

Estimation of Total Fission Events in the JCO Criticality Accident by Passive γ -Ray Measurement of ^{140}La in Radioequilibrium with Precursor ^{140}Ba

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The distinctive feature of the JCO criticality accident is the continuous release of fission neutron without appreciable release of fission products. Hence, the estimation of total fission events (TFE) is the most important task for evaluating environmental impact. The sampling of uranium solution in the precipitation vessel (PV) in which the criticality accident took place, was performed by the members of JAERI to know TFE and the analytical results were reported to the Science and Technology Agency (STA) and the Nuclear Safety Commission (NSC).¹

On Oct. 25, the Ministry of Education, Science, Sports and Culture decided to organize the Japanese university member to make research for the environmental impact due to the JCO accident and about 12 scientists and their coworkers started their research under the management of the group leader Prof. K. Komura, Kanazawa University. This research group has already performed five times the sampling of soil and plants in the JCO site. On the 2nd sampling on Oct. 26, the present authors carried a portable Ge detector and a 4096ch PHA to make the in-situ γ -ray spectrometry.

The detector was set on the top of the northwest corner of a shielding wall made of sandbag heaped up around the Uranium Conversion Building (UCB) in which the PV was placed. Intense γ -ray signals mainly due to ^{140}La that was in radioequilibrium with its precursor ^{140}Ba ($t_{1/2}=12.75\text{d}$) was recorded by the 4096ch PHA. The dead time ratio of the PHA system was about 20% and we noticed that the detector was at the nearest position practically allowed to access. Within about 30 minutes, three spectra were recorded and one of the spectra is shown in Figure 1. The main γ -ray at 1596 keV is very clearly observed with its sharp Compton edge, i.e., that implies the 1596 keV γ -ray comes from a single remote radiation source. Although scattering components are quite large at below about 500 keV and intense bremsstrahlung are recognized up to about 2000 keV, many characteristic γ -ray peaks of ^{140}La are observed. Rather weak γ -ray peaks of ^{131}I and ^{95}Zr - ^{95}Nb are also observed. The reference data of the 66th meeting of NSC¹ suggested that the majority of fission products was retained in PV. Therefore, TFE can be estimated without the sampling of the uranium solution in PV by passive γ -ray spectrometry of appropriate fission product radioactivity.

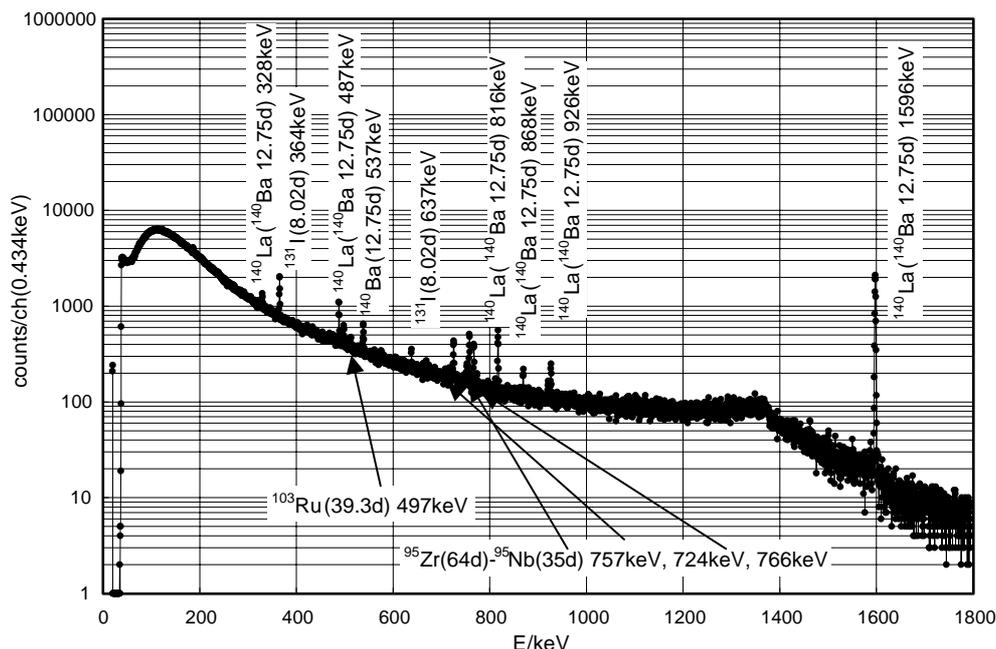


Figure 1. An in situ γ -ray spectrum obtained by a portable Ge detector set on the top of the northwest corner of a shielding wall made of sandbag heaped up around the Uranium Conversion Building.

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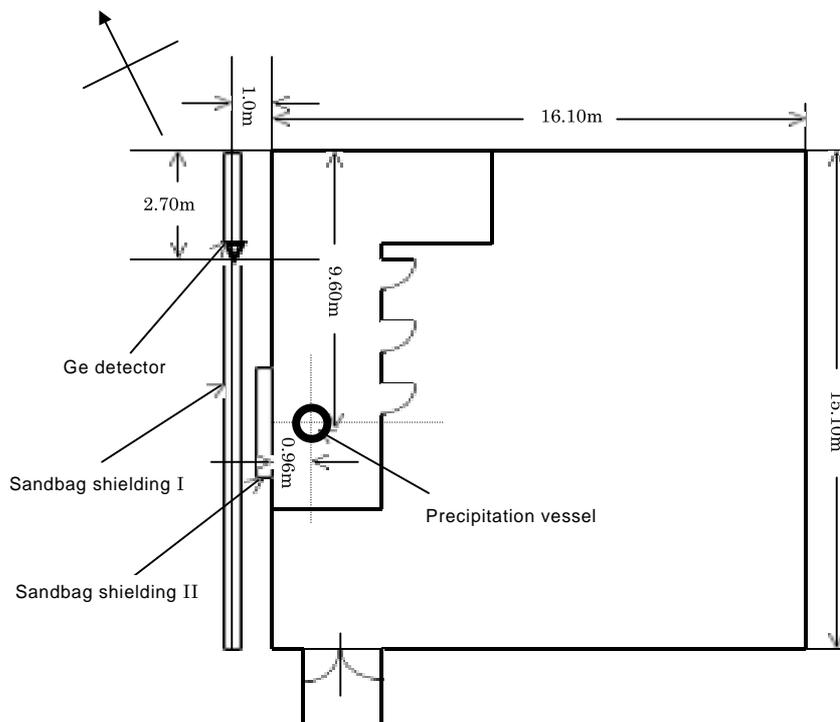


Figure 2. A schematic view of the Uranium Conversion Building of JCO Co. Ltd., and the Ge detector arrangement.

To calculate TPE from the observed γ -ray spectra, we estimated the attenuation of γ -ray by the wall of UCB, the uranium solution, SUS304 wall of PV, and the sandbag for shielding. Figure 2 is a plane figure showing a schematic arrangement of UCB, PV, the sandbag for shielding, and the Ge detector. The height of the sandbag shielding I is about 170cm and the head position of the Ge detector was about 175cm from the ground level. The bottom position of PV is estimated to be about 100cm from the ground level. From the geometric configuration, the γ -rays from PV penetrated the wall at an incident angle of 15.6 degree to its surface and at about 5.5 degree of elevation. The distance between the Ge detector and PV was determined to be 7.21m. The wall material of UCB is Siporex (ALC) that is calcined foam products of the mixture of silica, aluminum, quicklime, and cement. The density of Siporex is 0.65 g/cm³ and the wall thickness is 10 cm. From the angles, we estimated the penetration depth of the Siporex wall to be 37.3 cm. The concentration of the uranium in the solution and the penetration depth of the solution were evaluated to be 0.28 g/cm³ and 22.5 cm, respectively.¹ The thickness of SUS304 wall of PV and the cooling jacket is 3 mm. Thus, we assumed that the penetration depth of SUS304 was 6.7 mm. The height of the sandbag shielding II is about 2 m. The shielding II was heaped up at random and only its north end edge crossed the line between the detector and PV. We estimated the penetration depth of the sandbag to be 15 cm. Thus, the total attenuation factors of 329 keV, 487 keV, 816 keV, and 1596 keV γ -rays were estimated to be 0.000036, 0.00051, 0.0039, and 0.020, respectively.

The counting rate values of 329 keV, 487 keV, 816 keV, and 1596 keV γ -rays were 5.77 \pm 0.51, 11.48 \pm 0.77, 7.83 \pm 0.42, and 52.9 \pm 2.2 cps, respectively. The very large attenuation for 329 keV and 487 keV γ -rays compared with those for 816 keV and 1596 keV was mainly attributed to the uranium solution and sandbag shielding. The group leader Prof. K. Komura pointed out that the soil samples collected at the outside points close to the UCB wall were weakly contaminated with ¹⁴⁰La (Ref. 2), which would be explained by the activation of soil or

the trace release of the β^- -decay chain with mass 140 through ¹⁴⁰Xe ($t_{1/2}$ =13.6 sec).³ Thus, we omitted the 329 keV and 487 keV γ -rays from the TFE calculation since the traces of the existence of ¹⁴⁰La supported by ¹⁴⁰Ba in the outside of UCB greatly affected the results calculated under such large attenuation. Absolute counting efficiency values from a 7.21m point source for 816 keV and 1596 keV γ -rays were determined to be 2.74 \times 10⁻⁷ and 1.41 \times 10⁻⁷, respectively. The TFE values were calculated to be 2.95 \times 10¹⁸ and 1.96 \times 10¹⁸ for 816 keV and 1596 keV γ -rays, respectively, by adopting 0.0627 as the cumulative fission yield for ¹⁴⁰Ba (Ref. 4). This rough estimation for TFE of 2–3 \times 10¹⁸ was in fairly good agreement with Reference 1.

Our estimate of TFE based on passive γ -ray spectrometry is principally independent of the amount of uranium and the volume of uranium solution. It should be mentioned that we did not confirm the 1/r² dependency of the observed γ -ray intensity. In spite of the above fairly good agreement, the authors consider that the values are a rough approximation. Thus, our present result must be ensured by much more sophisticated measurement and such attempts are proposed to JCO, STA, and NSC by the present authors.

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References

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