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Imaging of cesium in water-containing plants using TOF-SIMS

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In Fukushima, Japan, the influence of radioactive Cs on agricultural crops is attracted attention as a problem. It is urgent task of elucidating dynamics of Cs in plants for securing the decontamination plan and safety of foods. Secondary ion mass spectrometry is elemental imaging technique for micro region with high sensitivity, and therefore it is suitable for the visualization of radioactive Cs in plant. However, it is difficult to analyze the original state of plants due to the evaporation of water under the vacuum condition in the apparatus. In order to investigate the behavior of radioactive Cs in plants, a new sample preparation method and imaging technique were developed. For fixing the distribution of Cs in living plants, an instrument for rapid-freezing and cutting of a plant stem in liquid nitrogen was made. In the imaging experiment, it was successfully shown that the water (ice) was maintained in the vacuum, and Cs distribution was clearly imaged over a cross-section with a high lateral resolution.

1. Introduction

By the accident in Fukushima daiichi nuclear power plant, a large amount of radioactive species was released. It is one of the serious problems that ^{137}Cs was absorbed in plants [1]. However, the mechanism of Cs absorption in plant was not clarified satisfactory. It is an urgent task of elucidating dynamics of Cs in plant for securing the decontamination plan and safety of food. Therefore, micro-scale visualization of Cs in plant is important. Radiography is widely used for visualization of radioactive Cs in plants [2]. Radiography visualizes only γ-rays. Therefore, it cannot distinguish Cs from the other species such as ^{40}K which exists in ordinary plants, and signal intensity (contrast of image) depends also on the thickness of the sample. Fluorescence microscopy and bulk analysis technique such as ICP-MS, AAS are also used for clarification of absorption mechanism, but these has large problems such as low lateral resolution and element-selectivity [3~5]. On the other hand, surface analysis technique is appropriate to the visualization of Cs with high lateral resolution. It is reported that the challenge of visualizing Cs in plant by Particle induced X-ray emission (PIXE) and scanning electron microscopy energy dispersive X-ray

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spectrometry (SEM/EDX) [6,7]. However, these techniques do not have very good detection limit (around a few % order). Moreover, since these techniques have no isotope selectivity, ^{137}Cs cannot be observed directly. In terms of both element- and isotope-selectivity, spatial resolution and detection limit, time-of-flight secondary ion mass spectrometry (TOF-SIMS) and resonant ionization laser sputtered neutral mass spectrometry (R-SNMS) are the most appropriate for the visualization of Cs in plants. In this study, a cooling system of apparatus and sample were introduced for preventing of evaporation of volatile compounds, and freeze-sectioning system was developed for visualization of Cs in plant.

It is important to fix the distribution including all kinds of liquids such as water in plants for the interior distribution imaging of Cs in plants. At the same time, smooth cross-section is needed for TOF-SIMS analysis of sample in order to prevent unanticipated contrast of image by shape effect. Therefore, we selected physical fixing method, instead of general chemical fixing method or freeze drying. In our previous work, we devised rapid cooling system and sectioning instrument under frozen condition [8)]. This system is equipped with a holder with a cap and the problem of air pollution such as frost adhesion on section has already been solved [9)]. However, can not analyze by selecting a region because there is a macro unevenness in a created cross-section. Therefore, It is difficult to elucidate dynamics of Cs in plants.

In this study, we aimed to develop the technique to select and analyze the arbitrary part of the sample surface to elucidate the dynamics of Cs in plants using TOF-SIMS.

2. Experimental

A. *Imaging apparatus and sectioning instrument under frozen condition.*

The TOF-SIMS apparatus used in this study is equipped with a gallium FIB, an electron beam (EB) for SEM observation, a TOF-MS and sample stage with 5-axes of movement [10)]. It also owns a spatial resolution of the world's best (40 nm). The sample stage can be cooled to -160 °C by circulation of liquid nitrogen.

A diagram of the sample sectioning instrument is shown in Fig. 1. In our previous study, the blade angle and movement speed were kept constant and cut in a frozen state in liquid nitrogen[8)]. This sectioning instrument sections the plant by moving the sample holder and can be adjusted the angle of the blade. It is also possible to be performed rapid cooling and cutting in under frozen.

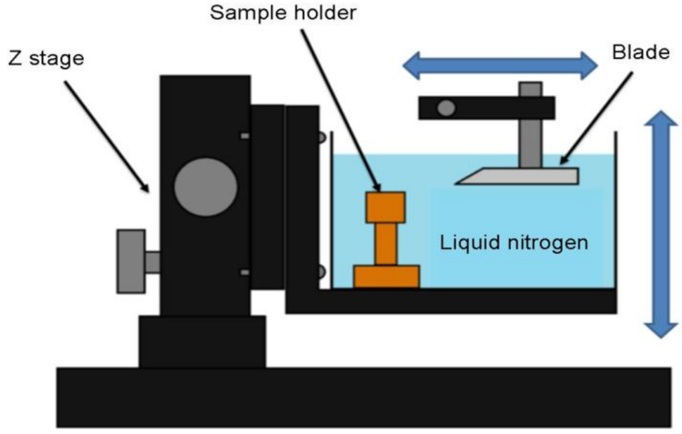


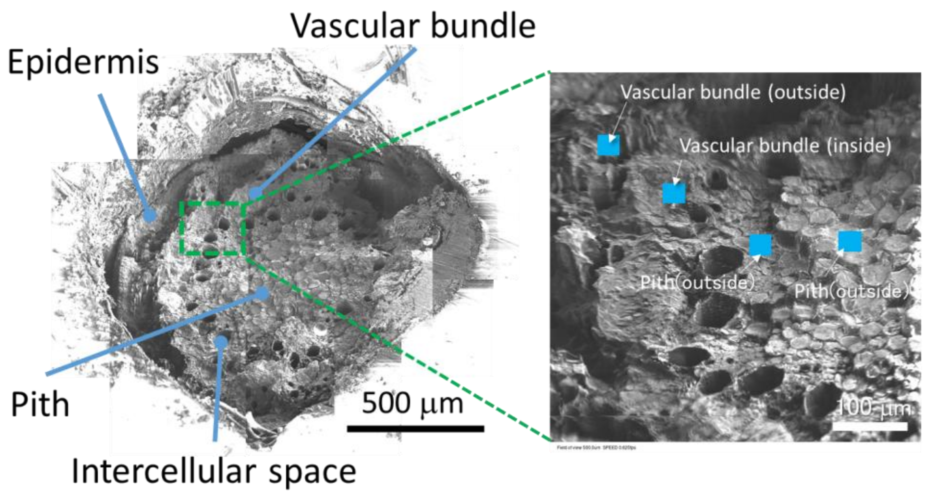
Fig.1. Sample stem sectioning instrument in liquid nitrogen

B. ***Sample preparation procedure***

First, a solution contained 0.5 mM cesium carbonate and 4.47 mM potassium chloride were absorbed into the stem of the plant for 30 minutes. The stems of the plant were cut at 1 cm, 15 cm and 30 cm from the position where the solution was absorbed. Next, these stems were wrapped with an indium board and inserted in aluminum pellets with holes 3 mm in diameter, respectively. The gap between the hole and the stem was filled with indium. Finally, that aluminum pellet (10 mm dia.) was attached to the sample holder. That sample holder was immersed in liquid nitrogen and quickly cooled. From these processes, the stems were cut in the freezing state by the current instrument.

3. Results and discussion

Figure 2 (a) shows the image of the sample surface with the current method by SEM observation integrated in the TOF-SIMS. From this SEM image, it is possible to be understood the arbitrary region of the internal anatomy in plants. Therefore, we considered that the internal anatomies are selectable, analyzable and depend on the fixed stem with indium and the use of the sectioning instrument.



(a) SEM image of sample surface (b) Analyzed points

Fig. 2. Cross-section image of the stem

Figure 2 (b) shows the image of the plant stem cross section at height of 1 cm by SEM observation integrated in the TOF-SIMS under frozen condition. These four points in the picture were analyzed with the TOF-SIMS for a field-of-view of 50×50 μm^2. In this study, the TOF-SIMS spectrums are used by things which the inside and outside at intercellular spaces were each the pith and the bundle. From obtained spectrums in the four areas, Figure 3 shows the graph of the count ratio of Cs and K at specific heights (1 cm, 15 cm and 30 cm). The contained amounts of K at the pith and the bundle are assumed constant values irrespective of the long from the absorbed points. In the ratio Cs and K of the pith, the largest value was obtained in case of 1cm, followed in order by 15cm and 30 cm. In the ratio Cs and K of the outside pith, the about same amounts are 1cm and 15cm, the minimum amount is 30cm. The career of Cs to the pith are considered two patterns how Cs was transported in the pith. Firstly, the way of the transport from the duct. Next, the pattern of the transport from the duct to the distributed each tissue. Based on these results, the ratio of Cs and K from these two transports show that first way of the transport is followed 1cm, 15 cm and 30 cm and next way is followed 30 cm, 15 cm and 1 cm and the expected ratio Cs and K is showed the transport from the duct. For that reason, we considered that Cs in plants are transported from the duct to the pith.

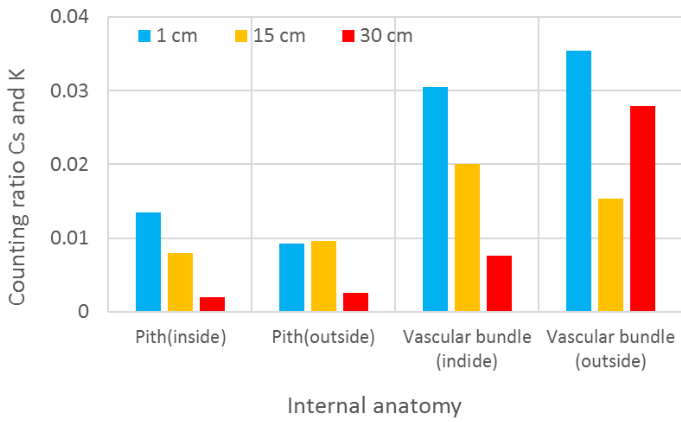


Fig.3. Count ratio Cs and K at each specific height by TOF-SIMS measurements.

4. Conclusion

The internal anatomies are selectable, analyzable and depend on the fixed stem with indium and the use of the sectioning instrument. According to the analysis of Cs in plant using this technique by TOF-SIMS, the dynamics of Cs in plants were confirmed. Therefore, SIMS analysis of Cs in plants to elucidate the dynamics became possible.

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References

(1) F. Sakamoto, *et sl. Atomic Energy Society of Japan*, **12**, 257 (2013) (in Japanese).
(2) T. Keitarou *RADIOISOTOPES*, **62,** 25 (2013) (in Japanese).
(3) M. Akamatsu, *et al. ACS Appl. Mater. Interfaces*, **6,** 8208 (2014).
(4) K. Tagami, S. Uchida, *BUNSEKI KAGAKU*, **65,** 511 (2016) (in Japanese).
(5) M. Sato, *et al. THETOHOKU BRANCH, THECROP SCIENCE SOCIETY OF JAPAN*, **57,** 29 (2014).
(6) H. Sugai, *et al. Nuclear Instruments and Methods in Physics Research B*, **318,** 191 (2014) .
(7) D. Yan, *et al. Journal of Environmental Radioactivity*, **126,** 232 (2013) .
(8) K. Kanenari, M. Morita, T. Sakamoto, *e-J. Surf. Sci. Nanotech*, **14,** 131 (2016) .
(9) K. Ohishi I. Kawakami, T. Okumura, and T. Sakamoto, *Bunseki Kagaku*, **63**, 391 (2014) (in Japanese).
(10) T. Sakamoto, M. Koizumi, J. Kawasaki, J. Yamaguchi, *Applied Surface Science*, **255,** 1617 (2008).