Comparison of calculated beta- and gamma-ray doses after the Fukushima accident with data from single-grain luminescence retrospective dosimetry of quartz inclusions in a brick sample

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ABSTRACT

To estimate the beta- and gamma-ray doses in a brick sample taken from Odaka, Minami-Soma City, Fukushima Prefecture, Japan, a Monte Carlo calculation was performed with Particle and Heavy Ion Transport code System (PHITS) code. The calculated results were compared with data obtained by single-grain retrospective luminescence dosimetry of quartz inclusions in the brick sample. The calculated result agreed well with the measured data. The dose increase measured at the brick surface was explained by the beta-ray contribution, and the slight slope in the dose profile deeper in the brick was due to the gamma-ray contribution. The skin dose was estimated from the calculated result as 164 mGy over 3 years at the sampling site.

Keywords: Fukushima accident; FDNPP; beta-ray dose; gamma-ray dose; cumulative dose; brick sample; Monte Carlo simulation; single-grain luminescence retrospective dosimetry

INTRODUCTION

The main fission products from the Fukushima Daiichi nuclear power plant (FDNPP) accident are $^{129m}$Te-$^{129}$Te, $^{131}$I, $^{132}$Te-$^{132}$I, $^{134}$Cs, $^{136}$Cs and $^{137}$Cs [1–4]. These radionuclides emit gamma rays and beta rays through $\beta^-$ decay. However, there are few studies about dose estimation from beta-ray irradiation following the FDNPP accident [5–7]. The beta-ray dose contributes to the whole-body dose among small biota, such as insects, plant leaves, and human skin. Therefore, beta-ray dose estimations are important for the risk assessment of the impact of the FDNPP accident (including on small biota) to clarify the effects of this large-scale radiological accident.

Retrospective dosimetry with brick samples has been used to evaluate the gamma-ray dose of the Hiroshima atomic bomb [8–10], the Chernobyl nuclear power plant accident [11–14], and the Semipalatinsk nuclear weapon testing [15, 16]. Recently, Stepanenko et al. [17] used retrospective dose evaluation of brick samples to estimate gamma-ray doses and perform beta-ray dose reconstruction for the FDNPP accident with a similar method to that used for a Hiroshima tile sample [18]. They used a single-grain quartz optically stimulated luminescence (OSL) method (similar to that of Ballarini et al. [19], although layer-by-layer consequences for very thin layers of the sample’s aliquots were used for analysis, with separate dose calibration for each quartz grain) with brick samples taken in 2014 from Odaka, Minami-Soma City, Fukushima Prefecture, Japan [17]. Dose enhancement near the surface of the brick was identified by the OSL measurements [17]. Stepanenko et al. suggested that the...
To establish the cause of the dose enhancement near the brick surface, we performed a Monte Carlo simulation of a small brick building with radionuclides uniformly distributed on the ground surface. The calculated results were compared with the data measured by Stepanenko et al. The depth profiles of the dose in the brick sample for beta rays and gamma rays were estimated separately, and the dose enhancement near the brick surface was discussed.

**MATERIALS AND METHODS**

**Particle and Heavy Ion Transport code System calculation**

The energy deposition as a function of depth in the brick wall of a small building was calculated using the Particle and Heavy Ion Transport code System (PHITS) Monte Carlo code Ver. 2.52 [20]. The calculation geometries are shown in Fig. 1. The calculation regions were 1 m × 1 m for beta rays and 21 m × 21 m for gamma rays. The calculation regions consisted of ground, air, and the small brick building (red region: 0.5 m × 0.5 m square, 1.5 m high, wall thickness of 10 cm). The brick building was located in the center of the soil surface. Beta- or gamma-ray sources were uniformly distributed in the 5-mm-thick soil surface (brown region). To save calculation time, the previously reported mirror condition was used for these calculations [21]. Figure 1a shows the geometry used to calculate the radiation that entered the calculation region (outer source calculation) via the mirror boundary. First, the histories for the particles were accumulated near the mirror boundary (green lines) without the brick building. Second, the particles were generated from the mirror boundary (back line) in Fig. 1b according to the accumulated histories. The generated particles were transported to the brick wall cells (yellow box) of the brick building. Third, radiation was generated from the surface of the 5-mm-thick soil layer (brown region) in the calculation region (inner source calculation) in Fig. 1b. The energy deposition in brick cell layers of 10 mm × 10 cm and thicknesses of 0.1, 0.2, 0.3, 0.4, 0.5, 1, 3, 5, 7.5, 10, 20, 40, 60, 80 and 100 mm were obtained by summing the outer and inner source calculations corrected with the number of particles generated per unit area.

Beta and gamma rays from $^{129m}$Te, $^{129}$Te, $^{131}$I, $^{132}$Te, $^{132}$I, $^{134}$Cs and $^{137}$Cs were calculated separately. Beta-ray energy spectra were taken from the literature [8], and the internal conversion electrons of $^{137}$Cs were taken from the website of the National Nuclear Data Center [21]. The gamma-ray energies and emission rates for the radionuclides were taken from the National Nuclear Data Center [22].

The elemental composition of the brick sample was Si: 28.9, Si: 50.4, Al: 17.5, Fe: 1.4 and Ti: 1.8 wt %, and those of soil and air were taken from the literature [8].

**Air dose and tissue dose calculation**

The air and tissue dose rates at the i-th depth per unit deposition density of 1 Bq/m², $D'_k$ (Gy Bq⁻¹ s⁻¹ m²⁻¹), for beta and gamma rays, were calculated from the calculated results of the energy deposition in brick as:

$$D'_k = \sum_j f_j \int_0^{E_{jk}} \frac{dE}{m_j} (j = \beta, \gamma; k = 129m_{\text{Te}}, 129_{\text{Te}}, 131_{\text{I}}, 132_{\text{Te}}, 132_{\text{I}}, 134_{\text{Cs}}, 137_{\text{Cs}}),$$

where $E_{jk}$ is the energy deposition (J) at the i-th depth by beta or gamma rays from the k-th radionuclide, $m_j$ is the brick sample mass (kg), and $a_j$ is the area of the source (0.75 and 1 m² for inner and outer beta calculations, 440.75 and 441 m² for the inner and outer gamma calculations, respectively). $I_j$ is the emission rate for beta or gamma rays per Bq and $f_j$ is the conversion factor of the stopping power ratio [23] for beta rays and the kerma ratio [24] for gamma rays between air or tissue and brick to convert from the brick dose to the air or tissue doses.

**Cumulative dose estimation**

The dose rate at the sampling point can be calculated by the measured deposition density, $A_{ks}$, for each radionuclide at the sampling point of Odaka, Minami-Some City by multiplying the calculated result by Eq. 1. The change in dose rate over time is assumed to depend only on the half-lives of the radionuclides. Therefore, the cumulative dose, $D'_{tot}$ for the i-th depth can be integrated by:

$$D'_{tot} = \sum_k \sum_j \int_0^\tau A_k \cdot D'_k \left(1 - e^{-\frac{T_k}{\tau}}\right) dt,$$

where $T_k$ is the half-life for each radionuclide of $k = 129m_{\text{Te}}, 129_{\text{Te}}, 131_{\text{I}}, 132_{\text{Te}}, 132_{\text{I}}, 134_{\text{Cs}}, 137_{\text{Cs}}$ (Table 1), and $\tau$ is the time period from deposition to the brick sampling date.

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Fig. 1. (a) Mirror condition calculation, (b) top view and (c) side view of the calculation geometry.
RESULTS AND DISCUSSION

Calculated dose rate for beta and gamma rays

A $^{137}$Cs deposition density of 308 kBq/m² and the ratio of each radionuclide to $^{137}$Cs deposition density taken from the literature \cite{1} were used to obtain $A_k$ for each radionuclide. The deposition densities for the seven radionuclides are listed in Table 2. The beta-ray dose rates on the brick surface and gamma-ray dose rate at a depth of 0.5 mm in the brick at a height of 80 cm are shown in Fig. 2a and b, respectively. $^{129m}$Te contributed less to the gamma-ray dose rate, and accounted for the third and fourth largest contribution to the beta-ray dose rate. This is due to the small gamma-ray emission rate per decay of $^{129m}$Te of $<10\%$. The gamma- and beta-ray doses decreased by $\sim10\%$ and $\sim30\%$, respectively, over 1 month. The calculated beta-ray dose rate decreased slower than the calculated gamma-ray dose rate.

Beck reported conversion factors for various radionuclides to estimate the air dose rate at a height of 1 m from the unit deposition density of radionuclides \cite{25}. The initial gamma-ray air dose rates (15 March 2011) at a height of 80 cm from the ground for each radionuclide obtained by our calculations were compared with the values estimated by Beck conversion factors \cite{25} interpolated at a relaxation depth of 0.65 g/cm² (Table 2). The present dose rates were estimated to be 57\% lower than those calculated by Beck conversion factors. The present dose rates were in-brick values in one of the walls of the brick building, whereas the Beck conversion factor values were free-in-air values. Therefore, the difference of 57\% can be explained by shielding effects, whereby gamma rays from behind the building are neglected.

Cumulative dose

The cumulative dose over 3 years, from 12 March 2011 (Unit 1 explosion) to 19 March 2014 (brick sampling by Stepanenko et al.) and the dose rate change over time are shown in Fig. 3. The solid line shows the calculation result, the dashed histograms are the averaged calculation values for the measured sample thickness, and the open circles are Stepanenko et al.'s data \cite{17}. The calculation agreed well with the data measured by Stepanenko et al. in the region deeper than 10 mm. The results indicated that the cumulative dose deeper in the brick was due to gamma rays, and that the dose enhancement at the surface was dominated by the beta-ray contribution. The difference between the calculated and measured doses at the surface was about 2 standard deviations. A possible explanation might be connected with the contributions of low $\gamma$ emission rate radionuclides, such as $^{89}$Sr, $^{127m}$Te, $^{127}$Te, $^{146}$Ba, $^{146}$La, etc. However, the trend in the dose increase at the brick surface was supported by the calculations. Therefore, the single-grain OSL measurement by Stepanenko et al. shows the advantage of dose estimations not only the cumulative gamma-ray dose but also the cumulative beta-ray dose. Thus, we concluded that the single-grain OSL method is a good tool for retrospective beta-ray dose estimation.

Table 1. Half-lives of the calculated radionuclides

| Radionuclide | Half-life |
|--------------|-----------|
| $^{129m}$Te  | 33.6 d    |
| $^{129}$Te   | 33.6 d a  |
| $^{131}$I    | 8.04 d    |
| $^{132}$Te   | 3.204 d a |
| $^{132}$I    | 3.204 d a |
| $^{134}$Cs   | 2.062 y   |
| $^{137}$Cs   | 30.0 y    |

*aHalf-life of parent radionuclide.

Table 2. Deposition density, initial gamma-ray dose rate in air obtained by Beck conversion factors (Beck 1980) (relaxation length of 0.65 g/cm²), and the present calculations

| Radionuclide | Deposition density (kBq/m²) | Gamma-ray air dose rate ($\mu$Gy/h) | Conversion factor ($\mu$Gy/h)/(Bq/m²) |
|--------------|-----------------------------|-----------------------------------|--------------------------------------|
| $^{129m}$Te  | 438                         | 0.017                             | 0.0981                               |
| $^{129}$Te   | 304                         | 0.027                             | 0.185                                |
| $^{131}$I    | 6130                        | 4.57                              | 7.91                                 |
| $^{132}$Te   | 2377                        | 1.24                              | 1.63                                 |
| $^{132}$I    | 2205                        | 10.0                              | 16.3                                 |
| $^{134}$Cs   | 308                         | 0.94                              | 1.58                                 |
| $^{137}$Cs   | 308                         | 0.34                              | 0.58                                 | 1.871                               |

Fig. 2. Air dose rates of (a) beta rays and (b) gamma rays over time.
The calculated tissue dose at a brick depth of 50 μm was assumed to be a skin dose, and would be similar to a 70-μm tissue dose. The skin dose was estimated to be 164 mSv for 3 years at the sampling location.

CONCLUSION
To confirm the cause of the dose enhancement near the surface of a brick sample taken from Odaka, Minami-Soma City, Japan, a Monte Carlo calculation was performed using PHITS code and the calculated results were compared with measurements. The calculated results agreed well with previously published measured data. The dose enhancement at the brick surface in the measured data was explained by the beta-ray contribution, and the gentle slope in the dose profile deeper in the brick was due to the gamma-ray contribution. The calculated result estimated the skin dose to be 164 mGy over 3 years at the sampling location.

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CONFLICT OF INTEREST
The authors declare that there are no conflicts of interest.

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