ICANS XIX,
19th meeting on Collaboration of Advanced Neutron Sources
March 8 – 12, 2010
Grindelwald, Switzerland

DISCUSSIONS ON SPALLATION PRODUCTS BEHAVIOUR BY MEASURING MERCURY AND SUBSTANCES ADHERING INSIDE THE MERCURY CIRCULATION SYSTEM IN MLF/J-PARC

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ABSTRACT

Some spallation products are found to adhere to inner surface of a mercury circulation system in the Materials and Life science experimental Facility (MLF) in J-PARC. We obtained specimens of mercury and adhering substances from a pipe of the circulation system, and carried out gamma-ray spectroscopy aiming at obtaining information for discussions about how to manage high radiation from the adhering substances. Mercury of 150 g in weight and pieces of small paper smeared inner surface of the pipe were obtained after proton beam operations of 130 kWh. In the mercury specimen, twelve nuclei were identified: Y-88, Ag-110m, Sn-113, Eu-146, Lu-172, Lu-173, Hf-175, Os-185, Ir-188, Ir-189, Pt-188, Hg-203. Eleven of them, except for Ag-110m, were also found in the adhering substances specimen. The radioactivity of each nuclide in the mercury specimen was more than several percentage of a calculated value. The rate of radioactivity, except for Hg-203, in the adhering substance specimen to the calculated total radioactivity agreed each other within one order of magnitude. It was concluded that Hg-203 and Ag-110m were hard to adhere to the pipe, other spallation products adhered to the pipe with almost same rate.

1. Introduction

Mercury is used as target material for neutron production in the Materials and Life science experimental Facility (MLF) of Japan Proton Accelerator Research Complex (J-PARC [1]). Twenty-ton of mercury is circulated to remove heat generated by irradiation of 1-MW proton beam. All components of the circulation system are located on a target trolley in a hot cell of MLF. Mercury is activated so high that human access near the target trolley should be strictly limited. In order to reduce the radiation from mercury, mercury was designed to be drained into tanks surrounded by 30-cm thick iron shields. Many efforts had been paid to reduce the amount of mercury remained inside the components, and the estimated dose rate was reduced to be 1 mSv/h at 3 m from the circulation system at 10-day cooling after 1MW operation [2].

It was expected that the dose rate was acceptable after draining mercury in the commissioning period operated by 4 kW of proton beam power. The fact was, however, far from the expectation. The radiation around the circulation system surely increased after draining. Gamma-ray spectroscopy was carried out before and after draining at 10 m behind the target trolley by using a high-purity germanium (HPGe) detector to investigate the reason for the radiation increase. The counting rates of peaks corresponding to ^{203}Hg decreased and became unrecognized after 10 minutes from the start of draining. On contrary, those to ^{185}Os and ^{188}Ir increased by a factor of about 2. It is concluded that some

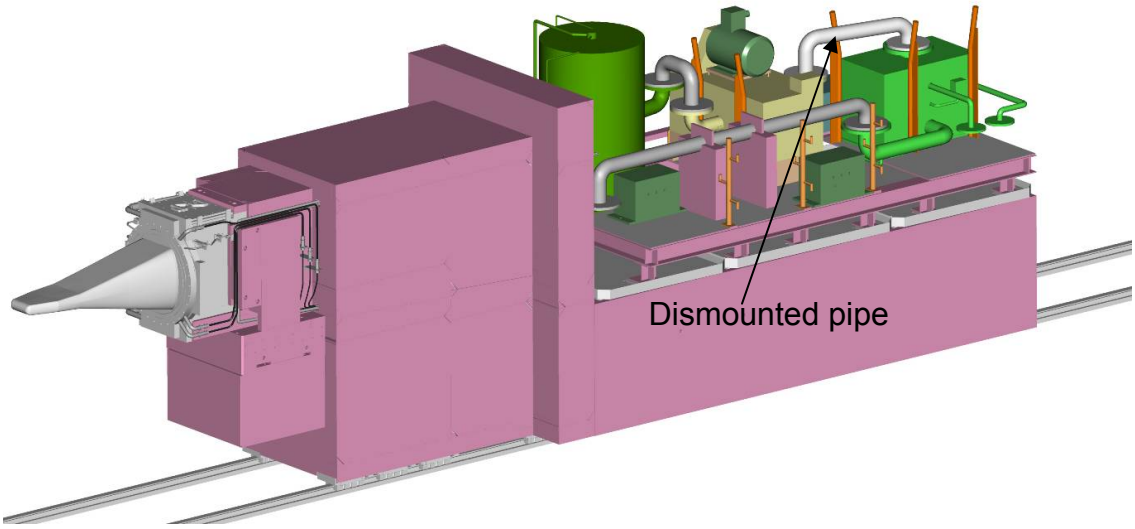

Fig. 1 The depiction of the target trolley and the mercury circulation system in J-PARC

of spallation products produced in mercury adhere to the components of circulation system. [3]

In this study, we observe inside the pipe, where the highest dose rate was found, visually by using a camera, then take and analyze specimens of both mercury and adhering substances from the circulation system aiming at obtaining more information to discuss how to manage the adhering substances. The results are compared with calculated radioactivity.

2. Measurements and calculation

2.1. Visual observation and taking specimens

The pipe of about 2 m (Fig. 1) in length was dismounted to observe the inner surface and to take the specimens of mercury and adhering substances at 57 days after 130 kWh beam operation. Both ends of the pipe were covered by plastic sheets. Then the pipe was moved onto a stand in the hot cell by using a crane. One side of the pipe had a bellows structure. Some amount of mercury was expected to remain in the bellows. A small 0.3 M pixel CCD camera, being putted into a plastic bag to avoid contamination, was used for the observation. There was nothing to remark although small drops of mercury were found as shown in Fig. 2. The plastic bag was highly

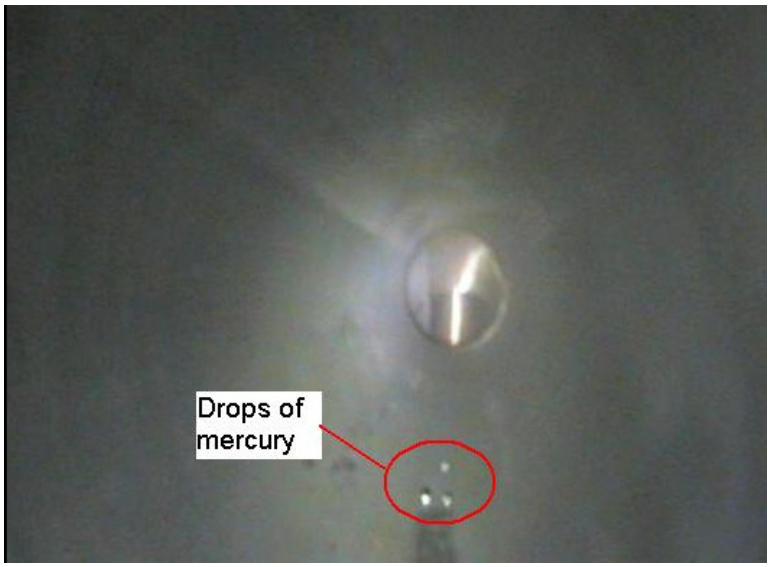

Fig. 2 Photograph of inner surface of the pipe in the mercury circulation system. Some drops of mercury are seen, but nothing special is found.

contaminated after the observation.

The inner surface of the pipe was smeared by small pieces of paper. Then each piece of paper was putted into a plastic bag that can be easily sealed by hand. Something like black dust was found on the paper as shown in Fig. 3. After the smearing, the pipe was tilted, and spilt mercury on the plastic sheet was taken into a polyethylene bottle by cutting the sheet. About 150 g of mercury was obtained. The dose rate at the surface of mercury was 60 μSv/h.

Fig. 3 Photograph of a piece of paper smeared inner surface of the pipe of mercury circulation system.

2.2. Calculation

The radioactivity of mercury was calculated by using the PHITS [4] and DCHAIN-SP [5, 6] codes. A calculation model had been commonly used not only for this radioactivity calculation but also for neutronic performance studies of the Japan spallation neutron source [7-9]. All components of the neutron source such as mercury target inner and outer vessels, cooling water, hydrogen moderators, reflectors and gaps, were included in the calculation model as detailed as possible.

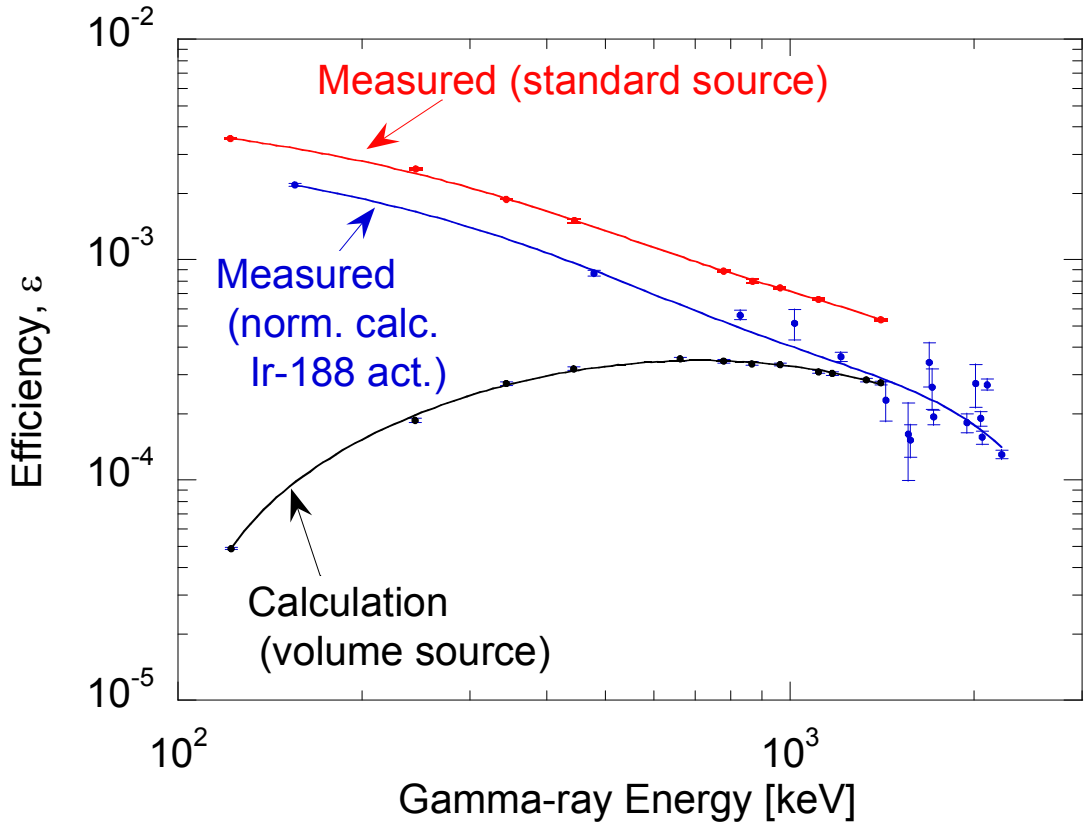

Fig. 4 Efficiency curves of HPGe detector obtained by using standard sources, Monte Carlo calculation assuming homogeneous distribution of source in mercury and Ir-188 in mercury. The efficiency by Ir-188 was normalized to the calculated radioactivity assuming all produced Ir-188 was included in mercury.

2.3. Efficiency of high-purity germanium detector

Commonly, the efficiency of HPGe detector is obtained by measuring gamma rays from standard sources that can be treated as a point source. A curve indicated as 'Measured (standard source)' in Fig. 4 shows the result determined by standard sources. This curve was applied to the gamma-ray spectroscopy for the smeared specimen. In the case of mercury, it is not adequate to assume a point source because gamma rays are absorbed in mercury itself especially at low energies. We performed Monte Carlo calculations to obtain efficiency for the mercury specimen. In the calculation, gamma-ray sources were assumed to be distributed homogeneously in mercury. The result is indicated as 'Calculation' in Fig. 4.

It is also possible to obtain an energy dependency of detector efficiency by using gamma-ray peaks from ^{188}Ir in mercury since ^{188}Ir emits many gamma rays in wide range of energy. A blue curve indicated as 'Measured (norm. calc. Ir-188 act.)' in Fig. 4 shows the energy dependency normalized to the calculated radioactivity of ^{188}Ir assuming all produced ^{188}Ir remained in mercury. This curve was quite different from the efficiency for the mercury specimen calculated by Monte Carlo simulation. Low energy gamma rays were not absorbed in mercury. It indicated that ^{188}Ir was not distributed homogeneously in mercury. It might be natural to think ^{188}Ir stuck to the bottom of the polyethylene bottle, it was not easy to know a distribution of each nuclide in mercury. Although there would be large ambiguity in results, we assumed the efficiency was between two curves: the efficiency obtained by the standard sources and that calculated for the mercury specimen.

3. Results of gamma-ray spectroscopy

3.1 .Mercury specimen

Figure 5 shows gamma-ray energy spectra of the mercury specimen measured at cooling times of 86, 126 and 505 days. In the spectrum at 86-day gamma rays from ^{188}Ir and ^{203}Hg were dominant. The gamma rays from ^{188}Ir were almost invisible in the spectrum at 126-days since ^{188}Ir decayed with half-life of 10.2 day, which corresponded to the half-life of the parent nuclide ^{188}Pt. At 505-day, the gamma ray from ^{203}Hg, having half-life of 46.6 day, was no longer dominant. Twelve nuclides were identified from these spectra: ^{88}Y, ^{110m}Ag, ^{113}Sn, ^{146}Eu, ^{172}Lu, ^{173}Lu, ^{175}Hf, ^{185}Os, ^{188}Ir, ^{189}Ir, ^{188}Pt, ^{203}Hg. As an example, the radioactivity of ^{185}Os, ^{188}Ir, ^{203}Hg are shown in Fig. 6 as a function of the cooling time, and are compared with calculations assuming all produced radioactivity are contained in mercury. Large errors of the results were caused by the ambiguity of the detector efficiency as mentioned in section 2.3. Each nuclide decayed along with the calculated curve. Thus the average of ratio to the calculated value for each nuclide is shown in Fig. 7 in increasing order of gamma-ray energy used to be derive the radioactivity. Since the ambiguity of efficiency is the smaller in the higher energies, the ambiguity of results goes down in rightward. A benchmark study of proton-induced radioactivity production for the calculation system [10] shows calculation accuracies for production of these nuclei (except ^{188}Pt, ^{189}Ir, ^{188}Ir and ^{110m}Ag) in mercury are between 0.4 and 0.9 in ratios of calculation to experiment (C/E). Thus, it was indicated that at least more than several percentages of spallation products remained in mercury.

3.2 Smeared specimen

Figure 8 shows averages of ratios of measured radioactivity in the smeared specimen to calculated radioactivity in whole (20 tons of) mercury. Same nuclides as shown in the mercury specimen are plotted in Fig. 8. The ratio of ^{203}Hg was lower than other nuclides by three orders of magnitude, and ^{110m}Ag was not recognized in the smeared sample. The ratios of other nuclides agreed each other within one order of magnitude. It was indicated that mercury and silver were hard to adhere to the pipe.

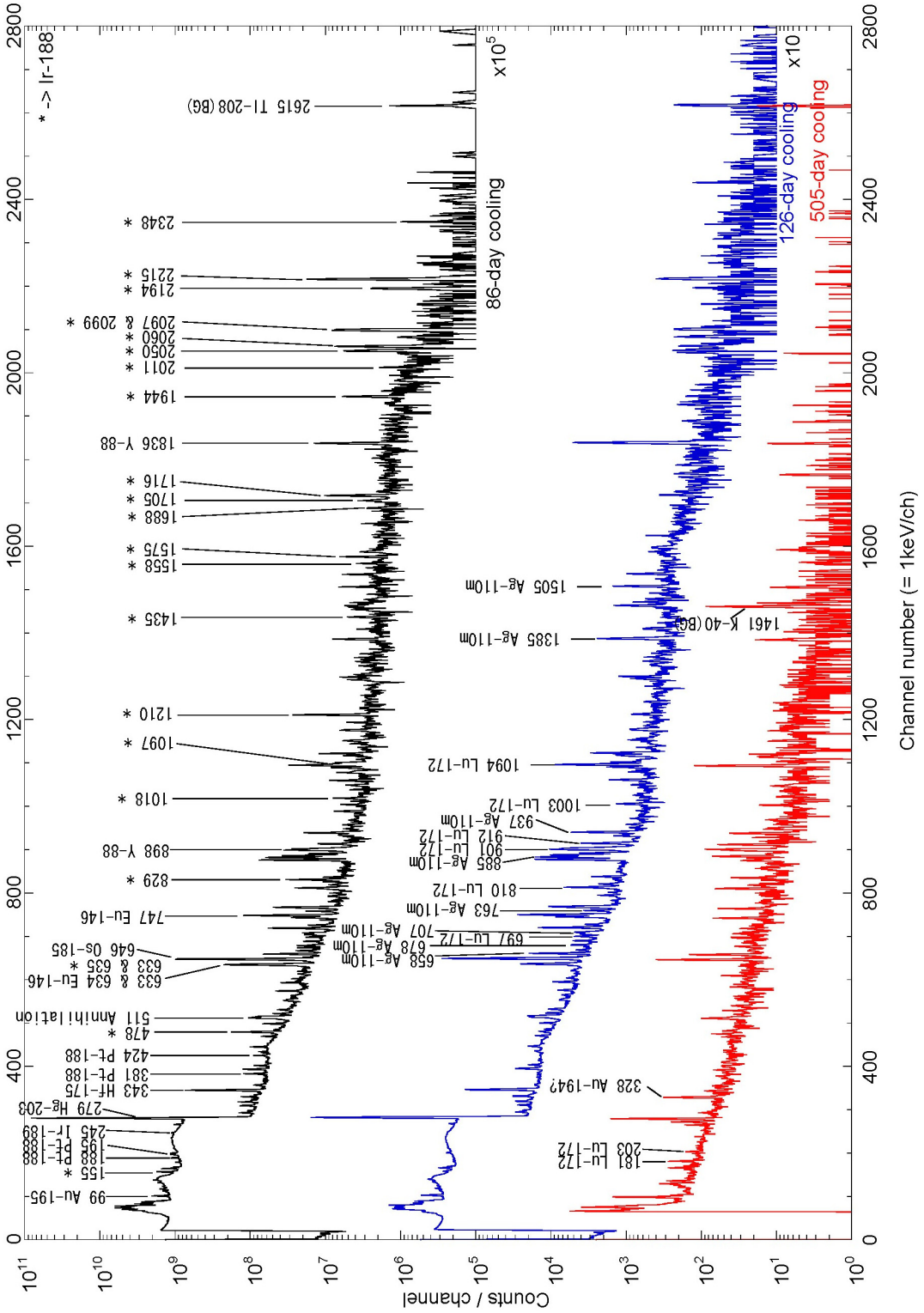

Fig. 5 Gamma-ray energy spectra of the mercury specimen measured at 86, 126 and 505 days cooling.

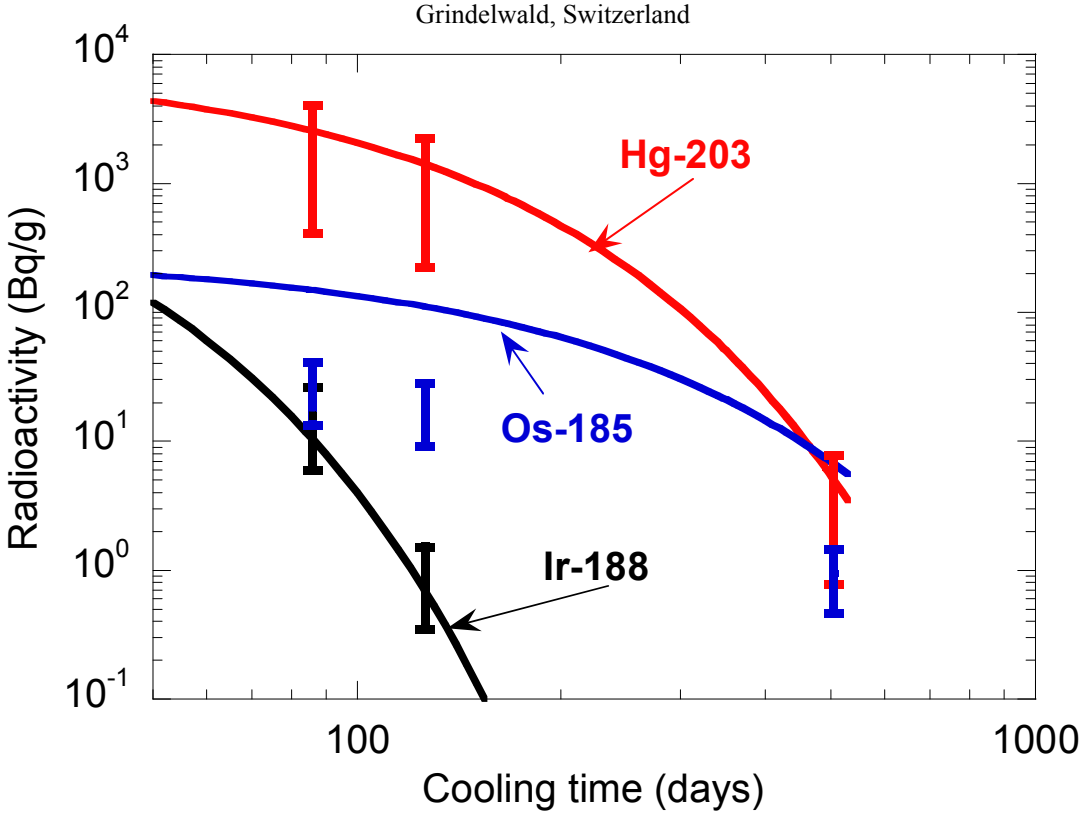

Fig. 6 Measured radioactivity of ^{185}Os, ^{188}Ir and ^{203}Hg in the mercury specimen are compared with calculation (solid lines). In the calculation all produced radioactivity was assumed to be contained in mercury.

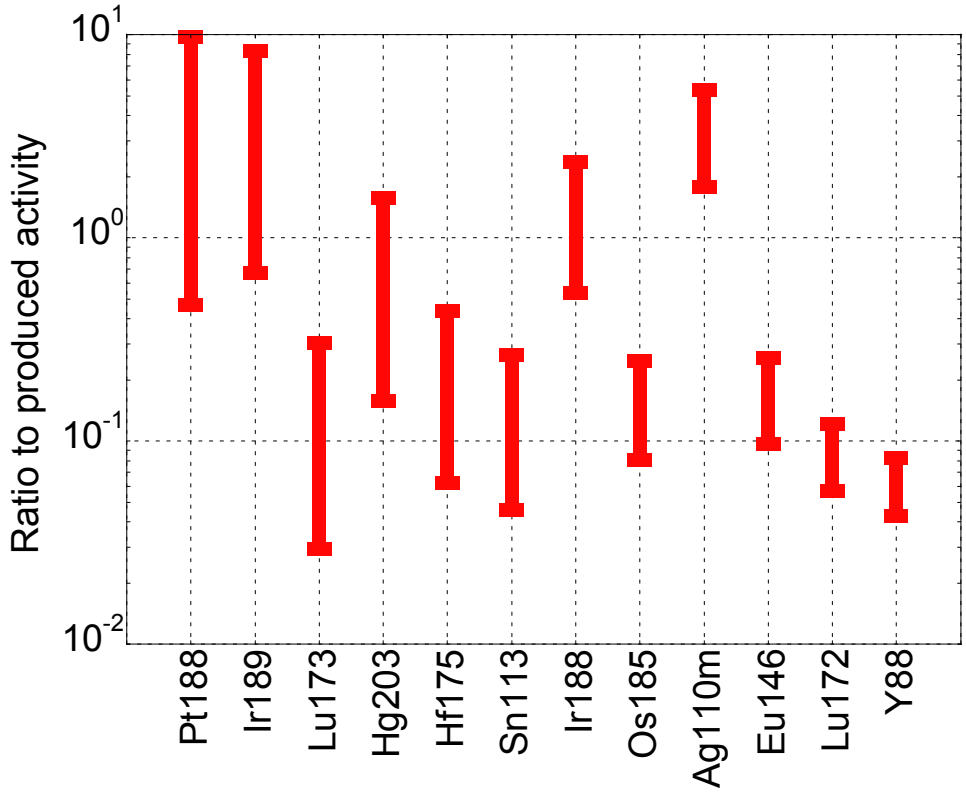

Fig. 7 Ratio of radioactivity in the mercury specimen to calculations assuming all produced radioactivity is contained in mercury.

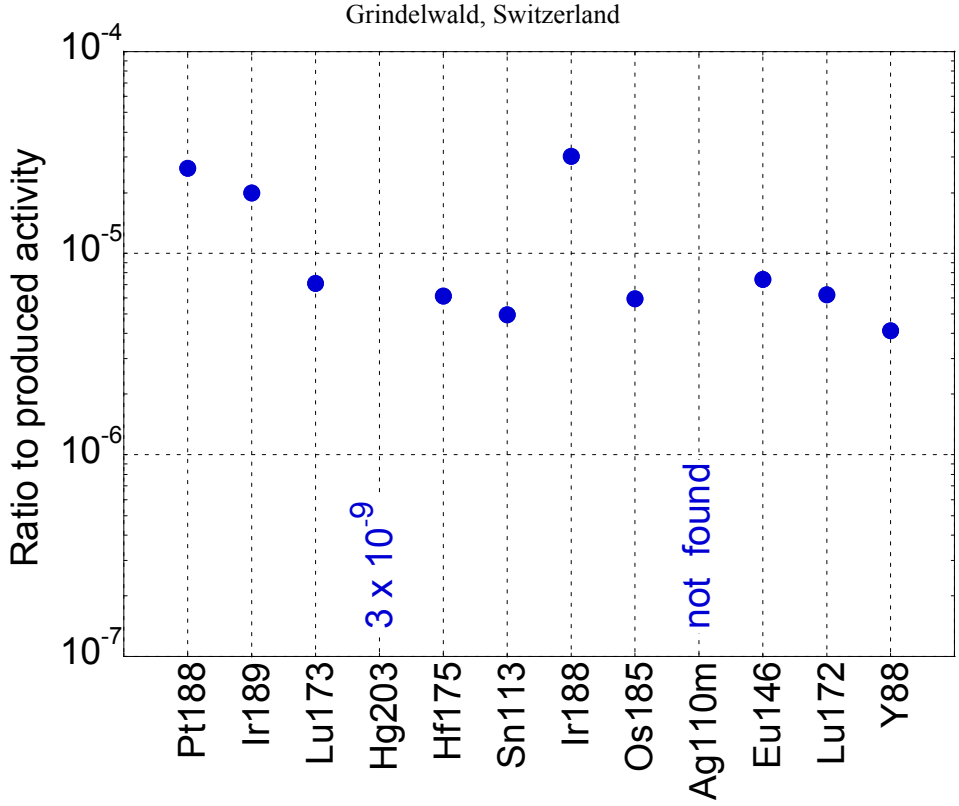

Fig. 8 Ratio of radioactivity in the smeared specimen to calculated radioactivity in whole (20 tons of) mercury.

4. Summary

It was directly confirmed that spallation products adhere to inner surface of the pipe of the mercury circulation system. Although nothing special was viewed inside the pipe by optical observation with the CCD camera, the inner surface was highly contaminated.

Gamma-ray spectroscopy was performed for mercury and adhering substances to the inner surface of the pipe. Twelve nuclides were found in the mercury specimen. The radioactivity was more than several percentages of the calculated ones.

All nuclides found in mercury except for ^{110m}Ag were also recognized in the adhering substances. The rate of radioactivity to the calculated total radioactivity was agreed each other within one order of magnitude except for ^{203}Hg. From these results, it was concluded that mercury and silver were hard to adhere to the pipe, and other spallation products adhered to the pipe with almost same rate.

Acknowledgements

The authors thank staff of the radiation safety section in J-PARC for extensive assistance in taking the specimens.

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