Estimation of ‘dose-depth’ profile in the surface layers of a quartz-containing tile from the former Hiroshima University building indicates the possible presence of beta-irradiation from residual radioactivity after A-bombing

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ABSTRACT

The problem of differentiating between primary irradiation and exposure due to residual radioactivity following A-bombing (including beta-exposure), is the subject of special attention and discussions in order to understand the health effects following the Hiroshima and Nagasaki A-bombings, especially among newcomers to cities soon after the detonations. In this work, the method of single quartz grain luminescence retrospective dosimetry was applied for a retrospective estimation of the ‘dose-depth’ profile in a quartz-containing tile extracted from the building of former Hiroshima University (HU), which was a ‘witness’ of the Hiroshima atomic bombing on the 6 August 1945. It has been shown that results of retrospective estimates of the ‘dose-depth’ profile using the method of optically stimulated luminescence (OSL) from inclusions of quartz grains in very thin layers of the sample, in combination with the calculations of the ‘dose-depth’ profile using the Monte Carlo method, indicates the possible presence of beta irradiation of thin layers of the sample located near the surface of the tile facing the air, where there is no electronic equilibrium from gamma radiation.

Keywords: Hiroshima; A-bombing; residual radioactivity; beta-irradiation; optically-stimulated retrospective luminescence dosimetry; depth-dose profile in quartz-containing samples
INTRODUCTION

Initial gamma- and neutron radiation exposure of the population as a result of A-bombing in Hiroshima was reevaluated in the Dosimetry System 2002 (DS02) [1] in comparison with the Dosimetry System 1986 (DS86) [2, 3]. In conventional dosimetry systems DS86 and DS02 only brief considerations were related to the residual radiation exposure—as a small contribution to the total whole body doses. The problem of differentiating between primary irradiation and exposure due to residual radioactivity following an A-bombing, is still the subject of special interest and discussion in order to understand the health effects following the Hiroshima and Nagasaki A-bombings [4, 5], especially among newcomers to cities soon after the detonations [6]. The estimation of the beta-exposure is the subject of special attention [7]. It should be specially noted that only gamma-rays have been considered in the conventional dosimetry systems DS86 and DS02. But on the other hand all residual radionuclides have the beta-radiation emission, which should be accounted for dosimetric estimations.

The presence of the exposure to beta-irradiation deriving from residual radioactivity can be indicated by 'depth-dose' profile measurements using the method of optically-stimulated luminescence (OSL) from single quartz grain inclusions in very thin layers of an exposed ceramic sample ('witness' of A-bombing)—in combination with Monte Carlo calculations of the dose profile from gamma-irradiation. One of the important advantages of the retrospective OSL single grain dosimetry method is the possibility to separate single quartz grains for OSL measurements and for the following analysis of the results of the measurements. As a result it was possible to exclude the luminescence signal from other materials than quartz.

The following steps of our study were performed:

- Measurements of the 'dose-depth' profile in the thin layers of the tile from the Hiroshima University (HU) building by OSL single grain (quartz) retrospective dosimetry technique;
- Calculations by Monte Carlo method of the 'dose-depth' profile in the layers of HU tile using gamma-spectra at the moment of A-bombing;
- Estimation of 'dose-depth' profile near the sample surface as a result of irradiation by beta-particles only: measurements by stimulated luminescence technique of the dose profile in the 'quartz equivalent' Al2O3::C crystals (irradiation by 90Sr/90Y source);
- Comparison of 'dose-depth' profiles from beta and gamma irradiations with measured 'dose-depth' profile in HU tile sample.

MATERIALS AND METHODS

Hiroshima quartz-containing sample

Location of tile sample collected from the roof of the former HU building (Hiroshima city, Japan) is the following: building—'A'; code of the sample from HU—'H4'; location of tile: eaves; height from the roof: 3.3 m; height from the ground surface: 15.82 m; ground distance from the hypocenter: 1.314 m; angle between perpendicular to the sample’s surface and direction to the hypocenter (degrees): 26.7 (according to [8]). GPS coordinates of former HU building: N 34°22’59.2” E 132°27’29.8”. Description of sample: ‘age’ of the sample (till 2014 y): 73 ± 1 years; surface size: 36 cm × 18 cm; thickness: 26 mm; texture of the surface: smooth (covered by 0.1-mm thick black glazed layer); the density of glaze is equal to 2.2 g/cm³; the density of tile is equal to 2.25 g/cm³.

OSL measurements

The OSL technique of retrospective dosimetry using single microcrystals of quartz as well as the method of samples’ preparation for the undertaking of the OSL measurements are described in detail in our previous publication [9]. Shortly the equipment and technique used are as follows.

The TL/OSL-DA-15 reader with special attachment for measurements of OSL from single quartz grains (DTU Nutech, RISO Nat Lab, Denmark) was used for OSL measurements. The reader equipped by 10 mW diode pumped solid-state Laser ND: YVO4 (532 nm) with high accuracy positioning and focused to a spot < 20 µm. Single quartz grains are loaded into the separate 0.3 mm holes (100 holes per disc, 48 discs). Registration of stimulated luminescence was performed by built in bialkali photomultiplier EMI 9235QA with ‘Hoya U-340’ 280–370 nm (340 nm max) filter. The built in 90Sr/90Y calibrated source (6.71 mGy/s) was used for obtaining dose calibration curve of each analyzed quartz grain. Analysis of calibration curves and dose estimation for each quartz grain was performed using standard ‘Analyst’ software of TL/OSL-DA-15 reader.

The sample preparation procedures consist of the following steps:

- Lateral side of low speed reciprocating saw with diamond-coated blade was used for removal and crushing of thin layers of tile;
- The outer 0.1 mm layer of the sample was removed just before measurements. This outer 0.1 mm layer consists of black glaze with 2.2 g/cm³ density;
- The subsequent layers of the sample with a thickness of 0.3 mm were removed and crushed (the micrometric control of the layers’ thicknesses was performed).

Aliquots of microparticles from each layer of the sample were subjected to the following procedures to separate fractions of quartz microcrystals

- Treatment by 7% HCl during 15 min at 50°C;
- Magnetic separation;
- Washing in distilled water, ethyl alcohol and acetone;
- Drying at 50°C, 24 hours;
- Etching in an ultrasonic bath by 40% HF, 15 min;
- Treatment in 25% AlCl3, 15 min, 50°C;
- Washing in distilled water, ethyl alcohol and acetone;
- Drying at 50°C, 24 hours;
- Separation of quartz grains by a set of sieves into different fractions by grain’s size;
- Fraction of quartz grains with 0.075–0.106 mm size was used for OSL measurements.

Sequence of irradiation and registration of OSL signal from each analyzed quartz grain was as follows

1) Measurements of OSL signal from natural (unknown) dose were performed with the following sequence: preheat, heat, registration, test dose irradiation, preheat, heat, registration [9];
2) Measurements of OSL signals after irradiation to known doses using built in 90Sr/90Y source (for construction of dose calibration curve)
were performed with the same sequence: preheat, heat, registration, test dose irradiation, and preheat, heat, registration;
3) Estimation of unknown dose for each quartz grain using calibration curve and measured values of OSL signal, estimation of uncertainty was performed using 'Analyst' software.

**Estimation of 'dose-depth' profile near the sample surface as a result of irradiation by beta-particles only**

In order to estimate the 'dose-depth' profile near the sample surface as a result of irradiation by beta-particles only, the measurements of the dose profile in assemblies of Al₂O₃:C crystals irradiated by ⁹⁰Sr/⁹⁰Y source were carried out. Al₂O₃:C crystals were considered as 'quartz-equivalent.' The irradiated assembly of Al₂O₃:C was a cylinder consisting of nine tightly connected crystals with a thickness of each crystal of 1 mm and a diameter of 5 mm. Irradiation was carried out with a hermitical standard ⁹⁰Sr/⁹⁰Ybeta-particles source located at the end of the Al₂O₃:C crystals assembly. Dose measurements by the stimulated luminescence technique were performed separately for each crystal.

**Calculations by the Monte-Carlo method of the 'dose-depth' profiles from gamma-irradiation in the layers of the quartz-containing sample**

MCNP-4C code [10] with MCPLIB02 library was used for calculations. The fluence and spectra of primary and secondary gamma-quanta corresponding to the location of the former building of the HU were taken from DS86 [2, 3] with accounting for the DS02 data [1]. The geometry of irradiation of the tile was adopted in a similar manner to that presented in the work [11]. Compositions of tile, concrete and air were taken from [11]. Calculation of the absorbed dose in the sample was performed from 0.1 mm to 1 mm depth with a 0.1 mm step, and from 1 mm to 25 mm depth with a 1 mm step. The absorbed doses were calculated in layers of the sample with a thickness of 0.1 mm.

**RESULTS**

Figure 1 shows the results of the undertaken OSL measurements of the profile of the accumulated absorbed dose depending on the depth in the analyzed sample. The background dose has been subtracted. The background dose was estimated in accordance with the procedure described in [12–14], using information about the known 'age' of the measured sample.

The average measured cumulative dose over the entire depth of the tile is 1060 mGy. This is quite close to the previously published average dose of gamma irradiation over its entire depth, equal to 1130 mGy for the same tile location [8]. It should be noted here that the authors of the cited work [8] did not analyze the distribution of the dose versus the depth in the sample, but gave only its average value over whole volume of the sample, associating this value with gamma irradiation. Meanwhile, near the surface of the sample, there are no conditions for electron equilibrium. Therefore, the dose from gamma radiation should be significantly lower in the sample layers close to the surface facing the air. It is noteworthy that according to the data of measurements presented in Fig. 1, there is no dose reduction near the sample surface facing the air. Perhaps that is suggests that in the vicinity of the sample surface there was an additional exposure from short-range beta radiation and, possibly, from low-energy quantum radiation. In order to check this assumption, we have calculated the distribution of the dose from gamma radiation versus the depth in the tile sample for the real spectrum of gamma rays at the location of the sample, at the time of the nuclear explosion in Hiroshima. The method of stochastic modeling of the interaction of ionizing radiation with matter (also known as the 'Monte-Carlo' method) was used for these calculations, with the calculation conditions given in the Materials and Methods section. Figure 2 shows the results of these calculations.

**Fig. 1. Results of OSL measurements of accumulated absorbed dose (D, mGy) of gamma-beta irradiation (vertical axis) vs the depth in the analyzed sample (mm, horizontal axis). The background dose has been subtracted. Sample: roof tile from the former building of HU. Point ‘0’ on the horizontal axis corresponds to the surface of the tile facing the air.**

**Fig. 2. Results of the Monte-Carlo calculation of the 'dose-depth' profile in the layers of the studied tile. D—absorbed dose, in mGy. Depth in the tile is indicated on horizontal axis (mm). Point ‘0’ on horizontal axis corresponds to the surface of the tile facing the air.**
Estimation of 'dose-depth' profile in the surface layers of a quartz-containing tile

Figure 2 shows that, starting from a depth in the tile of about 3 mm, the calculated dose from gamma irradiation does not significantly differ from the dose measured by the OSL method (within the 2SD uncertainty of measurements). On the other hand, based on the results of these calculations (Fig. 2), the absorbed dose of gamma-irradiation near the surface of the tile facing the air is significantly lower in comparison with the calculated dose at a depth more than 3 mm, and in comparison to the measured dose (see Fig. 3).

This is due to a lack of electron equilibrium near the surface at the energies of gamma radiation that took place at the time of the explosion. Meanwhile, the 'dose-depth' profile, as measured by the OSL method (Fig. 1), shows quite a uniform distribution of the dose along the depth 0.25 to 3 mm. This indicates that, possibly, the thin surface layers of the sample were subjected to additional short-range irradiation from residual radioactivity after the explosion (beta-particles and, presumably, some low-energy quantum radiation).

In order to estimate the possible contribution to dose near the sample surface as a result of irradiation by beta-particles only, we carried out measurements of the 'dose-depth' profile in assemblies of Al$_2$O$_3$:C crystals irradiated by a $^{90}$Sr/$^{90}$Y source (see section 'Materials and Methods' above). Al$_2$O$_3$:C crystals were considered as 'quartz-equivalent,' as far as the $Z_{eff}$ for Al$_2$O$_3$:C is 11.3, and for quartz (SiO$_2$) the $Z_{eff}$ is 11.8. Figure 4 summarizes the results of these measurements.

Figure 4 shows that the dose from beta irradiation is significantly decreasing along the depth of the 'quartz equivalent' Al$_2$O$_3$:C crystals when irradiated by $^{90}$Sr/$^{90}$Y beta-particles. Therefore, the results of the OSL measurements of the accumulated absorbed dose in the analyzed sample presented in Fig. 1 can be interpreted as a superposition of two types of irradiation: (i) beta-irradiation from residual radioactivity after nuclear explosion, that makes the main contribution to the irradiation of layers close to surface of the sample faced to the air, and (ii) penetrating gamma-irradiation at the moment of explosion (Fig. 2), that gives the main contribution to dose in the deeper layers of the sample. In this way, the measurements of 'dose-depth' using the OSL method from single quartz grains inclusions in thin layers of the sample, in combination with the Monte-Carlo calculations of dose profile from gamma-irradiation, indicates the possible presence of beta irradiation of thin layers of the sample located near the surface of the tile facing the air, where there is no electronic equilibrium from gamma radiation.

DISCUSSION

1. The findings presented in this article can be partially confirmed by available results of the TL/OSL measurements of quartz inclusions in the brick samples from the territories irradiated as a result of nuclear tests in Semipalatinsk Nuclear Test Site (SNTS) [14]. According to the cited publication [14], one of the quartz-containing samples (from the Kanonerk settlement, which is located near the trace of the nuclear test in SNTS) has provided a subset of data in 'dose-depth' profile related to the surface layers, which is not typical for gamma-irradiation. The subset of these results was obtained for depths less than 5 mm where significantly higher values of dose were indicated, and the slope was significantly steeper than that at greater depths. The concordance of results obtained within each subset using both TL and OSL procedures suggests that the experimental dose evaluation procedure is not the main cause of the differences. The steeper component of the profile has a ‘half depth’ of several mm. This could result from the presence of low penetration irradiation such as beta-particles, e.g. beta-emitting radionuclides, adhering to the front surface of the brick.

2. Another example of such kind of findings is related to the radioactive contamination following the accident at Fukushima-1 NPP: it was presented in publication [15] that was devoted to comparison of Monte-Carlo dose calculation with data obtained by single-grain retrospective luminescence dosimetry of quartz inclusions in a brick sample from the Fukushima Prefecture [9]. In fact, it was demonstrated that the dose increase measured at the brick surface was explained by the beta-ray contribution, and that the slight slope in the dose profile deeper in the brick was due to the gamma-ray contribution.

3. As for our study, the main objective was to show, that for the analyzed quartz-containing ceramic tile sample ('witness' of the atomic
bombarde ment of Hiroshima), the measured profile 'absorbed dose versus depth in the sample' can be interpreted as a superposition of short- range beta radiation and penetrating gamma radiation. The $^{90}Sr/^{90}Y$ source was used in this study not for experimental simulation of the beta-radiation spectrum at the moment of the atomic bombing, but because it is quite pure beta emitter. Using this source, it was possible to show that the instrumentally measured spatial gradient of the dose profile from the beta source near the surface of the analyzed ceramic sample differs from the absorbed dose profile from penetrating gamma radiation. This is due to the fact that, in the absence of electronic equilibrium, the gamma absorbed dose at the surface is much lower than in its depth, and the main dose in thin layers near the sample surface is caused by short-range beta irradiation. It should be emphasized once again that at this stage of research authors have not intention to simulate experimentally the real spectrum of beta-radiation from residual radioactivity after atomic bombing in Hiroshima. It is difficult and practically impossible to simulate this situation experimentally. This work is just the first step of the solving of the task. It is in our plans to use computational methods to consider the details of this task.

5. As it was noted in [16], retrospective dosimetry data are necessary for understanding and interpreting biological effects caused by uncontrolled radiation exposures, which occurred in the past.

The atomic bomb dropped on Hiroshima city on 6 August 1945 derived its explosive power from the nuclear fission of $^{235}U$. There were two main sources of residual radioactivity following the atomic bomb explosion in Hiroshima: fission products and neutron activated radionuclides [17].

According to [18, 19] the cumulative dose by the fallout of fission products has been estimated to be about 0.010 Gy in Hiroshima city. Based on these data, it can be concluded that the contribution of residual radioactivity from the fission products to the total radiation dose can be considered insignificant for the site of sampling of analyzed quartz containing ceramic tiles (the building of the old HU located in the Hiroshima city).

Another source of residual radioactivity following the detonation of the atomic bomb in Hiroshima is neutron-activated radionuclides. According to [17], the list of main radionuclides from neutron activated soil at Hiroshima, which were considered as a subjects of dosimetric interest, is the following: $^{28}Al$, $^{31}Si$, $^{44}Na$, $^{46}Sc$, $^{48}Ca$. In general this is consistent with the lists, which are presented in [7, 20]. However, the list presented in [20] is more complete. The summary of the radiation emission characteristics of these various radionuclides is provided in [5].

It should be specially noted that only gamma-rays have been considered in the conventional dosimetry systems [1–3]. But on the other hand all considered neutron activated radionuclides have the beta-radiation emission, which should be accounted for dosimetric estimations [5]. The major neutron-activated radionuclides in soil contributing to the external dose of the atomic bomb survivors are: $^{24}Na$ ($T_{1/2} = 15$ h), $^{26}Al$ ($T_{1/2} = 2.24$ m), $^{28}Si$ ($T_{1/2} = 2.62$ h), $^{30}P$ ($T_{1/2} = 14.3$ d), $^{31}Cl$ ($T_{1/2} = 0.622$ h), $^{44}K$ ($T_{1/2} = 12.4$ h), $^{48}Ca$ ($T_{1/2} = 165$ d), $^{44}Sc$ ($T_{1/2} = 84$ d), $^{50}Mn$ ($T_{1/2} = 2.58$ h), $^{56}Fe$ ($T_{1/2} = 45$ d), $^{60}Co$ ($T_{1/2} = 5.25$ y), $^{137}Cs$ ($T_{1/2} = 2.1$ y), and the maximal energy of beta-radiation emission of these radionuclides is 1.5 MeV-decay$^{-1}$ [5], with maximal range of beta-particles in quartz containing ceramics equal to 3.5 mm [21]. The maximal energy to beta-radiation emission of $^{90}Y$ is 0.934 MeV with the range of beta-particles in quartz containing ceramics equal to 2.45 mm [21], which is comparable to maximal energy and range of beta-particles irradiated by above mentioned neutron-activated radionuclides. These characteristics are important to interpret the results of estimation of 'depth-dose' profile in analyzed quartz containing ceramic sample in terms of relative contributions to 'depth-dose' profile from short range beta-particles and more penetrated gamma-irradiation.

Tanaka et al. [7] published the results of calculations of possible radiation doses to the skin from beta-particles and gamma-quanta irradiated by neutron-activated radionuclides. It has been demonstrated that among early visitors to the Hiroshima city just after atomic bombing, the accumulated during 1 week after detonation skin dose is to be about 0.8 Gy with 99% of the total skin dose caused by neutron activated radionuclides in the ground soil located at the distance of 1 m to the skin surface. The dose 0.19 Gy was estimated to be due to beta-irradiation and 0.63 Gy was caused by gamma-component, respectively. Skin irradiation by neutron-activated concrete dust from the crushed concrete buildings was considered as well [7]. The main constituents of this concrete dust, which caused the exposure during the first several days, were: $^{28}Al$ ($T_{1/2} = 2.24$ m), $^{31}Si$ ($T_{1/2} = 2.26$ h), and $^{137}Ba$ ($T_{1/2} = 82.7$ m). In this case, skin dose was estimated to be about four times higher than in a case of neutron activated soil material [7]. It was noted that the possibility of more extreme exposure scenarios, for example, among persons who received much heavier soil contamination on their skin cannot be excluded. Sure, it is not so simple to interpret these results in application to the situation with irradiation of quartz containing tile sample considered in our study. But anyway, it should be noted, that contribution to total dose from beta-particles (0.19 Gy) is comparable (at least within the one order of magnitude) with gamma-component of dose (0.63 Gy) [7].

The problem of estimation of the beta-exposure from neutron-activated radionuclide’s is the subject of special interest and discussion devoted to understanding the health effects following the Hiroshima A-bombing [4, 5], especially among newcomers to cities so soon after the detonations [5, 6].

This is important as well to understand the peculiarities of biological effects of internal irradiation by micro particles with beta-emitting radionuclides [16].

It seems to be especially important, as far as only gamma-rays have been considered in the conventional dosimetry systems DS86 and DS02 — without accounting for contribution from beta-radiation [1–3, 17, 22–25].

The peculiarity of our study is that 'depth-dose' profile measurements using the method of OSL from single quartz grain inclusions was performed in very thin layers of the sample located near the surface of the tile facing the air. These layers can be subjected to exposure from solar UV. As it is known, solar UV could confound the results of luminescence measurements with quartz grains located close to the surface.

It would be useful to consider the matters related to the depth of penetration of solar ultraviolet radiation into the surface layers of the solid sample. As it was noted in the section 'Material and Methods,' the analyzed quartz containing tile sample was covered by 0.1-mm thick black glazed layer with the 2.2 g/cm$^3$ density (the density of tile is 2.25 g/cm$^3$).

The electromagnetic spectrum of ultraviolet radiation, defined most broadly as 100–400 nanometers, can be subdivided into a
number of ranges recommended by the ISO standard ISO 21348 [26]; 'ultraviolet C' with wavelength range 100–280 nm (completely absorbed by the ozone layer and atmosphere), 'ultraviolet B' with wavelength range 280–315 nm (mostly absorbed by the ozone layer in atmosphere), 'ultraviolet A' with wavelength range 315–400 nm (most penetrating and not absorbed by the ozone layer). According to [27] the depth of penetration of UV radiation with wavelength about 400 nm (the most penetrating 'ultraviolet C') into the solid silicon (Si) with 2.3 g/cm³ density is equal to 0.03 mm. In a case of our study, the 0.1-mm thick black glaze layer of analyzed tile has the 2.2 g/cm³ density and the density of the tile is equal to 2.25 g/cm³, which is close to silicon density. It means that expected depth of penetration of solar UV radiation with wavelength up to 400 nm into the surface layers of analyzed tile sample will be less than 0.1 mm.

It should be noted in addition that as a result of exposure to light the luminescence signal from quartz grains should be reduced or zeroed due to 'bleaching effect.' Meanwhile, Fig. 1 shows that it is not a case in our study: the dose measured in the sample's layer located close to the surface (just behind of the removed 0.1 mm thick glaze) was not reduced in comparison with dose measured in deeper sample’s layers. This indicates that the quartz grains in the layer located close to the surface were not bleached due to UV exposure. It is looks like 0.1 mm thick black glaze served as shielding from UV exposure.

We are going to undertake further research into these matters: the OSL single-grain measurements of 'dose-depth' profiles in additional quartz-containing samples from Hiroshima and Nagasaki; as well as the comparative measurements of the 'dose-depth' profiles in thin layers of Hiroshima tile samples irradiated under experimental conditions separately by gamma-sources only, and by beta-sources only.

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