THERMOLUMINESCENCE DATING OF POTTERY OBJECTS FROM TELL AL-HUSN, NORTHERN JORDAN

S. Khasswneh[1], Z. al-Muheisen[1], R. Abd-Allah[2]

[1]Faculty of Archaeology and Anthropology, Yarmouk University, Irbid 21163, Jordan

[2]Conservation Dept., Faculty of Archaeology, Cairo University, Orman 12611, Giza, Egypt

ABSTRACT

During the excavation works adopted in 2008 by Yarmouk University team, with the support of the Jordanian Department of Antiquities at Tell al-Husn archaeological site located in northern Jordan, a considerable collection of pottery objects and shards were found. Dating these pottery objects was very important to reveal the historical and archaeological aspects of this significant site in northern Jordan. Thermoluminescence (TL) techniques is one of the most accurate and absolute method for dating pottery. An appropriate collection of pottery shards was collected and prepared for measurement using this technique. It shown that pottery objects are dated back around to 3400 to 3700 years BP (Late Bronze Age). This age is consistent with that estimated by archaeological (typology) studies.

INTRODUCTION

Archaeological background

The first season of excavations at Tell al-Husn was conducted in the summer of 2008 by the Faculty of Archaeology and Anthropology at Yarmouk University, with the support of the Jordanian Department of Antiquities, under the directorship of Prof. Zeidoun al-Muheisen.

During this excavation works, a considerable collection of pottery objects and shards were found. Dating these pottery objects was very important to reveal the historical and archaeological aspects of this significant site in northern Jordan.

Geographic location

Al-Husn, the ancient and modern village is located nearly 8 km south of Irbid and 70 km north of Amman (Fig. 1). The highway which connects Irbid with Amman intersects the town of Husn, dividing it into eastern and western sections. The geographical coordinates of al-Husn are 32º 27' N, 35º 37' E and it is located about 680 m. above sea-level. The moderate, warm climate of this region, fertile soil, and strategic location have been instrumental in making it a settlement area of choice since the Chalcolithic period (before 4000 BC), throughout the Bronze, Iron, Persian, Hellenistic, Roman, Islamic periods, and even up to the present.

Al-Husn encompasses a total surface area of about 58.5 km² , out of which about 20 km² constitutes the main city. The remaining surface area is used for agriculture and has been cultivated with all kinds of crops and olive trees especially on the heights, where stone quarrying is also carried out (Al-Muheisen, 2009).

Naming

'Al-Husn' means a 'well-fortified place' or 'castle'. It is generally assumed that the name of Tell al-Husn was derived from the Umayyad fort which had been built on the Tell located in the north of the town. Some parts of this fort were uncovered during the first season of excavation. A mosque, built during the Umayyad period, was also excavated (Al-Muheisen, 2009).
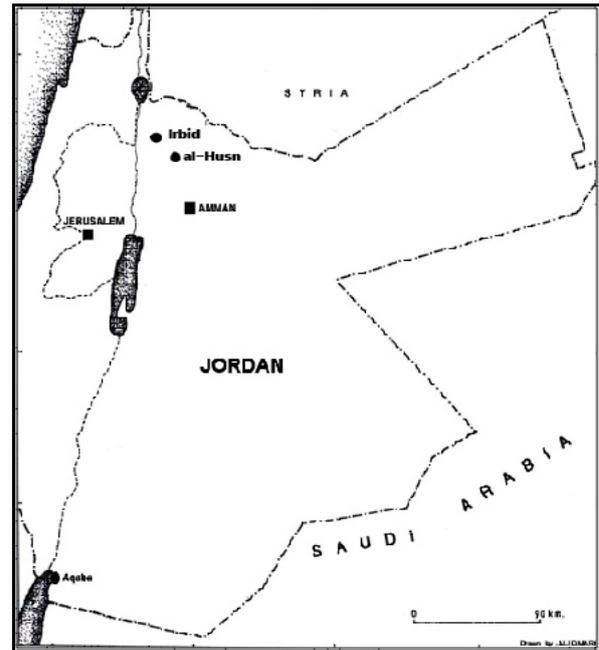

Fig. 1: Location map of the site of al-Husn

Tell al-Husn

Tell al-Husn is located at the north of al-Husn modern village and dominates the plains of Hawran from the northern and eastern sides. It is large in comparison to other towns, with a surface area of about 90000 m² , and an elevation of 26 m above street level (Figs 2 and 3).

Fig. 2: Air-photo of Tell al-Husn before excavation works

The Tell obviously played an important role since the Calcolithic Age (4000 BC), and developed in the Late Bronze Age (1550-1200 BC), as indicated by findings of dense settlements. The site began to grow in significance during the

reign of Thutmose IV (Menkheperure) (1419-1386) BC, and developed during the second phase of the Late Bronze Age (1410-1340 BC) (Mittmann 1966, Al-Muheisen 2009).

Fig. 4: A view of excavation works at Tell al-Husn 2008

Fig. 3: A contour map of Tell al-Husn

EXCAVATION RESULTS

The first season of excavation in 2008 took place in the central part of the site which contains three areas: A, B, and C in order to determine the successive periods of settlement at the site. Eleven 5x5 meter squares (N3, N4, N5, N6, N7, M4, M5, M6, M7, E 7, E8, F7, and F8) were excavated in three locations in Area A.

Distinctive architectural remains consisting of a number of rooms and small Umayyad mosque were uncovered (Fig. 5).

On the other hand, twelve 5x5 meter squares (D20, D21, D22, E20, E21, E22, F21, F22, D26, D27, E26 and E27) were excavated in Area B, which consisting of the architectural remains of walls and fortress dated back to Byzantine and Umayyad periods.

A big pottery *Taboon* 120 cm in diameter was also uncovered. In the northern squares (E22 and F22) some architectural remains, which can be dated back to the Hellenistic and Roman periods, were found. Only two 5x5 meter squares (B40 and C40) were excavated in Area C, which is located on the EN side of the Tell's foot.

From this Area a considerable collection of pottery findings was excavated inside some remains of rooms and walls dated to the Iron and late Bronze Age.
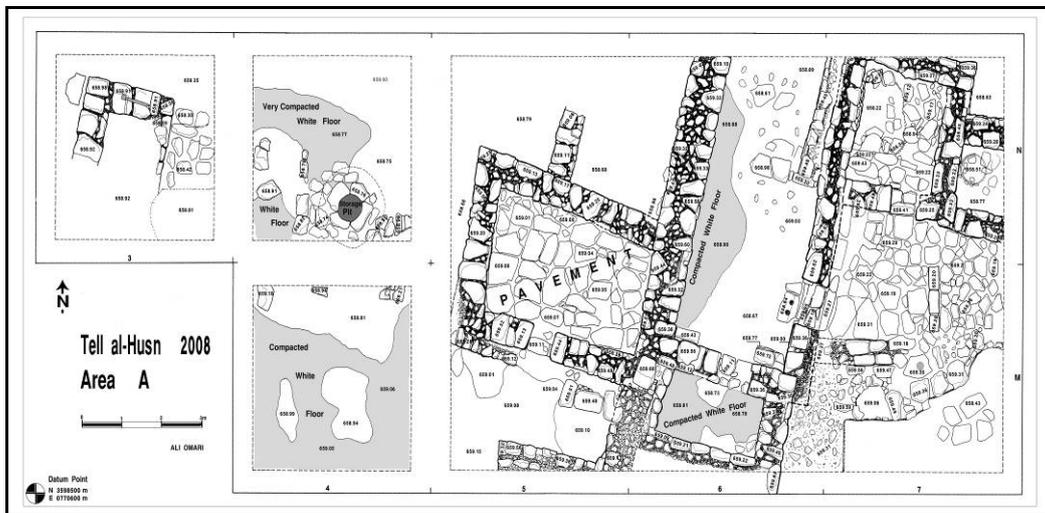

Fig. 5: plan of the site and excavated remains

The most significant works were carried out at a distance of 50 meters north-east of Tell al-Husn where the cave or shaft tomb was uncovered (Fig. 6).
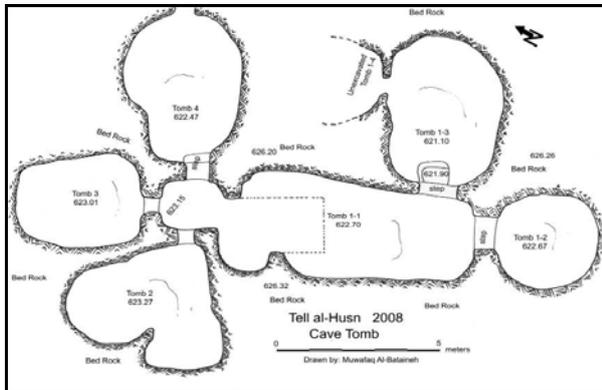

Fig. 6: Plan of the shaft tomb, in which a considerable collection of pottery objects were found.

Four cemeteries consisting of numbers of chambers were excavated, in which a great collection of intact pottery objects were found. According to the associated finds such as bony skeleton, metal and stone tools, the tomb was certainly dated back to the late Bronze Age (1410-1340 BC) (Al-Muheisen, 2009).

Pottery finds from Tell al-Husn

Addition to the great numbers of pottery shards (Fig. 7), a considerable collection of intact pottery objects were found during the first season of excavation at Tell al-Husn (Fig. 8). These objects are of different types (Bottles, vessels, plates, cooking stoves, *taboons*, pots, jars and lamps) and colors (red, brown, yellow and black). Paddle and hand-forming was the most technique used for making these objects, whereas stamping and engraving were the decoration methods used. Few glazed bottles were found in the shaft tomb. The objects are varying in size, so their walls ranging between 2-7 mm in thickness. Typologically, these pottery finds were dated back to end of the Late Bronze Age (1410-1340 BC) (Al-Muheisen, 2009).

Fig.7: Pottery shards of different sizes and types spread on the surface of Tell al-Husn

Fig.8: Pottery artifacts typologically dated to end of the Late Bronze Age from Tell al-Husn

Pottery in archaeology and archaeometry

Pottery in archaeology is an important material for archaeologists due to its ability in providing information on many aspects of the past, including chronology (Gregg *et al.*, 2009). In the last century scientists worked to establish new methods for dating pottery, different relative techniques used to determine the chronological sequence of pottery artifacts. Such methods are stratigraphy where artifacts are arranged according to their position in strata and the typological sequencing which is a systematic classification for pottery artifacts into types based on similarities in form, construction, style, and content. The relative dating techniques had dominated the archaeologists' works in the first half of last century, however these methods considered as inadequate methods of dating, there was a need for more accurate and precise dating method to give certain and numerical age for pottery artifacts. Daniels (1953) suggested a new method of dating geological materials that depends on the luminescence energy released of minerals by heating or being subjected to light.

Since then major efforts lead to the development and employment of thermoluminescence (TL) dating in archaeology. Aitken (1974, 1985) established the basic principles and protocols for TL dating. In Jordan TL dating lab was first established at the Faculty of Archeology and Anthropology labs in Yarmouk University at the end of 2005, to be part in excavations carried out by the Faculty and other excavation expeditions. The lab is equipped with all necessary facilities needed for dating process; it is composed of three main parts, the chemical preparation lab that used in pottery sample preparation, the TL lab and the gamma scintillation lab which used for measuring radioisotopes concentrations existed in the soil.

The principle of luminescence dating based on the thermally stimulated emission of light from crystalline materials such as quartz when heated to 500ºC; this emission is referred to the release of trapped electrons in defects of the mineral's crystal lattice structure. For archaeological ceramics and pottery, the first firing for quartz minerals drains out all the TL energy stored in quartz and sets the age clock to zero, afterward by prolong exposure to the radiations made by the radioisotopes Th-232, U-238 and K-40 present in the soil and with contribution of comic's rays; thermoluminescence energy are restored in the crystals, where more electron traps are created and hence more energy is deposited in the crystals (Fleming, 1976, 1979; Aitken, 1985; Liritzis 2000). The age is defined by the formula:

Age (kilo years) = Equivalent Dose ED (Gy) / $aD_\alpha + D_\beta + D_\gamma + D_{cosmic}$ (Gy/ kilo year)

Where the equivalent dose (ED) is the laboratory irradiation dose that reproduces the natural TL signal, D_β and D_γ is the annual beta and gamma dose rate respectively and D_{cosmic} is the annual cosmic dose rate, aD_α is the annual effective alpha dose rate, the *a* factor is represent the ratio between thermoluminescence sensitivity from beta particles and thermoluminescence sensitivity from alpha particles. The annual dose is measured by specifying the concentration of radioisotopes Th, U and K in the surrounding soil in term of (Bq/kg), the concentrations are then evaluated and converted in terms of energy absorbed per unit of time (Gy/kilo year) based on measurements of the energy of emitted particles and radiations as published in nuclear data tables, the conversion factors are listed in different references to ease the process of calculating the annual dose for dating specialists, earlier use of nuclear data was by Aitken 1974, Bell (1976 and 1979), Nambi and Aitken (1986) used the Evaluated Nuclear Structure Data File database (ENSDF) published by the International Commission on Radiation Protection to present the values of energy released in terms of dosimetry values, reassessment was presented by Liritzis and Kokkoris (1992) and Ogoh et al (1993) and later update for the conversion values presented by Adamiec and Aitken (1998), without significant differences.

EXPERIMENTAL

Sample collection

The samples were collected under the reduced red light conditions, number of pottery shreds were excavated from two main positions labeled as H1 and H2, from each position three groups of pottery shreds were taken and contained within opaque bags that do not allow light transition. Soil samples were obtained with each group of pottery shreds sample.

Sample preparation

The quartz inclusion technique was applied. The first millimeters of the pottery shreds were scrubbed off and discarded using dental drill to avoid soil contamination because of its high level of geological Thermoluminescence, and beta contribution which is transitional between that corresponding to the pottery radioactivity and that corresponding to the soil radioactivity Aitken (1985), shreds were crushed using the agate pestle and mortar, grains less than 45μm in size were obtained by sieving the crushed material with suitable mesh size.

The grains were later chemically treated to remove calcites and organic matters using 10% HCl and 30% H_2O_2 independently for 24 hours. To extract grains between four and 11 micrometer we applied the water floatation process (Zimmerman 1978; Wedia and Junding, 1989) which based on separating grains under the gravitational field, 20 minutes is needed to allow grains bigger than 11μm to deposit in a 50cm tube in room temperature, obtaining the suspended material and repeat the process several times to enrich the 11μm and discard any existing bigger size grains, the last sedimentation was for 40 minutes to discard grains that are less than 4μm which exist in the first 6 cm of the tube, the whole process of preparation and extraction were carried out under reduced red light to minimize bleaching for the TL signals. Finally the fine grains were plated over aluminum disks with 1mm thick and 10mm in diameter, this step were performed by setting the disks in 40mm glass tube and pour the suspension of grains over and allow for evaporation in oven fixed at 60ºC.

TL instrument

All measurements were accurately carried out in TL lab located in the Faculty of Archae- ology and Anthropology at Yarmouk University, The TL reader used is Daybreak 1100 Automated TL system, it is equipped with a sample-holder which can carry 20 discs and is coupled to a PC loaded with the software to control the whole reading process. Artificial irradiation for the samples was performed using Sr-90 beta source, the nominal activity of the source is 100 mCi($3.7×10^9$ Bq) as reported by AEA Technology, the source was calibrated in may 2008 using Co- 60 gamma ray facility to deliver reference dose; the calibrated dose rate of the source was specified as of $5.64*10^{-2}$ Gys^{-1} (Khasswneh, 2009).

RESULTS AND DISCUSSION

Annual Dose

The annual dose evaluated by the contribution of the radiation of cosmic rays and the contribution of gamma radiation, beta and alpha particles taken from the radioisotopes Th- 232, U-238 and K-40 from the soil surrounding the pottery samples. 500 gm of soil surrounding pottery fragments were collected and measured using High Purity Germanium Detector, the results of the radioactive elements and the water content of the soil and pottery samples are listed in Table 1.

Tables 2 and 3 represent the evaluated dose rate for gamma and beta in term of (Gy/ka) respectively after applying the correction made of water content and water uptake *F*, Table 4 represent the dose rate evaluated by the contribution of alpha particles, but since the effect of alpha particles are less than the effect of gamma radiation and beta particles Aitken (1985); the *a*-values were derived for the five samples and inserted in evaluating the effective alpha dose rate.

Table 1: Soil water content and the concentration of Th-232, U-238 and K-40 as measured using High Purity Germanium Detector.

Sample	Th_232 (Bq/Kg)	U_238 (Bq/Kg)	K_40 (Bq/Kg)	Water content (soil)	Water content (pottery)
H1a	17.25±0.04	66.55±3.86	407.32±0.24	0.57014	0.22631
H1b	18.99±0.04	70.6±4.11	455.51±23.39	0.423844	0.167
H2a	14.45±0.05	72.71±4.18	330.71±17.4	0.712531	0.28964
H2b	14.17±0.05	72.34±3.4	351.34±18.57	0.124193	0.667
H2c	18.18±0.04	72.67±4.1	426.07±21.96	0.44776	0.8669

Table 2: Evaluated dose-rate of gamma rays (Gy/ka) after corrections for the water content of soil and pottery and Water uptake of soil = 0.8 and uncertainty = ±0.2.

Sample	H1a	H1b	H2a	H2b	H2c
Th-232	0.161±0.0066	0.187±0.00717	0.128±0.00603	0.157±0.00595	0.177±0.0069
U-238	0.485±0.0455	0.544±0.04843	0.504±0.0494674	0.629±0.0435	0.554±0.0488
K-40	0.26±0.046	0.307±0.02176	0.201±0.01609	0.267±0.01715	0.284±0.02041
sum	0.906±0.00982	1.038±0.8380313	0.833±0.049834	1.053±0.04393	1.015±0.0493

Table 3: Evaluated dose rate of beta particles (Gy/ka) after corrections for the water content of soil and pottery and Water uptake of soil = 0.8 and uncertainty = ±0.2.

Sample	H1a	H1b	H2a	H2b	H2c
Th-232	0.104±0.0035	0.118±0.00384	0.085±0.002934	0.071±0.002878	0.085±0.003677
U-238	0.707±0.051	0.771±0.0547	0.751±0.0558	0.642±0.0477	0.6±0.05492
K-40	0.969±0.032	1.113±0.0717	0.765±0.053	0.698±0.05654	0.787±0.0204
sum	1.78±0.0398	2.002±0.923	1.601±0.0558	1.411±0.0478	1.472±0.0550

Table 4: Dose rate of alpha particles per U, Th per sample (Gy/ka) for the soil after corrections for the water content of pottery and Water uptake of soil = 0.8 and uncertainty = ±0.2, a-values and the calculated effective alpha dose rate.

Sample	H1a	H1b	H2a	H2b	H2c
Th-232	2.73828±0.1	3.11093±0.103	2.21966±0.078	1.82524±0.077	2.15754±0.0986
U-238	13.24±0.98	14.5028±1.0453	14.0054±1.066	11.6845±0.91	10.81439±1.05
sum	15.98± 0.98	17.6±1.050	16.22±1.068	13.509±0.9151	12.97±1.054
a-value	0.091739	0.09263	0.08987	0.09465	0.0876
Effective alpha dose	0.906257± 0.98	1.008885±1.050	0.89761±1.068	0.786994±0.9151	0.701849±1.054

Evaluation of the Equivalent Dose

The equivalent dose ED was determined using the additive dose method for multiple aliquots; the radioactive source Sr-90 beta source was used to deliver the artificial lab dose specified as 10, 20 and 30 Gy. A pre heating was performed for 200°C for 120 seconds and the ramp rate control was fixed to 10°C/s, to obtain the equivalent dose (ED); TL glow curves of natural TL and natural TL plus the generated thermoluminescence after and the laboratory additive doses were recorded, Figure 9 provides a typical glow curve of the sample H1a.

TL signals were integrated in the range 200 – 500°C, the plateau test was applied (fig 10), and the growth curve was plotted on the basis of the measurement of the natural TL signals and the additive doses using four samples for each dose (Fig 11). Table 5 shows the evaluated equivalent dose for each sample and the calculated age considering the cosmic rays equals to 0.156 (Gy/ka) that calculated based on the depth of the samples by Prescott and Hutton (1994).

Sample	Temperature range(°C)	Equivalent Dose(Gy)	Age (ka)
H1a	375- 445	11.8± 0.23	3.418± 0.070
H1b	370- 445	15.0± 0.20	3.569± 0.058
H2a	375- 435	13.0± 0.33	3.728± 0.104
H2b	365- 440	12.3± 0.17	3.610± 0.070
H2c	365- 385	12.0± 0.31	3.586± 0.118

Table 5: The evaluated ED per temperature region (TL peak) and the calculated age (BP) of each pottery sample.

Fig 9: Glow curves of the unirradiated disks (N) , the irradiated disks with ⁹⁰Sr beta radiation source at three different doses (N+10Gy, N+20Gy and N+30GY), all for sample H1a.
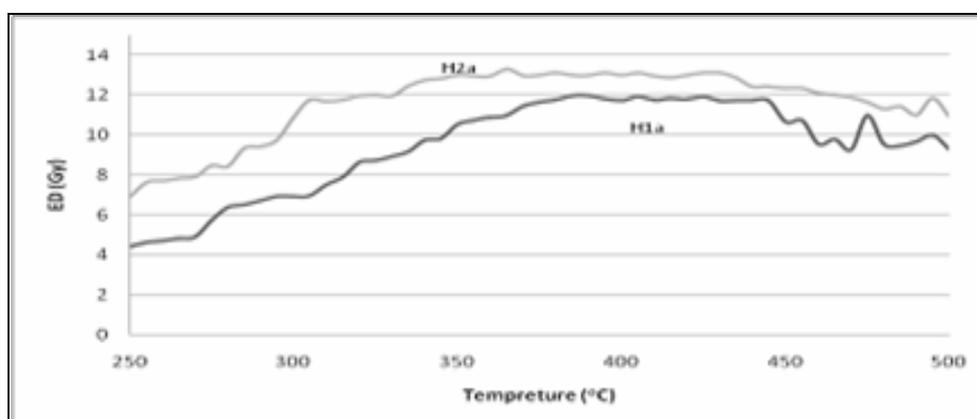

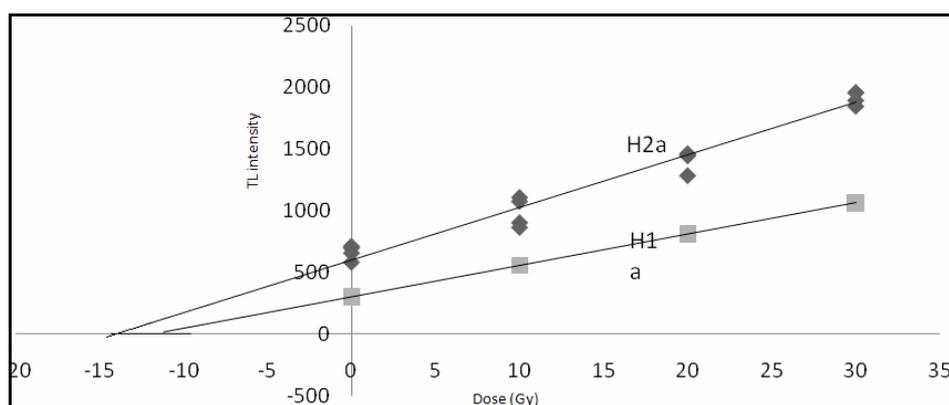**Fig 10: Plateau test for samples H1a and H2a showing the stability region.**

Fig 11: The growth curve of samples H1a and H2a representing the variation of the TL intensity as a function of the added dose.

CONCLUSION

The paper deals with the Thermolumines-cence (TL) dating of ancient pottery excavated in summer 2008 from Tell al-Husn archaeologi-cal site, northern Jordan, this is the first attempt of using luminescence as a dating method for archaeological artifacts in Jordan. The annual dose rate and archaeological dose were ob-tained for number of pottery shards and dated back around to 3400- 3700 years BP. This age is consistent with the estimated age by archaeo-

logical studies performed on this archaeological site (Al-Muhesin, 2009), which point out that this site was extensively settled in Late Bronze Age (1550-1200 BC) and during the second phase of Late Bronze Age (1410-1340 BC) to which the pottery artifacts have typologically been dated.

The agreement between archaeological studies and TL dating results make a potential for TL dating to contributed significantly for establishing the chronology of Tell al- Husn site.

AKNOWLEDGEMENTS

REFERENCES

Adamiec, G. and Aitken, M. (1998) Dose-rate conversion factors: update. *Ancient TL* 16 (2): 561 – 572.

Aitken, M. (1974) *Physics and Archaeology*, Clarendon Press, Oxford.

Aitken, M. (1985) *Thermoluminescence Dating*, Academic Press, Oxford.

Al Khasswneh, S. (2009) Dose Rate Calibration of Sr-90 for Thermoluminescence Dating Using the Additive Dose Method. *Abhath Al- Yarmouk "Basic Sci. & Eng."* 18:139-146.

Al-Muhesin, Z. (2009) Preliminary report of the first season of excavations at Tell al-Husn 2008, *Newsletter of Faculty of Archaeology and Anthropology* (38): 1-5.

Atlihan, M., and Meric, N. (2008) Luminescence dating of a geological sample from Denizli, Turkey. *Applied Radiation and Isotopes* 66: 69-74.

Bell W. (1976) The assessment of the Radiation doe- rate f or Thermoluminescence dating. *Archaeometry* 18: 107- 110.

Bell W.T. (1979) Thermoluminescence dating: Radiation dose- rate data, *Archaeometry* 21*:* 243- 246.

Daniels, F., Boyed, C. A. and saunders, D. F. (1953) Thermoluminescence as a research tool. *Science* 117: 343- 349.

Fleming S. (1976) *Dating in Archaeology*, J.M.Dent & Sons, London.

Fleming S. (1979) *Thermoluminescence Techniques Archaeology*, Clarendon Press, Oxford.

Gregg, M., Banning, E., Gibbs, K., and Slater, G. (2009) Subsistence practices and pottery use Neolithic Jordan: molecular and isotopic evidence. *Journal of archaeological science* (36): 937-94.

Guibert, P., Bechtel, F., Schvoerer, M., Müller, P., Balescu, S. (1998) A new method for gamma

Dose–rate estimation of heterogeneous media in the TL dating, *Radiation Measurements*, 29: 561 – 572.

Liritzis Y. and Kokkoris M. (1992) Revised Dose rate data for Thermoluminescence/ ESR dating. *Nuclear Geophysics* 6: 423- 443.

Liritzis I. (2000) Advances in thermo- and opto- luminescence dating of environmental materials (sedimentary deposits): part I: techniques, *GLOBAL-NEST,* vol.2, No 1, 3-27, & part II: applications. *GLOBAL-NEST, vol.2, No 1, 29-49*

McKeever, I. (1989) *Thermoluminescence of Solids*, Cambridge University Press, Cambridge.

Mittmann, S. (1966) Studies in the history and archaeology of Jordan, *ADAJ 11*: 6587.

Nambi K.S.V. and Aitken M.J. (1986) Annual dose conversion factors for TL and ESR dating. *Archaeometry* 28: 202-205.

Ogoh K., Ikeda S. and Ikeya M. (1993) Advances in ESR applications 9: 22-29.

Prescott, J.R., Hutton, J.T. (1994). Cosmic ray contributions to dose rates for luminescence and ESR dating: large depths and long-term variations. *Radiation Measurements* 23:497–500.

Zimmerman, D. W. (1978) Introduction to basic procedure for sample preparation and TL measurement of ceramics, *PACT* 2: 1-6.