Note

PIXE (Proton Induced X-Ray Emission) Analysis of Caatinger[†]

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Caatinger (Mimosa acutistipula Benth), which has been a popular wood among the people in north east part of Brazil, was analyzed using PIXE (proton induced X-ray emission). The wood sample was divided into three parts, center, inner and outer side of the wood. Using the wood slice of each part, Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, Mn, Fe, Cu, Zn, As, Se, Br, Rb, Sr, Y, Ru and Pb were detected. When the wood slices were decomposed with nitric acid, Co and Ni were able to be determined. Two samples, non-destructive and chemically treated sample, of heartwood and sapwood showed the similar concentration of the elements. There was a discrete change of the accumulated elements between heartwood and sapwood. Most of the elements detected were tended to be concentrated at sapwood, whereas Ca, Cu and Sr were concentrated at heartwood. At center part, especially, Na, Mg, Fe and Sr were accumulated. Since most of the heartwood and sapwood is dead, discrete change of the element concentration suggests some element movement across the annual ring at heartwood formation.

Key Words : caatinger, PIXE (proton induced X-ray emission), element distribution

Introduction

Caatinger, which is a popular wood at north east of Brazil has been used widely as materials for house and furniture, as solid fuel or to feed domestic animals. From this year ecological survey of caatinger has begun. One of the purpose of the study is to know the water and mineral tolerance of the wood with the change of environmental conditions reflected in soil. Since

caatinger is one of the most important wood for people's life in Brazil, the afforestation of the wood is now planned. Weather condition in Brazil has been vigorously investigated¹⁾, therefore, to plant the wood, other environmental conditions should be studied. One of the other approach for the environmental study is to know the nutritional change of the wood as well as soil conditions. Since the element distribution in the wood according to the annual ring is expected to reflect the mineral change in the soil, the authors tried to apply PIXE (proton induced X-ray emission) method to know the wide spectrum of the element concentration in the wood. In Japan a lot of baby cyclotrons have been constructed. Most

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of them are to produce short half-life isotopes for the medical purposes. The cyclotrons are not used all the time for the therapy purposes, therefore, a lot of cyclotron facilities are able to set the analytical system for PIXE analysis²⁾. Using human or animal samples, element distributing data by PIXE method have been accumulated. However, there is not enough papers analyzing plant samples by PIXE method^{3), 4)}. Since PIXE method seems to be widely spread in near future, basic technological data, especially in plant research, should be accumulated. Therefore the authors tried to analyze the elements, first using the wood slices itself to know to which degree and what kind of elements could be detected. Then the wood slice was chemically decomposed to reduce the background in the measurement to perform PIXE analysis. Since many samples have to be analyzed, in the future, for survey study of the wood, analytical condition have to be tested. We report the preliminary study of the wood using PIXE method.

Materials and Methods

A caatinger (*Mimosa acutistipula Benth*) slice, which is estimated to be 24 year old, was cut into two along with the line across the annual ring and center. The one half of the wood slice was shaved with a plane to get the thin radial section. The section was cut into seven parts, center (C), three parts from the heartwood (H1, H2 and H3) and three parts from the sapwood (S1, S2 and S3). The heartwood was clearly distinguished from the sapwood by the darker color.

Each samples was fixed at the center of a polypropylene film (4 μm , in thickness), by a tape, set at a polyester holder (600 mm $\times$ 250 mm $\times$ 3 mm), and irradiated with proton beam from the cyclotron installed at Nishina Memorial Cyclotron Center, Japan. In one run, seven holders were able to be measured. Monitoring the proton beam current during the measurement was not

able to perform, since the sample was too thick to detect the beam by a Faraday cup equipped behind the sample. Therefore the proton beam irradiated was estimated from the beam current before and after the sample setting. Irradiated proton beam energy was 2.9 MeV. Counting the X-ray from the sample was carried out for 600 to 900 s for each sample. Two Si(Li) detectors were used for the X-ray measurements. One is set beside the target with a 300 μm myler absorber to detect the heavy elements ($>\text{Ca}$). Another detector with no absorber was used for the detection of light elements ($\leq\text{Ca}$).

After non-destructive analysis, three samples, center, heartwood and sapwood were chemically treated to reduce the background in the measurement. Each sample was decomposed with 1 – 2 ml of nitric acid in a teflon vessel, using an electric oven. Then all of the solution was dropped on the film fixed at a holder and dried. Indium standard solution (500 $\mu\text{g/g}$) was added with the sample as an inner standard⁵⁾. The amount of the indium standard added to the sample was listed in Table 1. The determination of each element was performed using the program, especially developed for the measurement⁶⁾.

Table 1

	Sample (mg)	In standard (μg)	Av. current (nA)	Irradiation (nC)
S	275	137	50	72 000
H	155	77	50	100 000
C	144	72	50	75 000

Results and Discussion

The element distribution in the wood by non-destructive measurement was shown in Fig. 1. Since there was no standard to determine each element, in non-destructive analysis, first, Ca content was set as 1 000 ppm and the relative amount of each element was obtained. Then, Ca content, which was obtained from the chemically treated sample, was regarded as an absolute amount of Ca

Fig. 1 Non-destructive analysis of caatinger by PIXE.

(a) Elements less than Ca, detected by a Si(Li).

(b) Elements higher than Ca, detected by the Si(Li) covered with a 300 μ m mylor.

C, H and S are center, heartwood and sapwood, respectively. The numbers indicate the distance from the center, that is, H1 or S1 are closer to heartwood than H2 or S2, respectively.

in each sample. Based on the absolute Ca content, the relative amount of each element was converted to the absolute amount. Therefore, the

amounts of calcium measured from the chemically treated sample, 2 830, 2 400 and 1 560 ppm for center, heartwood and sapwood, respectively,

Fig. 2 PIXE analysis of the wood when chemically treated.
C, H and S are center, heartwood and sapwood, respectively.

were used to obtain other element concentration. H and S stand for heartwood and sapwood, respectively. The numbers indicate the distance from the center, that is, H1 or S1 are closer to heart wood than H2 or S2, respectively.

Figure 1(a) is from Na to Ca and Fig. 1(b) shows the elements from Ti to Ru. In almost all of the elements detected showed the drastic change of the element abundance between heart wood (H) and sapwood (S). There was no element which accumulating pattern showed smooth change from the center to sapwood. Most of the elements were tended to be concentrated in sapwood. However, Ca, Cu and Sr were accumulated in heartwood. The difference within heartwood (H1 to H3) or sapwood (S1 to S3) was tended to be smaller than that between heart and sapwood. Though H1, H2 and H3 are within the same part of the wood, K and Zn showed drastic change of the abundance within the heartwood. At H3 part, where the part is closer to sapwood, amount of Cu

was more than twice as high as the other part. Similarly, in S2 part, Cl abundance was extremely high. At the center part(C), especially Na, Mg, Fe and Sr were accumulated. Though the data were not plotted, trace amount of Al and Pb were detected only at the center part. Though the accumulating pattern of Ca and Sr was similar, because of the trace amount of Rb, the similar pattern between K and Rb could not be found.

Figure 2 shows the data from chemically treated sample. When the sample was treated, Na, Mg, Si and some other trace elements were not detected, which might be lost during the treatment. However Co and Ni were able to be detected due to the decrease of the background value during the measurement.

The elements detected in both samples, non-destructive and chemically treated, showed the similar tendency. It was interesting that most of the elements did not distributed in uniform pattern in the wood. Both measurements showed that

there was a discrete change of the element accumulating pattern between sapwood and heartwood. At the center of the wood, Ca and Sr were concentrated. It was interesting to note that, in animals, Ca or Sr are concentrated mainly at bone, namely, constructing tissue.

Two samples from two kinds of wood parts, heartwood and sapwood, showed similar element content. None of them at the same wood part showed drastic change of the element content as shown between heartwood and sapwood, suggesting that there was not any nutritional difference from year to year. Therefore it is reasonable to estimate that the drastic change of the element content between heartwood and sapwood seems to be derived after the necrosis of the center part of the wood had begun. If the element concentration occur during the heartwood formation, the chemical reaction across the annual ring is necessary. Though there is no data, so far, to support any chemical reaction across the annual ring, when neutron radiography of the wood slice was taken, water distribution across the annual ring at outer part of the wood was observed⁶⁾. which might support the hypothesis. It is known that there are only a few living cells at sapwood, however, which cannot be the reason to cause the large difference of the element abundance.

To discuss in more detail the difference of the element content, each sample along with each annual ring should be analyzed. To estimate the environmental change of the wood occurred, only

inside the heartwood or sapwood sample should be compared, since the discrete change of the element abundance between heartwood and sapwood does not seem to be derived from the environmental change. It was also noted that at the center, there was another discrete element pattern, different from other parts. The center of the wood does not grow across the annual ring. The tissue only grows with the height of the wood, therefore the center part reflects the original tissue of the wood of the year, showing the different element concentration from either sapwood or heartwood.

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