

Residual Radio-activity of the Kyoto University Cyclotron

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Received January 9, 1974

The residual radioactivity of the Kyoto University Cyclotron was surveyed at about one year after its shutdown and before its reconstruction. The radioactive nuclides remained in the electrodes of the cyclotron and in the dust were also assigned.

I. INTRODUCTION

The Kyoto University Cyclotron accelerated mostly protons and α -particles, and less frequently deuterons for more than ten years. The cyclotron was operated severely throughout the period, though it had some troubles in later years. The induced radioactivities in every part of the cyclotron were still remained to some extent after one year's cooling time following the accidental breakdown in December 1969 due to the failure of the cooling pipe of the deflector electrode. At the time of dismantling the cyclotron, the radioactivities around the machine had to be surveyed to insure safety of the workmen who were engaged in the reconstruction.

II. MEASUREMENT OF RESIDUAL RADIOACTIVITIES AROUND THE CYCLOTRON

1) Dose Rate of the Induced Radioactivity at the Surface of the Pole and the Coil Tank

After the acceleration chamber was removed, some residual activities were found in the pole and in the tank of exciting coil of the magnet. This activity must be induced by the fast neutrons generated during acceleration of the particles.

The dose rate was measured using a TEN SM-102 survey-meter with a GM tube for γ -ray detection after about sixteen months after the breakdown of the cyclotron. The distribution of the dose rate at the surface of the pole and the tank of the coil which excites the magnet is shown in Fig. 1. The activities amount to several mR/hr and show ten-fold difference from place to place. The most intense one was found at the place just outside the front edge of the deflector electrode.

The distribution of the radioactivities in two directions along median plane between the two poles is shown in Fig. 2. The activities in both directions are roughly

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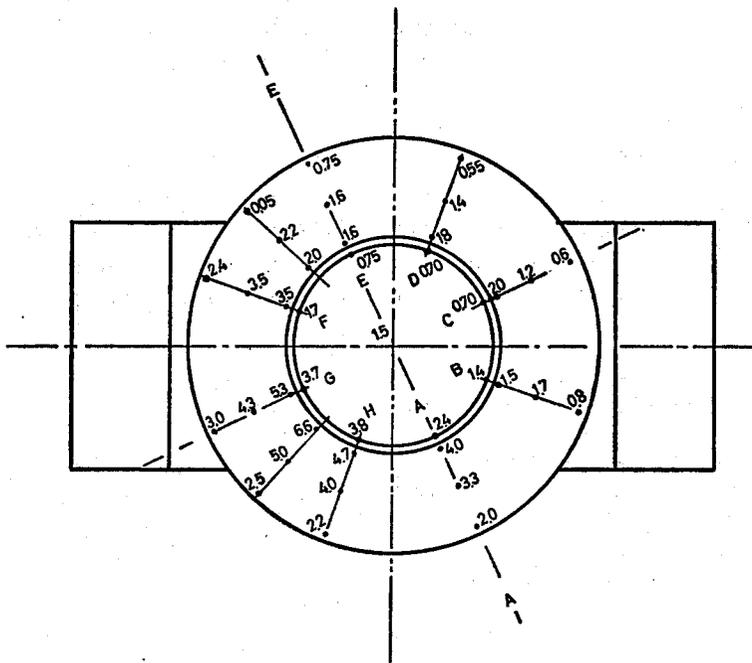


Fig. 1. Dose rate at the surface of the pole and of the coil tank on the 21 st of April 1971. The numbers denote the dose rate in mR/hr unit.

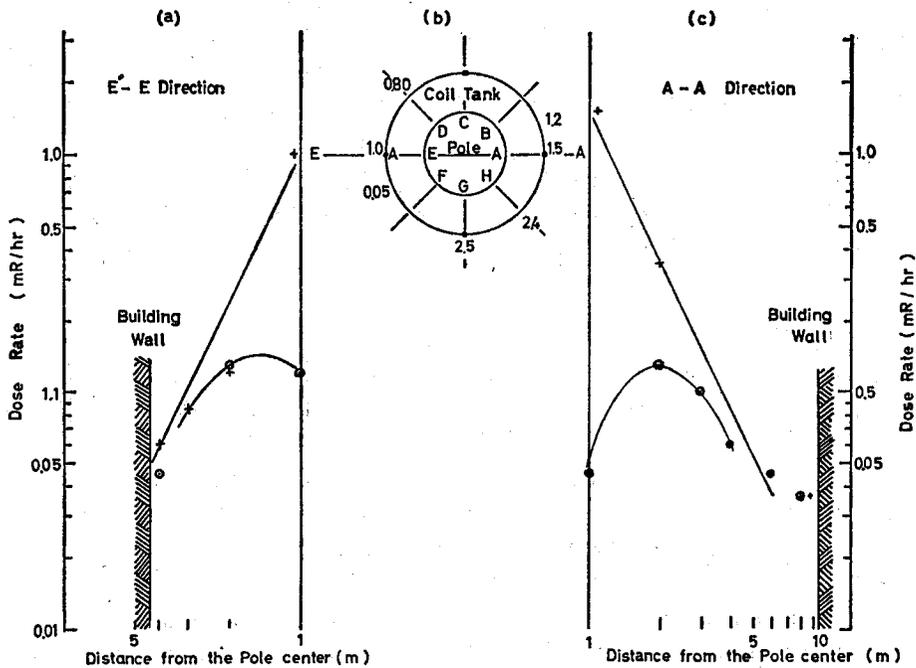


Fig. 2. Dose rate along the median plane between the two poles on the 13 th of March 1971. The numbers denote the dose rate in mR/hr.

inversely proportional to the square of the distances from the center of the pole. In the same figure are also shown the activities around the side wall of the coil tank which has the radius of 1.087 m.

2) Assignment of the Radio-Nuclides

The activity was measured with a 50 cm³ Ge(Li) detector and a 4 k P.H.A. on the 13th of March 1971. The detector was set in two directions between the two coil tanks; the one was in the direction between *G* and *H* shown in Fig. 1 (position I) and the other is in the opposite direction (position II). Both spectra show only γ -ray peaks of ⁵⁴Mn and ⁶⁰Co. The relative intensity of the two nuclides in the two directions is shown in Table I.

Table I. Activity Ratio of the Radio-Nuclides in the Pole and the Coil Tank.

| Nuclide | Position I | Position II |
|------------------|------------|-------------|
| ⁶⁰ Co | 1.0 | 0.361 |
| ⁵⁴ Mn | 0.164 | 0.125 |

⁵⁴Mn is produced by the reaction ⁵⁴Fe(n, p)⁵⁴Mn ($Q=0.0890$ MeV) and ⁶⁰Co is by the reaction ⁶⁰Ni(n, p)⁶⁰Co ($Q=-2.0370$ MeV) and ⁵⁹Co(n, γ)⁶⁰Co ($Q=7.4900$ MeV) in the coil tank made of stainless steel. ⁵⁴Mn is also produced by the ⁵⁴Fe(n, p)⁵⁴Mn reaction in the pole and the yoke of the magnet. ⁶⁰Co is also produced by the reaction ⁶³Cu(n, α)⁶⁰Co ($Q=1.7149$ MeV) in the copper wire of the coil. The fluctuation of the relative activities of the two nuclides from place to place must be due to the difference of neutron intensities and its energy dependence.

III. RESIDUAL RADIOACTIVITY IN THE ELECTRODES

The electrodes in the acceleration chamber suffered from the damage brought about by the intense bombardment of the accelerated particles and retained in them most of the radioactivities which were produced as a result of nuclear reactions induced by them.

1) The Septum Electrode

The front part of the septum electrode was made of a tungsten metal strip of 32 mm \times 200 mm \times 0.7 mm and was slid into the frame of the main electrode. The gap distance between the electrode and the deflector electrode was set to 0.6 mm.

The front edge of the tungsten strip was melted and completely disappeared leaving a wedge-shaped deficit of about 6 cm long as shown in the lower part of Fig. 3. In the rear part of the electrode there was a hole of 3 mm \times 10 mm as a result of the bombardment of the accelerated ions.

The surface opposite to the deflector electrode was covered by a metallic copper film which was sputtered from the latter. The other surface of the electrode was roughened but had a metallic luster.

A small piece of the electrode fragment of about 500 mg by weight had a radio-

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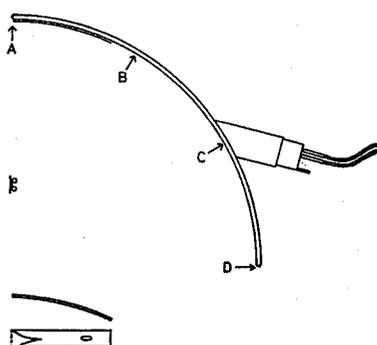


Fig. 3. The deflector electrode and the septum electrode.

activity of several microcuries. Relative intensity of the nuclides retained in it is shown in Table II together with others. All of them are produced by the nuclear reactions such as (p, xn), (α , xn), and (d, xn) reactions (in this case $x=0\sim 2$), with tungsten isotopes of mass number ranging from 182 to 184.

Table II. Relative Intensity of the Radio-nuclides Smearred out from Several Parts of the Deflector Electrode. (Alphabetical Sample Name Corresponds to the Part Shown in Fig. 3.) Measured on the 13th of November 1970.

| Sample name | Relative intensity (μC) | | | | | | |
|-------------------------|--------------------------------------|------------------|------------------|---------------------|-------------------|---------------------|-------------------|
| | ^{22}Na | ^{60}Co | ^{65}Zn | $^{110}\text{Ag}^m$ | ^{183}Re | $^{184}\text{Re}^m$ | ^{185}Os |
| A silver solder | | 2.80 (-1) | 8.14 (1) | 1.05 | | | |
| copper | | 1.41 (-2) | 4.86 | 6.30 (-3) | | | |
| black scale | | 1.0 (-2) | 1.11 (1) | 1.99 (-2) | | | |
| smearred sample | | 6.97 (-3) | 4.22 | 8.81 (-3) | | | |
| B smearred sample | 3.10 (-3) | 1.31 (-3) | 1.56 | 1.23 (-3) | 1.69 (-1) | 2.63 (-1) | 1.13 (-1) |
| C smearred sample | 3.15 (-4) | 5.90 (-4) | 8.40 (-2) | | | | |
| D smearred sample | | | 2.63 (-2) | | | | |
| Small piece of tungsten | | | | | 4.30 | 5.33 | 7.83 (-1) |

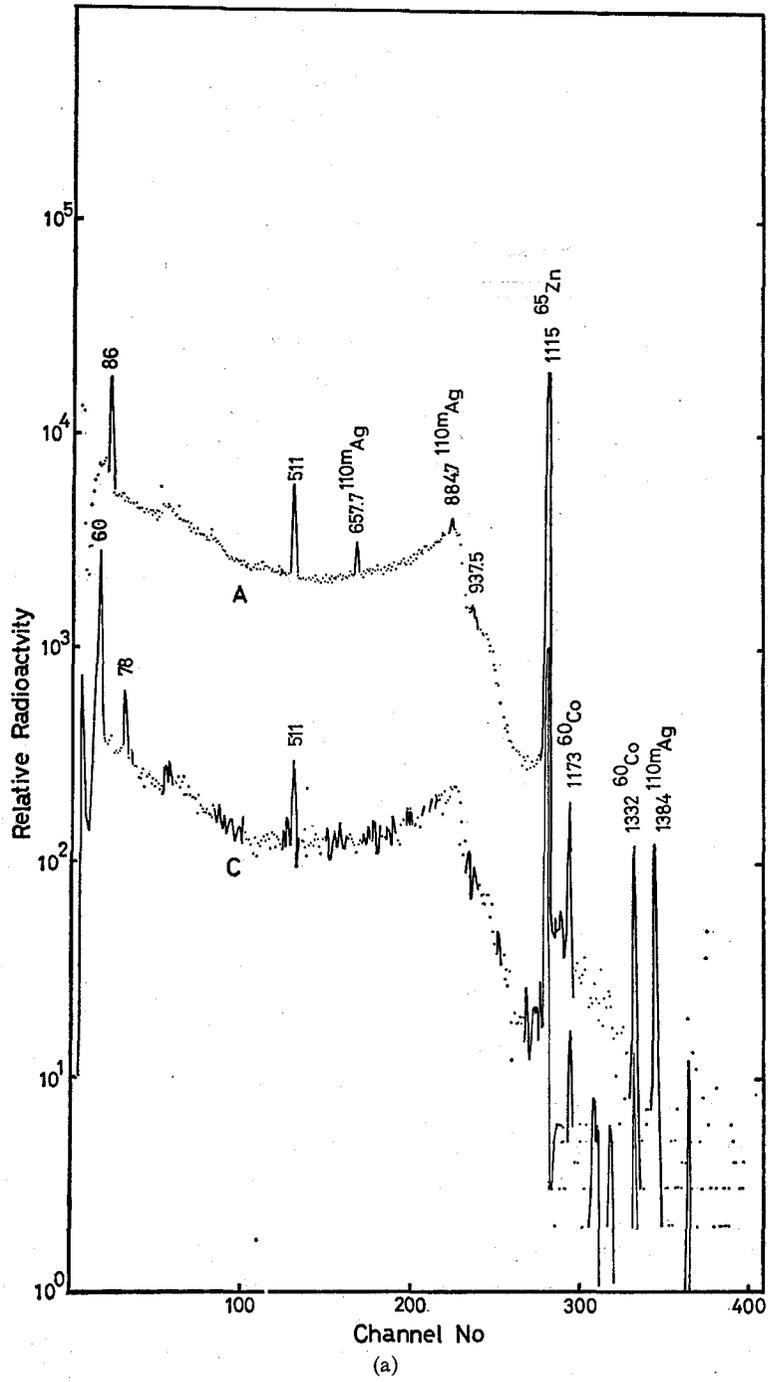
2) The Deflector Electrode

The deflector electrode which was originally made of a copper strip of 25 mm \times 1250 mm \times 1 mm and a cooling coil of 8 mm outside diameter silver soldered to the circumference of the former.

The electrode suffered from the intense bombardment of the accelerated ions, and at last, at the end of 1969, a small pin hole was generated at the front end of the cooling pipe.

The surface of the deflector electrode opposite to the septum electrode was covered with a thick black rust.

The γ -ray spectrum of small pieces of shavings and of black powder smearred out from several parts of the electrode is shown in Figs. 4(a) and (b), and the activities of the assigned nuclides in them are also shown in Table II.



(a)

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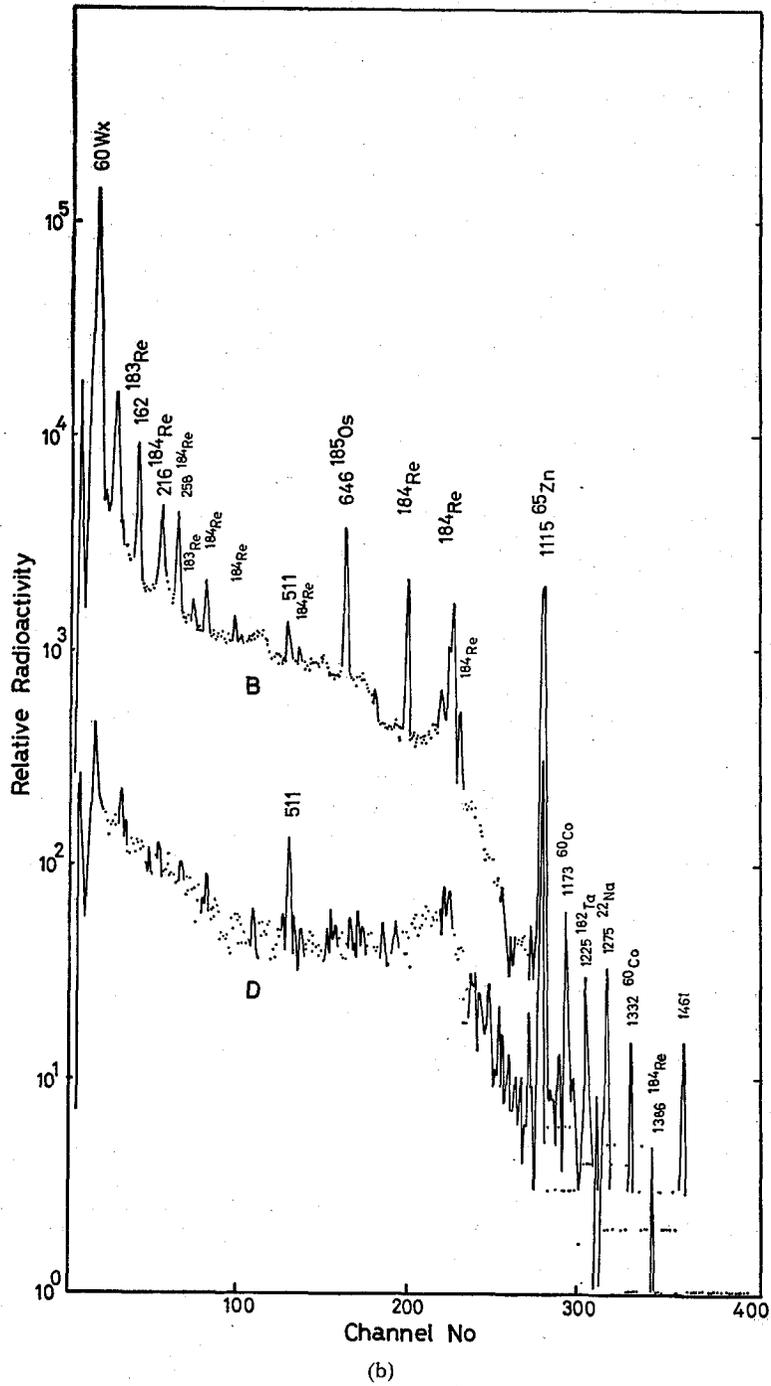


Fig. 4. γ -ray spectrum of the sample smeared out from several portions of the deflector electrode.

Though the activities are not quantitative as the weight of the smeared sample is not known, the relative activities of the nuclides from the different parts of the electrode differ appreciably.

At the front edge of the electrode, the activities are originated from the electrode material itself, copper and silver solder.

At about 25 cm from the front edge of the electrode, activities due to the sputtered ions from the septum electrode or the recoil atoms of the reaction products in the latter are remarkable.

On the other hand, a small piece of tungsten from the septum electrode one side of which is covered with copper, has only the activities due to tungsten itself and no Zn activity. Furthermore, activity of ^{65}Zn is found all over the inner surface of the acceleration chamber. As easily supposed, zinc must have a large diffusion velocity in copper metal, and evaporate out from the surface of the hot electrode.

IV. RESIDUAL ACTIVITY OF THE DUST IN THE ACCELERATION CHAMBER

There are fine black dusts in the acceleration chamber. Some of them are carbon soots produced from oil vapour from the diffusion pump, small fragments of the quartz insulators, and aggregates of sputtered ions.

Radioactivities of the nuclides found in a small portion of the gross soots are shown in Table III.

Table III. Relative Radioactivity of the Dust in the Accelerator Chamber. Measured on from the 4th to the 6th of February 1971.

| Sample | Relative activity (μC) | | | | | | | |
|---------------------------|-------------------------------------|------------------|------------------|------------------|---------------------|-------------------|---------------------|-------------------|
| | ^{22}Na | ^{60}Co | ^{65}Zn | ^{95}Nb | $^{110}\text{Ag}^m$ | ^{183}Re | $^{183}\text{Re}^m$ | ^{185}Os |
| Gross dust | 1.98(-4) | 6.46(-4) | 1.43(-1) | | | 1.71(-3) | 8.09(-3) | 2.21(-3) |
| Chemically separated dust | | | | | | | | |
| Cu-group sulfide | | | 4.72(-3) | | 1.65(-3) | 5.01(-3) | 1.06(-2) | |
| Cd-group sulfide | | | 6.68(-3) | | | 1.21(-3) | 3.25(-3) | 6.55(-4) |
| Zn fraction | | | 1.58(-1) | | | | | |
| Co fraction | | 2.23(-3) | | | | | | |
| Ta fraction | | | | 1.1(-2)* | | | | |
| R.E. fraction | | | | | | 7.83(-3) | 2.39(-2) | |
| Insoluble | | | 2.71(-2) | | | | | |

* Activity of Ta fraction was measured on another sample, and is roughly one thousand-fold intense relative to the other figures.

As there are many nuclides in the dust, chemical separation was performed to find any minute amount of radioactive component if it would be remained.

The dust was dissolved in aqua regia and separated into several groups following the method used in the usual quantitative analysis. The results are shown in Table III together with those of the gross dust. Though only a faint activity of ^{95}Nb was found, all others were those of the nuclides already found. So we conclude that there were

no appreciable radioactive nuclides except those mentioned above.

V. CONCLUSION

Dose rates around the cyclotron and the constituent nuclides were found before the dismantling of the Kyoto University Cyclotron at about one year after the shutdown of the machine. As was expected, we found only long lived nuclides, because the cooling time is so long to ensure the safety of the workmen who would be engaged in the reconstruction.

Though the radioactivities are not appreciable, most of them are due to ^{65}Zn and found all over the machine. This nuclide is supposed to be produced by the reactions such as $^{65}\text{Cu}(p, n)$, $^{63}\text{Cu}(d, \gamma)$, $^{65}\text{Cu}(d, 2n)$, and $^{63}\text{Cu}(\alpha, 2n\beta^-)$, as there were no parts made of zinc alloys which were bombarded directly by the accelerated ions. Some of it, however, would be produced in the acceleration chamber made of brass by the reaction induced by the scattered ions or the secondary fast neutrons with ^{64}Zn , and evaporated into the vacuum chamber. We are sure that any parts of the machine should not be made of brass, even though it is easy to work.