1. Introduction

Mulberry has been cultivated as a resource for the sericultural industry and bolsters the production of silk fibers. Mulberry growth rate is comparatively fast and is easy to cultivate in the field. In addition, the leaves and wood of the mulberry are rich in alkaloid components including 1-deoxynojirimycin (1-DNJ), which is known as a $\alpha$-glucosidase inhibitors which acts on preventing glucose absorption in the small intestine [1]. 1-DNJ is used as a dietary supplement for preventing diabetes because blood sugar level is decreased. Studies on 1-DNJ extraction from mulberry and the effect of 1-DNJ on $\alpha$-glycosidase inhibitory activity has been reported [1-3].

The bark of kozo (Broussonetia species), called paper mulberry, has been used to produce Japanese paper. The bark of mulberry for sericulture was also used to produce Japanese paper in the past in Japan [4]. Functional paper having conductive properties using Japanese paper has been studied [5]. Japanese paper has also been applied to fiber field as paper yarn [6].

As noted above, mulberry is not just used in sericulture, but is a potentially useful biomass resource. In this study, with the aim of investigating the characteristics of paper sheets prepared from mulberry bark for sericulture, we performed the preparation of paper sheets using the bark of kozo and mulberry (Morus alba L.), which are the general mulberry cultivated for sericulture in Japan, and examined the structure and properties of the prepared paper sheets.

2. Experimental

2.1 Materials

The bark of mulberry (Morus alba L.) and kozo (Broussonetia species) belonging to mulberry were used to prepare the paper. The mulberry and kozo were cultivated at the Experimental Farm, Faculty of Textile Science and Technology, Shinshu University. The bark of each sample was ringed from the branches which were boiled in hot water at 120 °C for 20 min in an autoclave.

2.2 Pulping

The prepared bark was cooked for 8 hour in a 0.8 wt% sodium hydroxide aqueous solution. The amount of water was equal to 10 times the weight of the dry bark. The cooked bark was thoroughly rinsed in water, and unevenly cooked parts and discolored parts were removed by hand. The treated bark was dried at 110 °C for 24 hours, and was cut in lengths of 10 mm to facilitate beating. The pulp obtained was beat in a beater. The freeness (Canadian Standard Freeness ; CSF) of the pulp was determined according to the Japanese Industrial Standard (JIS) P 8121(ISO 5263-compatible). The values of freeness are shown in Table 1.
2.3 Preparation of the paper sheets

The pulp was disintegrated by a pulp disintegrator, and paper sheets were prepared with a square type (250 mm by 200 mm) sheet machine (Yasuda Seiki seisakusho LTD.). The wet sheets were pressed under 0.4 MPa for 15 min and dried at 109 °C using a rotor dryer (ROTER DRYER DR-200, KUMAGAI RIKI KOGYO CO., LTD.).

2.4 Scanning electron microscope observations

The fibers of the paper sheets were observed by a scanning electron microscope (SEM ; S-2380N, Hitachi High Technologies Co., Ltd.). The fiber diameters were calculated by the SEM images.

2.5 Wide angle X-ray diffraction measurement

Wide angle X-ray diffraction (WAXD) measurement of the obtained paper sheets was carried out by an X-ray diffractometer (RU-200, Rigaku Corporation) with a goniometer. The wavelength of the X-ray was 0.154 nm, and the range of the scanning angle (diffraction angle) was 5 to 35 degrees by 2 degree/min scanning speed.

After the data of the WAXD profiles were fitted to a curve using the Gaussian function, the crystallinity index \( CrI \) of (200) reflection was calculated as follows [7]:

\[
CrI = \frac{I_{200} - I_{am}}{I_{200}} \times 100
\]

where \( I_{200} \) is the intensity of (200) reflection of cellulose and \( I_{am} \) is the intensity of amorphous of cellulose.

In addition, Semicrystalline size of (200) reflection was determined by Scherres’s equation [8] as follows :

\[
D = \frac{K \times \alpha}{\beta \times \cos \theta}
\]

where \( D \) is the semicrystalline size of (200) reflection, \( K \) is constant (0.9), \( \alpha \) is the wave length of the X-ray (0.154 nm), \( \beta \) is full-width of half maximum (FWHM) of the peak, and \( 2\theta \) is the diffraction angle of the peak of (200) reflection.

2.6 Molecular orientation measurement

The molecular orientation of the paper samples was measured by a microwave molecular orientation analyzer (MOA6000, Oji Scientific Instruments CO., Ltd.). The principle for determining the molecular orientation of paper is based on the interaction between the dipole moments of molecules constituting a paper [9, 10]. Molecular orientation ratio, \( MOR \), is a ratio of the maximum to the minimum values of the transmitted microwave intensities. If the \( MOR \) value is 1, molecules in a paper do not orientate. The \( MOR \) needs to be corrected for thickness because the \( MOR \) depends on the thickness of the paper. The corrected molecular orientation ratio, \( MOR_c \), was calculated with a standard sample thickness, \( t_c \), of 200 µm, and a sample thickness, \( t \), as follows :

\[
MOR_c = \frac{t_c (MOR - 1)}{t} + 1
\]

A total of ten measurements were performed for each sample.

2.7 Measurement of physical properties

The thickness, wet strength, tearing strength, folding endurance, smoothness, air resistance, and stiffness of the paper sheets were determined according to JIS P 8118 (ISO 534-compatible), JIS P 8135 (ISO 3781-compatible), JIS P 8116 (ISO 1974-compatible), JIS P 8114 (ISO 5626-compatible), JIS P 8119 (ISO 5627-compatible), JIS P 8117 (ISO 5636-5-compatible), and JIS P 8143 [11], respectively.

The tensile properties of the paper sheets were measured at room temperature using a tensile tester (RTC-1250, Orientec Tensilon). The initial sample length and width were 20 mm and 5 mm, respectively, and the extension rate was 100 % min\(^{-1}\). 10 measurements were performed for each sample.

3. Results and Discussion

3.1 Structure of the paper sheets

The diameter of the fibers in the paper sheets prepared from the mulberry and kozo bark were calculated from the SEM images (see Fig. 1) and are listed in Table 1. The fiber diameter of paper sheets

<table>
<thead>
<tr>
<th>No.</th>
<th>Raw material</th>
<th>CSF (ml)</th>
<th>Fiber diameter in paper sheet (µm)</th>
<th>Ave.*</th>
<th>S.D.**</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Mulberry</td>
<td>355</td>
<td>17.5</td>
<td>4.4</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Kozo</td>
<td>348</td>
<td>15.2</td>
<td>1.8</td>
<td></td>
</tr>
</tbody>
</table>

*) Average  
**) Standard deviation

Table 1 Paper making conditions, Canadian Standard Freeness (CSF), and fiber diameter in paper sheets.

<table>
<thead>
<tr>
<th>No.</th>
<th>Raw material</th>
<th>Crystallinity (%)</th>
<th>Semicrystalline size (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Mulberry</td>
<td>82</td>
<td>6.3</td>
</tr>
<tr>
<td>2</td>
<td>Kozo</td>
<td>85</td>
<td>6.3</td>
</tr>
</tbody>
</table>

Table 2 Crystallinity and semicrystalline of (200) reflection of paper sheets prepared from bark of mulberry and kozo.
obtained from the mulberry bark was larger than that obtained from the kozo bark.

The WAXD profiles of the paper sheet samples are shown in Fig. 2. A diffraction peak was observed at around 22.6° for all samples. The peak corresponds to (200) reflection on a crystal type of cellulose I [6].

As shown in Table 2, the results of the crystallinity index and the semicrystalline size did not depend on the difference between the mulberry and the kozo paper samples.

3.2 Physical properties of the paper sheets

Thickness of the paper sheet samples is plotted against the basis weight in Fig. 3. The mulberry paper sample was thicker than the kozo paper sample against the basis weight. This result means that the paper density of the mulberry sample was smaller than that of kozo sample.

In the tensile properties shown in Table 3, the tensile strength and the Young’s modulus of the mulberry paper sample were lower than those of the kozo paper sample.

The orientation angle and molecular orientation of the paper sheet samples are also shown in Table 3. The orientation angle means the angle of the highest molecular orientation on a plane of the paper sheet samples, and the value of a positive and negative of the orientation angle corresponding to the angle in a clockwise direction and a counterclockwise direction on the plane, respectively. The mulberry paper sample generated larger average and deviation in molecular angle and lower molecular orientation in comparison with the kozo paper sample.

Tensile direction for the tensile test is equal to 0° of the orientation angle determined by the molecular orientation measurement. The larger average and deviation of the orientation angle and the lower molecular orientation of the mulberry sample observed in the results of molecular orientation measurement correspond to the low anisotropy of the tensile direction. Therefore, the poor tensile properties may be attributable to the low

![Fig. 1](image1.png)  
**Fig. 1** SEM images of paper sheets for the conditions of mulberry bark (a) and kozo bark (b).

![Fig. 2](image2.png)  
**Fig. 2** Wide angle X-ray diffraction profiles of paper sheets prepared from mulberry and kozo bark.

<table>
<thead>
<tr>
<th>No.</th>
<th>Raw material</th>
<th>Tensile strength (MPa)</th>
<th>Young’s modulus (MPa)</th>
<th>Breaking elongation (%)</th>
<th>Angle of molecular orientation (degree)</th>
<th>MOR_c</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ave.*</td>
<td>S.D. **</td>
<td>Ave.*</td>
<td>S.D.</td>
<td>Ave.* S.D.</td>
<td>Ave. S.D.</td>
</tr>
<tr>
<td>1</td>
<td>Mulberry</td>
<td>7.8</td>
<td>1.7</td>
<td>91.1</td>
<td>32.8</td>
<td>2.2</td>
</tr>
<tr>
<td>2</td>
<td>Kozo</td>
<td>11.3</td>
<td>3.6</td>
<td>159</td>
<td>48</td>
<td>7.1</td>
</tr>
</tbody>
</table>

*) Average  
**) Standard deviation
anisotropy of the mulberry sample.

The tearing strength, folding endurance, smoothness, air resistance, and stiffness of the paper sheets are summarized in Table 4. The weight of each sample was about 40 g/m². For the value of the smoothness and the air resistance, the mulberry paper sample was lower than the kozo paper sample. This indicates that the mulberry paper sample has higher air permeability and lower smoothness in comparison to the kozo paper sample. It may be corresponded that the paper density of the mulberry sample was smaller than that of kozo sample as mentioned above.

4. Conclusions

From results of WAXD measurement, the paper sheets of mulberry bark were similar in the crystallinity and semicrystalline size of (200) reflection of to the paper sheets of kozo bark.

In comparison with paper sheets of kozo bark, paper sheets of mulberry bark shows larger fiber diameter and lower air resistance, smoothness and paper density. The paper sheets of mulberry bark also have lower tensile strength, Young’s modulus, and anisotropy than the paper sheets of kozo bark.

Fig. 3 Dependence of paper thickness on basis weight.

Acknowledgements

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