Time dependent effect of the isotope ratio of the beam from the sputter ion source

Masanobu Nakamura, Hiroshi Matsumoto, Masanori Hirose,
Department of Physics, Graduate School of Science, Kyoto University
Koya Ogino, Department of Nuclear Engineering,
Graduate School of Engineering, Kyoto University
Toshimichi Nakanishi, Department of Geophysics, Graduate School of Science, Kyoto University

Abstract

^{14}C was measured using the Kyoto University accelerator mass spectrometry system. In the measurements, a remarkable time dependent effect of the isotope ratios of $^{13}C/^{12}C$ and $^{14}C/^{12}C$ were observed. The cause is discussed.

The high sensitivity measurement of the long life isotope by accelerator mass spectrometry (AMS) began in 1970', and the measurement of many isotopes is developed from the light element such as ^{10}Be to actinide at present. As for ^{14}C which is still dominant AMS isotope, the high precision of 0.3% is achieved by many facilities. But, it is difficult to find the absolute value of the isotope ratio generally, and the ratio is being derived in comparison with the standard well-known sample. It is because it expects that the isotope ratio may change in the treatment of the sample, the ionization process in the ion source, the acceleration efficiency, and so on. Though the measurement of the standard sample and the measurement sample are done without changing a condition as much as possible, it is important to examine the factor from which the isotope ratio is changed.

The Kyoto University tandem accelerator is an 8UDH Pelletron (the maximum terminal voltage 8MV) of NEC. We have developed AMS as a tool for dating and environment measurements [1]. We started the AMS program with ^{14}C measurements because many kind of ^{14}C standard samples were available. A sputter ion source of with 59 samples made by HVEE (model 846A) was installed . The layout of the AMS system is shown in figure 1. The measurement of ^{14}C by the actual sample was started in 2001.
In the measurements, a remarkable time dependent effect of the isotope ratios of $^{13}C/^{12}C$ and $^{14}C/^{12}C$ were observed.

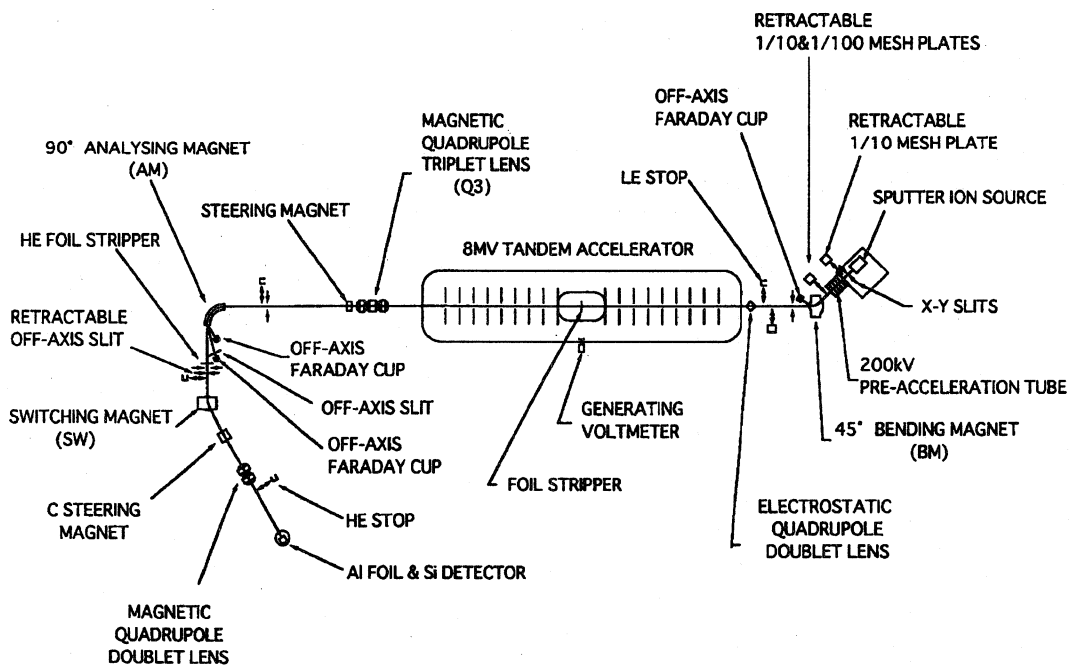


Figure 1: Layout of the Kyoto University AMS system

The example of C-6 of IAEA that it was measured for the comparison is shown in the figure 2. Though the $^{13}C/^{12}C$ ratio was saturated in about 20 minutes, the $^{14}C/^{12}C$ ratio was not saturated even after 2 hours.

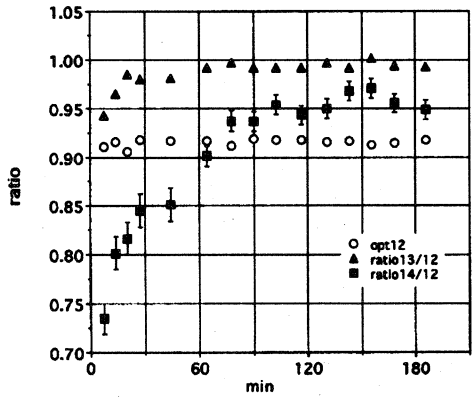
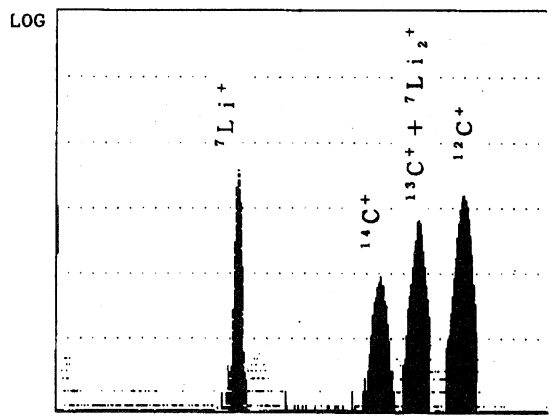


Figure 2: Variation of the ^{12}C transmission and the ratio of $^{13}C/^{12}C$ and $^{14}C/^{12}C$.

a



b

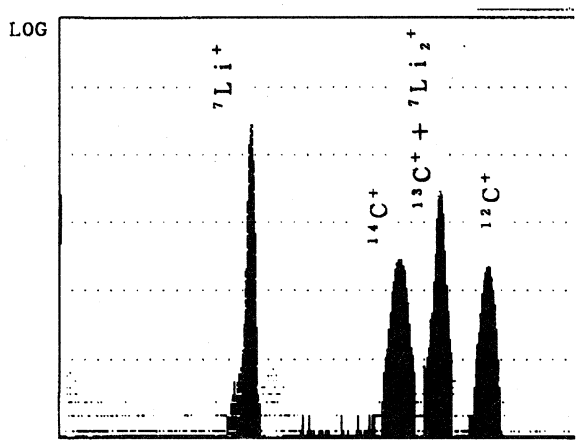


Figure 3: Energy spectra of Si detector. a:start b:end.

From the variation of the negative ion current after the sample exchange, this effect can be thought to happen with the sputter source. After the sample exchange, the sample is sputtered for a short period of time, and an influence such as pollution on the surface is removed. During the measurement it is usually moved through the position of the sample in every fixed time, and the formation of the crater is prevented. But, this observed effect is hard to think the pollution on the surface. Moreover, the crater on the surface of the sample after the measurement of two hours was shallow, and it is hard to think a cause, too

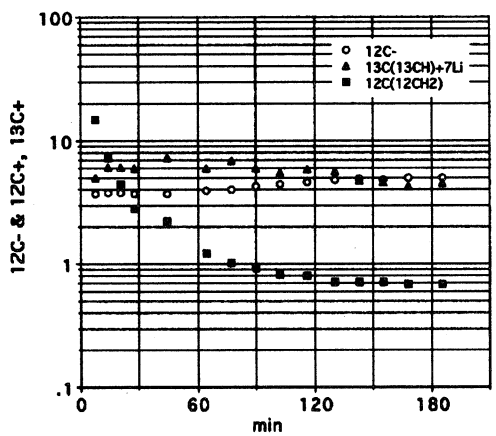


Figure 4: Variation of the $^{12}C^{-}$, $^{12}C^{+}$, $^{13}C^{+}+^{7}Li_{2}$.

The spectra of the Si detector are shown in the figure 3. The background of $^{7}Li_{2}$ is separated by using an Al foil of 6 μm with the peak of ^{14}C, and it piles up to the peak of ^{13}C. It is compared with the peak of ^{14}C from the spectra at the start and the end, and it is seen that the relative height of ^{12}C is decreased.

The variation of $^{12}C^{-}$, $^{12}C^{+}$, and $^{13}C^{+}+^{7}Li_{2}$ is shown in the figure 4.

The variation of $^{14}C/^{12}C$ and $^{12}C^{+}$ are similar time scale, and these correlation is large as shown in the figure 5.

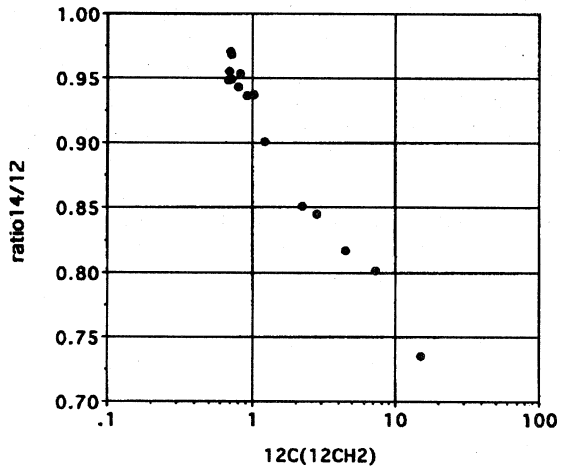


Figure 5: Relation between $^{12}C^{+}$ and the $^{14}C/^{12}C$ ratio.

When ^{14}C was injected, $^{12}CH_2$ was also injected. This molecular ion was stripped and scatted, and ^{12}C which had the momentum that was the same as ^{14}C passes through the high energy side beam analysis magnet, and it became the peak of $^{12}C^{+}$. When ^{12}C is sputtered, $^{12}C^{-}$, $^{12}C_2^{-}$, $^{12}C_3^{-}$, ..., $^{12}CH^{-}$, $^{12}CH_2^{-}$, a neutral atom and a positive ion are formed. These ratios of ions can be affected many things, for example: H_2O contained in the sample, the pressure of press to pack a sample into the cathode hole, a form on the surface, temperature of the cathode, sputter voltage.

Next, the condition which is effective in the measurement of the isotope ratio will be examined.

REFERENCES

[1] Masanobu Nakamura, Yuji Tazawa, Hiroshi Matsumoto, Masanori Hirose, Koya Ogino, Nucl. Instr. and Meth. B 172 (2000) 124.