

THE TEMPERATURE DEPENDENT EFFECT OF EXTRACTIVE CONTENT ON REDWOOD SHRINKAGE¹

Larry A. Demaree
Masonite Corporation
Chicago, Illinois

Robert W. Erickson
Department of Forest Products
University of Minnesota
St. Paul, Minnesota

Introduction and Literature Review

There is evidence in the literature of a direct relationship between extractive content and shrinkage (or collapse) of wood when drying at high temperatures. Erickson (7) found a highly significant positive correlation between hot-water soluble extractives and thickness shrinkage in redwood. In western redcedar Meyer and Barton (16) found collapsed boards to average 18.8% acetone-soluble extractives while the uncollapsed averaged 12.6%. Ellwood, in correspondence to Guernsey, suggested that extractives may act as plasticizers during drying to induce excessive shrinkage or collapse in incense cedar, redwood, and western redcedar. Narayanamurti (17) agrees that at high temperatures extractives could act as wood plasticizers. While densifying redwood heartwood, Tarkow and Seborg (24) discovered that the surface was easily densified with little springback. This behavior was attributed to the extractives softening at high temperatures and thereby plasticizing the wood.

In apparent contradiction to the above, the literature also (21) states that woods high in water-soluble extractives shrink less than expected due to bulking of the cell wall by the extractives. This may be true only when the drying temperature and/or stress situation is such that the plasticizing effect of the extractives is negated.

Objective

The primary objective of this research was to determine the relationship between extractive content and volumetric shrinkage for samples dried at various dry bulb temperatures.

Experimental Materials and Procedure

Six sets of wet redwood samples were dried, each at a different dry bulb temperature: 190°F, 175°F, 150°F, 130°F, 110°F, and room airdry. The relative humidity was held constant at 38 percent for all groups except the airdry.

The samples were obtained from four 4" x 4" cants (parent group 1) and four 2" x 6" planks (parent group 2). The samples for airdrying, 110, 130, and 175°F came from the cants, while those dried at 150 and 190°F came from the planks. A set contained 24 samples, six from either each cant or plank. The end-and-face-matching employed is exemplified in Figure 1.

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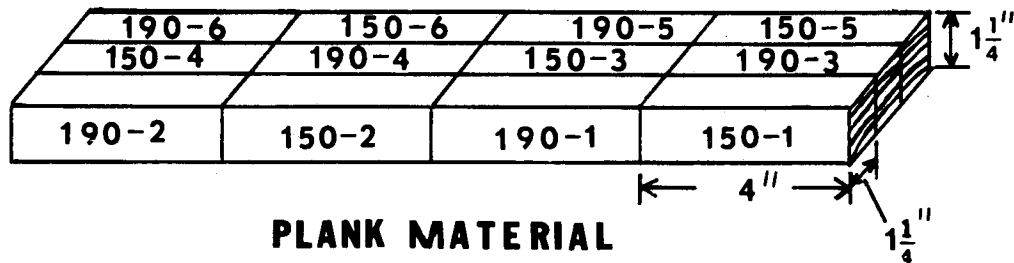
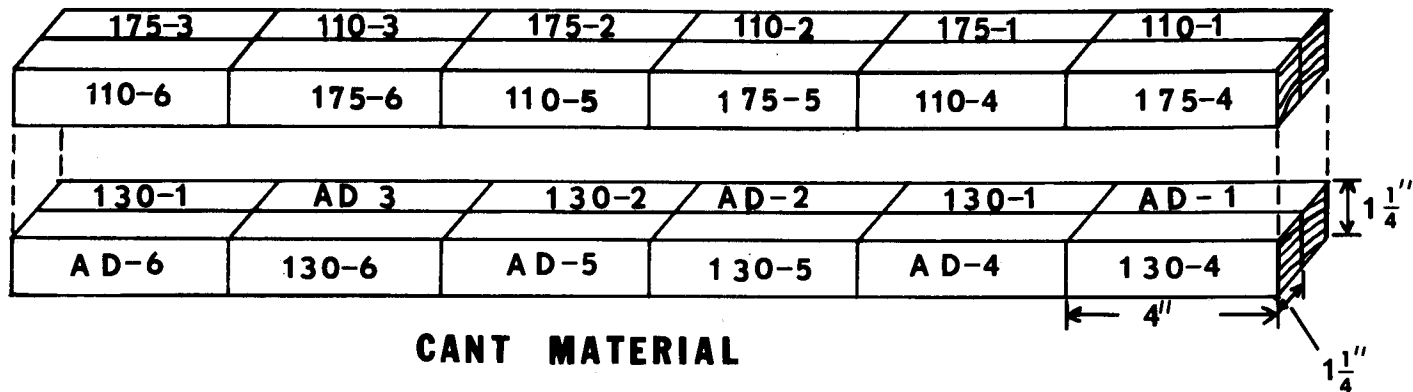


Figure 1. Typical sample allocation from a 4" x 4" cant or a 2" x 6" plank to the six drying temperatures. Two 1-1/4" boards were obtained from each cant as illustrated in the exploded drawing.

Each sample was 1-1/4 inches square and 4 inches long. As described by Kauman (11) a square sample was used to eliminate any bias in either the thickness or width shrinkage. The ends of each sample were coated with log sealer² plus aluminum foil to insure perpendicular-to-grain moisture movement.

The following is a description of the experimental procedure for each set:

Before drying, each sample's green volume, weight, and cross sectional dimensions were determined. Volume was obtained by the water displacement method. During drying the samples were periodically weighed and cross-sectional dimensions were determined. This procedure was continued until every sample in the set equilibrated. The equilibrium weights and dimensions were determined, after which the samples were oven-dried and again weighed. The percent shrinkage for both cross-sectional dimensions (hereinafter called width and thickness) and the percent moisture content were then calculated for the various times the samples were measured during drying. Length measurements were also taken on the 110 and 175°F sets, but the shrinkage was negligible. Consequently, longitudinal shrinkage was not derived for the remaining sets and volumetric shrinkage was determined from the change in cross-sectional area according to the formula: % volumetric shrinkage = $W + T - WT/100$ where W and T are the % width and thickness shrinkages. Future references to volumetric shrinkage refer to this area shrinkage. Also determined for each set for use in Figure 4 was the average volumetric shrinkage of all 24 samples at a given drying time.

Finally, each sample was milled into particles. A 2 gr. sample was randomly selected from the portion that passed through a sieve with holes 2mm. in diameter. This sample was oven-dried, weighed, and placed in a beaker with 300 ml. of distilled water. The mixture was frequently stirred during the next 48 hours and then filtered through a frittered glass crucible with the aid of a Venturi vacuum. The percent cold-water extractive content for each sample was calculated as follows: [unextracted oven-dry weight - extracted oven-dry weight/extracted oven-dry weight] 100. The specific gravity of each sample was also determined on the basis of extractive-free oven-dry weight.

Since the equilibrium moisture contents in this study varied both within and between temperature sets, it was necessary to adjust the final volumetric shrinkage for each sample to a common moisture content. This was accomplished by calculating a regression line for each sample using the plots of percent moisture content vs. percent volumetric shrinkage over the lower, nearly linear range of moisture content. (The moisture content range employed for this purpose increased with drying temperature.) The linear equation was solved for the shrinkage value at 6 percent moisture content.

Results and Discussion

Figure 2 displays the simple correlations and regression lines for volumetric shrinkage on extractives at 6 percent e. m. c. for the air-dry, 110, 130, and 175°F drying temperatures (parent group 1).

²SB525 Log Sealer, Mersman Bros., Celina, Ohio.

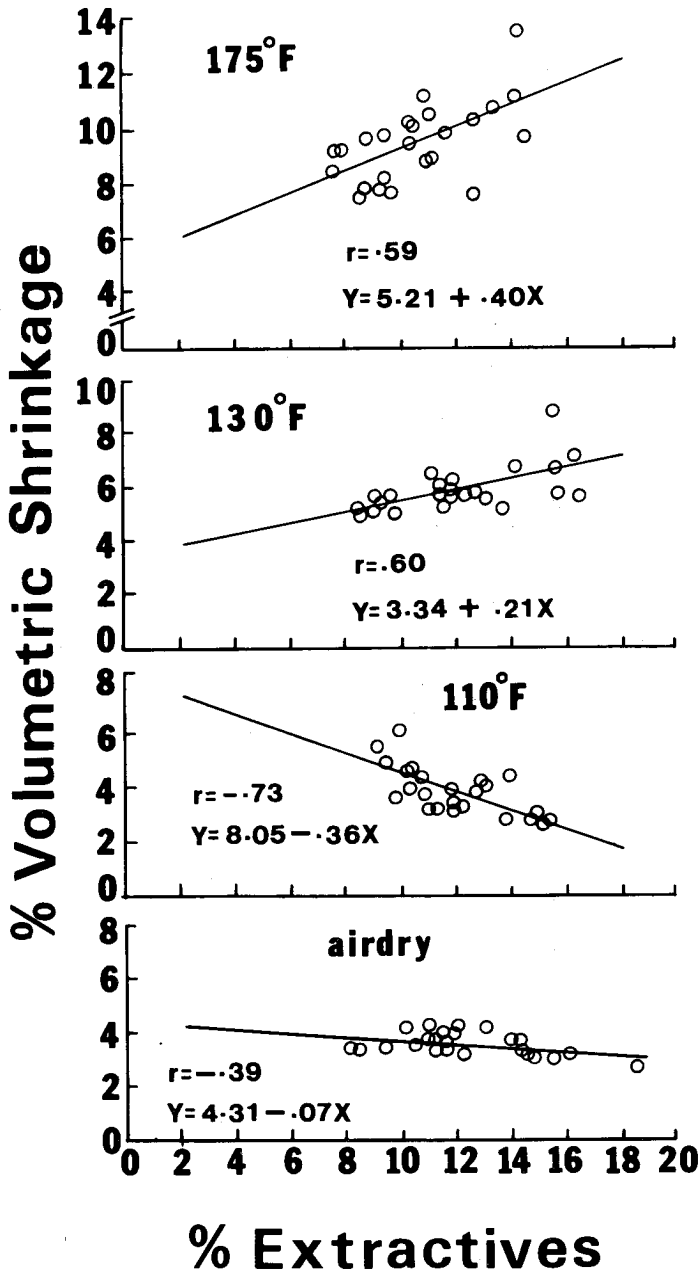


Figure 2. The relationship between percent volumetric shrinkage at 6% moisture content and percent cold-water extractives for the samples dried at airdry, 110°F, 130°F, and 175°F (parent group 1).

For air-dry and 110°F the percent extractive content is inversely related to shrinkage while at 130 and 175°F it is directly related to shrinkage. The reversal of this relationship between 110 and 130°F coincides with what is considered, by the redwood drying industry, as a critical temperature. Resch and Ecklund (20) have stated in relation to the kiln drying of redwood, "As long as the wood is above the fiber saturation point, increases in temperature above 120°F will cause collapse in a certain portion of the lumber." It would appear from Figure 2 that when redwood lumber is dried above 120°F a high extractive content promotes board shrinkage whereas when drying below 120°F the predominant extractive effect is one of bulking the cell walls resulting in below normal shrinkage for the lumber. Stamm and Loughborough (23), and Nearn (18), have verified this bulking phenomenon in low temperature drying experiments. Apparently, the shrinkage-inducing characteristics of the redwood extractives are dormant, or overshadowed by bulking, at drying temperatures below 120°F. At temperatures above this, however, they overshadow the bulking effect.

Figure 3 shows the regression of volumetric shrinkage on extractives for samples dried at 150 and 190°F (parent group 2). For each temperature, the correlation for all 24 samples was not significant. However, examination of the 190°F scatter diagram revealed a cluster of samples with atypically high shrinkages for their extractive contents in light of results from the 130 and 175°F runs. Inspection of these 8 samples revealed a common characteristic of wide growth rings. Following the 150°F run a similar situation was recognized, where ten samples with wide growth rings shrank excessively in view of their extractive content. When these "atypical" samples were removed from the analyses, the resulting correlations and regression lines were in keeping with the 130 and 175°F runs of Figure 2.

A summary of the statistical analysis is given in Table 1. The simple correlation of shrinkage vs. extractives was significant at the 1% level for four of the six drying temperatures. The air-dry coefficient (.39) just missed being significant at the 5% level (.40).

Shrinkage and initial moisture content were negatively correlated for air-dry and 110°F. This was probably due to a direct relationship between initial moisture content and extractive content (8).

In the case of partial correlations, shrinkage vs. extractives was again significant for 4 of the 6 drying temperatures. However, with extractives held constant only 3 of the 12 correlations were significant.

Shrinkage of wood under stress-free drying conditions typically shows a straight-line relationship with moisture content (21). All shrinkage in this situation is below the fiber saturation point and is associated with bound water loss. However, when moisture loss occurs perpendicular to the grain, as is the case with all lumber, a moisture gradient develops through the cross section of the board and results in a complex build-up of stresses from the surface to the core of the board. Under this condition significant board shrinkage will occur at average moisture contents above the f. s. p., i. e., during "apparent" free water loss.

The curves of volumetric shrinkage vs. moisture content for the various drying temperatures are in Figure 4. The air-dry and 110°F groups shrank comparatively little above the f. s. p. and therefore more nearly approached the condition of stress-free drying. However, as temperature was increased, more shrinkage occurred above the f. s. p.

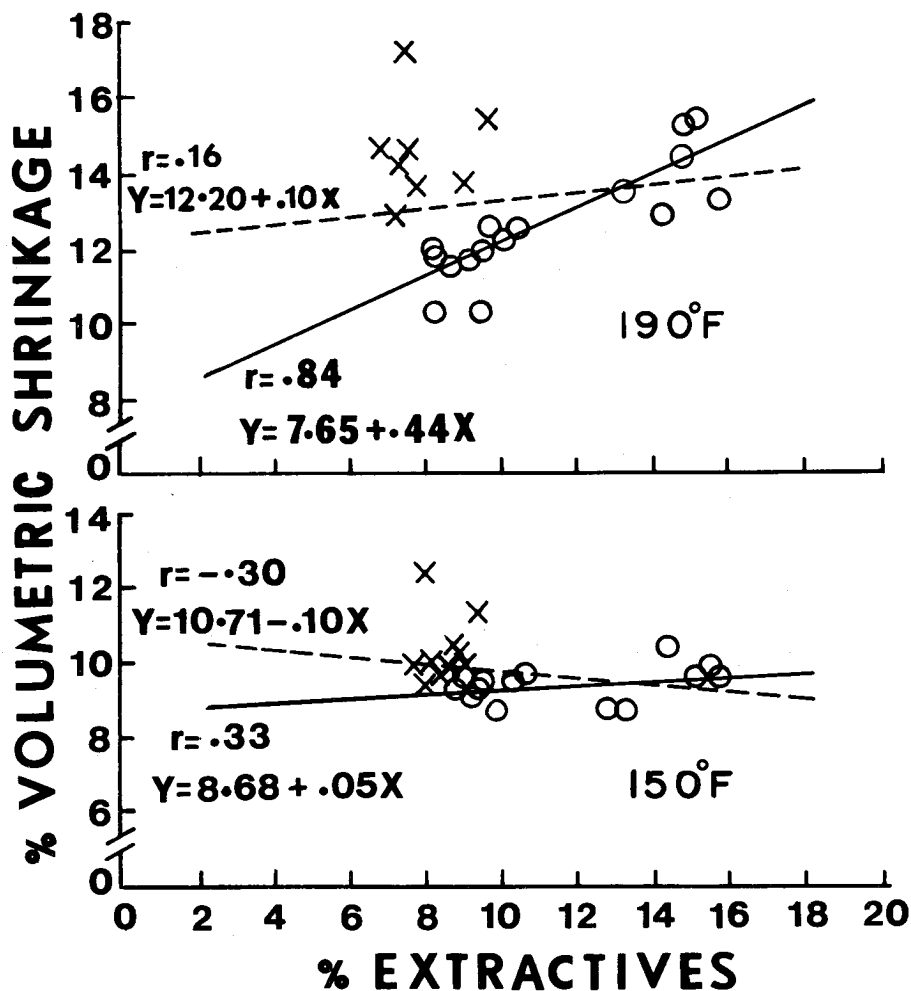


Figure 3. The relationship between percent volumetric shrinkage at 6% moisture content and percent cold-water extractives for the samples dried at 150°F and 190°F (parent group 2). The X symbols identify atypical samples. The dashed regression line is based on all 24 samples and the solid line is with the atypical samples removed.

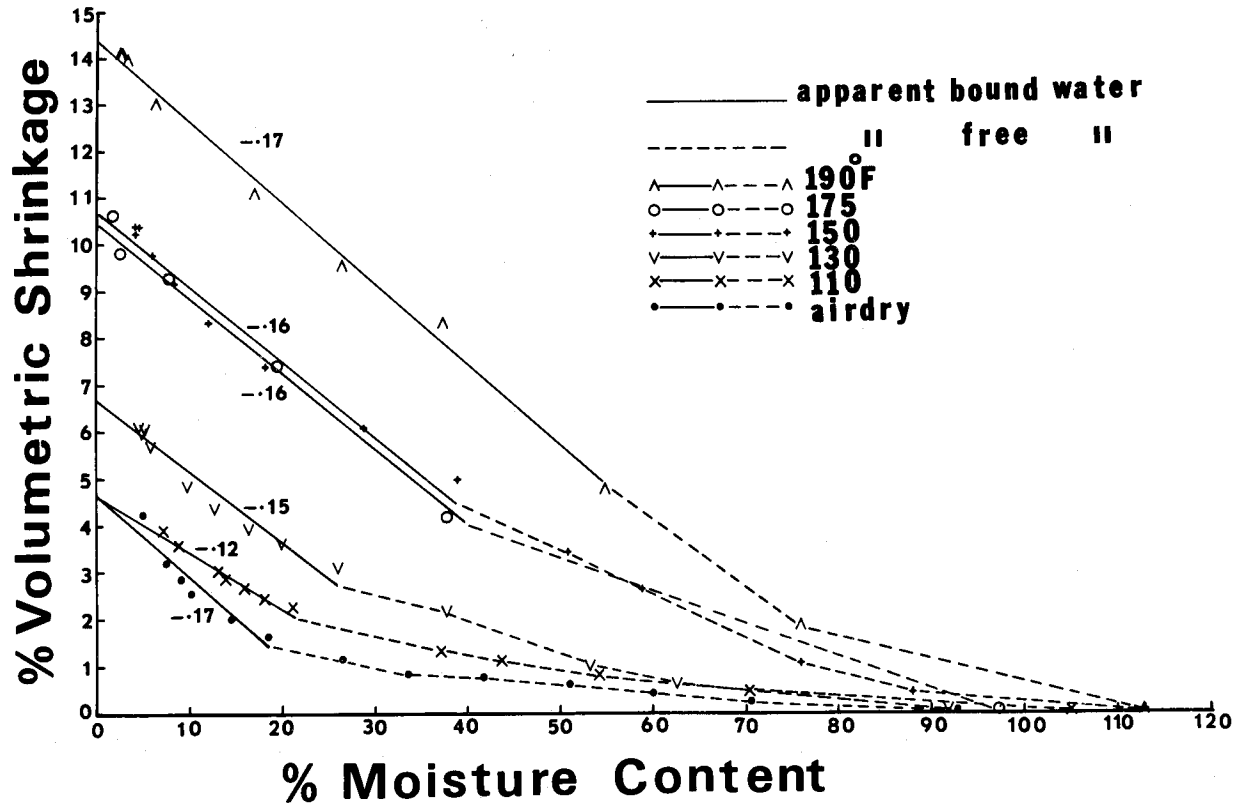


Figure 4. The relationship between average moisture content and average volumetric shrinkage for the 6 drying temperatures. The slopes of the regression lines for the apparent bound water loss are indicated. Each coordinate is an average for 24 samples.

Also, approximate straight-line shrinkage, normally associated with bound water loss, commenced at progressively higher average moisture contents. We defined this approximate straight-line shrinkage as occurring during apparent bound water loss, and shrinkage previous to this was defined as occurring during apparent free water loss.

Examination of the regression lines in Figure 4 indicates that the final differences in volumetric shrinkage between temperature sets developed during apparent free water loss. The apparent bound water slopes were tested in pairs to see if they were significantly different from one drying temperature to another. They were not. Since the final shrinkages were different, the differences must have developed during the loss of apparent free water. This is not particularly surprising since excessive shrinkage or collapse is known to occur early in drying (12). It is interesting to note, however, that the quantity of apparent bound water increased as the drying temperature increased.

Proposed Mechanism

Since tannins constitute nearly 50% of the extractives in redwood (1), and are water soluble, there is good reason to suspect they could influence shrinkage. Kollman and Cote' (13) state "the content of tannins and of substances found in heartwood may affect plasticity."

It is well known (2, 3, 4, 5, 14, 15) that the addition of minute quantities of tannins or simple phenols to a hydrophilic colloidal system greatly reduces the system's viscosity. The drastic reduction of viscosity is produced by a dehydration (15) of the system's colloidal particles. Tannins, because they lower surface tension considerably, are strongly absorbed by the colloidal particles, and hence, replace the adsorbed hydration shell normally found around the colloidal particles. Bungenberg de Jong (4, 5) believes this dehydration must be more than a simple adsorption of tannin molecules, for he has observed that when the tannin concentration in a hydrophilic colloidal system is increased to a certain level, the viscosity of the system suddenly increases again. This implies that rehydration has taken place. It is also interesting that the higher the temperature, the lower the tannin concentration required to induce the rehydration. Bungenberg de Jong has therefore hypothesized that at the boundary surface of the colloidal particle during dehydration, the dispersion medium separates into two liquid phases, the tannin-rich layer immediately surrounding the particle and the tannin-poor dispersion medium.

In the wet heartwood of redwood, this type of temperature and concentration sensitive tannin-water colloidal system could be present at two sites, the cellulose network of the cell wall and the cell lumen sap. Wood, meaning the network of the cell wall, has been defined as a limited swelling colloidal gel (13, 21) and, therefore, can be classified as a hydrophilic colloid. The cell lumen sap may also have the necessary constituents to qualify as a hydrophilic colloid. An indication of this is the lower surface tension of heartwood sap in redwood (22) as compared to sapwood sap or distilled water. Therefore, it is not unreasonable to assume that a tannin-water system, as described by Bungenberg de Jong, does exist in redwood and could very well be responsible for the typically high shrinkage encountered during apparent free water loss in this experiment whenever the extractive content and drying temperature were high. The following mechanism is offered to demonstrate the potentialities of this system in relation to shrinkage rather than developing an infallible model.

Examination of a redwood board before kiln drying would probably reveal the two liquid phases, which Bungenberg de Jong hypothesized as forming during the tannin dehydration of a colloid, to be present both on the microfibrils in the amorphous regions of the cell wall and on any colloidal particles in the cell lumen sap. Upon exposure to kiln conditions, a temperature gradient develops through the board, while simultaneously the outer surface starts to lose moisture. Since redwood is a relatively impervious species, the majority of the water movement would be bound water diffusion through the cell walls plus vapor diffusion through the cell cavities (21). In addition to moisture movement and loss, a sufficiently high temperature is necessary in a redwood board before collapse or excessive shrinkage will occur. As recognized by the industry and confirmed by this experiment, about 120°F and above is required. Kemp (12), in his study with aspen, observed the time before collapse started to occur was 44 minutes at 170°F and 150 minutes at 140°F. These times to collapse correspond quite well with times required to heat the interior of redwood boards, under similar conditions, above the critical drying temperature of 120°F (8). The exact critical temperature could be dependent on the concentration of tannins in the board.

As the temperature of the board increases above 120°F, rehydration occurs in the cell lumen sap. That is, the tannin molecules are no longer intimately associated with colloidal particles of the sap and are free to diffuse into the amorphous regions of the cell wall. This could be envisioned as similar to the phase of leather tanning where the emulsoid impurities of the tanning liquor yield their weakly adsorbed tannin to the more strongly adsorbing skin when the two are brought in contact (2). As more and more tannin diffuses into the cell wall and adsorbs into the microfibrils, a situation could develop where only tannin molecules are the adsorbed medium between the microfibrils. This would be similar to the clotted precipitate of tannin and hydrophilic particles which Bungenberg de Jong (5) observed as forming in a tannin-gelatin-water system when both the temperature and tannin concentration were high. Subsequent high temperatures and stirring would not disperse the clot. Now, when the drying stresses of the board are imposed on such tannin-enriched areas, they would logically behave differently than an area containing only adsorbed water. In discussing the introduction of tannins into leather, Jirgensons (10) states, "The polymers introduced into the framework of the tanned leather act as plasticizers." Similarly, it is not unreasonable to assume that in these tannin-rich areas at higher temperatures, the usually rigid cellulose network of the cell wall would be modified, so that under stress, "plastic flow" of the cell wall could occur. The formation of these tannin-rich amorphous regions would be expected to occur in areas of a board containing a high extractive content plus free water. These areas are also known as the most collapse susceptible.

Conclusions

When redwood heartwood samples were dried at room air dry and 110°F, the quantity of extractives was inversely related to volumetric shrinkage. When samples were dried at 130, 150, 175, and 190°F, the extractive content was directly related to shrinkage. The difference in final volumetric shrinkage for the different drying temperatures developed during the removal of what was defined as apparent free water.

Table 1. Summary of the Statistical Analysis.

Simple Correlations	Dry Temperature					
	Airdry	110°F	130°F	175°F	150°F ¹	190°F ¹
Shrinkage vs. extractives	-.39(n)	-.73**	.60**	.59**	.33(n)	.84**
Shrinkage vs. specific gravity	-.08(n)	-.51*	-.24(n)	.27(n)	-.26(n)	-.29(n)
Shrinkage vs. initial moisture content	-.68**	-.67**	.45*	.33(n)	.34(n)	.77**
<u>Partial Correlations</u>						
Shrinkage vs. extractives (initial moisture content and specific gravity held constant)	-.07(n)	-.51*	.55**	.49*	.03(n)	.76**
Shrinkage vs. specific gravity (initial moisture content and extractives held constant)	-.08(n)	-.43*	-.37(n)	.07(n)	-.08(n)	.76**
Shrinkage vs. initial moisture content (specific gravity and extractives held constant)	-.59**	-.35(n)	.39(n)	-.02(n)	.09(n)	-.01(n)
<u>Multiple Correlation</u>						
Shrinkage vs. extractives, specific gravity and initial moisture content	.68**	.82**	.71**	.59**	.00(n)	.91**

n = not significant

* = significant at the 5% level

** = significant at the 1% level

¹ Values obtained after removal of atypical samples (see text)

It appears that as low drying temperatures the extractives act primarily as bulking agents but at higher temperatures they increase the plasticity of the cell wall, causing increased shrinkage and collapse during apparent free water loss. The tannins of the extractive fraction are believed to be responsible for this phenomenon.

Abstract

Rectangular redwood heartwood samples were dried from the green condition to equilibrium moisture content at 38% relative humidity and the following dry bulb temperatures: 110°F, 130°F, 150°F, 175°F, and 190°F. A room air dry run was also included. Resulting shrinkage data was related to moisture content, specific gravity, and percent cold-water extractives. The results show that when redwood heartwood was dried at room air dry and 110°F, the quantity of extractives was inversely related to shrinkage. At 130, 150, 175, and 190°F, the extractive quantity was directly related to shrinkage. The difference in final volumetric shrinkage for the different drying temperatures developed during the removal of apparent free water. It was hypothesized that at low drying temperatures the extractives act as bulking agents but at higher temperatures they become heat sensitive and make the wood cells collapse-susceptible during apparent free water loss. A mechanism involving the tannin extractive fraction was suggested as being responsible for the latter phenomenon.

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