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Thermoplastic behavior of wood powder compacted materials

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Abstract To investigate the possibility and problems in recycling techniques for wood powder compacted material (WPCM) obtained by steam compression, a dynamic mechanical thermal analysis (DMTA) was conducted in dry and wet conditions. The results obtained show that mechanical properties such as the static Young's modulus and bending strength of WPCM increased with an increasing steam temperature up to 170 °C during the compression of the wood powder. It is emphasized that WPCM having a bending strength of 80 MPa with a Young's modulus of 8 GPa can be prepared by steam compressing of wood powder only due to a auto-condensation of wood components. The DMTA showed that the relative storage Young's modulus of WPCM dramatically decreased in water exposure by heating, although it slightly increased in the dry condition. This indicates that WPCM is softened under heat in the presence of water, but it becomes harder by heating without water. The loss tangent peaks showed that the softening behavior of WPCM seems to result from lignin. Consequently, it is thought that WPCM can be shaped by compressing in water, and after obtaining the desired shape, the drying process should be conducted to fix the shape and harden the WPCM.

Introduction

Due to the remarkable increase in the quantity of carbon dioxide (CO_2) emitted to the atmosphere, environmental problems associated with global warming have become a serious problem. The development of new techniques and materials that will enable the reduction of CO_2 emissions in the atmosphere has drawn the attention of many researchers in various fields. In the field of material development, environmentally friendly materials as well as eco-materials have been the focus of research.

Wood-based products promise a good future because their uses lead to decrease in the amount of CO_2 in the atmosphere. Long-time use of wood products containing CO_2 as their components results in the reduction of CO_2 in the atmosphere. During the disposal stage of these products, although CO_2 is generated, it is needed by trees for their photosynthesis activity. For such reasons, wood-based materials are sustainable resources as well as environmentally friendly material, and wood resources can be an abundant quantity on the earth if appropriate afforestation and deforestation are performed. As future industrial materials, materials should be developed from sustainable resources such as wood biomass instead of oil-based products.

Recently, composites made from mixtures of wood or natural fibers and polymers have been developed [1–6], and such combined products, which are called wood polymer composites (WPC), have expanded the market of building structural materials in the USA [7]. These composites may be a good way, in terms of eco-material, to use wood resources effectively; however, these products have been produced by using a lot of oil-based plastics or binders. Due to such oil additives, recycling techniques including secondary forming have been hardly developed.

A plastic-like material can be obtained by the compression of wood powder only under an appropriate temperature and pressure [8, 9]. It is considered that this change in surface texture comes from auto-condensation of

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the wood components. The mechanical properties of the wood powder compacted material (WPCM), such as the bending strength, is roughly the same as that of plastics such as ABS (Acrylonitrile Butadiene Styrene), and due to the auto-condensation of wood components, WPCM can be enhanced and hardened. These positive outcomes mean that WPCM might be applied as a substitute material for existing plastics. However, present studies concerning the compaction of wood biomass resources tend to discuss the mechanical properties of WPCM without additional processing using deformation ability. Little research has considered the behaviors under heating and moisture conditions with respect to the recycling and secondary forming of these products.

In this study, the compaction of wood powder was performed by the steaming compression process, and the mechanical properties of WPCM obtained in the experiments were measured. Then, for considering the thermoplastic behavior of WPCMs, a dynamic mechanical thermal analysis (DMTA) was conducted in dry and wet conditions using a thermal mechanical analyzer (TMA). The effect of the vapor steaming temperature during WPCM production on the mechanical properties are discussed, and the storage elastic modulus (E') behavior of WPCMs was obtained from the dynamic mechanical thermal analysis (DMTA) from 30 °C to 100 °C in wet and dry conditions. The possibilities and problems in developing recycling techniques including secondary processing of the WPCM are discussed.

Material and methods

Preparation of WPCM

Coniferous wood powder consisting of a mixture of Japanese cedar and cypress, was used as the raw material. The powder was prepared by crushing mixed cedar and cypress chips (1:1 in weight) with a Wiley crusher machine and screening (less than 355 μm). The powder passing through the 355-μm mesh was defined as the mixed wood powder. A microscopic photograph and the size distribution of the mixed powder are given in Fig. 1 and Table 1, respectively. As seen in Fig. 1, the powder has mainly a fibrous shape of various sizes, and the aspect ratio is a maximum of 12. The initial moisture content (M.C.) of the wood powder was conditioned in air (M.C. = about 10%). Any special treatments such as the addition of an adhesive were not carried out in the experiment.

A compressing machine with steaming made by HI-SAKA Corporation is schematically illustrated in Fig. 2. This steaming compressing machine consists of a boiler, a chamber and a compressing machine; the compressing machine is installed in the chamber. The saturated vapor for steaming has a temperature of more than 100 °C and can be produced under a vapor pressure of 0.1–0.8 MPa in the boiler. This vapor is transported into the chamber, in which metal molds that pack the wood powder (25 g) are compressed under the constant pressure P = 36.5 MPa. The inside temperature of the chamber, which was filled with vapor during compression of the wood powder, is defined as the steaming temperature T and varied from 100 °C to 170 °C by controlling the quantity of saturated vapor. In this experiment, the compressing pressure was kept constant for 30 min, and the removal of WPCM was carried out after cooling and unloading.

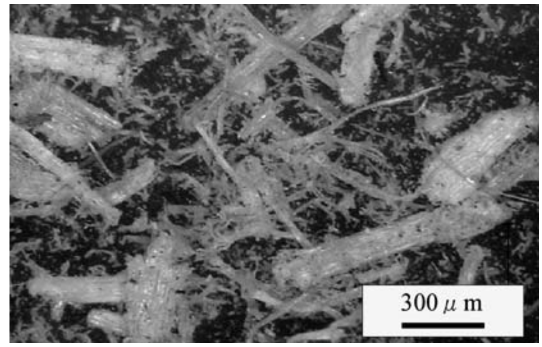

Fig. 1 Micrograph showing mixed wood powder

Table 1 Powder particle size distribution

Size μm	Weight %
Less than 53	20.52
53–90	8.76
90–106	3.43
106–150	8.15
150–180	7.47
180–250	13.27
250–355	14.18
More than 355	22.27

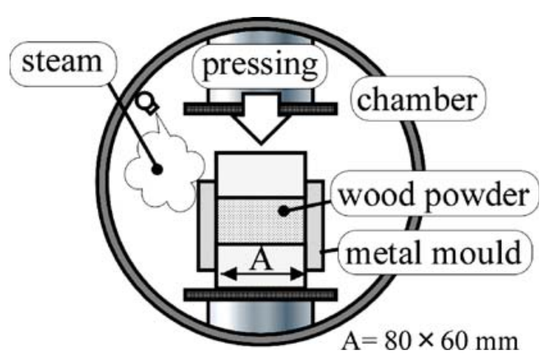

Fig. 2 Schematic diagram of steaming compression process

Mechanical properties of WPCM

To understand the effects of the production conditions on the mechanical properties of WPCM, bulk density measurements, static bending tests and Charpy impact tests were performed using specimens cut into 10-mm-wide sections according to Japan Industrial Standard (JIS) K 7171, 7111. For each specimen, the bulk density ρ, static bending strength σ, and Charpy impact strength ac were obtained. Also, for the rigidity of WPCM, Young's modulus of bending was obtained from the bending stress and strain curve at a small strain rate up to 0.4%, which can be assumed to be linear.

These mechanical tests were conducted for specimens obtained under the same production conditions of WPCM at least five times.

Dynamic mechanical thermal analysis of WPCM

The thermal mechanical analyzer (TMA/SS6100, Seiko Instruments Inc.), interfaced with EXSTAR 6000, which is an automatic dynamic viscoelastometer, was used to determine the storage Young's modulus E' and loss tangent tan δ. A schematic diagram showing the DMTA of the WPCM is shown in Fig. 3. To conduct the DMTA in a wet condition, a sample bottle filled with distilled water was installed in the TMA. The WPCM specimen for the DMTA was cut into a round shape less than 10 mm in diameter. The sample bottle, bottle case and probe were entirely made of quartz because of its small thermal expansion. An experimental compressive oscillation was applied to the WPCM through the quartz probe within 30–100 °C in dry (air) and wet (distilled water) conditions. The average oscillation load of 70 g in compression was set with an amplitude of 30 g and frequency of 0.1 Hz. The temperatures of the air and water heated by the jacket were measured by the thermopile, which was set near the WPCM. From the change of the oscillation load and displacement when heating the air or water, the storage Young's modulus E' and loss tangent tan δ were obtained in relation to the measured temperature T_d. As E' changes, E' at a given temperature divided by the E'_{in} value at 30 °C in both the air and water; this feature will be discussed for understanding the thermoplastic behavior of WPCMs. Furthermore the set of dry-wet cycle of the DMTA was performed three times to provide data on its influence on thermoplastic behavior and water resistance.

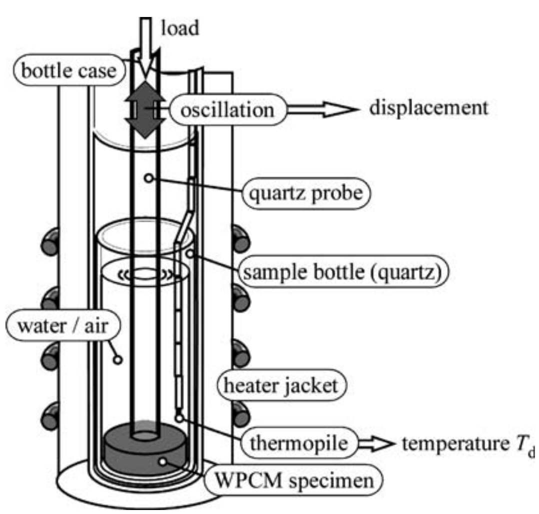

Fig. 3 Schematic diagram showing dynamic mechanical analysis of wood powder compacted material in dry/wet conditions

Results and discussions

Mechanical properties

Figure 4 shows a photograph of the appearance of WPCMs made by the steaming compression process of wood powder only. There is a great difference in surface texture due to the steaming temperature T. The material surface is darker when temperature T is higher, and a superficial gloss is generated at the black portions. This smooth surface comes from the transfer of the metal mold surface and it results from the degradation and condensation of the wood components. The black portion of the material expands when increasing the steaming temperature T.

Figure 5 shows the effects of the vapor steaming temperature T on the bulk density, static bending strength, bending Young's modulus and Charpy impact strength of the WPCMs. The bulk density ρ increases with the steaming temperature T under constant compressive pressure $P = 36.5$ MPa, and it reaches the maximum at 150 °C. This tendency is similar to those of the static bending strength and Young's modulus. By this steaming compression process, WPCM with the static bending strength of 75 MPa and Young's modulus of 8 GPa can be obtained with comparably smaller pressure than a heating compaction process without steaming [8]. This difference is caused by the existence of moisture during the compaction of the wood powder. Basically, the compressive deformability of wood is affected by the moisture content and heating temperature in the compression. This can result from the softening phenomenon of wood components such as lignin and hemicelluloses, which are softened with heat and moisture [10]. Furthermore, in the steaming compression process, since saturated vapor can be provided for the wood powder above the temperature of 100 °C, the moisture content will not decrease, unlike normal heating compression.

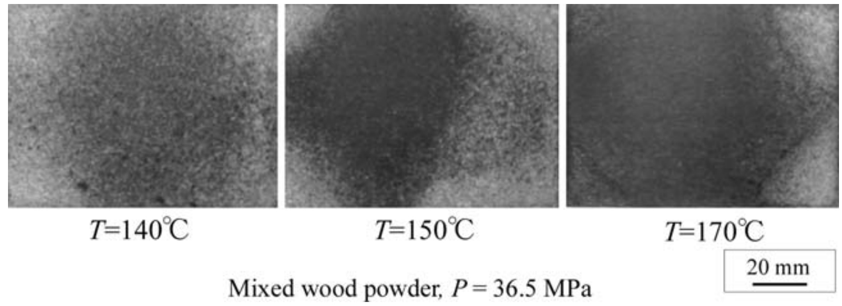

Fig. 4 Appearance of wood powder compacted materials

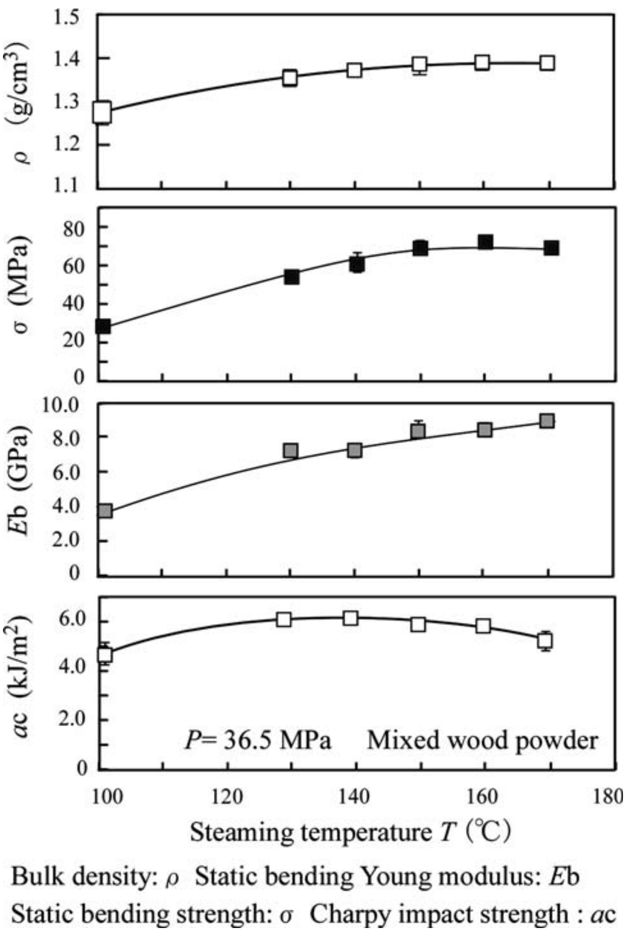

Fig. 5 Effects of steaming temperature on mechanical properties of WPCM

Therefore, the steaming compression of the wood powder is effective in producing WPCM with lower pressure. However, in terms of the Charpy impact strength, it slightly decreases with increasing temperature. This means that a static mechanical property such as bending strength can be improved when the steaming temperature T is set higher, but a dynamic property such as impact strength becomes worse due to the occurrence of brittleness. Thus, there should be optimum vapor temperature conditions in the production of sound WPCM.

Thermoplastic behavior of WPCM in dry and wet conditions

Figures 6 and 7 show the effect of the measured temperature T_d of the DMTA in the dry condition on the relative storage Young's modulus E'/E'_{in} and loss tangent tan δ, respectively. E'_{in} means the initial storage Young's modulus at 30 °C for each WPCM obtained by the given steaming temperature T, and E' is a measured value at elevated temperature T_d in the DMTA. It is seen from Fig. 6 that E'/E'_{in} increases by 10% when the measured temperature T_d is increased in the dry condition. This indicates that WPCM in the dry condition becomes hardened at a higher temperature, up to 100 °C. The hardening rate of WPCM becomes large when the steaming temperature is set higher, and it reaches the maximum at 160 °C. In contrast, when the steaming temperature is set too high, that is, as much as 170 °C, the hardening rate becomes smaller. This hardening seems to be derived by a level difference in the auto-condensation of the wood component

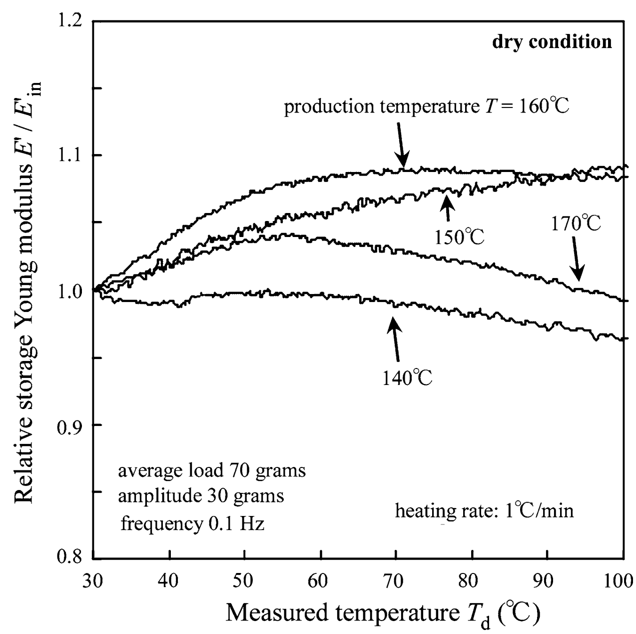

Fig. 6 Effect of measured temperature on relative storage Young's modulus of WPCM in the dry condition

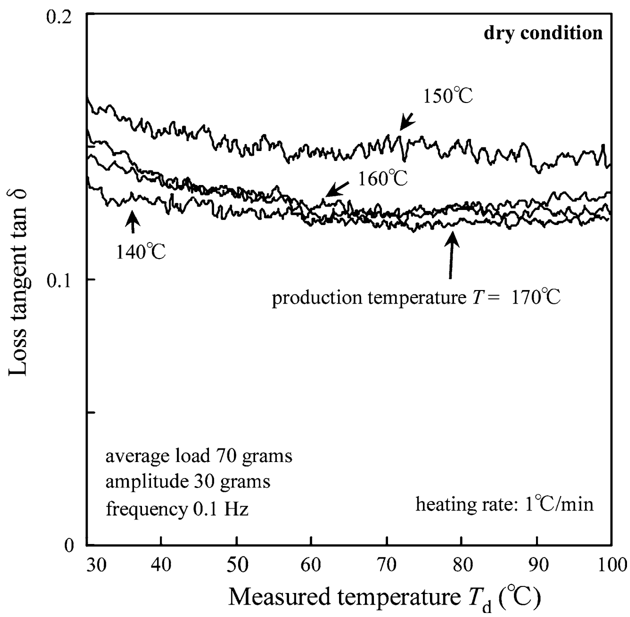

Fig. 7 Effect of measured temperature on the loss tangent of WPCM in the dry condition

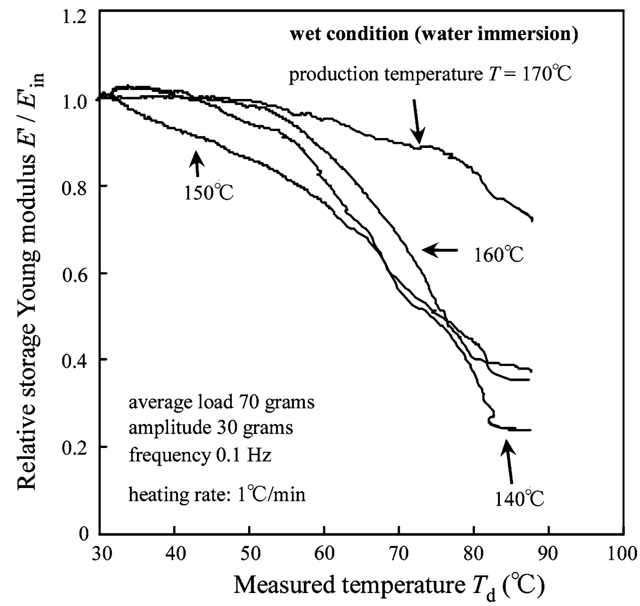

Fig. 8 Effect of measured temperature on the relative storage Young's modulus of WPCM in the wet condition

in production, as well as the decreasing moisture content of the WPCM. As seen in Fig. 5, the bending strength increases when the steaming temperature is increased up to 160 °C. This increased bending strength is caused by autocondensation of wood components such as hemicelluloses and lignin. However, the steaming temperature of 170 °C leads to too much degradation of the components. Thus, the Charpy strength decreases due to insufficient molecular binding. The decreased moisture content can provide increased strength in wood materials; therefore, WPCMs obtained at 150 °C and 160 °C would harden at an elevated temperature in the DMTA. In the case of T = 140 °C, the hardening is hardly observed because the shape fixation would be generated by mainly mechanical binding, like an anchor effect, and not by condensation of the components. In the case of T = 170 °C, hardening also occurs due to decreasing moisture, but softening also begins from roughly 60 °C to 100 °C in the measured temperature T_d due to small molecule chains resulting from too much degradation. It is found from Fig. 7 that the loss tangent tan δ slightly decreases with increasing temperature regardless of the steaming temperature T in the production of WPCM. In the dry condition, WPCM does not show any softening properties even if the temperature reaches as high as 100 °C. It is concluded that WPCM in the dry condition is hard to be deformed.

Figures 8 and 9 show the effect of the measured temperature T_d of the DMTA in the wet condition on the relative storage Young's modulus E'/E'_{in} and loss tangent tan δ of the WPCM, respectively. It is obviously seen from Fig. 8 that E'/E'_{in} in the wet condition rapidly decreases with increasing temperature regardless of the steaming temperature T. When the steaming temperature in the production of WPCM is lower, the decrement of E'/E'_{in} becomes larger; especially, the WPCM of T = 140 °C shows almost 80% of the E'/E'_{in} decrease while the WPCM of T = 170 °C shows less than a 30% decrease. This tendency of the E'/E'_{in} decrease can be seen in the change of loss tangent tan δ given in Fig. 9. The loss tangent tan δ increases with increasing temperature T_d from 40 °C, and

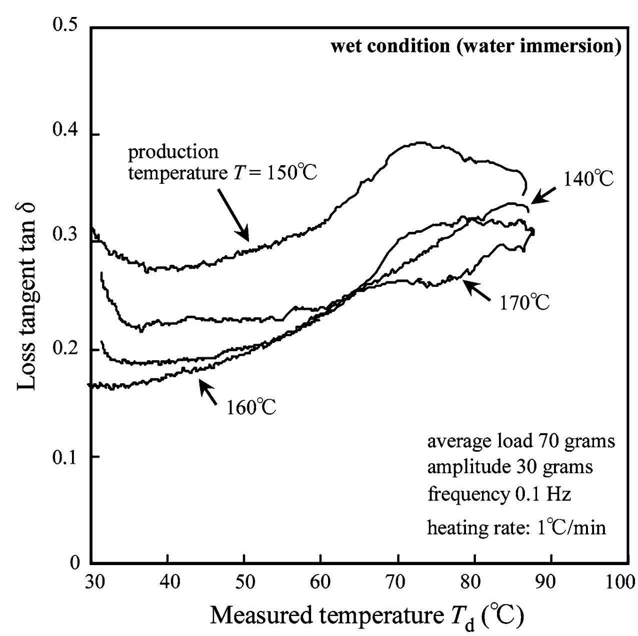

Fig. 9 Effect of measured temperature on the loss tangent of WPCM in the wet condition

the measured values of tan δ for given temperatures T_d in the wet condition are entirely larger than those in the dry condition. There seems to be a broad peak of tan δ for each WPCM of T = 140 °C, 150 °C and 160 °C. Especially, it is clearly recognized that tan δ of the WPCM obtained at T = 150 °C has a local maximum at around T_d = 70 °C in the wet condition. This indicates that the mechanical properties of WPCM are dramatically affected by heat and water as with natural wood properties, and it softens. Therefore, the WPCM can be formed by compressing in a wet condition at elevated temperature.

Repetitive DMTA in dry and wet conditions

To investigate changes of the thermoplastic behavior of WPCM subjected to dry-wet cycles during heating, where a wetting condition follows a drying in a cycle, repetitive dynamic mechanical thermal analyses in dry and wet conditions were performed. By this measurement, multi-deformability as well as durability of the WPCM against repeated dry and wet conditions can be determined.

In the experiment, the WPCM was completely dried by an oven conditioned at 105 °C for 2 h between the cycles.

Before the DMTA measurement in each wet condition, the setup displacement of the probe changed due to deformation of WPCM under the constant load of 70 g. The changes in the displacement during the given period seem to depend on the steaming temperature T in the production of WPCM. This can be summarized in Fig. 10 as the effect of the steaming temperature T on the deformation displacement under constant pressure in the repetitive measurement. It is recognized that deformation is hardly seen in the dry conditions, namely the 1st, 3rd, 5th drying regardless of the temperature T of the WPCM, even if the DMTA is conducted after the wet conditions, namely, the 2nd, 4th and 6th wetting. By repeating the dry-wet cycle by three times, the displacement in wet conditions gradually increases. In particular, the WPCM made at 140 °C fractured due to excessive expansion in the wet condition of the 6th. However, the deformation displacement decreases when the production temperature of WPCM is set higher. This indicates that WPCM made in high-temperature steaming has improved durability related to its dimensional stability against exposure to water. Also, without water, it is difficult for WPCM to be deformed at room temperature.

Figure 11 gives the effect of the measured temperature T_d on the relative storage Young's modulus of WPCM in the repetitive DMTA. In this figure, the relative storage Young's modulus E'/E'_{1st} of the WPCMs is defined based on the storage Young's modulus of the WPCM obtained at T = 160 °C of the production condition at T_d = 30 °C of the measured temperature in the dry condition. It is seen that E'/E'_{1st} in the 1st dry condition slightly increases as the temperature is increased within 30–100 °C, as shown in Fig. 6. In the 2nd wet condition, E'/E'_{1st} is higher compared with the 1st dry condition up to about T_d = 70 °C, and it rapidly decreases to half the value above 70 °C. The reason is not clear, and further work is necessary to investigate this behavior. In the 3rd dry condition after the 2nd wet condition, E'/E'_{1st} becomes higher by 1.5–1.8

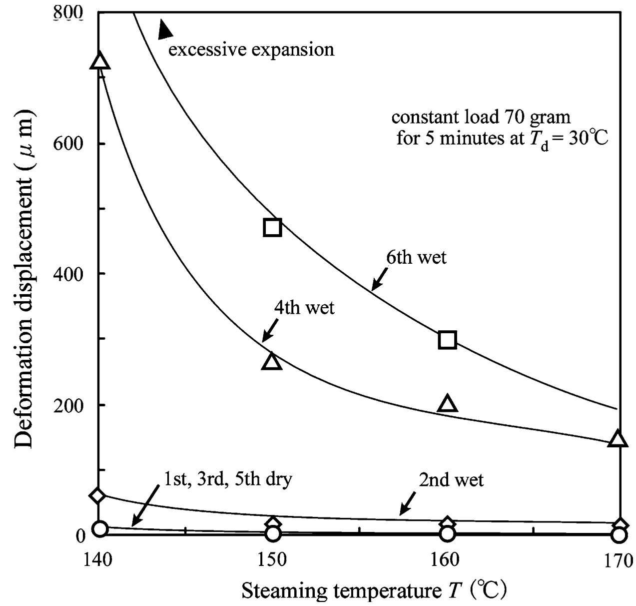

Fig. 10 Effect of steaming temperature of WPCM on the deformation displacement under constant pressure in repetitive DMTA

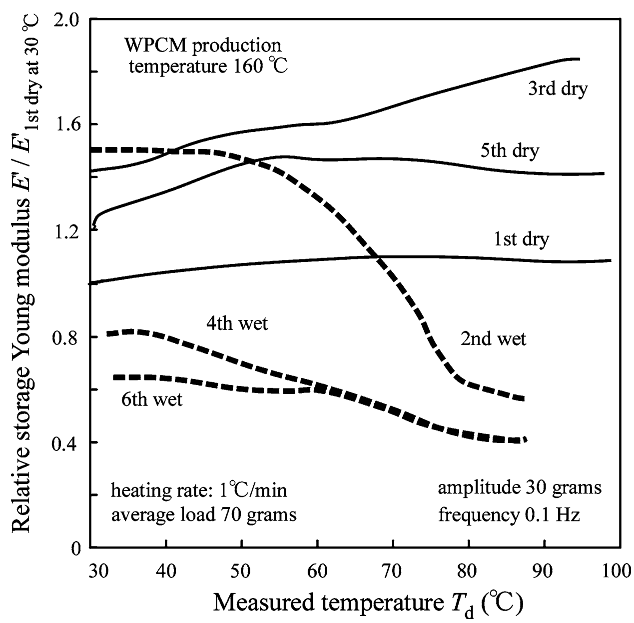

Fig. 11 Effect of measured temperature on relative storage Young's modulus of WPCM in repetitive measurements

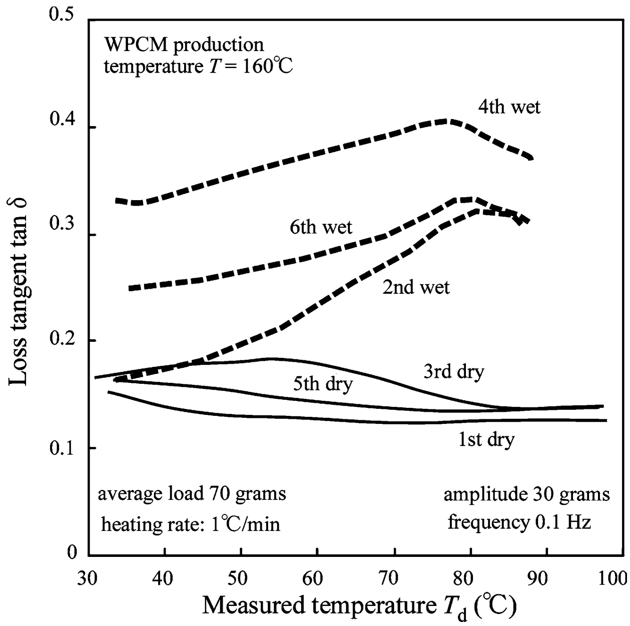

Fig. 12 Effect of measured temperature on loss tangent of WPCM in repetitive measurements

times of that for the 1st dry condition. This trend can also be seen in the 5th dry condition. Since the water-soluble substance, which was generated from the WPCM in the sample bottle, was observed after the 2nd wet experiment, the increment of E'/E'_{1st} might occur due to the removal of the substance. This substance will be investigated in accordance with the mechanical properties of WPCM in the future. However, in the 4th and 6th wet conditions, E'/E'_{1st} decreases by heating, and above T_d = 60 °C it shows almost the same values. This indicates that WPCM can be softened and deformed with several cycles of water. Thus, the following can be considered for forming techniques: WPCM can be shaped by a type of compressing in water since formability can be applied in water, and after obtaining a shape, the drying process of WPCM should be conducted to fix the shape and harden it.

Figure 12 shows the effect of the measured temperature T_d on the loss tangent tan δ of WPCM in the repetitive measurements. The tan δs in the dry conditions (1st, 3rd and 5th) show relatively smaller values, ranging from 0.1 to 0.2, and the tan δs in the wet conditions (2nd, 4th and 6th) give larger values. Furthermore, the tan δs in the wet conditions have peaks at around 80 °C. Many reports have stated that the thermal softening phenomenon of lignin, which is a main component of wood, can be seen around 80 °C [11–15]. Therefore, it is concluded that the softening mechanism of the WPCM results mainly from the thermal behavior of lignin.

Conclusion

To investigate the possibility and problems in the development of recycling techniques of wood powder compacted material (WPCM), a dynamic mechanical thermal analysis (DMTA) was conducted in dry and wet conditions using a thermal mechanical analyzer (TMA) for WPCM obtained by various vapor compression processes. Mechanical properties such as the static Young's modulus and bending strength of WPCM increased with an increasing steaming temperature during compression of the wood powder up to 170 °C. It must be emphasized that WPCM having the bending strength of almost 75 MPa with the Young's modulus 8 GPa can be prepared only by steam compressing of the wood powder. It is found from the DMTA that WPCM is dramatically softened with water exposure by heating up to 100 °C, although in the dry condition it becomes hard by heating. As determined from the loss tangent peak of WPCM, the softening behavior seems to come from the lignin behavior. For forming techniques, WPCM could be shaped by compressing in water since formability can be applied with water exposure, and after obtaining a shape, the drying process of WPCM should be conducted to fix the shape and harden it.

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