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## Effect of thermoplastic binder on flow deformation behavior of wood

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### Abstract

A processing technique was recently developed to realize plastic deformation in wood impregnated with a thermoset binder. This paper proposes a new flow forming technique where a thermoplastic binder is used instead of a thermoset binder with the expectation that the formability and recyclability of the products will be improved. To clarify the effect of the thermoplastic binder on the flow deformation behavior of wood, capillary fluidity tests were performed using impregnated wood with various contents of thermoplastic binder (polymer). The extrusion load of the impregnated wood through the capillary decreased with an increase of the polymer content in the wood. Results of the second fluidity test using the first extruded material reveals that the recycled impregnated wood can flow again. The extrusion load of the second extrusion was equal to or lower than the first. These results indicate that the recyclability of the wood impregnated with a thermoplastic binder is highly promising. The internal configuration of the impregnated wood during extrusion was also dependent on the polymer content.

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## 1. Introduction

Wood is a natural resource and a sustainable material with potential as a countermeasure against global warming by fixing carbon dioxide. Wood has been conventionally shaped by cutting processes, which results in the generation of much waste until a final product is obtained, and is thus an inefficient process. Wood can also be formed into simple-shaped products by deformation processes such as bending and compression. However, more complex-shaped products are not typically formed in industry, because of the poor formability of wood and the inefficiency of the wood forming.

Recently, we have reported interesting flow deformation behavior of wood (Yamashita et al., 2006; Miki et al., 2012; Miki et al., 2013). This flow deformation behavior of wood can be applied using the various plastic forming techniques used for industrial materials such as metals and polymers, and is currently being developed with wood flow forming techniques. In the techniques, a thermoset binder such as phenol formaldehyde resin and melamine formaldehyde resin are impregnated into the wood to reduce the forming pressure and improve the durability of the product. However, it is difficult to control the fluidity of wood formed with a thermoset binder because the impregnated wood becomes hardened by heat during the forming process. Moreover, the products obtained when using a thermoset binder are non-recyclable.

This paper proposes a new flow forming technique that employs a thermoplastic binder and could enable wood to be processed into more complex forms and with improved recyclability. The objective of this study is to clarify the effect of the thermoplastic binder on flow deformation behavior of wood and examine the feasibility and problems of using a thermoplastic binder in forming processes. A thermoplastic monomer (methyl methacrylate) was impregnated into wood and polymerized using heat. Capillary fluidity tests were conducted on the impregnated wood with various contents of thermoplastic binder (polymer). The capillary fluidity test was conducted twice using the same impregnated wood to examine the fluidity of recycled wood materials impregnated with polymer. Finally, the configuration of the extruded material was investigated by observation, and the effect of the polymer content in the impregnated wood is discussed.

## 2. Materials and experimental procedure

### 2.1. Materials

Japanese cypress (*Chamaecyparis obtusa*) was the wood used in this study. To evaluate the polymer content of the impregnated wood during production of the material, specimens were cut into 10 mm (longitudinal direction, L) × 9 mm (tangential direction, T) × 10 mm (radial direction, R) cuboids. Furthermore, rotary-cut veneers (ca. 3 mm thick (R)) were cut into 25 mm diameter (L×R) disks for the capillary fluidity test. All specimens were dehydrated in an oven at 105 °C for more than 3 h.

### 2.2. Production of impregnated wood materials

Methyl methacrylate was used as the thermoplastic binder in this study. To increase the affinity between hydrophilic wood and hydrophobic monomer, and to control the content of thermoplastic binder in the wood, methyl methacrylate monomer was diluted with ethanol to produce various concentrations and then introduced into the wood specimens under vacuum (Furuno et al., 1978). A mass equivalent to 1% of the methyl methacrylate monomer of a polymeric initiator (V-601, Wako Pure Chemical Industries, 10 h half-life decomposition temperature: 66 °C) was added to the monomer solution. After soaking in the solution under atmospheric pressure at 30 °C for 20 h to achieve impregnation, the three dimensions (L, R, and T) of the cuboid specimens were measured. The impregnated specimens were then hermetically sealed in aluminum foil to suppress monomer volatilization and were placed in an oven at 85 °C for approximately 24 h to polymerize the monomer. The specimens were then vacuum dried at 50 °C for 5 h to remove unreacted monomer and ethanol. The polymer contents of the impregnated wood specimens were evaluated based on the oven-dried weight of untreated wood and the vacuum-dried weight of impregnated wood.

### 2.3. Capillary fluidity test

Capillary fluidity tests were conducted using a press machine (Saginomiya, Series V-1815) and the mold shown in Fig. 1. The diameter of the barrel was 30 mm and a die having a capillary with 10 mm in length and 4.0 mm in diameter was used to study the fluidity. Impregnated wood materials (25 g) were stacked in the same direction of fiber orientation and placed manually into the barrel, followed by the punch, and the die was then placed in the testing instrument. The die block surface was heated to 150 °C, which can soften the impregnated wood and is sustainable for thermal degradation (Okumura et al., 1977). After 10 min of equilibration, the punch was moved by displacement control (5 mm/min) until the distance from the capillary die surface was 2.0 mm. The impregnated wood materials were compressed and extruded through the capillary while the force loaded to the punch ( $F$ ) was recorded. The sampling time was 500 msec..

To estimate the fluidity of recycled wood materials impregnated with thermoplastic binder, 20 g of extruded material was supplied into the barrel again. The recycled material was pre-compressed with a 10 kN load at room temperature and the load was then released. The temperature of the die was elevated to 150 °C and held for 10 min to equilibrate. The second fluidity test was then conducted under the same conditions as the first test.

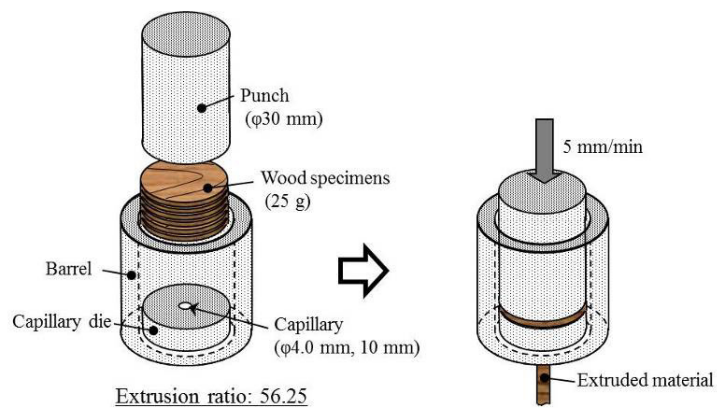


Fig. 1. Schematic illustration of capillary fluidity test (first test) apparatus.

## 3. Results and discussion

### 3.1. Polymer content and internal configuration of impregnated wood

The polymer content in various mole fractions of methyl methacrylate monomer solution are shown in Table 1. The polymer content was slightly different between the cuboid (a) and disk (b) specimens. Both specimens showed a similar increase in the polymer content with an increase in the monomer concentration. These results indicated that the polymer content in the impregnated materials and the product can be controlled by changing the concentration of the monomer solution.

Fig. 2 shows representative scanning electron microscopy (SEM) micrographs of transverse sections of polymer-impregnated cuboid specimens and untreated specimens. Wood is a porous material that has many voids, such as the cell lumen (black part in Fig. 2(a)). As the impregnated polymer content increases, the cell lumens fill with polymer (▼ in Figs. 2(b) and (c)) also increased. At the highest polymer content (Fig. 2(c)), the polymer was densely filled in a large majority of the cell lumens. Furuno et al. (1978) reported that polymer was present not only in the cell lumen, but also in the cell wall, when wood swelled during impregnation with a methyl methacrylate solution in ethanol. Therefore, it is considered that the cell walls of specimens impregnated with methyl methacrylate mole fractions of 0.125 to 0.75 are also impregnated with the polymer.

Table 1. Polymer content of impregnated wood specimens.

Mole fraction of methyl methacrylate solution in ethanol	0.125	0.25	0.375	0.5	0.75	1
(a) Polymer content of cuboids (%)	23.8	34.5	45.6	49.6	54.5	57.0
(b) Polymer content of disks (%)	19.4	32.2	42.2	50.8	55.5	58.0

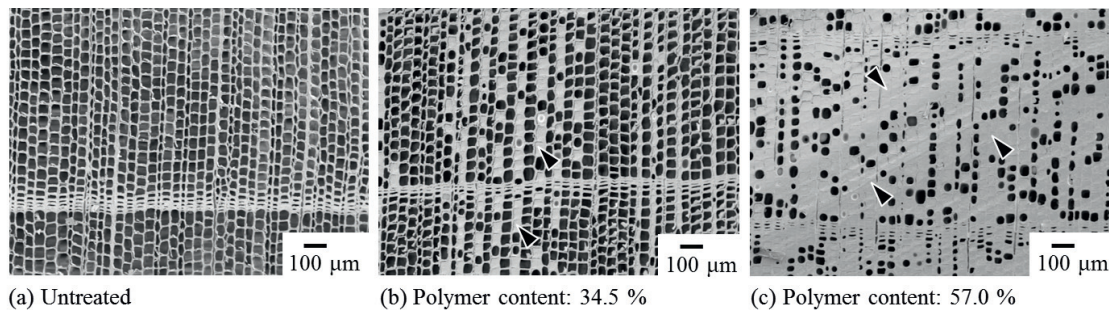


Fig. 2. Transverse section SEM micrographs of representative polymer-impregnated cuboid wood specimens. (▼: Cell lumen filled with polymer)

### 3.2. Flow deformation behavior of wood impregnated with thermoplastic binder

Fig. 3 shows the variation of load during the capillary fluidity test of wood impregnated with various polymer contents, where load ( $F$ ) is plotted against the distance between the punch and the die,  $x$ . At first, the load remained lower, during which time the materials were significantly deformed and packed. Eventually, the load rapidly increased until the material in the barrel began to flow.

Fig. 3(a) shows the results for the first fluidity test using the disk materials. The untreated wood did not flow, but was only compressed until the end of the test. The impregnated wood started to flow at yield points of load (▼). After the yield point, the load vibrated and stayed relatively steady during extrusion of the material. The load starting point of flow and the load during extrusion differed significantly, especially with lower polymer content (<42.2%), where the load was higher with a lower polymer content in the material. Fig. 3(b) shows the results for the second fluidity test using the recycled extruded materials obtained from the first test. The material with a polymer content of 19.4% did not flow during the second test, whereas the materials with other polymer contents began to flow at a lower load than that in the first tests. The results indicate that the wood structure of the disk specimens was destroyed to small chips, which enabled flow at the yield point during the first test. In the second test, the surfaces of the wood chips in the recycled material could easily slide against each other, which is the weak point of these materials.

Fig. 4 shows the influence of the polymer content of the materials on the yield point of load ( $F_Y$ ) for both the first and second fluidity tests.  $F_Y$  for the first test decreased with increasing polymer content and became constant above approximately 50%. For the second test,  $F_Y$  was decreased continuously with increasing polymer content. Thus, the extrusion load was affected significantly by the polymer content of the material.

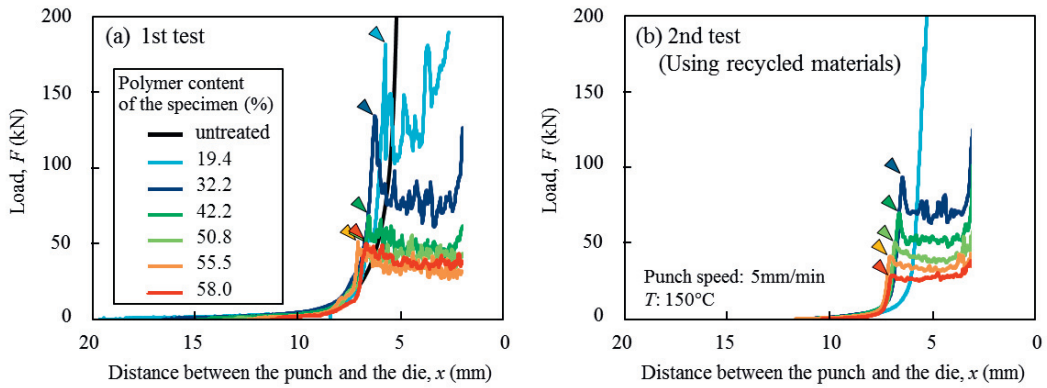


Fig. 3. Variation of load during capillary fluidity tests using impregnated wood materials with various polymer contents.

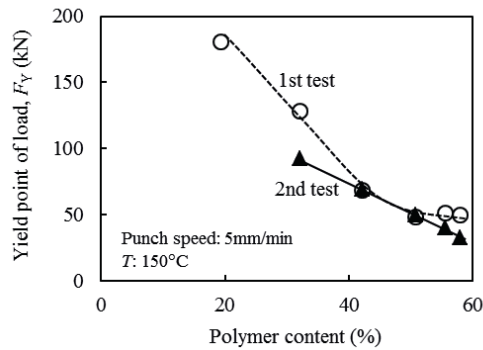


Fig. 4. Effect of polymer content in impregnated wood materials on the yield point of load ( $F_{\gamma}$ ) during extrusion for both the 1st and 2nd fluidity tests.

Fig. 5 shows photographs of typical extruded specimens with various polymer contents from the first capillary test. There was no significant difference in the form for polymer contents above 32.2% ((a) through (c)). However, the surface of the extruded material with a polymer content of 32.2% (c) was rougher than the materials with higher polymer contents. In contrast, the extruded material with a polymer content of 19.4% (d) collapsed and chip-shaped materials were obtained.

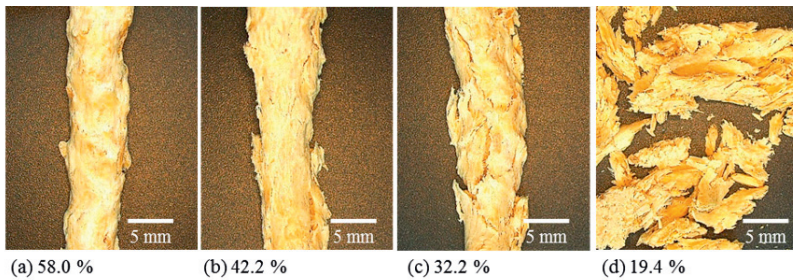


Fig. 5. Extruded wood materials (first capillary fluidity test) with various polymer contents.

Fig. 6 shows representative transverse section optical micrographs of polymer-impregnated wood materials that remained in the capillary die during the first fluidity test. Each micrograph indicates that the material configuration during extrusion is completely different from that of the material before extrusion shown in Fig. 2; most of the wood cells are deformed and some of the wood cells changed their relative positions. The wood cells of the higher polymer content materials (ca. >50 % (a) and (b)) were deformed, but not completely compressed. Because the materials with high polymer contents originally had a few voids, and a large majority of the cell lumens were filled with the impregnated polymer (Fig. 2(c)). On the other hand, the lower polymer content wood materials (c) were consolidated; most of the cell walls were buckled and the volume of the cell lumen is decreased. These results indicate that the internal configuration of the extruded material is significantly dependent on the polymer content.

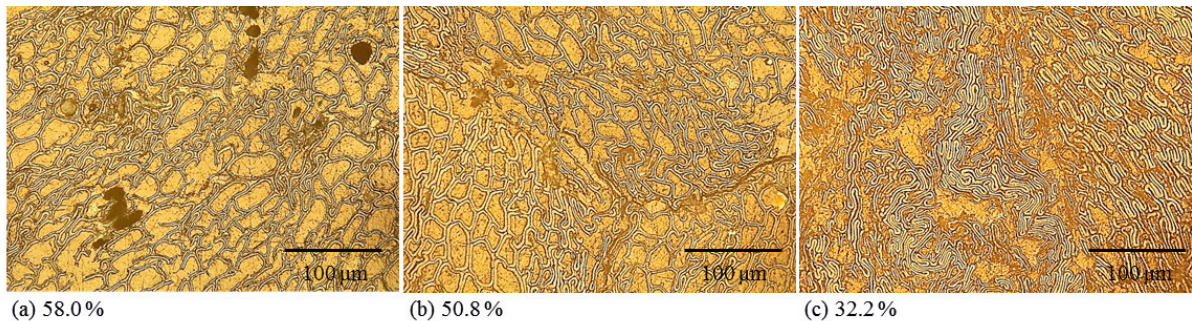


Fig. 6. Transverse section optical micrographs of representative polymer-impregnated wood materials that remained in the capillary die.

#### 4. Conclusions

A thermoplastic binder instead of a thermoset binder was introduced into wood in an attempt to develop new wood flow forming techniques that could enable wood to be processed into more complex forms and improve recyclability. The fluidity of wood impregnated with the thermoplastic binder can be controlled according to the binder content in the materials. The extrusion load decreased with an increase of the polymer content in the material. The recyclability of the impregnated wood is highly promising, especially for polymer contents greater than approximately 30 %. The internal configuration of the extruded material differed significantly with the polymer content of the material; therefore, the physical and mechanical properties of the products would be influenced by the polymer content.

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