Survey Methodology for the Activation of Beamline Components in an Electrostatic Proton Accelerator

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To establish a systematic guideline for accelerator decommissioning, as a case study, beamline activation of 12 MeV-proton electrostatic accelerator was investigated employing a survey meter and γ-ray spectrometers. Beam loss points where reflected as high dose-rate area were identified, and generated nuclides and their activities were determined. Almost beamline components are made from stainless steel and 52Mn and 56Co were detected as principal induced activities. It was found that the 56Co activity significantly contribute to the dose rate value denoted on the survey meter. From the beam operation history and the monitor currents of Faraday-cups, we revealed the beam loss on a certain point significantly reflects the 52Mn activity on there. Induced activities of 52Mn and 56Co on the certain point of the beamline could be reproduced by the contact dose-rate on that point.

Key Words: electrostatic accelerator, activation, decommissioning, beam-loss, survey meter

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1. Introduction

Accelerators are widely used in science, engineering, and medical fields, and have played a significant role in the development of human society. However, beam loss during the accelerator operation provokes activation of various materials constituting the facility; these further lead to problems in waste management. Particularly, in the case of long-lived nuclides such as 60Co(half-life: 5.27 y) and 152Eu(half-life: 13.1 y), their effects must be considered over prolonged periods of time. Therefore, it is important to determine what region is activated, along with the type and quantity of radionuclide activity generated in the accelerator facility before decommissioning. Since the activation level and generated nuclides depend on the acceleration energy and the current in each facility, systematic study is required to comprehend the activation of the whole accelerator facility. To establish a reasonable procedure for the decommissioning of an accelerator facility, we conducted a large-scale activation survey in typical accelerator facilities in Japan. In this paper, we introduce the method and results of beamline activation investigation using a case study of an electrostatic accelerator.

Generally, the activation level of an electrostatic accelerator facility is expected to be quite low and the activated areas are limited because the beam energies and currents are relatively low. However, the activated areas and nonactivated areas should be segregated properly at each facility during decommissioning. The generated nuclides differ between the beamline and building due to differences in the activation mechanisms¹⁻³. Thermal neutrons generated during the accelerator operation influence concrete activation the most,
whereas in the case of beamline activation, the effects of primary particles must also be considered. We had already evaluated thermal neutron flux during accelerator operation using dosimeters and gold foils, and we established the evaluation method for activated concrete materials in various accelerator facilities in Japan\textsuperscript{4–10}. In this study, we evaluated the contact dose-rate (i.e. a value displayed as 1 cm dose equivalent on the survey-meter) of the beamline as an indicator of activation, and identified the generated nuclides at each activated area, employing a 6-MV tandem-type accelerator.

We also investigated other facilities, and we found that all the beamline components, except for the target and beam slit, are not activated in the case of an acceleration energy of less than 3 MeV for protons, except for the case at facilities that accelerate deuterons or intentionally generate neutrons.

2. Experimental

2-1. Facility

All experiments were conducted in the 6-MV Pelletron tandem accelerator at the university of Tsukuba (UTTAC), starting from 2016\textsuperscript{11}. The entire beamline is schematically shown in Fig. 1. A negatively charged hydrogen (H\textsuperscript{−}) is accelerated to 6 MeV by a tandem accelerator, then stripped of all electrons, and finally accelerated to its maximum energy of 12 MeV as a proton beam. Additionally, various elements from hydrogen to gold are available as ion beams. A total of 12 types of beamlines can be used depending on the processes in the experiments, such as bombardment, diffraction, and mass spectrometry.

2-2. Proton beam bombardment for a metal target

For simulating the activation of accelerator components, a metal test piece was irradiated with a proton beam in January 2018. A 1-mm-thick SAE grade 304 stainless steel (SUS) and tantalum (Ta) was placed at the center of the target chamber on an A3 beamline and bombarded with the proton beam. Since many beamline components, such as the beam pipe, joint, and vacuum chamber, consist of SUS, and Ta is used in a Faraday cup, these materials were selected. All the experimental conditions are summarized in Table 1. After irradiation, the test piece was withdrawn from the chamber and measured with a germanium (Ge) semiconductor detector (Canberra, GR2018), placed at a distance of 0.3 to 1 m. Neutrons generated from the

| Target | Particle/Energy/Current | Irradiation period |
|--------|-------------------------|--------------------|
| SUS    | p\textsuperscript{+}/12 MeV/1 μA | Jan. 18, 14:05–16:05 |
| Ta     | p\textsuperscript{+}/12 MeV/1 μA | Jan. 18, 18:20–20:20 |
| SUS    | p\textsuperscript{+}/6 MeV/1 μA | Jan. 19, 13:55–16:10 |

![Fig. 1. Beam line layout of 6-MV tandem accelerator at the university of Tsukuba (UTTAC). There are 12 branch beamlines for various ion experiment, depicted as A1–A7 and L1–L5. The dotted line indicates the beam stream for the experiment on Jan. 18. Measurements with γ-ray detectors were performed at points indicated by “A”–“V”. Points of “A”–“O” were determined by the pre-assessment, “P”–“V” were determined by the post-assessment.](image-url)
target during proton bombardment were measured by various methods, and the result has been discussed elsewhere.\(^7\)

2-3. Activation survey on the beamline

As a pre-assessment of activation, we scanned the contact dose-rate on an entire beamline using a sodium iodide (NaI) scintillation survey meter (Hitachi, TCS-171) before performing beam irradiation experiments, on Jan. 17th and 18th, to reveal an activation circumstance. Simultaneously, \(\gamma\)-ray spectrometry at high-dose areas was performed with a lanthanum bromide (LaBr\(_3\)) scintillation spectrometer (Mirion, InSpector1000), and the generated nuclides were identified.

Post-assessment was performed after 12 MeV proton beam irradiation to the SUS and Ta target, the entire beamline was scanned with a NaI survey meter and nuclide identification using a LaBr\(_3\) detector in high-dose areas was conducted. A Ge detector was also employed to identify the generated nuclides.

3. Results and Discussion

3-1. Pre-assessment of activation before the beam experiment

Before the experiment, the contact dose rates of all beamline components were measured with the NaI survey meter; some activated areas were found, and these are indicated as “A”–“O” in Fig. 1. The beam pipe and flange were activated rather than the yoke or coils in the magnet. This activation was caused by previous operations. The dose rates of other areas except these points were less than those at the background level. The nuclear identification in principal points was also conducted using the LaBr\(_3\) detector. The results are summarized in Table 2. It is suggested that the high-dose area would reflect the beam loss point where the trajectory of the beam changes, such as before and after the magnet, or a part where the cross-section for beam passage changes significantly, such as a flexible joint. Radionuclides of \(^{52}\)Mn (half-life: 5.59 d) and \(^{56}\)Co (half-life: 77.3 d) were detected at many activated points. These nuclides are considered to be generated by the \((p, n)\) reaction of chromium and iron, respectively. Radioisotopes of technetium (\(^{95m}\)Tc; half-life: 61 d, \(^{99m}\)Tc; half-life: 4.28 d, \(^{99}\)Tc; half-life: 6.01 h) were considered to be attributed to the \((p, n)\) reaction of molybdenum and were detected in the beam profile monitor (“K” in Fig. 1).

3-2. Generated nuclides in the bombarded target

Under the condition of 12 MeV proton bombardment for the SUS target, mainly production of two radionuclides of \(^{52}\)Mn and \(^{56}\)Co was found, as shown in Fig. 2. This was consistent with the estimation result of the radionuclides produced by 12 MeV proton irradiation for SUS, from the Q value, the Coulomb barrier, and the cross-section of the formation reaction. There is no concern for generation of pure \(\beta^-\) emitters such as \(^{63}\)Ni and \(^3\)H, in this experimental condition. Activation level of the Ta target was relatively low and \(\gamma\)-rays attribute to \(^{181}\)W, and \(^{182}\)Ta were observed. The 6 MeV for SUS target generated minimal detectable radionuclides. Measurement time of each condition was approximately 5–10 min. and detection limit was 1 kBq for \(^{56}\)Co.

3-3. Post-assessment of activation for beamline components

The result is summarized in Table 2. The activated areas, denoted by “P”–“V” in Fig. 1 were found on the A3 beamline following the entire beamline re-investigation after 20:30 on Jan. 18. The short-lived nuclide of \(^{56}\)Mn (half-life: 2.68 h) was detected at many measurement points, and \(^{52m}\)Mn (half-life: 21 m) and \(^{60}\)Cu (half-life: 23 m) were also found at some points. A peak attributed to annihilation was observed in many spectra, they immediately attenuated then almost disappeared the next day. Considering the half-life and \(\gamma\)-ray energy, it is suggested that the peak was derived from \(^{52m}\)Mn, \(^{60}\)Cu, and \(^{62}\)Cu (half-life: 9.7 m). This experiment would not have influenced the activation in the accelerator room components since the spectra obtained on Jan. 19th were consistent with the spectrum obtained on Jan. 17th, as shown in Fig. 3. The increase in the dose rate at the beamline was transient and supposed to be attributed to the short-lived nuclides.

3-4. Beam loss estimation

It was found that \(^{52}\)Mn and \(^{56}\)Co generated by the proton beam reaction in the SUS account for significant induced activity in the facility. Therefore, the activation of the beamline could be caused by the loss of primary beam particles. It is expected that the beam loss amount at the component made of SUS can be estimated from the activity generation rate for \(^{52}\)Mn or \(^{56}\)Co in an SUS material. Based on the analysis for the SUS target bombarded with 12 MeV \(p^+\), and \(\gamma\)-ray spectrometry on the beamline, the relative beam loss was calculated.

First, the recent operation history was investigated to reveal the influence of previous operations on the present activities of \(^{52}\)Mn and \(^{56}\)Co. A total of ten proton beam experiments were
performed between Sep. 1st, 2017 and Jan. 18th, 2018. There were no beam operations from Dec. 15th, 2017 to Jan. 16th, 2018. Acceleration energies of 3, 6, and 12 MeV were employed. The beam current ranged from 1 to 30 $\mu$A, and the total operation time was 122 h. Considering the above, the residual activity due to previous operations, “A”, was estimated from the equation (1).

$$A = I \cdot \sigma \cdot (1 - e^{-\frac{t}{T}}) \cdot e^{-\frac{t}{T}}$$  \hspace{1cm} (1)

Here, parameters “I”, “T” are the beam current and operation time for each experiment, respectively. “$\sigma$” is the cross section for $^{56}$Fe(p, n)$^{56}$Co or $^{52}$Cr(p, n)$^{52}$Mn reactions. Since the cross sections of both the reactions are very similar in the incident particle energy ranging from 3 MeV to 12 MeV\(^{12}\), a common

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### Table 2. Measurement place on the beam line and detected nuclides.

| Point | Component name | Contact dose rate \((\mu Sv/h: NET)\) | Detected nuclides | Relative beam loss (%) |
|-------|----------------|----------------------------------------|-------------------|------------------------|
| A     | Faraday cup    | 0.57 Jan. 17 1.02 Jan. 18             | $^{52}$Mn, $^{56}$Co | 54.6±4.0 -0.01±0.10 |
| B     | Stripper foil  | 26.2 22.1 Jan. 17 22.1 Jan. 18        | $^{52}$Mn, $^{56}$Co | 4.13±0.31 -0.006±0.009 |
| C     | Bending magnet | 6.52 6.65 Jan. 17 6.65 Jan. 18        | $^{52}$Mn, $^{56}$Co, $^{57}$Co | 0.0600±0.0061 0.0118±0.0011 |
| D     | Faraday cup    | 0.04 Jan. 17 0.27 Jan. 18             | $^{56}$Co         | 0.09±3.58              |
| E     | Bending magnet (in) | 0.16 5.35 Jan. 17 5.35 Jan. 18 | $^{56}$Co         | 0.0600±0.0061 0.0118±0.0011 |
| F     | Bending magnet (out) | 0.09 3.58 Jan. 17 3.58 Jan. 18 | $^{52}$Mn, $^{56}$Co | 0.058±0.041 0.0031±0.0024 |
| G     | Quadrupole magnet (in) | 0.02 1.19 Jan. 17 1.19 Jan. 18 | $^{52}$Mn, $^{56}$Co | 0.02±0.87 0.016±0.007 |
| H     | Quadrupole magnet (out) | 0.39 0.87 Jan. 17 0.87 Jan. 18 | $^{52}$Mn, $^{56}$Co | 0.08±1.09 0.016±0.007 |
| I     | Flexible joint | 0.87 1.09 Jan. 17 1.09 Jan. 18        | $^{52}$Mn, $^{56}$Co, $^{57}$Co | 0.558±0.041 0.0031±0.0024 |
| J     | Bending magnet (in) | 0.18 0.15 Jan. 17 0.15 Jan. 18 | $^{56}$Co         | 0.01±0.18 0.0031±0.0024 |
| K     | Beam profile monitor | 0.65 11.3 Jan. 17 11.3 Jan. 18 | $^{95m}$Tc, $^{96}$Tc, $^{99m}$Tc | 0.03±0.012 0.0050±0.0057 |
| L     | Flexible joint | 9.92 29.9 Jan. 17 29.9 Jan. 18        | $^{52}$Mn, $^{56}$Co, $^{57}$Co, $^{60}$Cu*1 | 5.83±0.42 0.107±0.015 |
| M     | Quadrupole magnet (in) | 0.78 2.43 Jan. 17 2.43 Jan. 18 | $^{56}$Co         | 0.10±0.011 0.0039±0.0013 |
| N     | Quadrupole magnet (out) | 0.49 1.65 Jan. 17 1.65 Jan. 18 | $^{52}$Mn, $^{56}$Co | 0.09±0.49 0.016±0.007 |
| O     | Switching magnet (in) | 5.92 29.9 Jan. 17 29.9 Jan. 18 | $^{52}$Mn, $^{56}$Co, $^{57}$Co, $^{60}$Cu*1 | 1.52±0.12 0.72±0.06 |
| P     | Faraday cup (BG) | 4.24 0.20 Jan. 17 0.20 Jan. 18 | $^{56}$Mn*1        | 2×10⁻⁵±8×10⁻⁵ |
| Q     | Quadrupole magnet (out) (BG) | 1.75 0.20 Jan. 17 0.20 Jan. 18 | $^{56}$Mn*1        | 0.0012±0.0028 |
| R     | Beam pipe (BG)  | 0.20 0.20 Jan. 17 0.20 Jan. 18        | $^{56}$Mn*1        | 0.0012±0.0028 |
| S     | Target chamber (in) (BG) | 0.65 0.65 Jan. 17 0.65 Jan. 18 | $^{52m}$Mn*1, $^{56}$Mn*1 | 0.06±0.05 0.0012±0.0028 |
| T     | Target chamber (BG) | 0.86 0.86 Jan. 17 0.86 Jan. 18 | $^{52m}$Mn*1, $^{56}$Mn*1 | -0.018±0.012 |
| U     | Target chamber (out) | 0.61 0.61 Jan. 17 0.61 Jan. 18 | $^{52m}$Mn*1, $^{56}$Mn*1 | -0.018±0.012 |
| V     | Gate valve (BG)  | 0.12 0.12 Jan. 17 0.12 Jan. 18        | $^{52m}$Mn*1, $^{56}$Mn*1 | 0.06±0.05 0.0012±0.0028 |
| SUS target | BG | 52Mn*1, 56Co*1 | 100 |

*1. Detected after proton beam irradiation to the SUS and Ta target (20:30 Jan. 18)
value was adopted for each energy—for 12 MeV: 0.44 b, 6 MeV: 0.05 b, and 3 MeV: 0 b. “λ” is the decay constant, “t” is the elapsed time from the end of the irradiation to 20:30 Jan. 18th. We found that the residual activity of 52Mn on the beamline on Jan. 18th was attributed only to the experiment on Jan. 16th, whereas that of 56Co was attributed to multiple operations. To simplify the analysis, the activity of 52Mn at each point was calculated, and the beam loss at the point was estimated.

The relative beam loss of principal points is summarized in Table 2. We assumed that 100% beam loss occurred in the SUS target. This activity generation rate was employed as a calculation standard. The activity generation rate of 52Mn is proportional to the beam loss rate. The detection efficiency of γ-rays is calculated using ISOCS13,14), assuming a disk-shaped SUS plate having a diameter of 10 mm and a thickness of 1 mm. The saturation factor for 52Mn was corrected based on the operation history. As expected, experiments performed on Jan. 18th did not contribute to the activation of the beamline, and it was revealed that the experiment on Jan. 16th significantly influenced the beamline activation. The results corresponded closely to the beam loss values obtained by Faraday cups at principal points on the beamline. The relative beam current losses between “A”–“D”, “D”–“K”, and “K”–“P” were 52%, 2%, and 2%, respectively. This indicates that the most recent beam loss can be estimated accurately by quantifying the 52Mn activity on the beamline.

3-5. Reproduction of induced activity from contact dose-rate

Finally, the correlation between the contact dose rate and activity was discussed. Assuming a Φ10 mm × t1 mm SUS disk, the present activities of 52Mn and 56Co were estimated from the spectrum measured with the LA detector and are represented in Fig. 4 as a hollow bar and a shaded bar, respectively. Furthermore, the correlations between 52Mn and 56Co for each activated point were deduced, and they were very good (r = 0.997). For all activated points, it was revealed that 56Co was the dominant nuclide.

The dose rate value denoted on a survey meter, “E”, can be expressed as equation (2), by employing an activity of existence radionuclides, “A”, and 1 cm dose equivalent rate constant, “T”15).
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Here, parameters “F”, “d” are the factor with respect to the shielding condition, and the distance from the radiation source, respectively. As, it is difficult to determine precise values of these parameters at actual activated points, we replaced the equation (2) as below.

\[ E = C \cdot \sum (\Gamma \cdot A) \quad (3) \]

The 1 cm dose equivalent rate constant for \(^{52}\text{Mn}\) (0.511) and \(^{56}\text{Co}\) (0.492)\(^{16}\), were employed, and the constant, “C”, was determined by the least square fitting. The reproduced contact dose rate of each activated point by equation (3) is depicted as a solid line in Fig. 4. These are similar to the measured values, which are represented by open circles in Fig. 4, at all points except for “K” and “O”. The deviation at “K” was due to the existence of the radioisotopes of Tc. In contrast, the deviation at “O” was caused by differences in the probe sizes of the NaI survey meter and LaBr\(_3\) detector, as the probe of the LaBr\(_3\) detector was too big to approach the activated point where in a narrow slit. In conclusion, by measuring the contact dose rate on the beamline, the activity of principal nuclides and beam loss can be reproduced.

4. Conclusion

We proposed a method to investigate beamline activation as a case study at the 6-MV tandem-type accelerator facility. The contact dose rate on an entire beamline was scanned to determine an activated area, and the generated nuclides were identified by \(\gamma\)-ray spectrometry using LaBr\(_3\) and Ge detectors. The activated area of the beamline was limited, and generated nuclides were also limited for \(^{52}\text{Mn}\) and \(^{56}\text{Co}\). We also discussed a method to evaluate beam loss from the activity of \(^{52}\text{Mn}\), which reflects the recent operation history. A correlation between the contact dose rate at a certain point on the beamline and the activity at that point was clarified, and the activity of principal nuclides and beam loss are expected to be estimated effectively. We are planning to publish a manual for a reasonable and effective decommissioning of an accelerator facility based on a series of our studies, including this one.
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