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THERMOLUMINESCENCE PROPERTIES OF IRRADIATED QUARTZ AND FELDSPAR AT DIFFERENT DOSE RATES

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Abstract: Thermoluminescent (TL) properties of irradiated quartz and feldspar have been investigated in this work. Samples were irradiated at two different dose rates of ^{60}Co source. TL intensity of both quartz and feldspar did not show dependence from the dose rate. Hence this property may be used for the dating of archaeological artifacts.

Keywords: Quartz, feldspar, thermolumiscence, dating

1. Introduction

The thermoluminescent (TL) properties of quartz and feldspar are widely used for retrospective dosimetry, environmental dosimetry, accidental dose assessment, geochronometry, and dating [1], [2], [3]. In these applications, it is assumed that the growth of TL under laboratory irradiation is exactly the same as in nature, despite the fact that the natural dose rate and laboratory dose rate differ by 10^8 times. However, the dependence of the TL intensity on the dose, as well as on the dose rate for these minerals, is quite complex and there are many conflicting data and models explaining such an important problem. Radiation sources that have different rates of energy transfer to the lattice can also lead to a change in the TL luminescence curves, even if the total dose remains unchanged.

Baijapurkar et al. [4] observed that the TL of the Rajasthan sand has a linear dose dependence from several cGy up to 20 Gy with a minimum detectable dose of 3 cGy, insignificant attenuation and independence from the dose rate. The thermally treated samples were irradiated with ^{60}Co gamma radiation at various dose rates, namely 2.05 mGy h-1, 0.2 Gy h-1, 21 Gy h-1, and 1.36 kGy h-1.

The dependence of the TL intensity on the dose rate of ^{60}Co gamma radiation was studied for Brazilian crystalline quartz and milky quartz at two dose rates ($2x10^{-2}$ Gy s^{-1} and $2x10^{-5}$ Gy s^{-1}). The total dose was 2.5 Gy and the TL intensity of the Brazilian crystals was higher at higher dose rates than at lower doses. Milky quartz had no dose rate dependence, and the TL response was within the experimental error [5]. On the basis of the experimental results, it was suggested that irradiation of quartz samples in the laboratory for TL dating with medium and high doses could lead to underestimation or overestimation of the age of the measured samples.

Directly opposite results have been obtained in other studies. Khazal studied the effect of the dose rate of ^{60}Co gamma radiation on the TL intensity of quartz rock crystals at dose rates ranging from 0.14 Gy s^{-1} to 2.2 Gy s^{-1}. A significant decrease in TL intensity was observed at higher dose rates. Groom et al. [6] measured a similar decrease in TL intensity to five orders of magnitude on the same quartz sample as Khazal, with an increase in the dose rate from $1.4x10^{-3}$ to 3.3 Gy s^{-1}. Changes in the thermoluminescence sensitivity under laboratory irradiation were also observed in natural quartz at doses exceeding 250 Gy.

Valadas and Fereira [7]studied the effect of the dose rate of ^{60}Co gamma radiation on TL intensity of rock crystals and milky quartz at dose rates of 0.15 Gy s^{-1} and $1.3x10^{-4}$ Gy s^{-1}. The total radiation dose was 32 Gy. Rock quartz crystals showed high TL emission at 300 °C at high dose rates. The behavior of the three main TL peaks at about 320, 360, and 370 °C (heating rate 5 °/sec) was studied using blue, ultraviolet, and green filters. This effect is more pronounced in rock quartz and tends to disappear as the sample temperature rises upon irradiation. The peak around 370 °C shows only slight dose rate dependence and therefore appears to be more suitable for dating.

Fujita and Hashimoto [8] investigated the dependence of the intensity of violet radio fluorescence (~ 400 nm) of quartz on the X-ray dose. They irradiated quartz with a fixed dose, varying the dose rate and hence the irradiation time for different aliquots. The integrated intensity of radio fluorescence increased nonlinearly for a dose rate of 0.1–8.0 Gy/min, but then sharply decreased at 10 Gy/min. After irradiating the quartz in this way, the authors measured the resulting blue OSL and found a negative relationship between the OSL intensity and the integrated RF. On this basis, they conclude that the radioluminescence emitted during irradiation causes a "self-discoloration" of the sample and therefore a decrease in induced OSL/TL. The insertion of quartz pieces of various thicknesses, absorbing wavelengths shorter than 420 nm on a filter between the X-ray source and the sample, confirmed the effect of self-decolorization of radioluminescence during their experimental setup. In terms of dosimetry and OSL dating, the authors, therefore, propose to pay attention to self-discoloration in order to avoid underestimating dose or age.

Friedrich et al. [3] analyzed the main signal of UV radiation from 3.37 - 3.47 eV (band C) during radiofluorescence in natural quartz with a dose rate in the range from 10 to 500 mGy/s. Their results confirmed the theoretical results, the direct proportionality of the initial C-band radiofluorescent signal with the dose rate and the direct proportionality of the initial slope with the square of the dose rate.

The effect of the dose rate on the TL spectrum has been studied not only for quartz or feldspar. A recent study was carried out for five different TL phosphors [9]. It was shown that the characteristics of the TL and the kinetic parameters of the TL do not depend on the dose rate for these materials. The study of thermoluminescence samples depending on gamma and beta irradiation with different dose rates was carried out for calcite samples. The samples were irradiated at room temperature with either a calibrated $^{90}Sr/^{90}Y$ beta source with a dose rate of 0.1470 Gy/s or exposure to a ^{60}Co gamma radiation source with a dose rate of 0.308 Gy/s. The difference in intensity was significant in both cases and higher for gamma radiation than for beta radiation. Different dose rates achieved with alternating gamma or beta rays result in different light emission efficiencies during TL. After examining the TL response from the Harshaw TLD 100, reported that in situations with more than one dose rate, the value of the final intensity is neither the sum calculated for individual exposures (even taking into account their respective TL efficiency) nor the value predicted from the total dose delivered at a single dose rate. The actual signal may even exceed that for dose prediction based on the TL efficiency of any of the mixed exposure components. In other words, the total TL effect was not a linear combination when irradiated using certain types of irradiation.

All these data cast doubt on the consistency of the TL method in determining the absorbed dose during irradiation. Aitken summarized the state of the art: "It is clear that it (referring to the dose rate) cannot be very pronounced since otherwise, it would not be possible to obtain good results when dating samples with a precisely known age" [10], p. 141. This is a very strong argument, but, unfortunately, it cannot explain the numerous experimental data.

2. Materials and methods

The feldspar samples were presented by the Institute of Inorganic Chemistry, and Catalysis of the Azerbaijan National Academy of Sciences. Large feldspar samples were crushed, treated with HCl and washed with deionized water, and then sieved fractions from 100 to 160 μm were used. In the context of this article, we have designated it as a “natural” feldspar specimen to distinguish it from thermally annealed samples. Then these samples were irradiated with different dose rates, but with the same total dose.

Sand samples taken from a beach on the Caspian Sea coast in the eastern part of Baku were sieved and a sand fraction of 100-160 μm was used to measure TL. They have been treated with H_2O_2 to remove organic matter. Hydrochloric acid (1N) was used to remove carbonates and then washed with distilled water. Magnetic separation was applied to remove any magnetic inclusions. The grains were etched with 40% HF for 40 min at 20 °C and treated with HCl again to dissolve any remaining soluble fluorides. The material was thoroughly washed with distilled water and dried overnight in an oven at 50°C. In the context of this article, we have designated it as "natural quartz" to distinguish it from thermally annealed samples.

Another set of experiments was also carried out using thermally annealed samples at equal doses. Samples of both quartz and feldspar were annealed at 600 °C in a muffle furnace for one hour. For each measurement, a new portion was used with the same number of samples, that is, five milligrams, which allowed a quantitative comparison of the results.

The samples were irradiated using a ^{60}Co source using sample containers. The dose rate of the ^{60}Co source was determined with a Magnettech Miniscope MS400 EPR spectrometer using BioMax alanine dosimetry films (developed by Eastman Kodak Company). To determine the effect of dose rate, we measured dose rates at several locations around the ^{60}Co source. Irradiation was performed at ambient temperature.

A Harshaw TLD 3500 Manual Reader was used to measure the characteristics of the TL samples. TL measurements were carried out in the temperature range from 50 ºC to 400 ºC at a linear heating rate of 20 ºC/sec. Three aliquots of 5 mg each of the samples were used for each measurement. So the TL data is the average of three different portions of the sample. A thin and uniform layer of quartz /feldspar grains was applied to the plate surface in order to obtain full contact, which ensures a uniform TL signal from the samples.

3. Results and discussion

The thermoluminescence process consists of the thermal release of electrons from traps and the capture of some of them in luminescent centers. Such capture occurs only for centers that are charged with a hole, and therefore the thermoluminescence sensitivity is proportional to the number of centers that are so charged. The TL sensitivity of samples depends on many factors, such as preliminary heat treatment and irradiation, irradiation temperature, heating rate during TL measurement, and many others. In order to reveal the effects of only the dose rate, the experiments were planned as follows: the samples of natural quartz and feldspar were not exposed to heat and preliminary laboratory irradiation.

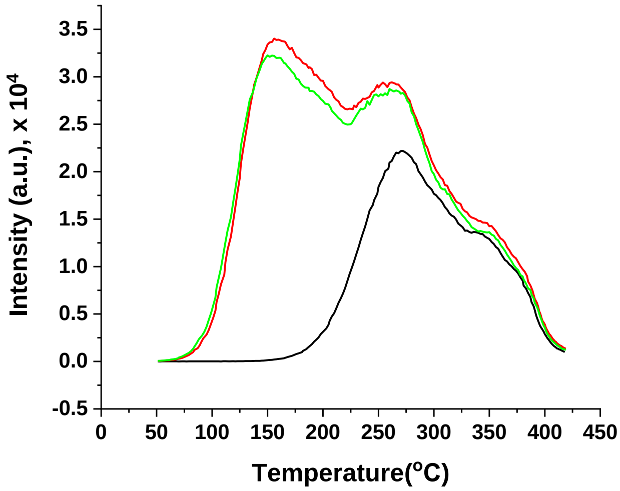

Fig. 1 TL intensity of irradiated natural feldspar with a dose rate of (2) 3.45 mGy/s; and (3) 92.07 mGy/s. The total dose for both samples was 33 Gy. Curve (1) represents the TL spectrum of natural feldspar not irradiated under laboratory conditions. The heating rate of TL measurements is 20°C/sec.

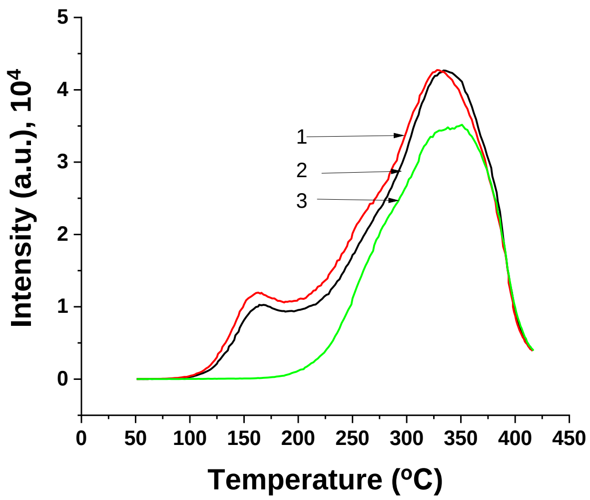

Fig. 2 TL intensity of irradiated natural quartz with a dose rate of (2) 3.45 mGy/s; and (3) 92.07 mGy/s. The total dose for both samples was 33 Gy. Curve (1) represents the TL spectrum of natural quartz unirradiated under laboratory conditions. The heating rate of TL measurements 20 °C /sec

After cleaning from impurities, these samples were irradiated as in natural conditions i.e. without any thermal treatment, but at different dose rates. The samples received the same dose, i.e. 33 Gr. The TL curves of the luminescence of the samples irradiated under laboratory conditions are shown in Figures 1 and 2. Each of these curves is the average of five or more measurements.

It can be seen from these figures that the TL intensity for both crystals increases with irradiation at different dose rates in comparison with the samples not irradiated under laboratory conditions and is the same within the experimental error.

Approximately the same behavior we have observed for the thermally annealed samples. Irradiation at different dose rates did not show any significant changes at the TL intensity of quartz and feldspar. For the purpose of simplicity, those figures are not referred in this paper. As was expected TL sensitivity was much higher for the thermally treated samples to compare with the natural samples. Such a picture can be explained by a very simple model [11], in which, upon irradiation, electrons rise from the valence band to the conduction band and then accumulate in traps, while the remaining holes in the valence band accumulate at the recombination center.

The authors of [11] showed that if we add to this model the stage of recombination of trapped electrons and holes during irradiation, this can lead to a noticeable effect on the dose rate. Then, the degree of the effect will depend on the relative values of the capture parameters, such as the initial and current concentrations of holes and trapped electrons, as well as on the total absorbed dose. For a given total dose, the effect would be strong in the high dose rate range and relatively independent of the dose rate at low dose rate levels. The calculation results show that the curves of the dependence of the TL intensity on the dose rate will flatten with a further increase in the dose rate[11]. In other words, a very flexible model has been proposed that can explain various experimental data, sometimes diametrically contradicting each other.

4. Conclusions

Thermoluminescence properties of irradiated quartz and feldspar have been studied at different dose rates. It has been shown that the thermoluminescent properties of these crystals are independent of the dose rate within the applied total dose range. This property suggests that these crystals may well serve the purpose of dating archaeological artifacts.

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ТЕРМОЛЮМИНЕСЦЕНТНЫЕ СВОЙСТВА ОБЛУЧЕННОГО КВАРЦА И ПОЛЕВОГО ШПАТА ПРИ РАЗЛИЧНЫХ ДОЗАХ

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Резюме: В данной работе исследованы термолюминесцентные (ТЛ) свойства облученных кварца и полевого шпата. Образцы облучали двумя разными мощностями дозы источника ^{60}Co. Интенсивность ТЛ как кварца, так и полевого шпата не зависела от мощности дозы. Следовательно, это свойство может быть использовано для датировки археологических артефактов.

Ключевые слова: Кварц, полевой шпат, термолюмисценция, датирование

MÜXTƏLİF DOZA GÜCÜNDƏ ŞÜALANMIŞ KVARS VƏ ÇÖL ŞPATININ TERMOLUMİNESSENT XASSƏLƏRİ

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Açar sözlər: Kvars, çöl şpatı, termoluminessensiya, yaş təyini