Directional Distribution of Radiation around an Accident at a Uranium Fuel Factory in Tokai-mura, 1999

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A beta-ray survey was carried out on concrete walls of the boundary and buildings after a criticality accident at a factory of JCO Co. Ltd. at Tokai-mura. A remarkable distribution of beta counts was observed on the walls depending on the complex internal and external structures of buildings surrounding a precipitation vessel containing uranium 23 days after the accident. The directional distribution function, based on the beta counts on the walls, was consistent with data concerning the neutron dose rate measured in several directions during the accident, suggesting an anisotropic neutron distribution to the residential area.

INTRODUCTION

A criticality accident, which occurred at the uranium conversion facilities of JCO Co. Ltd. in Tokai-mura, Japan on September 30, 1999, caused radiation exposure to not only workers, but also residents1). The critical state, which occurred at 10:35 am, was terminated by draining off cooling water around the precipitation vessel (PV) at 6:30 am on October 1. The evacuation of people within a 350 m zone began at 15:30 on the same day1). During the accident, neutrons and gamma rays exposed people around the factory. The gamma ray and neutron dose rates were reported to be 0.84 mSv/h (gamma) at 11:36, 0.5 mSv/h (gamma) and 4.5 mSv/h (neutron) at the boundary of the factory at 19:09 on Sep. 30, as the maximum values1,2). This neutron exposure to residents was completely different from that in the Chernobyl atomic power plant accident, and nuclear weapon tests where there was no direct

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neutron exposure to the population around the site \(^3\).

A dosimetry study for residents is urgently important under such circumstances. Japan Atomic Energy Research Institute (JAERI) reported external doses as a function of distance \(^2,4\). We focus here on the directional dependence of radiation from the criticality accident due to the complex structures of the buildings between the source and the residential area. The enriched uranium and fission products were kept in the PV (450 mm in inner diameter) of the building where the accident occurred during our measurements. Sand-bag walls were built on Oct. 2 for radiation protection of the residents located around the facility. Several concrete walls were built instead of sand-bag after our measurement on Oct. 26, 1999. A radiological survey was conducted mainly for beta counting on the wall of the boundary and of the building after the accident under the present condition.

**MATERIALS AND METHODS**

Beta counting was conducted at about 1-m height on the walls using a ZnS (Ag) plastic scintillator (Aloka TCS-352, active area of 72 cm\(^2\)). The detection efficiencies for beta rays (\(E_{\text{max}} = 310 \text{ keV}\)) and gamma ray (\(E_{\text{av}} = 1253 \text{ keV}\)) of Co-60 are 14.3 and 1.3 (%/2\(\pi\)), respectively. Each measurement was done for one minute. The main measurements were carried out on the boundary wall, which was made of concrete with 3 cm thick exists, the length between northeast and southwest was 880m, as shown in Fig. 1 on Oct. 23–25. The values at three points with 80 cm intervals, which were measured at each site of the boundary wall, were averaged. The sites on the outer wall of facility where measurements were made on Oct. 26 are shown in Fig. 2. The number of measurements was one for each site in this case. The

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**Fig. 1.** Local map of the facility for uranium fuels factory in Tokai where the accident took place. Beta counting was carried out on walls B1, B2 and B3 at the boundary of the factory.
entire wall is also made of the same material and thickness (autoclaved light concrete with 10 cm thickness, Siporex, Sumitomo Metal Mining Co., Ltd.).

RESULTS

A clear distribution of beta counts was observed on the western B1 and northern B2 walls, as shown in Fig. 3. In this figure, the net count was evaluated from the gross counts subtracted by the background value of 137 cpm, which was assumed to be average one for data beyond 300 m from the northwestern corner of the wall. Moreover, each point was recalculated by smoothing with neighboring points \(n_i = (n_{i-1} + n_i + n_{i+1})/3\). The maximum net values were about 3 sigma (12 cpm) of the raw background counts in the B1 and B2 walls. On the other hand, the level of net counts on the eastern wall (B3) was about within ± one sigma of the background count. Therefore, the distribution of beta count rate on this B3 wall was not clear due to the distant area about 500 m east away from the uranium source.

The distribution of beta counts in the B1 wall had a maximum at 85 m from the northwestern corner with full width at half maximum (FWHM) of 40 m. The count rates sharply
decreased toward southern side from this site. This may have been due to the shielding of the E-building for radiation, as shown in Fig. 2. The distribution of the beta count rate at the B2 wall had the maximum value at 50 m from the northwestern corner with a FWHM of 29 m.

An obvious distribution of beta count rates was also observed on the outer wall of several buildings, as shown in Fig. 2 and Table 1. The most remarkable site was No. 7 of the D-building with a beta count of 31 kcpm. The values were about 30-times higher than those on neighboring sites of both sides on the same wall. There was no concrete wall but two iron doors between site No. 7 and the uranium source in the C-building. Neutrons passing through the iron door from the source might strongly irradiate the wall at site No. 7. We notice that site No. 7 of the D-building is on one line between the source in the C-building and the maximum site of the B1 wall. Moreover, a remarkable distribution of beta counts was observed on the outer walls of the C-building of the facility where the accident occurred. Sites No. 7 and No. 8 at the northeastern wall of the C-building exhibited more than 10 kcpm. This wall has no glass windows. The distance between the source and the residents is more than 400 m, and several buildings exist in the northeastern direction. In the northwestern direction, there is free channel with a 7 m width among buildings.

The radiation survey for the walls of building and boundary of JCO suggested to us some channels of neutron beams to the residential area, at least in the southwest and northwest.
This depended on complex internal and external structures of the building surrounding the uranium source.

**DISCUSSION**

First, we have a question concerning the origin of beta counts on the wall. The present detector counts not only beta ray but also gamma ray with relatively small efficiency. Therefore we attempted the beta-ray range measurements on the boundary wall on Nov. 27. The range of beta ray from the wall was measured to be 0.071 g/cm² (0.5 mm) of plastic film in-situ on the wall, as shown in Fig. 4. This value is close to the value (0.062 g/cm²) of betas from Ca-45 (beta with 257 keV-100%, half-life of 164 days). To confirm Ca-45 as an origin of the beta counts, a measurement of the half-life must be the key. However a big change in gamma-ray background, which occurred due to the construction of thick concrete walls around the building where the accident took place after Oct. 26 1999, made an estimation of the half-life from the first and second data impossible.

Under this situation, a theoretical discussion on the creation of Ca-45 by thermal neutrons would be helpful. The total number of neutrons ($N_{tot}$) was evaluated to be about $3.6 \times 10^{20}$.
The maximum yield of Ca-45 is estimated to be 500 decay per minute (dpm) in a detector active area (72 cm²) of the concrete wall 70 m from the PV in the case of no shielding and using the number of thermal neutrons (in case of \(N_{\text{thermal}} = N_{\text{tot}}\) as the maximum estimation). Kofuji and Yamada evaluated Ca-45 < 5.8 dpm/g-Ca (as on Oct. 1) by liquid scintillation counting for a chemically separated Ca sample in a small specimen of concrete from the wall which contained the site of maximum beta counts in the B1 wall. The decay rate of Ca-45 is estimated to be < 728 dpm (as Ca of 0.581 g/cm³ in concrete) in the active area. This value is consistent with 500 dpm as theoretically maximum value. However, these arguments lead to a beta counting rate much less than 20 cpm in the detector (efficiency of 4% for beta of 257 keV) due to the short range of beta in the concrete. Therefore Ca-45 was not the main origin for the present beta counts on the concrete wall in our observation.

Another possible origin may be the conversion electrons in the boundary concrete wall with a thickness of 30 mm, generated by gamma-rays from fission products (FP) in the PV. Mitsugashira et al observed the gamma-ray spectra around the accidental building C where the accident occurred on Oct. 26 and Nov. 9 1999 by using a portable Ge detector. They estimated that the amounts of La-140 (T₁/₂ = 12.7d), Ru-103 (39d) and Zr-95 (64d) were \(1.4 \times 10^{11}\) Bq, \(2.1 \times 10^{10}\) Bq and \(2.6 \times 10^{10}\) Bq, respectively as on Oct. 1. The counting rate of gamma-rays (1596 keV) from La-140 is estimated to be 2900 cpm, as on Oct. 23, with 1% detection efficiency in the case of no shielding. Several concrete walls and other materials located between the gamma-ray source and our detector must decrease the counts. A further theoretical discussion may be needed by taking into account a detailed description of the complex structure of the building. However, the detection of conversion electrons which are generated in a concrete wall by gamma-rays from FP is acceptable as a main source of our
observations from the above discussion and beta-range measurement.

Data concerning the directional distribution of radiation during the accident is one of the most important types of information used to evaluate the external doses for the residents around an accident. However, there was no sufficient data on this question. The data of beta counting around the JCO factory may be a key in this situation.

We now evaluate the directional distribution \( F(\phi) \) of the net beta counts \( (N_b) \) on the boundary walls from the PV as the center. First, the counts should be normalized by the measurement angle \( (\theta_i) \) between the normal direction from the source and wall plane at the measurement site \( (i) \). (see Fig. 2) The detector plane was parallel to the wall-plane,

\[
N_b(r_i, \phi_i) = \frac{N_b(r_i, \phi_i, \theta_i)}{\sin \theta_i},
\]

where \( r_i, \phi_i \) are the distance from the source and the angle to the northern direction at measurement site \( i \). In this calculation \( r_i, \phi_i \) and \( \theta_i \) are determined for actual sites of measurement by using a local map.

Next, the counts should be normalized by distance \( r_i \) between measurement site \( i \) and the source,

\[
N_b(r_n, \phi_i) = B(r_n, r_i)N_b(\phi_i, r_i)
\]

where \( B(r_n, r_i) \) is a normalizing function for distance. In this study we applied an interpolation function, \( B (r_n, r_i) = \text{Exp}(−0.0113(r_n - r)) \), which is derived with \( R^2 = 0.98 \) from data concerning neutron dose rate measurement at 13 sites near to the boundary of the JCO campus at 7 p.m. on Sep 30\(^{1,2}\). Namely, the \( B(r_n, r_i) \) function is derived from the data set which was taken at almost the same site as our measurement. Therefore, the present normalizing function for distance may not be the best, but must be a good choice for analyzing our data at between 70 and 160 m.

Finally, we obtain \( F(\phi) \) by normalizing \( N_b(\phi_i) \) with \( N_b(\phi_0) \) at the site of maximum beta counts in the B1 wall,

\[
F(\phi) = \frac{N_b(\phi_i, r_n)}{N_b(\phi_0, r_n) = B(r_n, r_i)N_b(r_i, \phi_i) \sin \theta_i} \times \frac{B(r_0, r_i)N_b(r_i, \phi_i) \sin \theta_0}{B(r_0, r_0)N_b(r_0, \phi_0) \sin \theta_i}.
\]

The vector expression of \( F(\phi) \) clearly shows an anisotropy aspect of the beta counts especially in the southwestern direction in Fig. 5. There are two clear maximum directions, southwest and northwest. \( F(\phi) \) is not clear in the southeastern direction, since there is no concrete wall. The vector expression of the neutron dose rates, which were taken by a rem counter during the accident, can also be derived by \( B (r_n, r_i) \), as shown in the same figure. We should note that the present result concerning \( F(\phi) \) of beta counts from wall data on Oct. 23–25 is consistent with neutron dose-rate measurements at the boundary of the JCO campus in several directions on Sep. 30\(^{1,2}\).

It is not easy to understand the physics on this coincidence between the two sets of measurements. The rem counter is for measuring fast neutrons. On the other hand, beta counting may indirectly measure gamma-rays from FP after the accident. The fission products in which the most intensive gamma-ray was 1596 keV gamma-ray of La-140 was kept mainly in the
The dose rate ratio of neutrons/gamma rays was almost constant at several sites around the JCO factory. For example, the ratio for the dose in Sv units was to be $9.2 \pm 1.6$ at 13 sites near to the boundary of the JCO campus at 7 p.m. on Sep. 30\textsuperscript{1,2}). Therefore, this may be the reason for the coincidence. Moreover, this coincidence may support the rationality of our analysis.

The vector expression $F(\phi)$ of the beta counts on the wall after the accident must be meaningful under the state of lack of any real-time measurement of the dose rate in all directions during the accident. The coincidence between the two suggests that the present function ($F(\phi)$) should be available for dose reconstruction on residents\textsuperscript{8}).

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