Extrusion of Solid Wood Impregnated with Phenol Formaldehyde (PF) Resin: Effect of Resin Content and Moisture Content on Extrudability and Mechanical Properties of Extrudate

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A new deformation technique of wood flow forming is promising for industrial uses of solid wood. Flow deformability, size stability, and good mechanical properties were obtained by using wood impregnated with low molecular weight phenol formaldehyde (PF) resin. In this study, to clarify the effect of the PF resin and moisture contents in wood on the flow deformability (extrudability) and the mechanical properties of the product (extrudate), a lateral extrusion was conducted by using wood impregnated with various contents of PF resin and three levels of moisture content. The results indicated that the extrudability of wood impregnated with PF resin improved with an increase in both PF resin and moisture content. The mechanical properties of the extrudate worsened with increases in the moisture content of the wood impregnated with PF resin. Because most of the water in the wood remained in the mold during the extrusion process, chemical changes of the wood substance and PF resin occurred due to steam forming under the high temperature and pressure of the extrusion. The steam worsened the mechanical properties of the extrudate.

Keywords: Wood; Extrusion; Phenol formaldehyde resin; Moisture content

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INTRODUCTION

Wood is one of the largest biomass resources in the world. However, poor deformability of wood has limited its use in industrial products. Other materials such as metal and plastic materials have superior deformability and can be mass-produced by plastic deformation techniques (*e.g.*, extrusion, forging, and injection).

Conventional deformation techniques of wood, such as compressing and bending, have been used in many applications, for example, flooring and furniture (Stamm and Seborg 1955; Sandberg *et al.* 2013). In these techniques, only the wood cells deform. Therefore, only simple shapes can be produced.

In addition to the conventional techniques, a new deformation technique of wood flow forming has been developed (Yamashita *et al.* 2009; Miki *et al.* 2012, 2013, 2014a, 2014b; Seki *et al.* 2016). Wood flow forming enables the processing of solid wood into more complex shapes in a shorter production time by using the general techniques of metal and plastic forming. The mechanism of wood flow is the movement of wood cells by mutual position changes between adjacent cells in addition to deformation of the cells. The

water in cell walls is important for generating wood flow, and the fluidity of the wood increases with increased moisture content (Yamashita *et al.* 2009; Miki *et al.* 2014).

In a conventional wood deformation technique, which is generically referred to as *Compreg*, the raw material is wood impregnated with low molecular weight phenol formaldehyde (PF) resin to prevent dimensional instability of the product due to moisture (Stamm and Seborg 1955). The PF resin penetrates the cell walls. PF resin-impregnated wood softens under heat. It can be compressed under relatively low pressure (approximately 2 MPa), and it can be hardened by polymerization of the PF resin. This process gives high dimensional stability and improved mechanical properties to the wood (Yano *et al.* 1997; Shams *et al.* 2004). The impregnation of low molecular weight PF resin into wood is an effective method in wood flow forming and improves the flow deformability as well as the size stability of the product (Miki *et al.* 2012, 2013, 2014a, 2014b). Furthermore, the mechanical properties are affected by the temperature and fiber arrangements in the forming conditions, and strength properties similar to those of industrial engineering plastics such as polyvinyl chloride (PVC), acrylonitrile butadiene styrene (ABS), and polycarbonate (PC) can be attained in products *via* wood flow forming (Miki *et al.* 2012, 2014b). Thus, wood flow forming using low molecular weight PF resin is a promising technique for industrial uses of wood.

The objective of this study was to clarify the effects of the content of PF resin and moisture in wood on the flow deformability and the mechanical properties of the product. Wood impregnated with various contents of PF resin and three levels of moisture content was extruded by lateral extrusion, and the load was measured. The pressure change during the extrusion was evaluated as the flow deformability (extrudability), and the mechanical properties of the extrudate were measured by bending tests. The interactions between the PF resin, water, and wood substance during extrusion were examined.

EXPERIMENTAL

Wood Sample Preparation

Wood samples with dimensions of 4.2 mm (L; longitudinal direction) × 23 mm (R; radial direction) × 23 mm (T; tangential direction) were obtained from a similar portion of the sapwood with a straight grain of Hinoki (*Chamaecyparis obtusa*) which is a species of cypress native to central Japan. About 100 samples were successively cut from the wood sticks, and their average annual rings width was 1.5 mm. The wood samples were leached in ethanol to remove natural extractives and completely oven-dried at 105 °C, and then their weight and dimensions in the T direction were measured.

PF resin (PX-341, Aica Kogyo Company, Aichi, Japan; original solid content is 50%), which has an average molecular weight less than 500, were diluted with water to the resin concentration of 1%, 2%, 4%, 8%, and 16%. The dried wood samples were soaked in the each resin solution and in the water (resin concentration of 0%), placed in a vacuum at 0.01 MPa for 1 h, and then subjected to pressure at 0.8 MPa for 18 h. About fifteen wood samples were used for the each solution of water. The impregnated samples were conditioned at a relative humidity (RH) of 32% in a desiccator with saturated solutions of magnesium chloride ($MgCl_2$) at 35 °C for 51 h to mildly evaporate the water in the impregnated samples. Conditioned samples were then completely dried in a vacuum chamber (0.01 MPa) with phosphorus pentoxide (P_2O_5) at 35 °C. The weights and dimensions in the T direction of the dried impregnated wood samples were measured.

After impregnation and drying, the wood samples were conditioned for more than 1 week in a conditioning desiccator to prepare the different levels of moisture content (MC) at a temperature of 35 °C. To provide RHs of 33% and 59%, $MgCl_2$ and sodium bromide (NaBr), respectively, were used in saturated solutions. The weight and T dimensions of four samples were measured before the extrusion test.

Extrusion Test

Samples were subjected to a lateral extrusion test. Figure 1 shows a schematic illustration of the extrusion process. Figures 1(a) to (c) show the metal mold used for the extrusion test, which consists of an upper plate, a base plate, and a punch. The upper plate has a rectangular container in which the wood samples were neatly aligned. There is a cavity of 1.2 mm depth, 26 mm width, and 50 mm length between the upper and base plates, and it has a material escape hole at the end to release excess materials. Figure 2 show photographs of the extrusion mold (a) and the wood samples used (b).

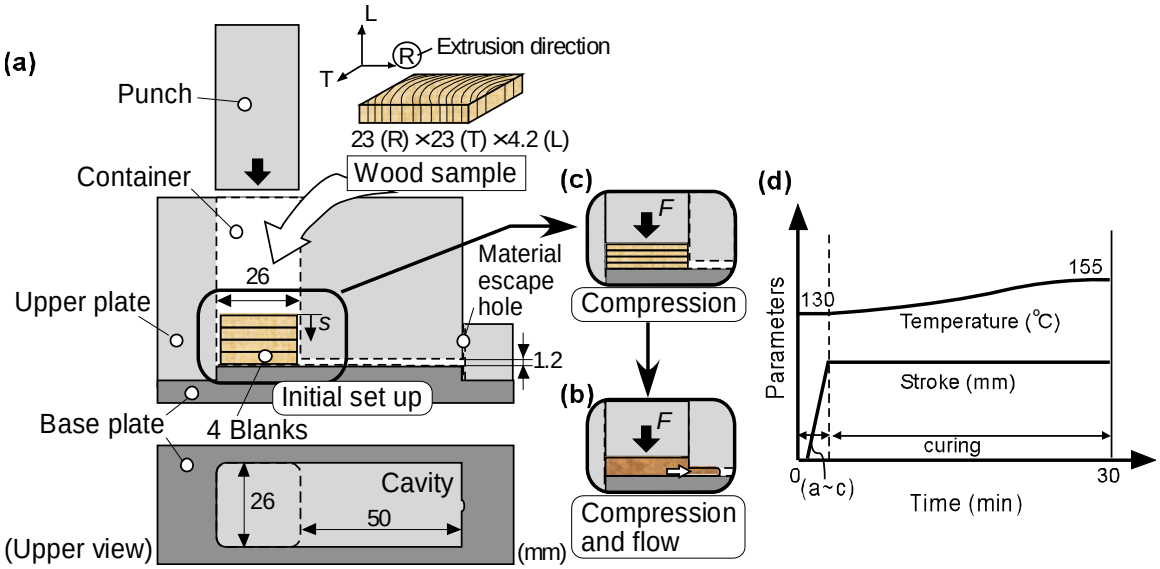

Fig. 1. Schematic illustration of the extrusion process. (a to c) Metal mold used for the extrusion test. (d) Wood deformation behavior and parameter changes during the extrusion process

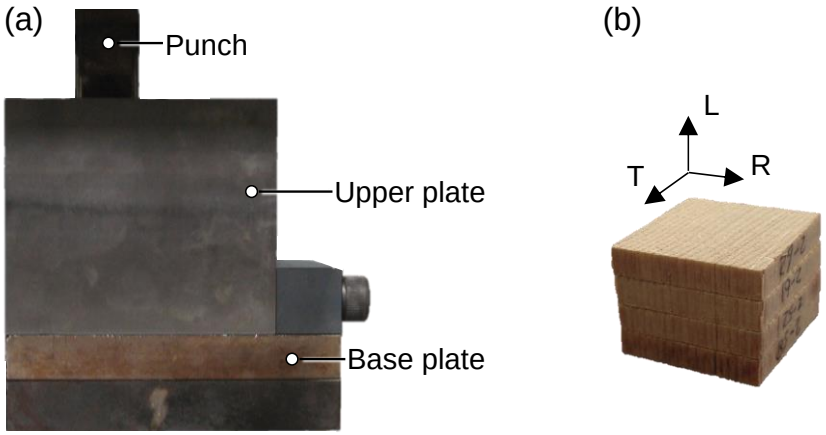

Fig. 2. Photographs of the (a) extrusion mold and (b) wood samples

The mold was placed in a servo press machine (V-1815, Saginomiya Seisakusyo, Inc., Tokyo, Japan) and heated by the heating plates of the press machine to a cavity surface temperature of 130 °C with a maximum variation of ± 2 °C. This temperature falls within the range at which the impregnated wood with PF resin softens (Miki *et al.* 2012). Prior to heating the mold, the mold was sprayed with a mold release agent containing a vegetable oil for commodity plastics (Pelicoat S, Chukyo Kasei Kogyo Co., Ltd., Aichi, Japan). In the initial setup condition (Fig. 1(a)), four wood samples were placed in the heated container, and the punch was inserted. The sample arrangement was in the R direction along the extrusion direction. At 1 min after the wood samples were inserted, the punch was lowered at 10 mm/min until the load applied to the punch became 150 kN (punch pressure: 225 MPa) or until the distance between the punch and base plate became 1.2 mm. The load on the lowering punch was measured while the material compressed and flowed (Figs. 1(b) and (c)). After the punch stopped and the forming was complete, the mold was heated to 155 °C to cure the PF resin in the formed material (Fig. 1(d)). At 30 min after the punch was inserted in the container, the mold was cooled to room temperature, and the material (extrudate) was removed from the mold. The extrusion test was conducted once.

Bending Test

In the bending test, four specimens of 5 mm (in the extrusion direction) × 26 mm were cut from the extrudate perpendicular to the extrusion direction. Before the test, cut specimens were maintained at 20 °C and 59% RH for more than one week. The static bending test was conducted per JIS K 7171 (2016).

FTIR Spectroscopy

To examine the chemical differences between the extrudates with different MC, Fourier transform infrared spectroscopy (FTIR) measurements were performed in attenuated total reflection (ATR) mode. FTIR is an analytical technique which obtains spectra from a very wide range of substances. The spectrum represents the presence of certain functional groups in a molecule. Before the measurements, a sample of approximately 0.5 mm of an extrudate sample was removed to obtain the internal chemical information. The sample was immersed in acetone for 24 h to elute the low-molecular substance and then completely dried in a vacuum chamber (0.01 MPa). The analysis was carried out with an FTIR spectrometer (Thermo Scientific Nicolet^{TM} 6700 spectrometer, Thermo Fisher Scientific, Waltham, USA). For each measurement, 32 scans were accumulated at 4 cm^{-1} resolution.

RESULTS AND DISCUSSION

WPG and Swelling of the Wood Sample

Table 1 shows the weight per gain (WPG) and swelling in the T direction of the wood samples due to the impregnation of PF resin. The WPG and swelling were based on the weights and dimensions of the dried samples before impregnation. Furuno *et al.* (2004) reported that PF resin with an average molecular weight of less than 500 penetrates cell walls, swelling the wood. In Table 1, the swelling result indicates that the amount of PF resin in the cell walls increased with the increasing PF resin concentration.

Table 1. Weight Per Gain (WPG) and Swelling in the T-Direction of Wood Samples due to Impregnation of PF Resin

PF Resin Concentration (%)	WPG (%)	Swelling in T direction (%)
0	0.0 (0.0)	0.0 (0.0)
1	11.1 (0.6)	2.3 (0.2)
2	17.6 (0.6)	3.6 (0.2)
4	27.8 (1.0)	5.0 (0.1)
8	43.1 (1.0)	6.2 (0.3)
16	66.6 (2.0)	7.4 (0.2)
Each value is the average of eight samples, values in parentheses are standard deviations.		

Figure 3 shows the relationships between the PF resin concentration and the MC and swelling in the T direction of the wood samples. The swelling was calculated based on the dimensions of the dried samples before the impregnation of PF resin. The MC of the wood samples decreased with increasing PF resin content. Swelling of the wood samples at 33% and 59% RH was larger than that at 0% RH; this result indicates that water as well as PF resin penetrates into the cell wall.

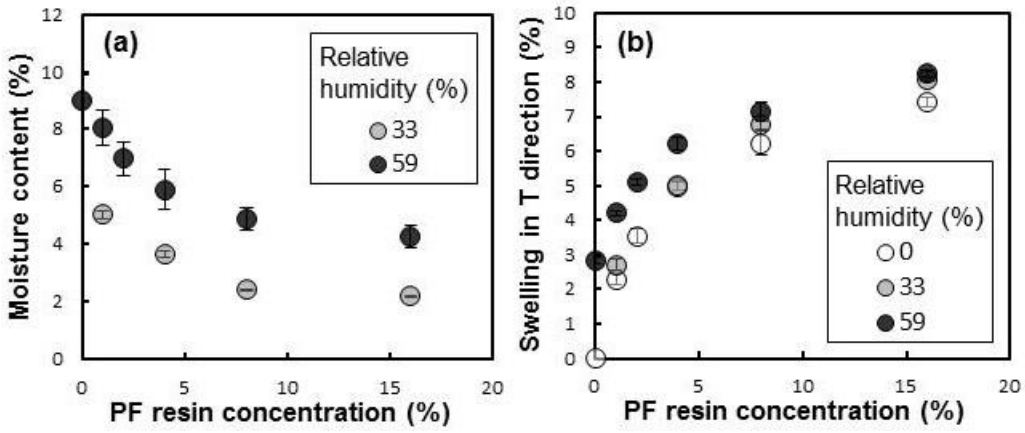

Fig. 3. Relationships between PF resin concentration and moisture content (a) and swelling in T direction (b) of the wood samples

Extrudability of Wood Impregnated with PF Resin

Figure 4 shows an example of the punch pressure and wood deformation behavior during the extrusion test using the wood samples impregnated with a PF resin concentration of 8% and an MC of 0%.

In the first stage, the pressure was kept low. Then, the pressure was rapidly increased, during which the wood samples were only compressed (Photo 1 on right-hand side of Fig. 4). After that, the pressure was relatively stabilized with some fluctuation (after the point indicated by the triangle in the plot shown on the left-hand side), during which the samples flowed and extruded into the cavity (Photo 2). Finally, the pressure was rapidly increased again until the punch stopped at the pressure of 225 MPa. At that time, the samples had already filled the cavity (Photo 3). From these results, the extrusion occurred when the pressure reached 136 MPa (at the triangle point in Fig. 4).

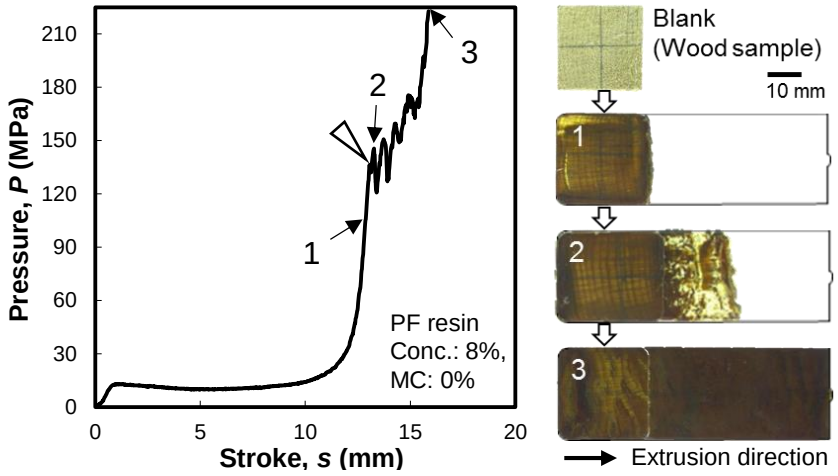

Fig. 4. Punch pressure and wood deformation behavior during the extrusion test (PF resin concentration: 8%, MC: 0%). A triangle denotes the starting point of extrusion.

Figure 5 shows a comparison of punch pressure behavior during the extrusion test of wood samples prepared with different PF resin concentrations and RHs. All results showed similar behavior to that in Fig. 4. However, the starting points of the pressure rising and the extrusion were different under each condition. The pressure at the starting point of the extrusion (triangle point) increased with decreasing PF resin concentration. The samples impregnated with a PF resin concentration lower than 2% and conditioned at RHs 0% and 33% never completely flowed and extruded during the extrusion test (Figs. 5(a) and (b)). In contrast, all wood samples conditioned at RH 59% flowed and extruded, even those without the PF resin (*i.e.*, PF resin concentration of 0% in Fig. 5(c)). The punch strokes at the pressure rise and the starting point of extrusion increased with decreasing PF resin concentration. These stroke differences are attributed to the amount of PF resin filling the cell cavities. A sample with higher PF resin concentration, which means a larger amount of PF resin existing in the cell cavities, was consolidated with a smaller stroke during the test.

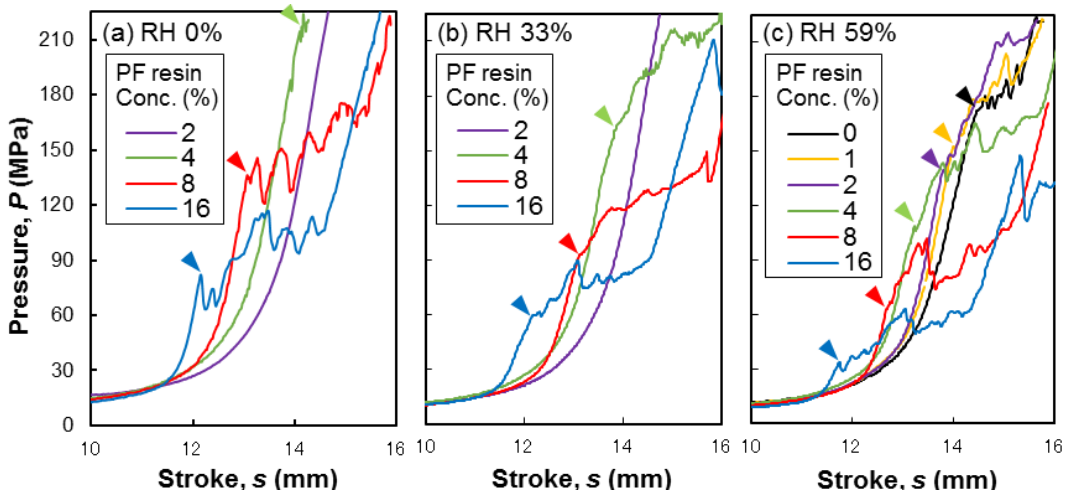

Fig. 5. Comparison of punch pressure behavior during the extrusion test of wood samples prepared with different PF resin concentrations and relative humidities (RHs). A triangle denotes the starting point of an extrusion.

Table 2. Extrusion Force and Mechanical Properties of the Extrudates Obtained from Wood Samples Prepared with Different Content of PF Resin and Moisture

WPG (%)	MC (%)	F_E (MPa)	γ	E (GPa)	σ_B (MPa)
0.0 (0.0)	9.0 (0.9)	171	1.252 (0.015)	2.66 (0.61)	27 (3)
11.1 (0.6)	8.1 (0.6)	152	1.367 (0.008)	5.14 (0.31)	79 (11)
17.6 (0.6)	7.0 (0.6)	137	1.393 (0.007)	5.43 (0.30)	82 (8)
27.8 (1.0)	0 (0.0)	218	-	-	-
	3.6 (0.1)	162	1.393 (0.003)	6.08 (0.17)	104 (5)
	5.9 (0.7)	106	1.384 (0.006)	5.27 (0.16)	85 (12)
43.1 (1.0)	0 (0.0)	136	1.385 (0.001)	6.87 (0.16)	116 (10)
	2.4 (0.0)	94	1.374 (0.006)	5.79 (0.20)	105 (16)
	4.9 (0.4)	63	1.374 (0.009)	4.89 (0.23)	80 (11)
66.6 (2.0)	0 (0.0)	83	1.378 (0.005)	5.99 (0.08)	118 (20)
	2.2 (0.0)	59	1.366 (0.005)	5.62 (0.10)	112 (6)
	4.3 (0.4)	33	1.370 (0.002)	4.51 (0.16)	76 (10)

WPG is the weight per gain of the wood sample due to PF resin impregnation, MC is the moisture content of the wood sample conditioned at different relative humidities, F_E is the extrusion force, γ is the specific gravity of the extrudate, and E and σ_B are Young's modulus and the bending strength of the extrudate, respectively. Values in parentheses are standard deviations.

The pressures at the starting point of extrusion (triangles in Fig. 5) were defined as the extrusion force (F_E). The values of F_E are shown in Table 2. Figure 6 shows the extrusion force (F_E) plotted against the WPG determined by the PF resin impregnation (listed in Table 1). The F_E values tended to decrease with increasing WPGs at all RHs. The result indicates that the extrudability of the wood improved with increasing PF resin content of the wood.

Wood samples were extruded under low force after they had been conditioned at a higher RH. This result indicates that the extrudability was higher for a higher MC (Fig. 1(a)).

The increases in the PF resin and moisture in wood contributed to soften the wood, resulting in improved the extrudability (Shams *et al.* 2004; Miki *et al.* 2012, 2013, 2014a). The differences of F_E between the RHs decreased with increasing WPG. These differences seem to be affected by the differences in MC of wood samples (Fig. 1(a)) which have a remarkable effect on the wood softening.

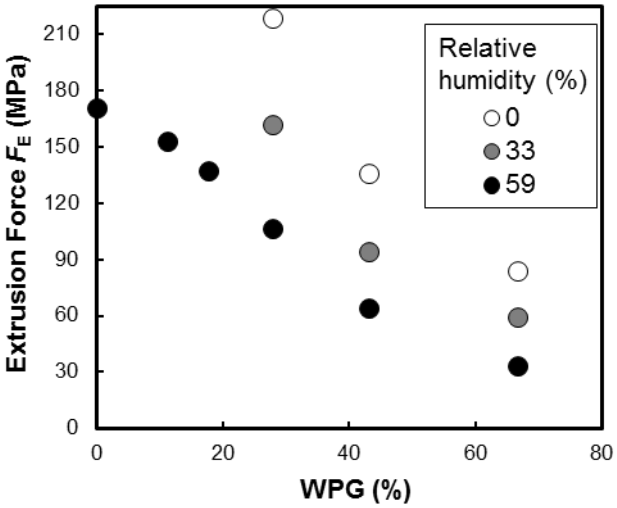

Fig. 6. Relationship between WPG of the wood samples due to PF resin impregnation and extrusion force (F_E) of wood samples prepared with various relative humidities (RHs)

Mechanical Properties of the Extrudate

The results for the specific gravity and mechanical properties of the extrudate are summarized in Table 2. Figure 7 shows the relationship between WPGs of the wood samples due to PF resin impregnation and specific gravity, Young's modulus, and the bending strength of the extrudate obtained from the wood samples conditioned at different RHs. The specific gravities of the extrudates increased until the WPG became 18%, then gradually decreased with increasing WPG. The specific gravities of the wood substance and cured PF resin were 1.45 and 1.30, respectively. These results suggest that the extrudates at the higher WPG (≥ 18%) had almost no voids, whereas the extrudates at the lower WPG (< 18%) contained large amounts of voids. Because the specific gravity of cured PF resin (1.30) is smaller than that of the wood substance (1.45), the specific gravity of the extrudates decreased with increasing WPG in the higher range (≥ 18%). No remarkable differences were seen between the conditioning RHs of the blank wood samples.

Young's modulus and the bending strength of the extrudates increased with increasing WPG in the lower range (< 18%) at 59% RH. However, Young's modulus decreased, and the bending strength approached a constant value at the higher WPG (≥ 18%) for all RHs. To discuss the reasons for these mechanical changes, the fracture surface of the extrudate were observed (Fig. 8). Long wood fibers were seen on the fracture surface of the extrudate without the PF resin (Fig. 8(a)), suggesting that there were gaps or weak points between the fibers. Further evidence for the existence of gaps is that the specific gravity of the extrudate was markedly lower than the others WPG (Fig. 7(a)), which seems worsened the mechanical properties. The gaps were also seen on the surface at the lower WPG (Fig. 8(b)). However, there is no fibrous form and gap on the surface at the higher WPG (Figs. 8(c), (d), (e), and (f)). It is suggested that the adhesion between the fibers was strong enough to separate each other, which contributed to improve the mechanical properties. The results of both bending test and microscopy have shown that the PF resin has improved the interfacial adhesion between the wood fibers, resulting in increased mechanical properties. The most effective WPG was found to be approximately 18%.

Both Young's modulus and the bending strength became lower with increases in the conditioning RHs of the wood samples (Figs.7(b) and (c)). The results reveal that the mechanical properties of the extrudate were reduced by increasing the MC of the blank wood samples.

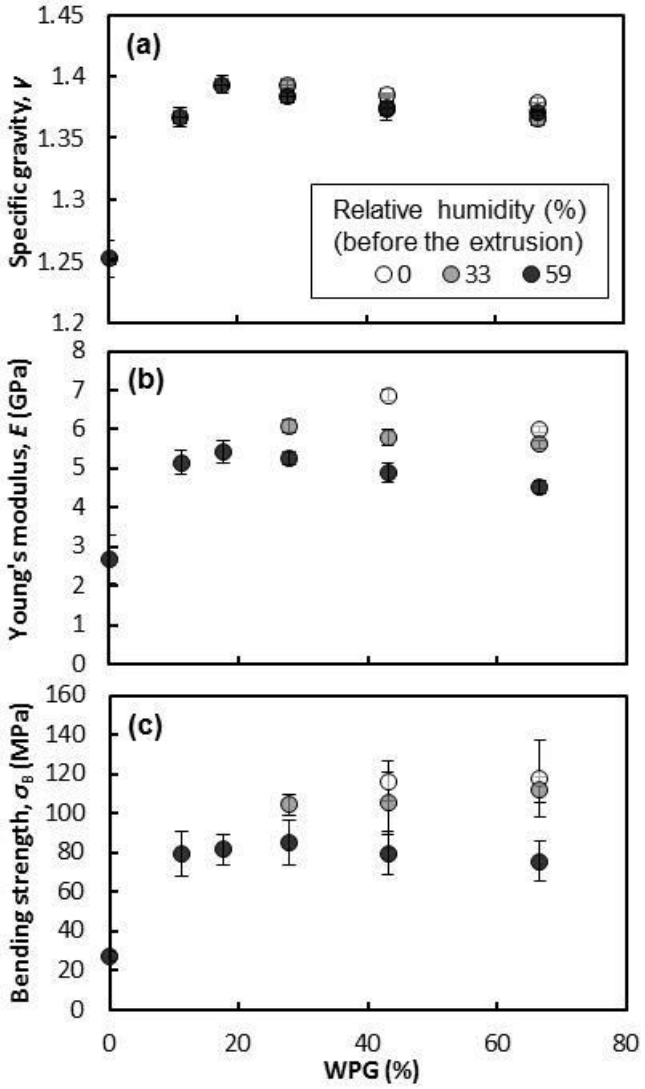

Fig. 7. Relationships between WPG of the wood samples due to PF resin impregnation and specific gravity (γ), Young's modulus (E), and bending strength (σ_B) of the extrudate obtained from the wood samples conditioned at different relative humidities (RHs)

The reasons why the mechanical properties of the extrudates worsened with increasing MC of the blank woods were considered as follows:

(1) The residual moisture content (RMC) of the extrudates was different between the extrudates obtained from different MCs of the blank wood samples.

(2) The water in the wood samples affected the chemical changes, for example, curing reaction of the PF resin and the wood during extrusion, because the wood samples were subjected to high temperature and pressure.

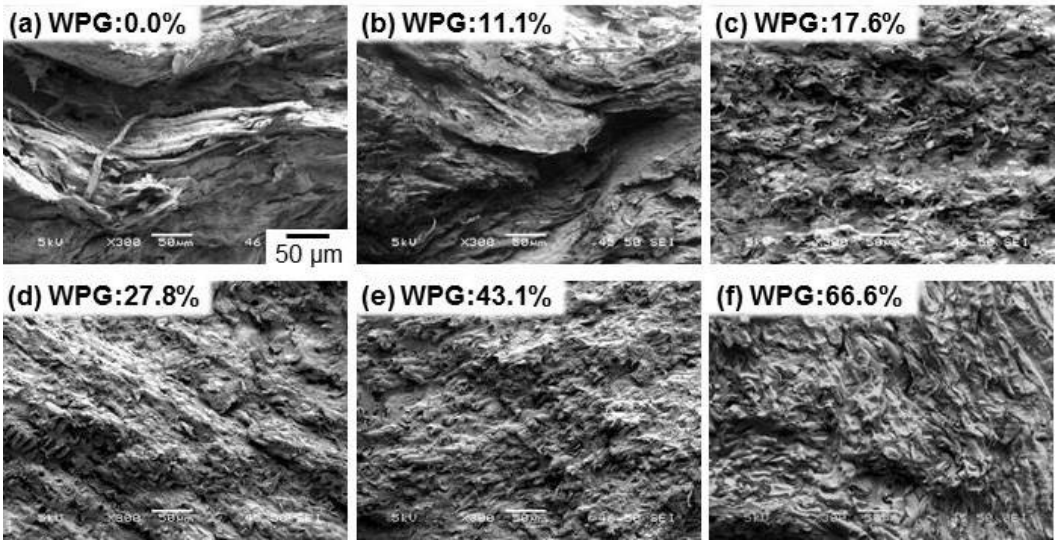

Fig. 8. SEM micrographs of fracture surface of extrudates obtained from wood samples conditioned at 59% RH

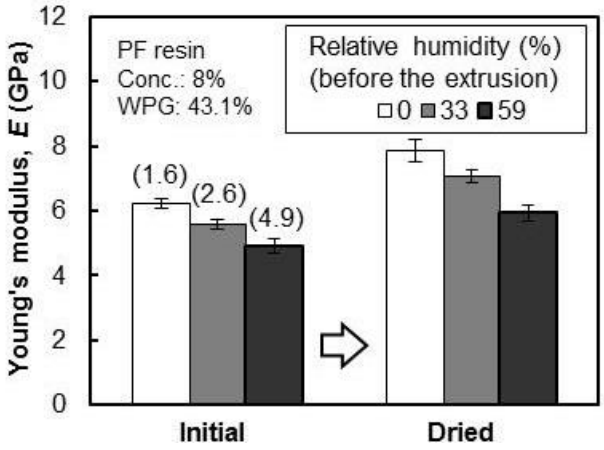

Fig. 9. Changes of Young's modulus of the extrudates before (Initial) and after drying (Dried) treatments. The extrudates were obtained from the wood samples impregnated with a PF resin concentration of 8% and conditioned at different relative humidities (RHs). The values given in parentheses are the residual moisture contents (RMCs) of the extrudates at the initial state.

To investigate the effect of the RMC on the mechanical properties of the extrudates (reason 1), the extrudates (conditioned under 20 °C at 59% RH; initial state) were completely dried in a vacuum chamber (0.01 MPa) at 50 °C for 48 h. The Young's modulus of the dried specimens was measured by the bending test. Figure 9 shows the changes of Young's modulus for the initial and dried extrudates obtained from wood samples conditioned at different RHs; RMCs of the extrudates at the initial state are also shown.

The MCs of the blank wood samples conditioned at RH of 0%, 33%, and 59% were 0%, 2.4%, and 4.9%, respectively (Fig. 1(a)). The RMCs of the initial extrudates (parentheses in Fig. 9) seem to be related to the MC of the blank wood samples. Because the PF resin had a dehydration reaction during the curing process, the RMC of the extrudate

became larger than the MC of the wood sample. The result indicates that most of the water remained in the mold during the extrusion process.

The Young's modulus of all extrudates was increased by drying. This is probably due to the decrease of water in the wood substances; the decreased water enhances the intermolecular interaction. The decreasing trend of Young's modulus in the dried extrudates with increasing the conditioning RHs of the wood samples was similar to that in the initial extrudates. The result indicates that the difference of RMCs of the extrudate have little effect on the difference of their mechanical properties.

To investigate the effect of water on the chemical changes of the PF resin and the wood substance during the extrusion (reason 2), FTIR measurements were performed on the extrudates obtained from the wood samples conditioned at different RHs (Fig. 10). The FTIR spectra were normalized by an absorption peak at 1508 cm^{-1}, which corresponds to the C=C stretching vibrations of the benzene ring, which is present in lignin (wood component) and PF resin.

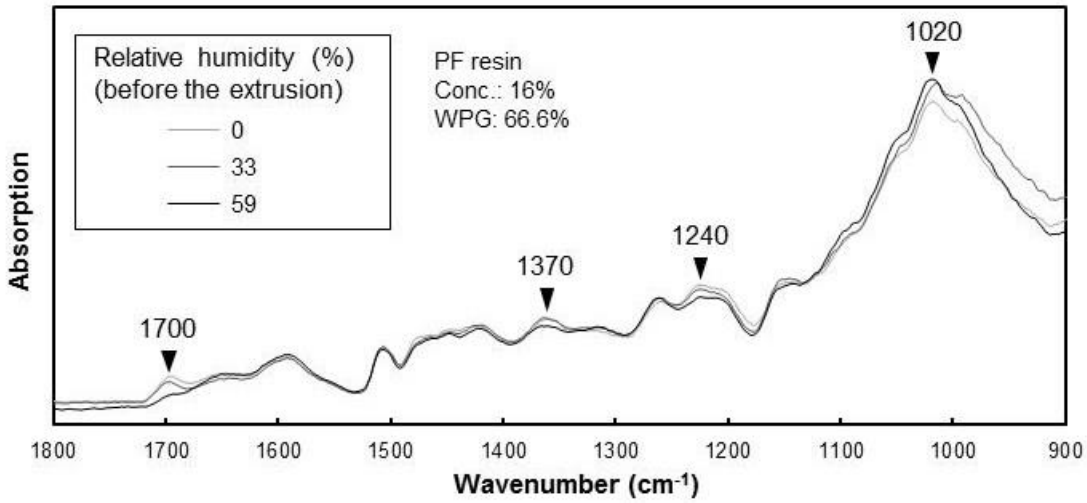

Fig. 10. FTIR spectra of extrudates obtained from wood samples conditioned at different relative humidities (RHs)

The absorbance peak at 1020 cm^{-1} decreased with decreases in the conditioning RHs of the wood sample. The decrease in absorbance at 1020 cm^{-1}, which corresponds to the hydroxymethyl C-O stretch of PF resin, probably occurred by polymerization (curing) of the PF resin due to dehydration (Poljanšek and Krajnc 2005; Valdez and Nagy 2010). The result indicates that the existence of water during the extrusion process disturbed the polymerization of the PF resin or promoted the degradation of the PF resin (Lorenz and Christiansen 1995).

The peaks at 1240 cm^{-1}, 1370 cm^{-1}, and 1700 cm^{-1}, which correspond to the C=O stretch in the acetyl group of hemicellulose, C-H bending vibrations of cellulose, and C=O stretch of hemicellulose, respectively, decreased with increases in the conditioning RHs of the wood sample. The decrease in the intensities of the bands probably resulted from degradation of the wood by the heat and steam (Temiz *et al.* 2006; Yin *et al.* 2011). The mechanical properties of wood worsen due to steam treatment under 200 °C compared with the heat treatment without water (Inoue *et al.* 1993).

From the results, the chemical changes of wood substances and PF resin due to the existence of water in the wood samples, which were subjected to high temperature and pressure during extrusion, worsened the mechanical properties of the extrudate. It is important to release the moisture from inside the mold during the extrusion process in order to improve the mechanical properties of the extrudate.

CONCLUSIONS

1. The extrudability of wood impregnated with PF resin improved with increases in both PF resin and MC of the blank wood due to the softening of the wood. The extrusion force of wood impregnated with PF resin conditioned at the higher RH (59%) was less than one-half that of the dried wood.
2. The mechanical properties of the extrudate worsened with increases in moisture content of the blank wood impregnated with PF resin; the Young's modulus of the extrudate obtained from the wood samples conditioned at the higher RH (59%) were about 30% lower than that of the extrudate obtained from the dried wood samples. Because most of the water in the wood remained in the mold during the extrusion process, the chemical changes of the wood substance and the PF resin occurred due to steam under the high temperature and the pressure of the process. The chemical changes worsened the mechanical properties of the extrudate.
3. Wood with a high moisture content (conditioned at 59% RH) could be extruded without PF resin. However, the mechanical properties of this extrudate were worse than those of the extrudate containing PF resin; the bending strength of extrudate without PF resin was 50% lower than that of extrudate contained PF resin.

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