Applied Radiation and Isotopes 101 (2015) 65–74

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

An investigation of the dosimetric and kinetic properties of sand using ESR and TL techniques

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HIGHLIGHTS

- Reporting of new additional results about the use of ESR and TL spectroscopy on irradiated sand sample.
- Detailed kinetic and dosimetric study on irradiated sand sample by using ESR and TL techniques.
- Calculation of sand sample kinetic parameters using various heating rates and deconvolution.
- Estimation of sand sample ESR activation energy using isothermal decay measurements.

ARTICLE INFO

Article history:
Received 15 December 2014
Received in revised form
19 March 2015
Accepted 25 March 2015
Available online 28 March 2015

Keywords:
Electron spin resonance (ESR)
Thermoluminescence (TL)
Sand
Kinetic
Radiation dosimetry

ABSTRACT

In this research, the general dosimetric and kinetic properties of sand from a beach in southern Turkey were investigated using electron spin resonance (ESR) and thermoluminescence (TL) techniques. The ESR dose response curve presents linear behaviour in the dose range of 250–1000 Gy followed by sublinear behaviour in the dose range of 2–8 kGy. Kinetic behaviors and activation energy of the free radical were also calculated using the data obtained from annealing studies performed at four different temperatures (220, 240, 260 and 280 °C). The activation energy value was calculated as 1.47 eV. The long-term fading of the ESR signal at room temperature turned out to be best described by a second-order kinetic decay function. The presence of measurable ESR signal intensity even after a storage period of 90 days was considered as providing an opportunity in the dose estimation of irradiated sand sample. Although the TL glow curve of the natural (unirradiated) sand sample only has a single broad peak at 317 °C, the glow curve of the irradiated sample has four glow peaks located at ~115 °C, ~156 °C, ~231 °C and ~308 °C and their intensity tends to be increased with absorbed dose. $T_{max} - T_{stop}$ and glow curve fitting results showed that presence of at least five peaks located at ~116 °C, 149 °C, 228 °C, 306 °C and 360 °C. This result suggests that the apparently single glow peak D may consist of two or more overlapping glow peaks. According to the thermal fading of the sand sample at room temperature, the TL signal intensities (23 °C and 308 °C) were found to be quite large after 30 days of storage this allows a more accurate measurement of the glow peak intensity. The additive dose method, variable heating rate method (VHRM), $T_{max} - T_{stop}$ and glow curve fitting method were used to number of peaks, dosimetric properties and kinetic parameters. This study shows that ESR and TL techniques could be successfully used to investigate the kinetics and dosimetric properties of sand sample. Furthermore, the results in this study plus the previous work done by the authors suggest that sand could, by using the ESR and TL techniques, be a suitable material for alternative dose measurement.

1. Introduction

Accidental radiation dose assessments using materials sensitive to radiation is still a very active research topic. These dosimetric materials should be easily found, are available in large amounts and are inexpensive. Sand is a natural material that satisfies these conditions. Although the composition of sand varies, its main component is quartz (SiO_2) which has certain thermoluminescence (TL) and electron spin resonance (ESR) properties that make sand suitable for general dosimetric purposes such as environmental dosimetry, retrospective dosimetry and, dating. The accuracy of dosimetry and dating information obtainable from the ESR and TL of investigated sand sample can be improved by understanding the main characteristics of ESR and TL glow peak responsible from radical and luminescence centers, respectively.

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http://dx.doi.org/10.1016/j.apradiso.2015.03.019
0969-8043/

Investigation of kinetic and dosimetric properties of sand sample is one way for characterizing of ESR and TL glow peaks.

Electron spin resonance (ESR) analysis is an effective technique to characterize the dosimetric and kinetic properties of dosimetric material on the basis of radiation-induced radicals. ESR dosimetry also determines the dose by measuring the relative concentrations of radiation-induced radicals. If the ESR and TL intensities corresponding to the number of paramagnetic centers are measured depending on the absorbed dose, the dose can be estimated. In the literature, there are many studies on the use of ESR to investigate the unpaired electron created by irradiation in quartz (Ikeya, 1993; Lee and Yang, 2007; Teixeira et al., 2005). However, most of these studies were undertaken for dating purposes only and the number of dosimetric and kinetic studies on sand samples using ESR is limited. One of these studies investigated the usability of sand samples from different Brazilian beaches as a high dose dosimeter using ESR (Teixeira et al., 2005, 2008; Caldas et al., 2006).

The luminescence of various minerals especially quartz and feldspar is very often used for dosimetric purposes. The luminescence signal is released when the grains are exposed to stimulation energy in the form of light or heat. The Signal intensity is proportional to the radiation dose that the material has received. This relationship allows several common minerals such as quartz, feldspar, zircon, and calcite to behave as natural radiation dosimeters, in other words, recording the amount of ionizing radiation to which they were exposed. On this basis, the TL technique is used for dose estimation of minerals. Quartz is abundant in nature and natural quartz exhibits TL properties, therefore it has been used widely for TL dating in literature (Falguères et al., 1994; Tatumi et al., 2006; Afouxenidis et al., 2007). Quartz has a complex TL glow curve structure and wide variety chemical form. This complexity is explained by the variety of defects in quartz that are either intrinsic (e.g., Si and O vacancies) or related to impurity atoms (e.g., Al or Ti). The concentration of impurity-related defects is dependent on the conditions of mineral formation or subsequent alteration (Preusser et al., 2009). In spite of complex TL glow curve; there have been more TL studies on this subject than those on ESR in the literature (Benny et al., 1997, 2000; Vaijapurkar et al., 1998; Soliman and Salama, 2009; Pitalua et al., 2013; Pandya et al., 2013).

There are some applications of sand as a dosimeter presented in the literature. Benny and Bhatt reported the application of sand as an *in-situ* dosimeter for estimating gamma dose absorbed by sludge during its sterilization (Benny and Bhatt, 1996; Benny et al., 1997). Sand samples have also been used for dating (Ikeya, 1993; Tatumi et al., 2006; Murray et al., 2015; Voinchet et al., 2015). Investigation of sand as a TL dosimeter for radiotherapy was undertaken by Pitalua et al. (2013). Coastal areas are preferred for establishment of nuclear power plants due to the massive water consumption. Sand near a nuclear power plant can be used for dose estimation in the case of nuclear emergencies. Sand samples can also be used as ESR dosimeters for different applications in medical, agricultural and industrial areas (Teixeira et al., 2005). As stated above, although there have been a lot of work related to application areas of sand samples in the literature, there are a limited number of studies on detailed kinetic and dosimetric properties of sand samples using different techniques.

The aim of the present work is to carry out a comprehensive kinetic and dosimetric analysis on the sand sample using ESR and TL techniques and to determine the kinetic parameters, dose response and stability of ESR and TL peaks. Different methods have been used to obtain kinetic parameters and dosimetric properties of the ESR and TL peaks for this sand sample. These are additive dose (AD), isothermal decay, $T_{max} - T_{stop}$, initial rise (IR), variable heating rate (VHR) and glow curve fitting methods.

2. Materials and methods

The sand sample used in our study was taken from a beach on the Mediterranean coast of southern Turkey. The samples were sieved and fractions of 125–180 μm of sand samples were used for ESR and TL measurements. The sand samples were washed with 1 N hydrochloric acid; after that, distilled water was used to remove the HCl and inorganic impurities. The samples were then allowed to dry at 100 °C. Any magnetic particles were removed with magnetic separation. After washing, drying and removal of magnetic particles, the sand samples were encapsulated in plastic pouches.

Irradiations were performed at room temperature in air using a ^{137}Cs gamma ray source (dose rate 500 Gy/h) and ^{90}Sr/^{90}Y beta ray source (dose rate 468 Gy/h) at the Sarayköy Nuclear Research and Training Centre of Turkish Atomic Energy Authority in Ankara. The dose rates of the gamma sources were measured by a Fricke dosimeter (ferrous sulfate solution). In all the irradiations in this work, the conditions of charged particle equilibrium were maintained. An estimated accuracy of $\pm 4\%$ was achieved in the determination of the dose delivered to the sample.

ESR measurements were carried out using a Bruker e-scan X-band ESR spectrometer interfaced with a computer data acquisition and analysis system with the following settings: modulation amplitude 0.1 mT, microwave frequency 9.8 GHz, microwave power 0.1 mW. Ten scans were accumulated. The receiver gain was adjusted as necessary to obtain the clearest spectra. All ESR measurements were obtained at room temperature and in air. Samples of 200 mg weight were inserted in a 3 mm internal diameter quartz ESR tube and centered in the cavity. Each sample was measured three times and an average of three such measurements was used for each data point. Between measurements, the sample tube was removed from the cavity, shaken and repositioned back into the cavity. Thus, experimental errors were estimated to be $\pm$ 2–4%. The *g*-values of the signals were determined using a MgO: Mn marker.

Isochronal annealing was performed in the temperature range of 100–500 °C by heating 3 kGy irradiated sand samples in a furnace at different temperatures for 15 min.

The kinetic feature of the radiation-induced resonance signal at high temperatures (220, 240, 260 and 280 °C) was also investigated by using the samples irradiated at a dose of 3 kGy. After the irradiation process, the sand samples were transferred to the furnace at temperatures mentioned above, then their ESR spectra were recorded regularly over a time interval of 0–270 min. after cooling them to room temperature following predetermined heating times.

All thermoluminescence measurements were carried out using an automated Risø TL/OSL reader (TL/OSL-DA-20) equipped with a ^{90}Sr/^{90}Y beta source, dose rate 468 Gy/h. Light detection was carried out with a photomultiplier tube bialkali EMI 9235 QA that has an extended UV response with maximum detection efficiency between 300 and 400 nm. The TL measurements were performed using a linear heating rate of 5 °C/s from room temperature up to 500 °C in a nitrogen (N_2) atmosphere. The radiation source was calibrated using quartz, which was given a well-known gamma dose using a ^{137}Cs secondary standard point source. Four aliquots of 5 mg each of the samples were used for each measurement. TL data points are the average of four different aliquots of the samples. In order to good thermal contact that ensures uniform TL signal from sand during the exposure to heat, a thin and uniform layer of sand grains was laid on the planchet surface. For TL measurement, it is possible to measure of the natural signal, thus ejecting all electrons from the traps. This then permits the repopulation of the electron traps by subsequent irradiation and can be repeated many times in order to construct a dose response