Ancient TL

www.ancienttl.org · ISSN: 2693-0935

Oppermann, F. and Tsukamoto, S., 2015. *A portable system of X-ray irradiation and heating for electron spin resonance (ESR) dating.* Ancient TL 33(1): 11-15.
https://doi.org/10.26034/la.atl.2015.485

Ancient TL

A portable system of X-ray irradiation and heating for electron spin resonance (ESR) dating

Frank Oppermann,[1*] and Sumiko Tsukamoto[1]

[1]Leibniz Institute for Applied Geophysics, Hannover 30655, Germany

[*]E-mail: Frank.Oppermann@liag-hannover.de and Sumiko.Tsukamoto@liag-hannover.de

Received: December 18, 2014; in final form: April 27, 2015

Abstract

In this study we present details of a portable system which contains an X-ray source and heater, designed to irradiate and heat samples within a quartz glass tube for electron spin resonance (ESR) dating. The spatial distribution of the X-ray beam was measured with a radiation sensitive film and the images were scanned and mapped using the Risoescan software. The reproducibility of irradiations was checked by multiple irradiations to alanine. The results confirmed that X-ray irradiations give reproducible doses to samples. The heating component can heat samples between 100°C and 600°C and can be used for preheating after irradiation and for resetting the natural signal for regeneration.

Keywords: ESR dating, X-ray irradiation, heater, spatial uniformity

1. Introduction

In electron spin resonance (ESR) dating the multiple aliquot additive dose (MAAD) method has been predominantly used for equivalent dose (D_e) estimation (see reviews of Grün, 1989; Rink, 1997; Schellmann et al., 2008). One of the problems of the multiple aliquots approach is scattering of signal intensity between different aliquots, which could lead to a large uncertainty in D_e. To avoid such problems single aliquot methods can be used, however, these methods are only practically possible if a radiation source is located close to an ESR spectrometer. In order to make a single aliquot procedure more feasible for ESR dating, we have developed a portable system which consists of a small X-ray tube (Varian VF-50J) with a maximum energy of 50 kV and a heater; the latter is both for preheating after X-ray irradiation and for signal resetting for regenerative dose estimations. The same type of X-ray tube has been used for automated luminescence readers (Hashimoto et al., 2002; Andersen et al., 2003). Here we report the design and performance of our mini X-ray/heating system. An X-ray irradiation system for ESR dating using a larger X-ray source with a maximum energy of 200 kV has been developed and reported by Grün (2012).

2. Apparatus and software

Fig. 1a shows a photograph and the schematic diagram of the system. The system is designed to irradiate and heat samples within Wilmad Suprasil quartz glass tubes with 3 mm outer diameter (2 mm inner diameter) and 159 mm long. The X-ray components are: a Varian VF-50J X-ray tube with tungsten target (4-50 kV high voltage, 5 mA emission current and 50 W maximum power) and a Spellman high voltage generator MNX50P50 which can produce high voltage (UHV) of 0-50 kV with an emission current (IE) up to 2 mA but limited to 50 W power (Fig. 1b). The power supply has a stability of 0.05% per 8 hours after 0.5 hour warm up with a temperature coefficient of 0.01%/°C (supplier information). To minimize any warm-up effects, the filament is always preheated at a current of 1.7 A. We also monitor the tube voltage, emission current, filament current and filament voltage.

The generated X-rays are emitted through a 76 μm beryllium end window of the tube. The X-ray components are located within a lead chamber (w x h x l = 80 x 120 x 40 mm) with 10 mm thick walls (Fig. 1b). The distance between the beryllium window and the sample tube is 30.5 mm. The X-ray beam has a diameter of 56 mm at the sample position. To reduce the effect of low energy components of X-rays a 200 μm aluminum filter has been inserted between the beryllium window and the sample position as suggested by

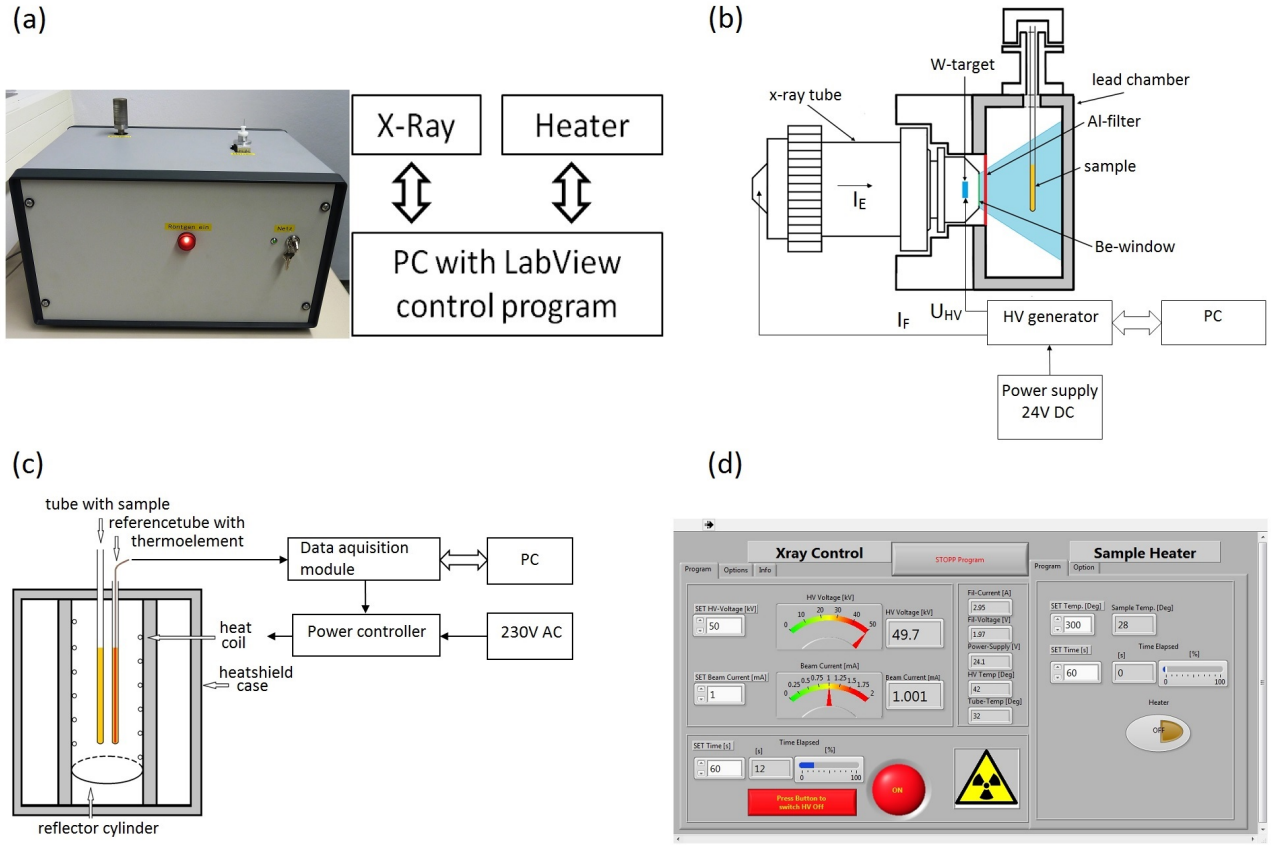

Figure 1. (a) Photograph and schematic diagram of the X-ray/heating system, and diagrams for (b) X-ray, (c) heater components and (d) software GUI.

Yawata & Hashimoto (2004) and Thomsen et al. (2006).

The heater can control the sample temperature within a tube between 100°C and 600°C. The sample tube is inserted beside a reference tube, which monitors the temperature within a steel reflector cylinder in a heat shield case (Fig. 1c). The heat energy is transmitted equally to the two tubes by a heat coil powered by a power controller, and a difference less than 1% between the two tubes was monitored. The reference temperature is measured with a data acquisition module using the thermoelement inside the reference tube.

The system is controlled by a Software-GUI written in LabView. Fig. 1d shows a screenshot of the program. In the X-ray control part (left) the high voltage [kV], the beam current [mA] and the exposure time [s] can be set by users. The actual high voltage, beam current, elapsed exposure time and other secondary parameters are monitored and shown on the screen. In the sample heater part (right) the sample temperature [deg] and the heating time [s] can be entered. The timer starts when the temperature reaches ± 5°C of set temperature. The reference temperature and elapsed heating time are monitored.

3. Spatial uniformity of the X-ray

The spatial uniformity of the X-ray beam was measured by taking images with a radiation sensitive film (Gafchromic HD-810) by irradiating for 3 different time intervals (50 kV, 1 mA for 200 s, 400 s and 600 s of irradiation) at the sample position without a sample tube and inside a quartz glass tube. These X-ray images were first scanned. The chromaticity of the images was calibrated into relative dose rate, and the distribution of the dose rate was mapped by the dosimetry software RisøScan (Helt-Hansen & Miller, 2004).

Fig. 2a shows the spatial dose distribution of the X-ray beam at the sample position for an irradiation time of 400 s. Fig. 2b shows the axial dose distribution inside the sample tube. The results indicate that the area of ∼ 1 cm diameter in the centre receives a relatively uniform irradiation of ± 1.6% precision. The position of the sample tube was chosen as to allow uniform irradiation of samples with a height of up to ∼ 1 cm (Fig. 2b). If ESR signals resulting from 1 cm sample height are too weak, it is possible to increase the height (and thus the amount of sample) up to 2 cm. The irradiation uniformity decreases to 3.1% for 1.5 cm sample height and 5.6% for 2 cm sample height, respectively. In order to avoid accumulation of non-uniformities by multiple irradiations of a single sample, and also to avoid signals originating

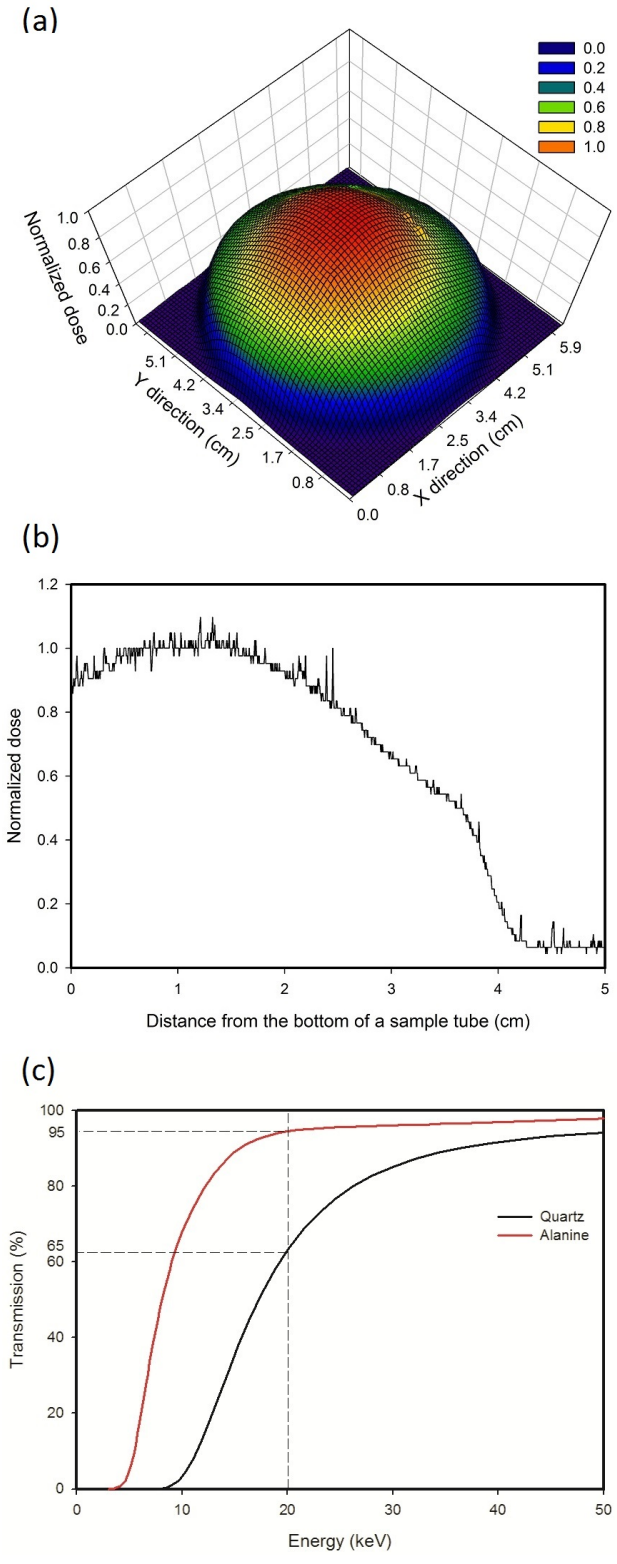

Figure 2. Spatial uniformity of the X-ray beam (a) horizontal cross-section at sample position (b) vertical cross-section thorugh a sample tube and (c) energy dependent transmission of X-ray intensity through 1 mm thick samples of alanine and quartz.

from the quartz glass tube itself, the tubes were sealed with parafilm and the tubes are turned upside down during each irradiation (Tsukamoto et al., 2015).

The comparison between the film images taken without a tube and inside the tube revealed that the wall of the quartz glass tube absorbed $\sim$ 60% X-ray dose. This was confirmed by a comparative irradiation experiment using 2 aliquots of 25 mg alanine; one was irradiated within a quartz glass tube the other was irradiated in a plastic straw with 3 mm diameter. The ESR signal intensity of the alanine irradiated inside the quartz glass tube was only 34% of the one irradiated in a plastic straw. Fig. 2c shows the energy dependent transmission of X-ray intensity through 1 mm thick samples (alanine and quartz) within a tube. The transmission was calculated assuming that a sample tube is filled with alanine or sand sized quartz with 30% porosity. At the peak energy of 20 kV the X-ray transmission (Andersen et al., 2003) between the inner surface and the centre of the tube is about 95% for alanine and about 65% for quartz. Therefore it would be better to rotate and/or vibrate tubes during irradiation in order to improve the uniformity of dose to sample grains and we plan to integrate such a mechanical function into the system in the near future.

4. Performance and calibration of the X-ray source

The dose response of X-ray irradiated alanine was compared with and without rotations in order to check the reproducibility of irradiations. One set of aliquots was irradiated with X-rays (50 kV, 1 mA) for 200 s without rotation. For the other set tubes were manually rotated 90° every 50 s during the 200 s irradiation. This process was repeated 4 times for each aliquot. Altogether 6 aliquots were measured (3 aliquots each with and without rotations). Each aliquot contained 50 mg of alanine. The 50 mg of alanine had a height of $\sim$ 2 cm in a quartz glass tube, which resulted in less homogeneous irradiations, however, signals for a smaller amount of material were too weak and irreproducible. ESR measurements were performed with a JEOL FA-100 with 2 mW microwave power, 0.3 mT modulation amplitude, and 20 mT sweep width. The scan time was 30 s and spectra were accumulated 3 times. The signal intensity around g = 2.003 was measured. Fig. 3a shows the increase of the alanine ESR signal intensity versus X-ray exposure time. Average intensities obtained from 3 aliquots (with and without rotations) and their 1-σ standard errors were plotted. Although a full penetration of X-rays throughout the thickness of samples was not expected (Fig. 2c), the intensities with and without rotations during irradiations are indistinguishable. We therefore conclude that rotations do not improve the homogeneity of our X-ray irradiations at least for alanine. The normalised increase of alanine ESR signals after each 200 s irradiation is shown in Fig. 3b as a histogram. The relative standard deviation of each measurement is 5.2%, including all other sources of measurement errors (e.g. adjusting heights of

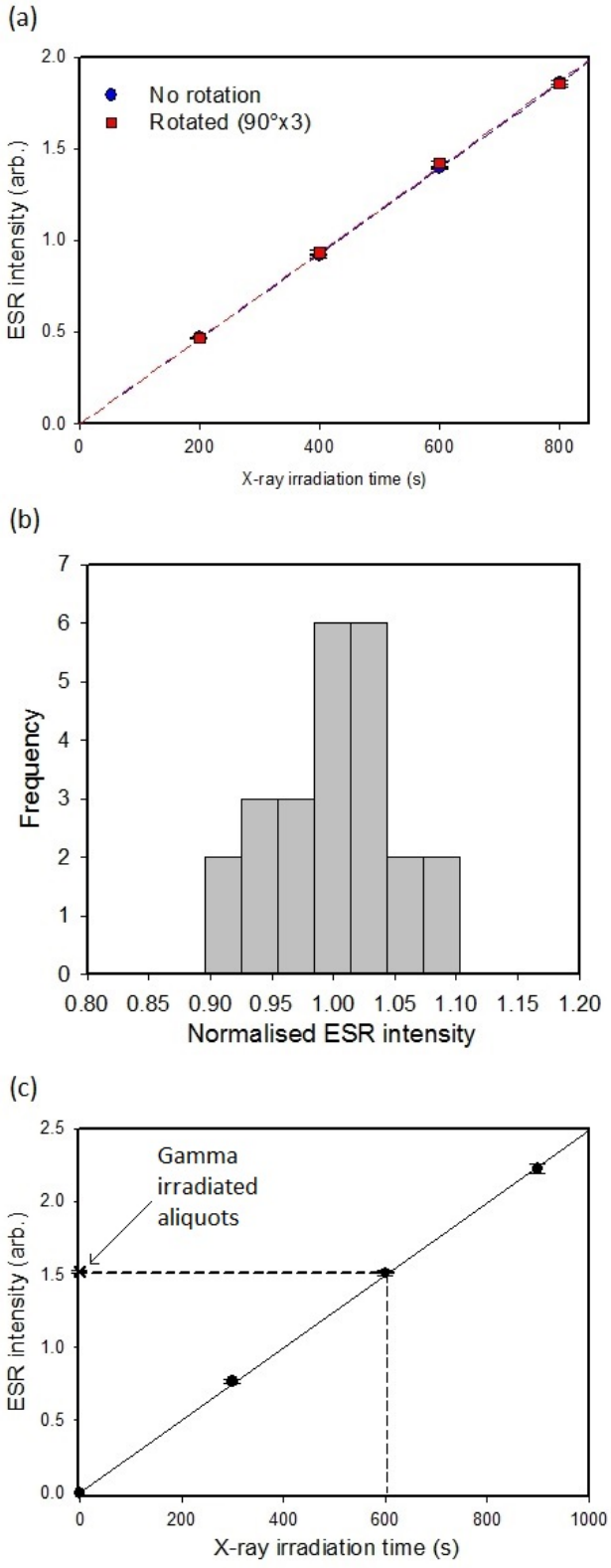

Figure 3. (a) Comparison of X-ray dose response curves with and without rotation during irradiations. (b) Histogram of relative alanine signal increase following 200 s irradiations. (c) Calibration of X-ray dose rate for alanine.

sample tubes at the irradiation and ESR measurement positions, and tuning of the microwave).

To calibrate the X-ray dose rate for alanine, 2 sets of aliquots were prepared. One set received a gamma dose in alanine of 75.2 Gy from a ^{60}Co source at the Technical University of Denmark. Another set of aliquots was irradiated with the X-ray and the intensities of the alanine radical for the gamma and X-ray irradiated aliquots were compared (Fig. 3c). Three aliquots each of gamma and X-ray irradiated aliquots were measured. The X-ray irradiation time which is equivalent to 75.2 Gy gamma dose was calculated as 603 $\pm$ 10 s. The resulting X-ray dose rate for alanine was thus 0.125 $\pm$ 0.002 Gy/s.

Our portable X-ray system provides the opportunities to conduct reproducible single aliquot ESR measurements, however, there is one disadvantage of using X-ray for artificial irradiation. Due to the low energy photons from the X-ray tube, the photoelectric effect dominates the energy transfer process, which depends on the atomic number of sample materials. Thus, calibration of the X-ray dose rate must be performed for each different material; the X-ray dose rate for quartz has been calibrated to 0.34 $\pm$ 0.01 Gy/s (Tsukamoto et al., 2015). Nevertheless this system provided us with the first opportunity to test the single aliquot regenerative dose method combined with preheat plateau tests using quartz ESR and therefore contributed to methodological developments in ESR dating (Tsukamoto et al., 2015).

5. Summary

This manuscript presents the details of our portable X-ray irradiation/heating system, which can be used to measure single aliquot equivalent doses in ESR dating. Although the experimental data with alanine showed that the reproducibility of the X-ray irradiation is not depending on the rotation of the sample tubes, for samples with higher density it might be advisable to rotate and/or vibrate the tubes. While using a higher energy X-ray tube is an alternative solution for the uniformity problem it would significantly increase cost and space requirements. The price for our system in total is approx.8000 Euro and it can easily fit on a desk. A higher energy system is about 20-30 times more expensive than the mini X-ray and needs much more space.

Acknowledgments

We thank Jakob Helt-Hansen, Torben Lapp, Jim Thorslund Andersen and Kristina Thomsen (Technical University of Denmark) for mapping the uniformity of X-ray irradiations and for gamma irradiations. Karsten Vollmer is thanked for his work on the lead chamber. Gudrun Drewes and Sonja Riemenschneider are thanked for their technical help. The constructive comments from David Sanderson and Regina DeWitt have greatly helped improve the paper.

References

Andersen, K., Bøtter-Jensen, L., and Murray, A.S. *A mini X-ray generator as an alternative to a $^{90}Sr/^{90}Y$ beta source in luminescence dating*. Radiation Measurements, 37: 557–561, 2003.

Grün, R. *Electron spin resonance (ESR) dating*. Quaternary International, 1: 65–109, 1989.

Grün, R. *Ionization efficiencies of alanine dosimeters and tooth enamel irradiated by gamma and X-ray sources*. Radiation Measurements, 47: 665–668, 2012.

Hashimoto, T., Nakagawa, T., Hong, D.G., and Takano, M. *An automated system for both red/blue thermoluminescence (TL) and optically stimulated luminescence (OSL) measurement*. Journal of Nuclear Science and Technology, 39: 108–109, 2002.

Helt-Hansen, J. and Miller, A. *RisøScan a new dosimetry software*. Radiation Physics and Chemistry, 71: 359–362, 2004.

Rink, W.J. *Electron spin resonance (ESR) dating and ESR applications in Quaternary science and archaeometry*. Radiation Measurements, 27: 975–1025, 1997.

Schellmann, G., Beerten, K., and Radtke, U. *Electron spin resonance (ESR) dating of Quaternary materials*. Quaternary Science Journal, 57: 150–178, 2008.

Thomsen, K.J., Bøtter-Jensen, L., Denby, L., and Murray, A.S. *Luminescence response to irradiation using mini X-ray generators*. Nuclear Instrumentas and Methods B, 252: 267–275, 2006.

Tsukamoto, S., Toyoda, S., Tani, A., and Oppermann, F. *Single aliquot regenerative dose method for ESR dating using X-ray irradiation and preheat*. Radiation Measurements, in press, doi:10.1016/j.radmeas.2015.01.018, 2015.

Yawata, T. and Hashimoto, T. *Availability of a small X-ray tube combined with Al-absorber as a new irradiator for luminescence researches*. Radioisotopes, 53: 207–212, 2004. (in Japanese with English abstract).

Reviewer

David Sanderson