, London, 1968.

[13] F. Preusser, M.L. Chinthambo, T. Gotte, M. Martini, K. Ramseyer, E.J. Sendera, G.J. Susino and A.G. Wintle, Earth Sci. Rev., **97**, 184, 2009.

[14] M. Aitken, Physics and Archaeology, Clarendon Press, Oxford. 1974.

[15] S. Fleming, Dating in Archaeology, J.M.Dent & Sons, London, 1976.

[16] S. Fleming, Thermoluminescence Techniques Archaeology, Clarendon Press, Oxford, 1979.

[17] A. Sankaran, K. Nambi and C. Sunta, Proc. Indian National Science Academy, **49**, 18, 1983.

[18] S. McKeever, Thermoluminescence of Solids, Cambridge University Press, Cambridge, 1989.

[19] M. Abdel-Wahab, S. EL-Fiki, M. EL-Fiki, M. Gomaa, S. Abdel-Kariem, N. EL-Faramawy, Radiat. Phys. Chem., **47**, 697, 1996.

[20] R. Bergmann, M. Hajek, M. Fugger, N. Vana, Radiat. Meas., **43**, 781, 2008.

[21] E. Bakraji, Nucl. Instrum. Method Phys. Res. B: Beam Interactions with Materials and Atoms., **269**, 2052, 2011.

[22] A. Carol. Redmount. Archaeological Research: The Egyptian modern pottery project: University of California pilot phase findings, 2003.

[23] M.A. Hamdan, S.M. Martinez, M.T. Garcia Valles, J.M. Nogues, F.A Hassan, R.J Flower, M.H. Aly, A. Senussi and E.S. Ebrahim. Ancient Egyptian Pottery from the subsurface floodplain of the Saqqara–Memphis AREA: ITS mineralogical and Geochemical implications Archaeometry., **56**, 987, 2014.

[24] http://josephandisraelinegypt.wordpress.com/2009/05/03/the-location-of-saqqara-in-egypt

[25] http://www.ancient-egypt.org/index.html.

[26] T Berger, M Hajek, W Primerano, N Vana Radiat. Prot. Dosim., **101**, 363 (2002).