Effects of Drying Temperatures on the Occurrence of Sticker Stain in Japanese Cedar (*Cryptomeria japonica* D. Don)

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Abstract

Sticker stain is a material defect that results from moisture migration during wood drying, often spoiling the appearance of the surface of wood products. The effect of drying temperatures on the occurrence of surface sticker stain was investigated using Japanese cedar (*Cryptomeria japonica* D. Don) and three types of stickers (air-dried Japanese cedar, aluminum, and stainless steel) under four drying temperatures (20°C, 50°C, 75°C, and 100°C). At lower drying temperatures, the air-dried wood sticker tended to suppress the occurrence of surface sticker stains, whereas higher temperature or metal stickers produced sticker stains with deep color. However, no definitive relation was shown between the initial moisture content and the extent of sticker stain with deep color regardless of drying temperatures. It was considered that the partial delay of drying happened around the contact area with stickers, especially in cases of metal stickers. It was also found that the use of metal stickers at higher drying temperatures induced depressions in Fourier transform infrared spectra related to the occurrence of hygrothermal conditions.

In the wood industry, drying is one of the essential processes in using wood resources as woody materials, structural materials, and so on. This process often accompanies some material defect caused by the loss of moisture content in wood cell walls, such as check, warp, twist, cup, and discoloration (Kollmann and Côté 1968). In the drying process of wood, stickers are usually placed between sawn pieces of lumber in order to stack them and ensure suitable ventilation. However, it is well known that such placement of stickers usually involves the occurrence of surface discolorations called *sticker stain* or *sticker mark* (Kollmann and Côté 1968). This surface stain not only spoils the appearance of wood but also reduces the commercial value of wood products.

Some factors are considered to have a role in the occurrence of sticker stain during wood drying, such as the condensation of extractives and their chemical changes. Ellwood et al. (1960) reported that moisture migration in wood cell walls produced the partial deposition of water-soluble extractives on the surface area of wood after drying. Similar phenomena in extractive migration due to drying were also reported in cases of hardwood species (Miller et al. 1990) and softwood species (Shibutani et al. 2006, Okuda et al. 2017). Previous studies on Japanese cedar also suggested that kiln drying under heating conditions at 70°C or more involved quantitative reduction in terpenes and norlignans (Shibutani et al. 2006, Okuda et al. 2017). In addition, some researchers investigated the suppression method for sticker stains by adopting chemical predrying treatments (Xu and Clement 2008) and fumigation processes with some fumigants (Amburgey et al. 1996; Schmidt et al. 1997, 2001).

Most lumbers are usually exposed under high-temperature conditions at around 100°C in the case of kiln-drying processes in the Japanese wood industry. For example, temperatures above 100°C are commonly given as the predrying treatment for boxed-heart timber of softwood species, such as Japanese cedar and hinoki cypress, after steaming at around 90°C for the purpose of suppressing

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Forest Prod. J. 71(3):209–215.
doi:10.13073/FPJ-D-20-00079

surface checks (Yoshida et al. 2000, 2004; Kuroda 2007; Yamashita et al. 2012). However, a few studies obtained detailed information about the effects of drying temperature on the occurrence of sticker stain under such severe conditions, even though the market for wood products in Japan demands better appearance, such as structural lumber without checks in all lateral surfaces.

The purpose of the present study was to investigate in detail the effect of drying temperatures on the occurrence of sticker stain on Japanese cedar. Therefore, the surface conditions of sticker stain arising as a result of drying treatments are discussed based on the results obtained after drying.

Materials and Methods

Materials

The studied specimens were prepared from the heartwood of boxed-heart square timber of Japanese cedar (*Cryptomeria japonica* D. Don) from Ibaraki Prefecture, Japan. Flat-sawn specimens with dimensions of 100 by 15 by 40 mm (longitudinal by radial by tangential) were sawn from the surface of boxed-heart square timber under green conditions. The ovendried density of specimens was 0.34 ± 0.03 g/cm^3 (mean ± SD). The initial moisture content of specimens, which was obtained by the ovendrying method, was 75.3 ± 26.9 percent.

Stacking and drying conditions

Eight specimens were stacked into two tiers with four lines in a sealed desiccator with the bark side up, as shown in Figure 1. One kilogram of weight was placed on the stacked specimens to ensure contact with the sticker during drying. Drying was done under 12 experimental conditions that differed according to three types of stickers and four temperature conditions as follows. Stickers of wood (air-dried Japanese cedar), aluminum (A5052), and stainless steel (SUS304) with 5-mm thickness and 10-mm width were used to stack the specimens. The sealed desiccator with stacked specimens was placed in a temperature-controlled room at 20°C or an electric oven at 50°C, 75°C, and 100°C to keep the temperature constant. A saturated salt solution was also placed in the desiccator to ensure constant relative humidity until the equilibrium moisture content reached 6 to 9 percent. Table 1 shows the experimental conditions for drying the specimens in this study. Saturated solutions of K_2CO_3, NaBr, NaBr, and NaCl were used under temperatures of 20°C, 50°C, 75°C, and 100°C, respectively. Relative humidity was retained at approximately 43, 50, 51, and 74 percent under each temperature condition, respectively (Kollmann and Côté 1968, Society of Polymer Science 1968, Saito and Shida 2016). The weight of specimens was intermittently measured to determine the end of drying. Reaching equilibrium moisture content took 76, 20, 8, and 3 days, respectively.

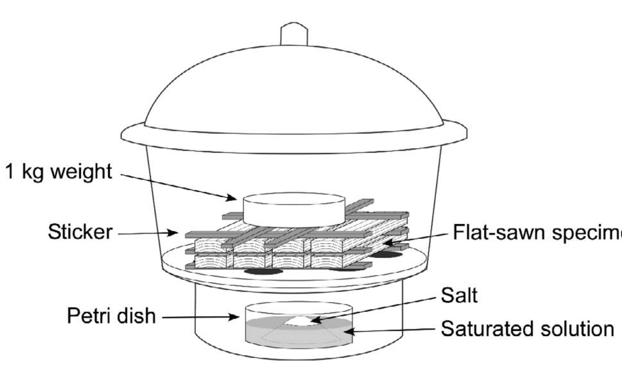

Figure 1.—Schematic diagram of the stacked flat-sawn specimens using sticker during drying.

Table 1.—Experimental conditions for drying specimen.

Drying temperature (°C)	Saturated solution	Relative humidity (%)	Drying period (days)
20	K_2CO_3	43	76
50	NaBr	50	20
75	NaBr	51	8
100	NaCl	74	3

Classification and evaluation of sticker stains

The surface of each specimen had two contact areas, with stickers on both bark and pith sides (Fig. 1). The occurrence of sticker stain was visually classified into three types after drying: no sticker stain, reduced shade, and deep color. Figure 2 shows typical examples. For sticker stain with deep color, the total area was also evaluated using image processing software (ImageJ, version 1.52a, National Institutes of Health) based on the binarized image. The surface images of specimens were collected using a scanner (MP490, Canon Inc., Tokyo, Japan). Image processing was not performed for stain with reduced shade because it was difficult to make an accurate determination.

Macroscopic observation of the cross section under the sticker

The central part of each sticker stain was sawn perpendicular to the fiber direction using a circular saw to obtain the cross section, and the macroscopic condition of the discoloration inside was confirmed in each specimen. The images of cross section were also obtained using the scanner (MP490, Canon Inc., Tokyo, Japan).

Fourier transform infrared spectroscopy

Fourier transform infrared (FTIR) spectra were obtained using a Fourier transform infrared spectrophotometer (FT/IR4700, Jasco Corporation, Tokyo, Japan) equipped with an attenuated total reflectance (ATR) device with a single-reflection diamond crystal (ATR Pro One View, Jasco Corporation, Tokyo, Japan) to investigate the effects of the drying temperature on the occurrence of surface sticker stains. FTIR/ATR analyses were conducted directly on the surface of each dried specimen. The incident angle of the infrared light through the diamond crystal was 45°. The spectral resolution, accumulation number, and measurement range were 4 cm^{-1}, 16 times, and 4,000–400 cm^{-1}, respectively.

Results and Discussion

Classification of sticker stains

Figure 3 shows the occurrence rate of surface sticker stains at each drying temperature. It was found that lower

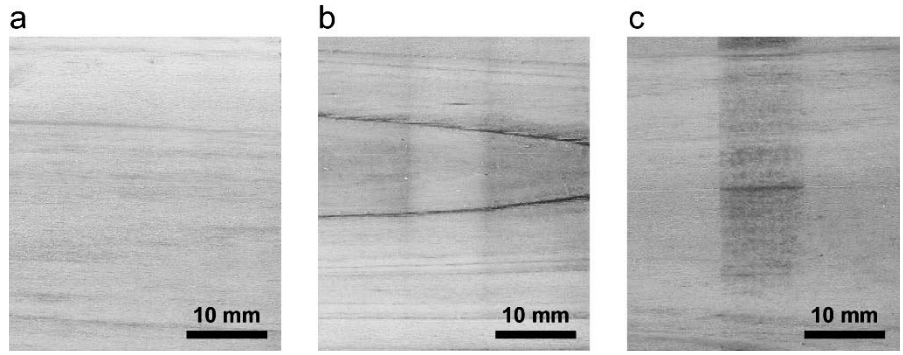

Figure 2.—Typical example of surface stains at the contact areas of sticker after drying: (a) no sticker stain, (b) reduced shade type, and (c) deep color type.

drying temperatures often provided clean surfaces without sticker stain regardless of the type of sticker. Similar surface conditions were often found in the case of wood sticker regardless of drying temperature. On the other hand, the occurrence rate of sticker stains became higher with increasing drying temperature, especially in the case of metal stickers, which tended to produced deep-colored stains (Fig. 2c). It is considered that the contact with metal stickers prevented wood specimens from drying under the usual conditions. The use of wood stickers made of air-dried Japanese cedar was considered to slightly suppress this condition due to the migration of free water from the wet specimen to the sticker of drying wood. However, certain tendencies were not found in the case of the reduced shade of sticker stain (Fig. 2b), and there was no difference in occurrence rates between the two surfaces (bark and pith sides).

Figure 4 shows the typical results of surface sticker stain and internal staining conditions. The use of wood sticker or lower drying temperatures (Figs. 4a and 4d) less likely involved internal stains, whereas the use of metal stickers or higher temperature resulted in the frequent occurrence of internal stains (Figs. 4b and 4c). However, a definitive relationship was never found between the surface and internal stains at this time. It should be noted that the partial bias of black color in the binarized image of cross sections that were not due to the effects of cell wall tissues (e.g., the difference between earlywood and latewood) was even found a few millimeters from the surfaces (Figs. 4b and 4c). This black color also indicates that the surface stain arising from the use of stickers might remain after surface finishing.

Relationship between initial moisture content and rate of discolored area

Figures 5 and 6 shows the relationship between the average initial moisture content of the specimen and the area ratio of deep-colored sticker stains on each surface (bark and pith sides). No significant correlation was found for all drying temperatures for both surfaces, although it was shown in a few cases that focused on the individual event (e.g., filled circles in Fig. 5d [$r = 0.75$]). Using higher drying temperatures or metal stickers resulted in larger area ratios in both surfaces as well as the occurrence of sticker stains (Fig. 3), although wood stickers always showed smaller values. This indicates that the use of metal stickers might frequently involve the occurrence of sticker stains over a wide range of contact area of piled timbers. It was also found that deep-colored sticker stain was more often seen in the surface of the pith side in all drying temperatures. Significant differences ($P < 0.05$) were often observed based on the comparison between sides using analysis of variance except in the case of wood stickers.

FTIR spectra of the contact area with stickers

Figure 7 shows the typical results of FTIR spectra at the contact area of each sticker on the surface of the pith side. It should be noted that gradual changes were found at 1,635–1,650 cm^{-1} with increasing drying temperature. In addition, depressions in absorbance were also found at 990, 1,160, 1,315–1,335, and 1,725–1,750 cm^{-1}, especially in using metal stickers at 100°C. It is known that those regions of wave numbers (990, 1,160, 1,315–1,335, 1,635–1,650, and 1,725–1,750 cm^{-1}) result from the stretching of C-O, C-O-C, bending of -CH$_2$-, adsorbed water, and stretching of C=O, respectively (Kataoka 2000). Therefore, it was possible that the changes in the spectrum shape were related to some sort of thermal influence on hemicelluloses, which are the first affected among the structural components of wood cell walls (Tejada et al. 1998, Esteves and Pereira 2009).

Figure 8 shows the FTIR spectra collected at the exposed internal region of specimens, which was not considered to be affected by contact with stickers. It was considered that this part retained more moisture than the surface of the specimen during drying. However, depressions in absorbance (1,160, 1,315–1,335, and 1,725–1,750 cm^{-1}) were found only in metal stickers at 100°C, as shown in Figures 7b and 7c, respectively. This suggested that the use of metal stickers at higher drying temperatures induced more severe thermal influences on hemicellulose in wood cell walls around the contact area with stickers than the internal region of specimens being uninfluenced by the stickers. It is possible that the contact with metal stickers induced the occurrence of the partial delay of drying. It is known that heating of wood with high moisture content usually involves some mechanical and chemical changes in wood cell walls, even at temperatures less than 100°C (Tejada et al. 1998, Clair 2012, Zhan et al. 2018). Conditions similar to these hygrothermal treatments are considered to be induced by drying using metal stickers at higher temperatures.

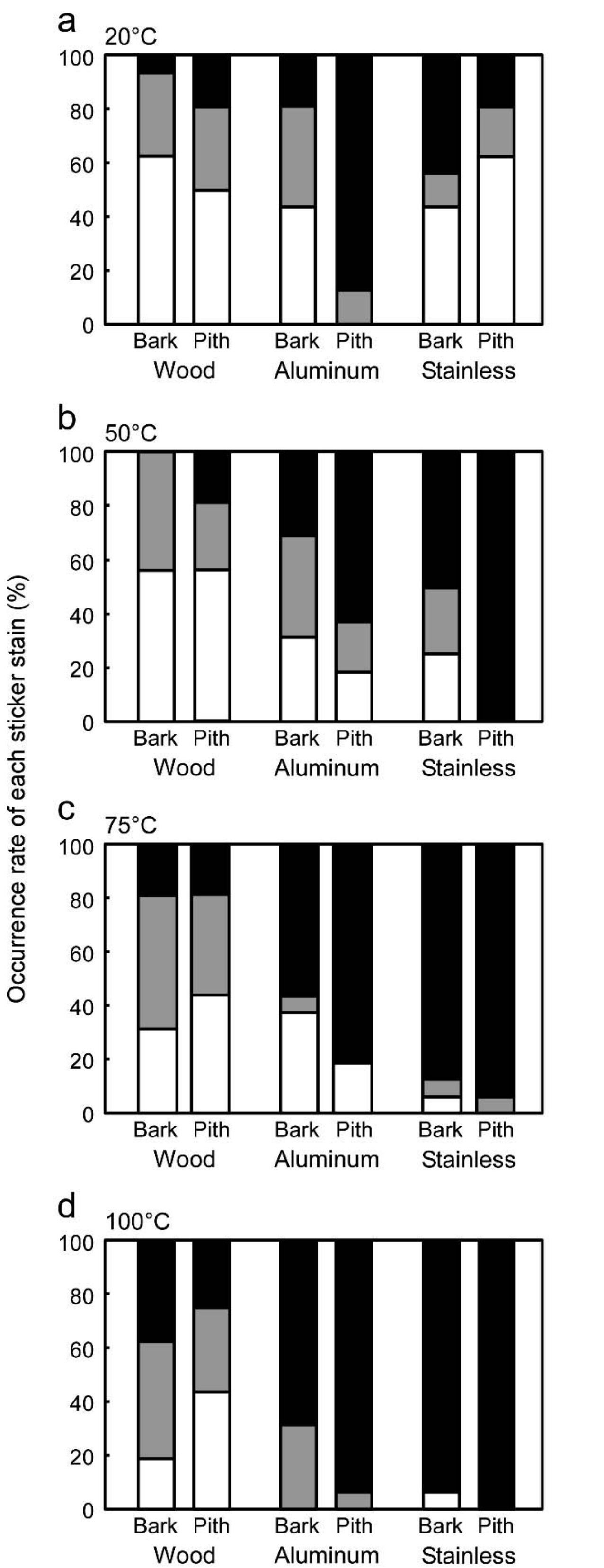

Figure 3.—The occurrence rate of sticker stains for each type of sticker at each drying temperature: (a) 20°C, (b) 50°C, (c) 75°C, and (d) 100°C. Open, gray, and filled columns show each type: no sticker stain, reduced shade, and deep color, respectively. "Bark" and "Pith" show sticker stain on the surface of the bark and pith sides, respectively.

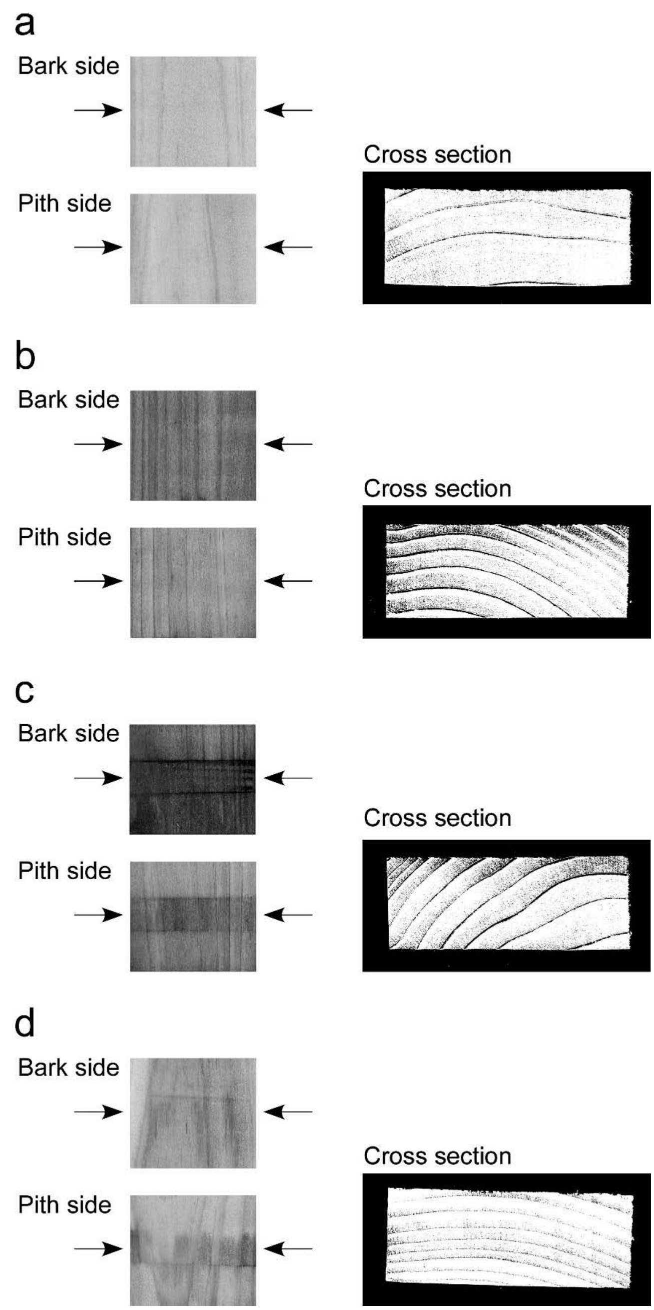

Figure 4.—Typical examples of surface sticker stain and internal staining conditions. (a) No stains were found on both the surface and the inside. (b) Clean surface with internal stains. (c) Stains were found on both the surface and the inside. (d) Stains were found on the surface, with no stain inside. Arrows arranged on both sides of all surface images indicate the cutting part. Images of the cross section are shown as binarized image.

Conclusions

This study investigated the effects of the drying temperature on the occurrence of surface sticker stains using Japanese cedar. The following conclusions were drawn based on the obtained results:

- Lower drying temperatures or the use of air-dried wood stickers provided clean surfaces without sticker stain,

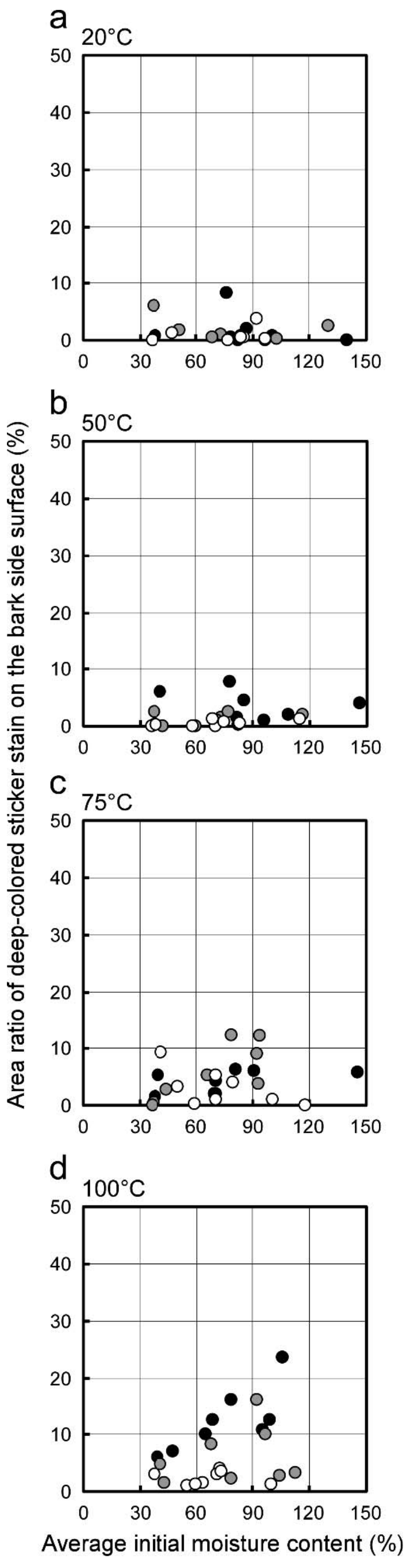

Figure 5.—The relationship between average initial moisture content and the area ratio of deep-colored sticker stain on the bark side surface. Open, gray, and filled circles indicate the data of stickers of wood, aluminum, and stainless steel, respectively.

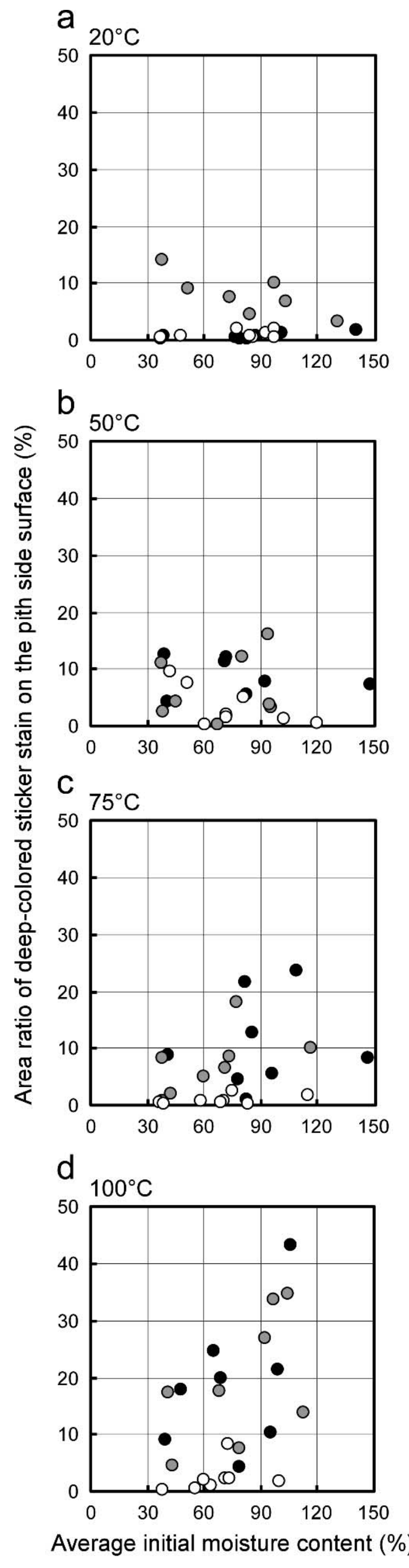

Figure 6.—The relationship between the average initial moisture content and the area ratio of deep-colored sticker stain on the pith side surface. All symbols here indicate the same as in Figure 5.

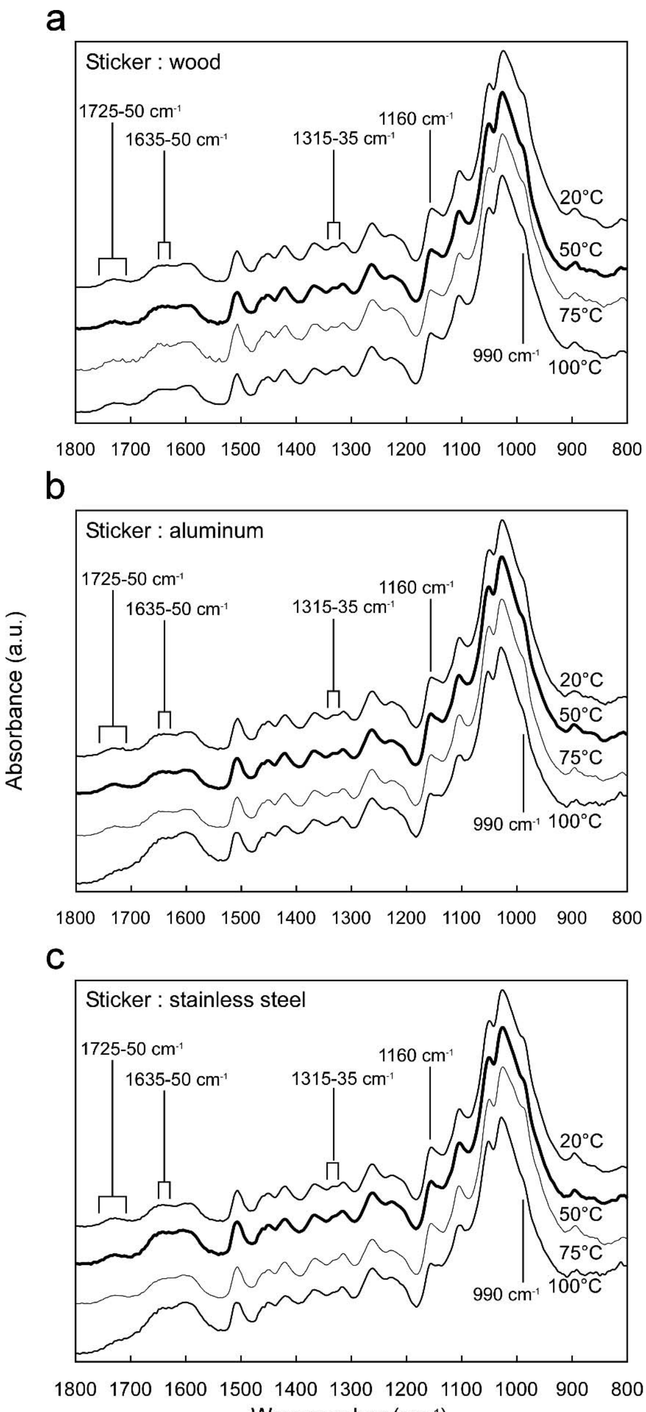

Figure 7.—Fourier transform infrared spectra of the contact area with each sticker on the pith side surface. (a), (b), and (c) show the result of wood, aluminum, and stainless-steel stickers, respectively.

whereas higher temperatures or metal stickers often produced deep-colored sticker stain.

- No definitive relationship was found between the initial moisture content and the extent of deep-colored sticker stain regardless of drying temperatures.
- FTIR spectra showed the depression in absorbance at some regions of hemicelluloses caused by the occurrence

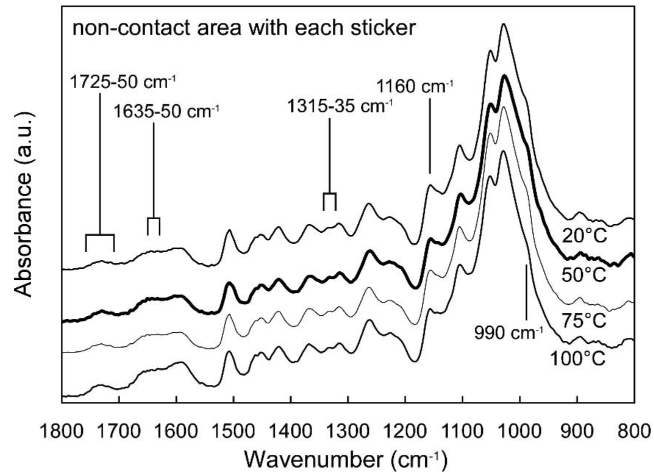

Figure 8.—Fourier transform infrared spectra of noncontact area with each sticker. The infrared light was irradiated on the internal region of the specimen with the exposed surface before measuring.

of a partial delay of drying around the contact area with metal stickers during drying.

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