

Measurement of Internal Structural Changes during Pyrolysis of Wooden Pellets under a Radiant Heating Field using Synchrotron X-ray CT

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1 Introduction

The pyrolysis and combustion rates of solid fuels are important properties of solid combustion. The pyrolysis rate and composition of polymers has been measured using thermogravimetric analysis (TG) and differential thermal analysis (DTA) [1-6]. However, determining the condition of the interior of a sample is difficult; in particular, the changing internal structure is unclear.

In Japan, the implementation of power generation facilities that use woody biomass as the main fuel has increased. Woody biomass is pyrolyzed during both heat use and power generation. Therefore, the elucidation of pyrolysis characteristics can further promote the utilization of woody biomass. Additionally, the active use of reproducible wood can contribute to fixing carbon, reducing fossil fuel consumption, and reducing carbon dioxide emissions. For example, the use of wood in buildings, particularly with the spread of wooden buildings, plays an important role in achieving a carbon-free society. Thus, it is important to understand the pyrolysis phenomenon of wood not only as a fuel but also from the perspective of disaster prevention.

In X-ray computerized tomography or computed tomography (CT), X-rays are incident on an object to obtain the spatial distribution of the linear absorption coefficient. In CT, time is generally required to obtain a transmitted image of a sample in multiple directions. If the sample changes while capturing the images, it cannot be successfully reconstructed; hence, capturing images under motion is difficult. However, when synchrotron radiation is used for X-ray CT, a high flux can be obtained even in a short time, making it possible to perform high-speed imaging compared with industrial X-ray CT apparatuses. Hence, the time evolution of a three-dimensional structure can be followed by continuously capturing images.

In this study, the microscopic visualization of the pyrolysis of wood biomass was conducted using the BL20B2 beamline at “SPring-8,” a large synchrotron radiation facility. X-ray CT imaging was used to observe the differences in the internal structures of the wood caused by thermal pyrolysis. To measure the sample using high-speed X-ray CT, an ultra-high-speed CT of less than 10 s per CT was performed.

A high heat flux was achieved using radiation as the heat source. A white pellet was used as the specimen. The internal structure of wood during transient pyrolysis was visualized under a nitrogen

atmosphere using ultra-high-speed X-ray CT. This research aimed to contribute not only to renewable energy but also to disaster prevention.

2 Experimental apparatus and method

The synchrotron X-rays used in this experiment were characterized by parallel light with high luminance and high directivity. Using a beam with high synchrotron radiation directivity, the refraction of X-rays by a substance can be processed; thus, it is effective for samples with small differences in the absorption coefficient. The SPring-8 BL20B2 beamline has a bending electromagnet as the light source [7]. "SPring-8" is an abbreviation of "super photon ring-8 GeV," where 8 GeV is the power output of the ring. With this beam line, an X-ray of 5–113 keV can be used, and an effective resolution of several micrometer to 100 μm can be achieved depending on the sample size. To obtain a high spatial resolution with parallel light synchrotron radiation, the transmitted X-ray image is converted to a visible light image using a thin-film fluorescent plate (scintillator), magnified by an optical lens system, and projected onto a CMOS camera.

Figure 1 shows an overview of the experimental apparatus. The sample installed on the apparatus was subjected to radiation heating on its surface using a radiation heater. The experimental apparatus was installed on a rotating stage between the X-ray irradiation unit and scintillator in the experimental hatch of the SPring-8 beamline. The sample was placed on the sample stage and inserted into the apparatus. Additionally, because the rotary stage with the apparatus and sample stage are independent, X-ray CT measurements are possible during heating. The X-ray transmission window was an aluminum foil. Heating was performed using a radiation heater in the direction perpendicular to the X-ray irradiation direction. The sample was rotated using a rotating stage; thus, it could be heated all around. The interior of the apparatus was nitrogen or air atmosphere. Flaming does not occur in a nitrogen atmosphere. The center of the wood was irradiated for 10 min at an output of 100 °C to remove moisture and then for 7 min at an output of 40 W for pyrolysis. X-ray CT was performed continuously for 17 min using a radiant heater. A commercially available white pellet was crushed once and molded again as a specimen. A few spherical ceramic particles were intentionally mixed into wood samples. Before pyrolysis, the specimens were circular columns (height: 5 mm; diameter: 5 mm). The X-ray source and radiant heating were directed radially at the specimen (Figure 1). The heat flux was extremely high owing to the thermally thick conditions. The specimen exhibited a temperature distribution and was deformed under the conditions of a large Biot number. The projection conditions are listed in Table 1. The time required for each CT was approximately 8.3 s. The energy value of the X-rays was 15 keV for the target carbon, which is the main element in woody biomass. As the spatial resolution of the detector, a pixel size of 6.6 μm was used for imaging.

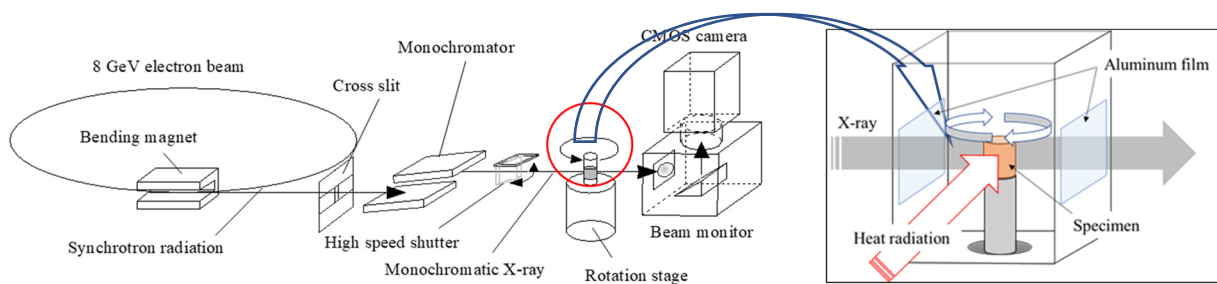


Figure 1: Projection tomographic X-ray system at BL20B2 of SPring-8.

Table 1: Experimental conditions used in X-ray CT.

Energy (keV)	15
Pixel size ($\mu\text{m}/\text{pixel}$)	6.6
Exposure time (ms)	5
Number of projections (/ 180°)	900 / 180

3 Results and Discussion

Figure 2 shows an example of an X-ray CT image of the specimen. The light regions in the images correspond to the regions where the X-rays were absorbed, and the dark regions correspond to the regions from which they were transmitted. The white circular regions in the specimen were spherical ceramic particles. We observed that the remolded pellets consisted of crushed wood chips.

Figure 3 shows an example of an X-ray CT image of a specimen during radiant heating. This figure shows the cross-section at the focal point of the radiant heater (340th slice) from the side view. The numbers in the figure represent the CT numbers. The crushed wood chips are clearly identifiable in the CT-001 image. Up to CT-071, crushed wood chips were observed during the drying process. After drying for 10 min, the specimen began to deform rapidly when the output of the radiant heater was set to 40 W. CT-076 showed the CT image at approximately 42 s after the output of the radiant heater was set to 40 W. The outer edge of the specimen could not be reconstructed. This was because the specimen was moving. This indicated that the pyrolysis progressed, and the outer part of the specimen was deformed. However, the structure of the crushed wood chips was observed in the interior of the specimen. In other words, structural changes did not occur within the specimen due to pyrolysis. For CT-120, approximately 408 s after the output of the radiant heater was set to 40 W, cavities of various sizes were observed. However, a few crushed wood chips remained.

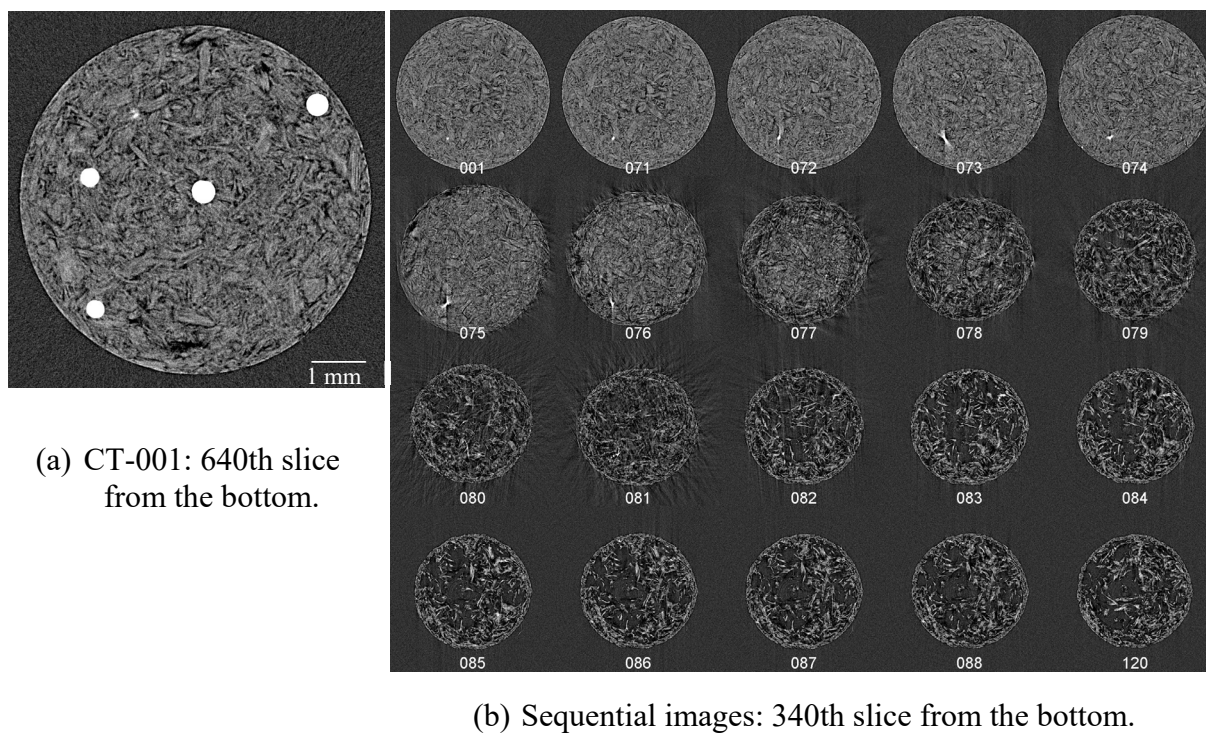


Figure 2: X-ray CT images of specimen in the process of radiant heating.

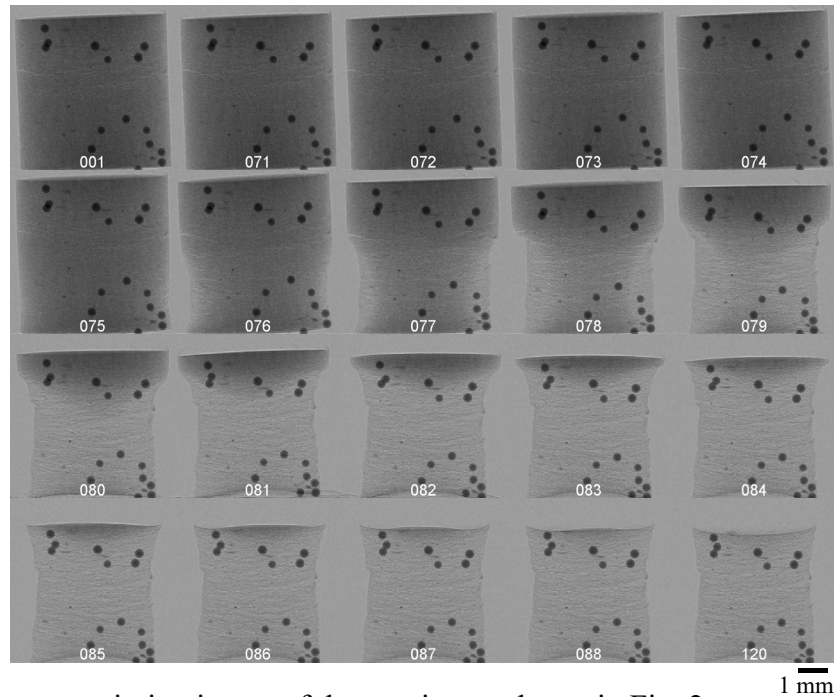


Figure 3: X-ray transmission image of the specimens shown in Fig. 2.

Figure 3 shows the X-ray transmission image corresponding to Figure 2(b). In contrast to the CT image, the dark regions absorbed the X-rays. Because it was before heating, the image-001 absorbed X-rays well. After 42 s of setting the heater output to 40 W, image-076 showed that pyrolysis progressed near the outer peripheral surface of the wood and X-rays were transmitted. The X-rays were absorbed at the center of the specimen, and pyrolysis had not yet progressed. This also corresponded to the fact that the structure of the crushed wood chip remained intact (Figure 2(b)). In the transmission image, approximately 408 s after the heater output was set to 40 W, according to image-120, the entire specimen easily transmitted X-rays, and the specimen contracted. Additionally, the spherical ceramic particles remained incorporated into the specimen without falling out.

Figure 4 shows the deformation trajectory of the specimen during pyrolysis. Transmission images were superimposed every 41.5 s beginning immediately before the start of the pyrolysis. The final position is indicated in a dark color. This figure clearly indicates that the specimen shrank owing to the pyrolysis. In addition, the ceramic particles in the specimen moved following the contraction. In our previous study [8], experiments using raw cypress wood confirmed that the specimen expanded during the early stage of pyrolysis. In this study, such expansion was not observed, and the pellet specimen shrank continuously. The cause appeared to be the absence of a continuous tracheid structure that resists the passage of pyrolysis gas. Under the conditions of the specimens used in this experiment, the Biot number was greater than 1, which is not a thermally thin condition. The high heat flux inside the wood has a significant influence on structural changes and breakage.

Figure 5 shows the changes in the coordinates of the center of mass with the progress of pyrolysis by stacking X-ray CT images and using image analysis. The color map represents the CT number and the time passed from 072 to 120 every 8.3 s. For the pellet specimen used in this study, almost no movement of the center of mass was observed in the radial direction, and the change in the height direction was remarkable. In the dehydration process, moisture was removed; thus, it changes by about 20 μm with time, but the blue and green plots almost overlapped. When the dehydration process was completed and radiation heating was initiated at 40 W, a sudden change in the height direction was observed. At this time, the center of the mass moved upward. Eventually, the center of mass decreased as the pyrolysis

progressed, and when the pyrolysis was completed, the change disappeared, as indicated by the red plot. In this experiment, the irradiation position of the radiation heater was slightly lower than that at the center of the specimen. Thus, the pyrolysis of the upper side of the specimen was delayed, and the center of mass moved to the upper side after heating began. In this experiment, the specimen was rotated for CT measurement, but we considered that pyrolysis progressed stepwise from the outside to the inside of the pellet specimen, which was uniformly heated from the surroundings in the radiation heating field.

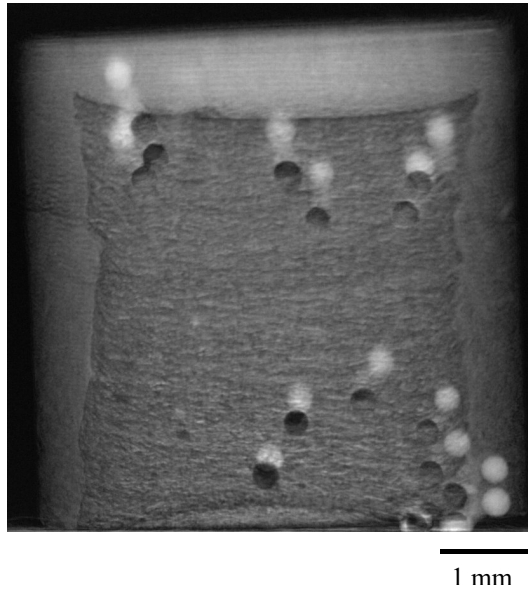


Figure 4: Trajectory of the deformation of the specimen during pyrolysis progress in X-ray transmission images.

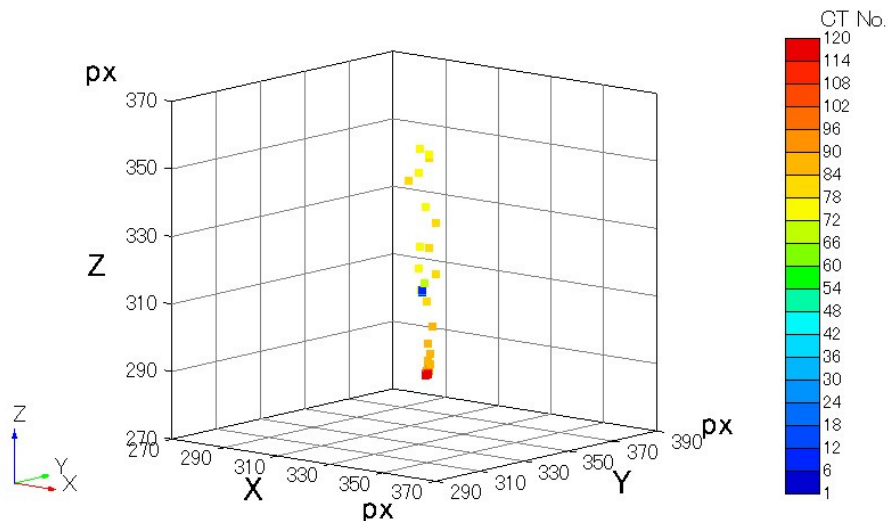


Figure 5: Changes in the coordinates of the center of mass that change with the progress of pyrolysis by stacking X-ray CT images.

4 Conclusions

Changes in the internal structure of wood pellets during transient pyrolysis under a high heat flux were studied using ultrahigh-speed X-ray CT imaging. The results are summarized as follows.

- 1) Despite the large difference in density, we were able to visualize fine wood chip structures and ceramic particles inside the samples.
- 2) Pyrolysis progressed to the interior, and cavities of various sizes were observed. And, the small amount of crushed wood chips remains.
- 3) In our previous study, cypress wood expanded during the early stages of pyrolysis, whereas pellet specimens shrunk continuously.
- 4) Under the conditions of the specimens used in this experiment, the Biot number was greater than 1, which is not a thermally thin condition. The high heat flux inside the wood has a significant influence on structural changes and breakage.
- 5) For the pellet specimen used in this study, almost no movement of the center of mass was observed in the radial direction, and the change in the height direction was remarkable.

Acknowledgements

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