Journal of Materials Processing Technology 239 (2017) 140–146

Contents lists available at ScienceDirect

Journal of Materials Processing Technology

journal homepage: www.elsevier.com/locate/jmatprotec

Forward extrusion of bulk wood containing polymethylmethacrylate: Effect of polymer content and die angle on the flow characteristics

Masako Seki[a,*], Soichi Tanaka[b], Tsunehisa Miki[a], Ichinori Shigematsu[a], Kozo Kanayama[b]

[a] Structural Materials Research Institute, National Institute of Advanced Industrial Science and Technology, Nagoya, Aichi 463-8560, Japan
[b] Research Institute for Sustainable Humanosphere, Kyoto University Gokasho, Uji, Kyoto 611-0011, Japan

ARTICLE INFO

Article history:
Received 1 November 2015
Received in revised form 27 June 2016
Accepted 11 August 2016
Available online 20 August 2016

Keywords:
Wood
Thermoplastic
Composite
Forward extrusion
Die angle

ABSTRACT

In this study, the forward extrusion of non-pulverized bulk wood containing polymethylmethacrylate (PMMA) was investigated to obtain new fundamental insights regarding the plastic forming of bulk wood. In particular, we studied the effect of the polymer content and die angle (20°–120°) on the extrusion forces and flow of various types of bulk wood during extrusion. Plastic-impregnated wood (composite) specimens with high (49%) and low (27%) polymer content were obtained by impregnating wood with solutions containing different concentrations of methyl methacrylate (MMA). Before the impregnation, the wood was hydrophobized by acetylation to allow the MMA to penetrate into the cell walls. The load during extrusion for the 49% composite was lower than that for the 27% composite at all die angles. For both composites, the extrusion force increased with decreasing die angle for small die angles (20°–40°) because of the increase in friction force at the die wall surface. For the 27% composite, the extrusion force increased with increasing die angle beyond 40° due to the formation of dead zones and shear zones in the wood. Therefore, the optimal die angle that minimized the extrusion force was near 40°. In contrast, the extrusion force for the 49% composite exhibited no significant difference for die angles larger than 40° in spite of the formation of dead zones and shear zones. Chevron cracking appeared in the 49% composites, whereas no cracks and voids were formed in the 27% composites.

1. Introduction

Wood is a sustainable natural resource, and long-term utilization of wood can alleviate global warming since wood-based materials contain carbon derived from carbon fixation as a constituent. Many attempts have been made to use wood-based materials as industrial products in an eco-friendly manner. Wood-plastic composites (WPCs) are an excellent example of such materials, which are generally produced by compounding wood flour with thermoplastic resin followed by forming processes such as extrusion and injection. WPCs are widely used in construction (Dittenber and GangaRao, 2012) and in the automotive industry (Ashori, 2008). One of the major problems of WPC is poor compatibility between the wood flour and the polymeric matrices, which results in a poor dispersion of the wood flour within the matrix and poor mechanical properties. Another disadvantage is the pulverization process for the wood, which requires considerable energy and makes it expensive to produce WPCs from bulk wood.

Sandberg et al. (2012) summarized techniques for non-pulverized wood shaping such as compression and bending processes. However, only products with simple shapes can be obtained using these processes, because the deformation of solid wood requires deformation at the cellular level. In recent years, Yamashita et al. (2009) have discovered flow phenomena for bulk wood to obtain more complex products from non-pulverized bulk wood, and our group has been developing plastic forming techniques based on these phenomena. The products obtained retain semblances of the wood grain to some extent and reduce fracturing of the wood fiber, so they offer the attractive appearance of wood grain as well as relatively strong mechanical properties (Miki et al., 2012). In order to generate wood flow, the wood is impregnated with some additives (thermosetting binder or thermoplastic binder). This causes the cells of the wood to soften during forming and allows slipping dislocation between neighboring cells (i.e., flow) under heat and pressure. Miki et al. (2013) reported that to some degree, it is possible to control material fluidity during form-

* Corresponding author.
E-mail address: m-seki@aist.go.jp (M. Seki).

http://dx.doi.org/10.1016/j.jmatprotec.2016.08.014
0924-0136/

ing as well as the product characteristics by selecting the type and quantity of additives.

Additionally, forward extrusion, which is a common plastic forming method used with metallic materials, has been applied to bulk wood forming. In a series of our studies, we (2016a) conducted a capillary extrusion, and showed that treating wood by acetylation and increasing the polymer content improve the fluidity of wood materials impregnated with polymethylmethacrylate (PMMA). We (2016b) also reported the effect of repetitive extrusion and polymer content on the mechanical properties of the extrudates.

Moreover, we have studied the effect of the geometry of the extruding die. It is well known that the die angle is an important factor for controlling the extrusion force and product characteristics in metallic materials forming. In a previous study, we (2014) investigated the effect of the metal die angle (20°–50°) on the extrusion force using bulk wood containing a large amount (55%) of PMMA, and the results showed that the extrusion load increased with decreasing die angle under no-lubricant conditions. However, there are no known investigations on the effect of the polymer content, which is an important factor that affects the fluidity of the wood and the characteristics of the final product, or the effect of using die angles of greater than 50° on the extrusion force.

The objective of this study was to identify how the wood polymer content and the metal die angle affect the flow of bulk wood during extrusion and the resulting extrusion forces. Specifically, we extruded bulk wood specimens containing small and large amounts of PMMA (27% and 49%) in the radial direction while widely varying the die angle between 20° and 120°, and measured the load on the punch. In the material preparation process, wood was impregnated with two solutions containing different concentrations of methyl methacrylate (MMA) to obtain the low (27%) and high (49%) polymer content specimens. Before the impregnation, the wood was hydrophobized by acetylation to allow the MMA to penetrate into the cell walls. In addition, the specimens left in the die after the extrusion test were observed using an optical microscope to evaluate structural changes caused by extrusion.

2. Materials and methods

2.1. Preparation of wood specimens

Pieces of Japanese cypress (*Chamaecyparis obtusa*) of dimensions 9.5 mm (longitudinal direction, L) × 69 mm (radial direction, R) × 15 mm (tangential direction, T) were used for the preparation of the wood specimens. They were successively cut from a larger stick with a transverse section of 69 mm (R) × 15 mm (T).

The wood was dried at 105 °C, weighed, and hydrophobized by acetylation in order to raise the affinity between the hydrophobic additives and the wood (Obataya and Shibutani, 2005). In the acetylation process, the wood was soaked in acetic anhydride under vacuum and heated to 120 °C for 24 h. The acetylated wood was then washed in water to remove the remaining acids, dried at 105 °C for 24 h, and weighed again. On average, the relative increase in weight due to acetylation was 16%. The dried acetylated wood was combined with PMMA during subsequent processing. To control the PMMA content of the wood specimens, MMA was diluted with ethanol and then impregnated into the acetylated wood under vacuum. The mole fraction of MMA in the solution was either 0.75 or 0.25 to provide a large contrast between the impregnated specimens; it was reported that the amount of the polymer in the cell lumens and cell walls is remarkably different between the two solutions, and this affects the fluidity (2016a). A mass of polymeric initiator (V-601, Wako Pure Chemical Industries, 10 h half-life, decomposition temperature: 66 °C) equivalent to 1 wt.% of the MMA was added to the MMA solution in advance. The impregnated wood was soaked in the solution at 35 °C under atmospheric pressure for 24 h to allow the MMA to penetrate into the cell walls. The soaked wood was then wrapped tightly in aluminum foil to suppress MMA volatilization, and then heated to 120 °C for 24 h to polymerize the MMA in the cell lumens and within the cell walls. The polymerized woods were then dried at 105 °C for 24 h to remove unreacted MMA and ethanol. The polymer (PMMA) content of the wood specimens after drying, as calculated from the post-acetylation weight, was about 27% and 49% for the 0.25 and 0.75 mole fraction MMA solutions, respectively.

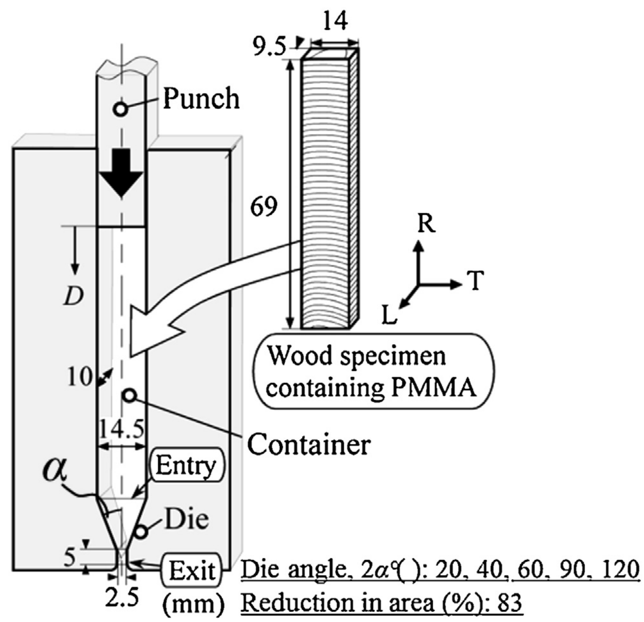

Fig. 1. Schematic illustration of the extrusion test.

2.1.1. Thermal analysis

A small test piece (1.5 × 1.5 × 1.5 mm^3) cut from the wood specimens containing PMMA was subjected to dynamic mechanical analysis (DMA; SII NanoTechnology Inc., TMA SS6100). An oscillating compressive load of 200 ± 100 mN in the R direction was applied to the test piece and dry nitrogen gas was purged during the measurement, while heating from 30 to 200 °C under a constant temperature ramping rate of 3 °C/min.

2.1.2. Extrusion test

The flow stress for wood exhibits remarkable anisotropy in mutually perpendicular directions; Yamashita et al. (2009) and Miki et al. (2012) reported that the force required for flow perpendicular to the fiber direction is substantially lower than that for flow in the fiber direction. To obtain flow mainly perpendicular to the fiber direction in this study, only one of the two pairs of die wall surfaces in the rectangular-shaped die was oriented in a taper during the extrusion test (see Fig. 1); the other pair was oriented in a parallel manner. Specifically, to suppress flow in the fiber direction during extrusion, the specimens were placed so that their end grain faces (TR faces) laid against the parallel die walls surfaces (i.e., the LR faces laid against the taper), and were forced in the R direction. The tapered dies were made of SKD11 alloy tool steel and their half-angles (α) were 10°, 20°, 30°, 45°, and 60° (i.e., the die angle 2α was 20°, 40°, 60°, 90°, and 120°). The widths of the die entry and die exit were 14.5 mm and 2.5 mm, respectively, for an extrusion ratio of 17.2. The stroke of the die from the entry to the exit was 34.0 mm for 2α = 20°, 16.5 mm for 2α = 40°, 10.4 mm for 2α = 60°, 6.0 mm for 2α = 90°, and 3.5 mm for 2α = 120°. The die surfaces were polished