Measurement of induced radioactivity in air and water for medical accelerators

K. Masumoto[1], K. Takahashi[1], H. Nakamura[1], A. Toyoda[1], K. Iijima[1], K. Kosako[2], K. Oishi[2], F. Nobuhara
[1]High Energy Accelerator Research Organization (KEK), Japan,
[2]Shimizu Co. Japan,
[3]Tokyo Nuclear Service Co. Japan

Abstract

Activation of air and water has been evaluated at the 10 and 15 MeV linear electron accelerator facilities. The air present in a glovebox and the *water present in* the *phantom were directly irradiated in front of the beam exit window. The typical irradiation condition was 50 Gy at the isocenter. No activity could be observed at 10 MeV irradiation. At 15 MeV irradiation, the activity of 10-min-half-life ^{13}N was observed in the case of the air in the glovebox, but no activity could be observed in the air sampled from the irradiation room and the maze. Aerosol was also sampled in the irradiation room and the maze by* the *dust sampler during irradiation and the activity deposited on* the *high-efficiency particulate air (HEPA) filter was measured using a GM-survey meter. Activity of ^{13}N was observed on the filters for 15 MeV irradiation. Air and water samples were also bombarded by 250 MeV protons and 400 MeV/u carbon, and the irradiation dose was 10 Gy at the isocenter. Upon the ion-chamber monitoring of the air sampled from the glovebox, ^{15}O, ^{13}N, and ^{11}C activities were mainly observed. Upon the dust sampling of the irradiation room and the maze,* low *activity of ^{13}N was* observed *on the HEPA filter. At the end of proton and carbon irradiation, the activity of the water was found to be about 10 kBq•cm^{-3} and several kilobecquerel per cubic centimetre, respectively. From the decay analysis of the induced activity in water, ^{15}O, ^{13}N, and ^{11}C were detected. The obtained results of air and water activation were then compared with data calculated by MCNP or PHITS code, coupled with DCHAIN-SP2001. At the 15 MeV linear accelerator, the observed activity of ^{13}N was higher than the calculated activity. A similar trend was observed in the case of proton and carbon irradiation. The reason why the calculated activity was lower than the experimental results needs to be investigated.*

Introduction

Accelerators have been widely used in medical applications, especially in cancer treatment. Electron linear accelerators have been used in hospitals for the X-ray irradiation of diseased organs. Particle accelerators have also been used in recent times as a powerful tool for cancer therapy. New regulations for disposing of activated-material components from accelerator facilities were established in Japan in 2012. Activation air and water should also be controlled in the same manner as the facility using non-shielded radioisotopes. In this work, we investigated the activation of air and water in treatment rooms in hospitals for radiation safety management. In the case of photonuclear reaction, several positron emitting radioisotopes such as ^{15}O and ^{13}N can be produced by the bremsstrahlung mechanism at higher than 15 MeV in air. In the case of hadron irradiation, many radioisotopes might be produced by high-energy particles. In this work, we measured the induced activity in air and water caused by the electron and hadron accelerators employed for medical use, in order to determine whether the concentration of radioactivity was below regulated levels.

Experimental

Activation of air and water

Electron accelerator

10 and 15 MeV irradiations for 10 min were performed by Varian Clinac 2 100C and Clinac iX, respectively. Doses were 40 and 60 Gy at the isocenter, respectively.

Proton accelerator

200 MeV proton beam was irradiated for 3.25 min on an area of 0.15 m×0.15 m, and 10 Gy at the isocenter.

Heavy ion accelerator

Carbon beams of 400 MeV/nucleon were irradiated for 2 min on an area of 0.15 m×0.15 m, and 10 Gy at the isocenter.

Measurement of radioactivity in air

Activity in air

A glovebox (0.5×0.5×1.0 m^3) covered with a polyethylene sheet was placed in front of the beam exit window. After irradiation, air in the glove box was collected in a vacuum ionisation chamber (volume 1.5 L). Air samples were also collected from near the glove box in the irradiation room and the maze to the irradiation room. Ionisation current of the ionisation chamber was monitored using a vibrating reed electrometer (Advantest TR8411 and TR8401). An ionisation current of 1.0×10^{-14} A corresponds to the beta activity of 0.063 Bq•cm^{-3}.

Activity in aerosol

Figure 1: Irradiation set-up for activation of air for the electron accelerator (left) and air and water for the proton accelerator (right)

A portable air sampler with a set of HEPA filter (HE-40T) (collection efficiency: 99.6%, particle size 0.09 to 0.8 μm) and charcoal filter (CP-20) was used for aerosol sampling in the irradiation room and the maze to the irradiation room during irradiation. Radioactivity collected on the filter was measured using a GM survey meter (window size: 50 mmϕ, TGS-146, Aloka Co.).

Measurement of radioactivity in water

In the case of the electron accelerator, water filled with a water phantom for dose calibration was set in front of the beam exit window and irradiated. After irradiation, the dose rate of the water surface was monitored using a NaI(Tl) survey meter (1.0 ϕ ×1.0 in, TCS-171, Aloka) and the gamma-ray spectrum was measured using a $LaBr_3$(Ce) scintillation spectrometer (1.5 ϕ×1.5 in, Canberra).

In the case of the hadron accelerator, two polyethylene tanks (volume: 20L, thickness: 19cm) were set after the glove box in order to stop the beam perfectly. After irradiation, the surface dose rate of the tank was measured using a $LaBr_3$ (Ce) scintillation spectrometer (1.5 ϕ ×1.5 in, Canberra) and the activity in becquerel per cubic centimetre was calculated. The conversion factor between the dose rate and the activity of water was obtained in advance.

The activity of the water sample was also measured using a Ge-detector (Canberra GR-2018) for detecting gamma-emitting nuclides and a liquid scintillation counter (PerkinElmer 3110TR) for beta-emitting nuclides.

Monte Carlo calculation procedure for determining level of activation of air and water

In the case of the electron accelerator, electron, photon, and neutron distribution were calculated by MCNP5. A plan of irradiation layout for calculation was drawn based on the thickness and density of each material used for the gantry. After calculating the gamma-ray and neutron spectrum using MCNP5, induced activities in the air and water

were calculated using DCHAIN-SP2001. In the case of the particle accelerator, the transportations of incident particles and neutrons were calculated using PHITS, and the activation was calculated using DCHAIN-SP2001.

Results

Results obtained using the electron accelerator

10 MeV irradiation

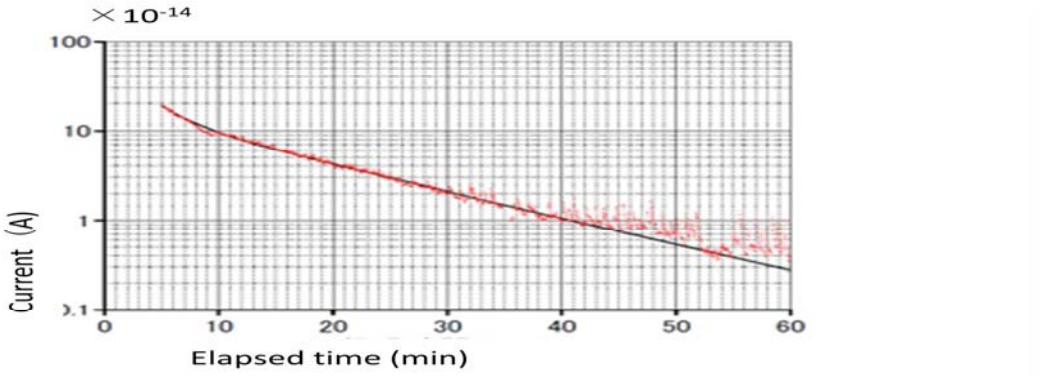
The ionisation current of air sampled from the glovebox was slightly noisy. However, the ionisation current of air sampled from the irradiation room and the maze to the irradiation room was the same as the background level. No activity was detected in the air filters sampled from the irradiation room and the maze to the irradiation room.

In the case of water, the dose rate of the water sample after irradiation was equal to the background level and no activity was detected by the measurement using a Ge-detector and a liquid scintillation counter. Cooling water was also sampled from the accelerator, and its activity was measured using a Ge-detector and a liquid scintillation counter. However, no activity was detected in the cooling water.

15 MeV irradiation

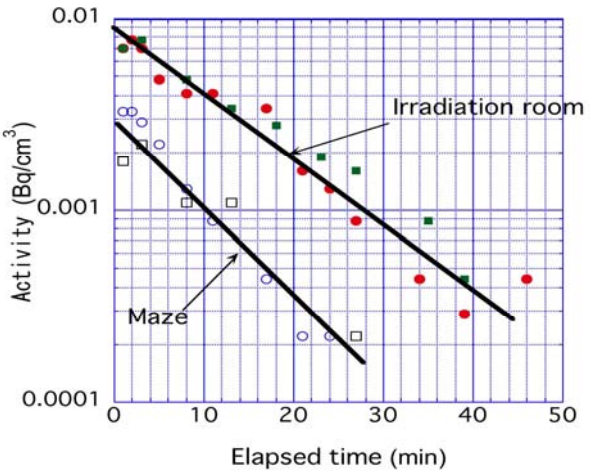
As a result of the decay analysis of the ionisation current, which is shown in Figure 2, a radioisotope with its half-life of 10 min was only observed in an air sample obtained from the glove box. It seemed to be ^{13}N, which is produced by the (γ, n) reaction of nitrogen in air. Its activity was estimated as 0.87 Bq•cm^{-3} at the end of the irradiation. This value is about a hundred times larger than the calculated value (0.009 Bq•cm^{-3}) obtained by the activation calculation using MCNP5 coupled with DCHAIN-SP2001. The ionisation current of the air sampled from the irradiation room and the maze to the irradiation room was the same as the background level.

Figure 2: Ionisation current of air irradiated by 15 MeV bremsstrahlung



Activities of the filters collected from the irradiation room and the maze to the irradiation room were also measured, and the half-life of the radioactive substance detected was about 10 min in the irradiation room, as shown in Figure 3. Activity concentration was 0.009 and 0.003 Bq•cm^{-3} at the end of irradiation in the irradiation room and the maze. An aerosol of radioactive nitrogen oxide (NO_x) may have been produced by the radiation effect.

Figure 3: Radioactivity concentration calculated by the activity collected on a HEPA filter



The dose rate of irradiated water, obtained using a NaI (Tl) survey meter, was found to be two times higher than the background level (0.08 μSv/h). A very small annihilation peak was observed using a $LaBr_3$(Ce) spectrometer. It was concluded that the structure material of water phantom was slightly activated.

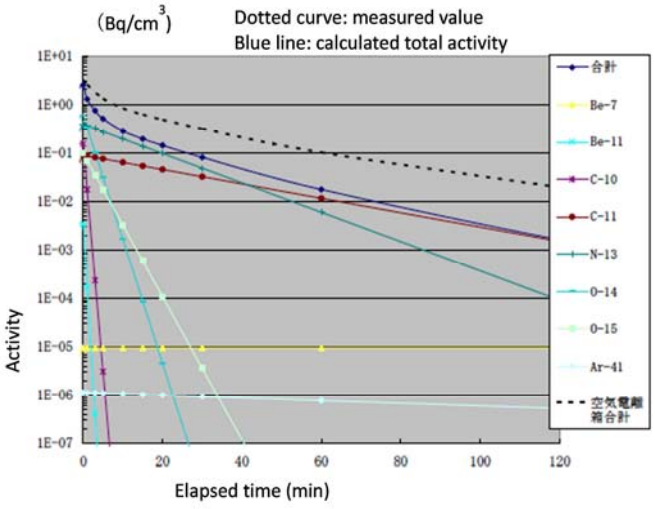
Results obtained using proton accelerator

Activation of air

The results of the measurement and calculation of the activity concentration of air are shown in Figure 4. Fitting the decay curve of the ionisation current observed in the air sample obtained from the glove box revealed that the following activities of ^{15}O, ^{13}N, and ^{11}C were 1.9, 0.6, and 0.6 Bq•cm^{-3} at the end of irradiation, respectively. On the other hand, the calculated activities of ^{14}O, ^{15}O, ^{13}N, and ^{11}C obtained using PHITS code coupled with DCHAIN-SP2001 were 0.6, 0.7, 0.4, and 0.09 Bq•cm^{-3}, respectively. The difference between the experimental and calculated values at the end of irradiation was small. However, the difference gradually became large because of the difference between the experimental and calculated ^{11}C activity. The ionisation current of the air sampled from the irradiation room and the maze to the irradiation room was equal to the background level.

The activities of the filters collected from the irradiation room and the maze to the irradiation room were also measured, and a half-life of these activities was about 10 min. The activity concentration was 0.05 at and 0.01 Bq•cm^{-3} at the end of irradiation in the irradiation room and the maze, respectively.

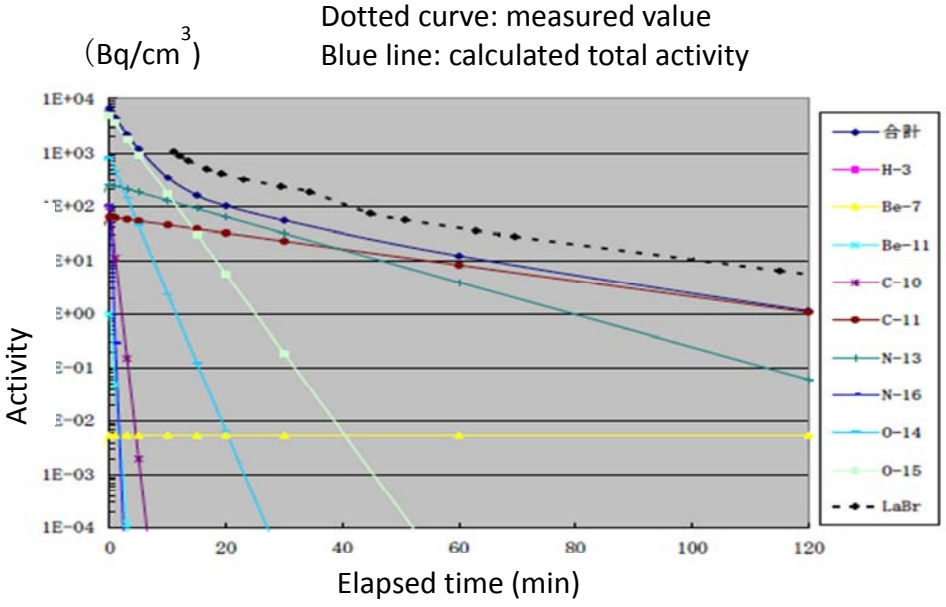
Figure 4: Decay curves and calculated values of radioactivity in air irradiated by 250 MeV protons



Activation of water

The results from the measurement and calculation of the activity concentration of water are shown in Figure 5. The decay curve analysis of the activity obtained from the surface dose rate of the polyethylene tank showed that the activities of ^{14}O, ^{15}O, ^{13}N, and ^{11}C were 91 000, 12 000, 1 100, and 190 Bq•cm^{-3} at the end of irradiation, respectively.

Figure 5: Decay curves and calculated values of radioactivity in water irradiated by 250 MeV protons



The activities of ^{14}O, ^{15}O, ^{13}N, and ^{11}C obtained using the PHITS code coupled with DCHAIN-SP2001 were 830, 5000, 250, and 63 Bq•cm^{-3}, respectively. The activity of ^{7}Be was 0.039 Bq•cm^{-3}, as determined using a Ge-detector, and it was calculated to be 0.0053 Bq•cm^{-3}. It was found that the calculated activity of each radioisotope was always smaller than its experimental result. No activity was detected using the liquid scintillation counter.

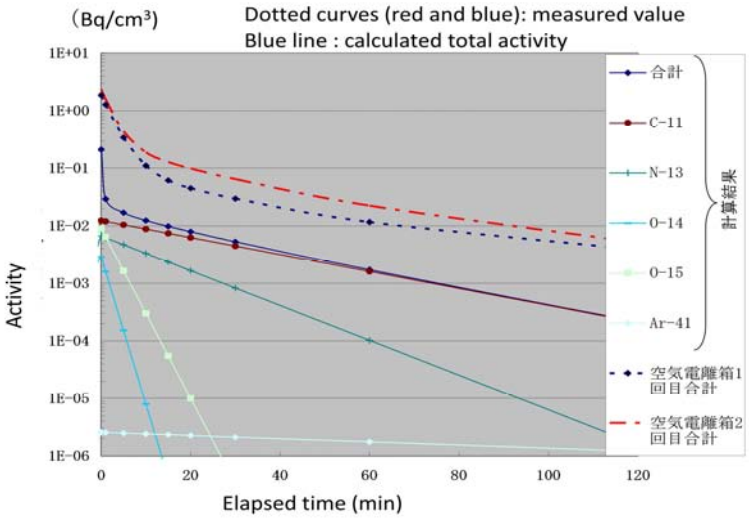
Results obtained from the heavy ion accelerator

Activation of air

The results of the measurement and calculation of activity concentration of air are shown in Figure 6.

We performed irradiation twice and fitted the decay curve for ^{14}O, ^{15}O, ^{13}N, ^{11}C, and ^{41}Ar. Average activities of ^{14}O, ^{15}O, ^{13}N, ^{11}C, and ^{41}Ar were, respectively, 0.8, 1.1, 0.08, 0.09, and 0.006 Bq•cm^{-3} at the end of irradiation. The activities of ^{14}O, ^{15}O, ^{13}N, and ^{11}C obtained using the PHITS code coupled with DCHAIN-SP2001 were 0.003, 0.009, 0.007, 0.012, and 0.000006 Bq•cm^{-3}, respectively. Determined activity levels were lower than those due to proton irradiation, and the activity was mainly due to ^{11}C. The calculated activity of each radioisotope was smaller than the experimental value.

Figure 6: Decay curves and calculated values of radioactivity in air irradiated by 400 MeV/u carbon

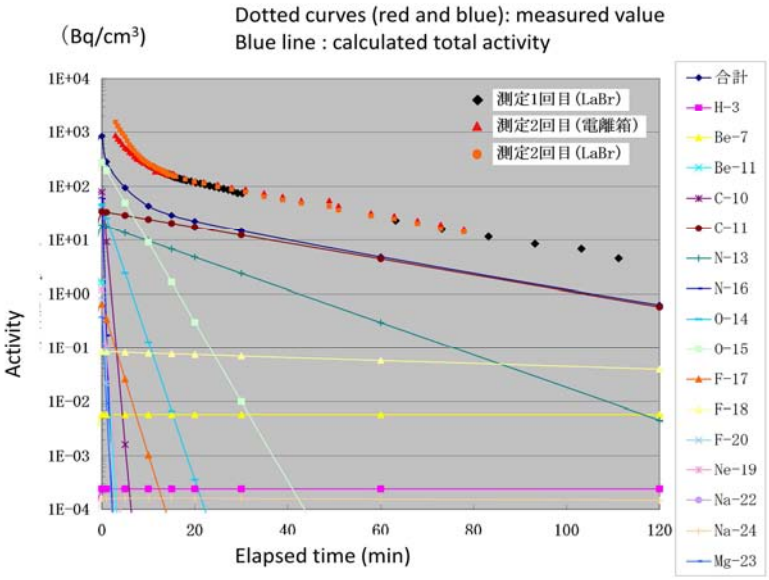


The activities of the filters collected from the irradiation room and the maze to the irradiation room were also detected and the half-life of these activities was 10 min. The activity concentration was lower than that of proton irradiation. At the end of irradiation, the activity concentration was about 0.02 Bq•cm^{-3} in the irradiation room.

Activation of water

The results of the measurement and calculation of activity concentration of water are shown in Figure 7. The decay curve analysis of the activity obtained from the surface dose rate of the polyethylene tank showed that the activities of ^{14}O, ^{15}O, ^{13}N, and ^{11}C were 2 100, 2 000, 100 and 180 Bq•cm^{-3} at the end of irradiation, respectively. The activities of ^{14}O, ^{15}O, ^{13}N, and ^{11}C obtained using the PHITS code coupled with DCHAIN-SP2001 were 46, 270, 19, and 34 Bq•cm^{-3}, respectively. In the case of carbon irradiation, the induced activity was lower than that of proton irradiation and ^{11}C became a major contributor to the activity after 20 min of the end of the irradiation. The activity of ^{7}Be was 0.39 Bq•cm^{-3} using a Ge-detector, and the calculated value was found to be 0.0058 Bq•cm^{-3}. The activity of ^{22}Na was also detected in water, and its concentration was 0.0004 Bq•cm^{-3}, which was only two times larger than the detection limit of our measurement (0.0002 Bq•cm^{-3}). Tritium concentration was found to be 0.28 Bq•cm^{-3} using liquid scintillation counting.

Figure 7: Decay curves and calculated values of radioactivity in air irradiated by 400 MeV/u carbon



Conclusion

In the case of 10 MeV irradiation residual activity could not be detected in air and water. In the case of 15 MeV irradiation, we observed ^{13}N in the air sampled from the glovebox irradiated in front of the beam exit window and on the filter collected by the air samplers in the irradiation room and the maze. The measured activity concentration of ^{13}N was very small, but it was higher than the value calculated by MCNP5 coupled with DCHAIN-SP2001. It is necessary to verify the irradiation energy for activation and the photonuclear reaction cross-section for the calculation.

In the case of 250 MeV proton activation and 400 MeV/u carbon irradiation, the activation of air was not excessively high and the induced activity was comparable with that induced by 15 MeV irradiation for the electron accelerator. The activity concentration of ^{11}C was seven times higher than the value calculated by PHITS coupled with DCHAIN-SP2001.

The induced activity of water was extremely high. The main radionuclides detected were ^{13}N and ^{11}C, 10 min after irradiation with protons and ^{11}C, 20 min after irradiation with carbon. The activity concentrations of water irradiated by protons and carbon were also higher than the value calculated by PHITS coupled with DCHAIN-SP2001. In these cases, activation cross-sections of light elements induced by the high-energy hadron or reaction models for activation calculation need to be checked.