Comparison of the effects of gamma irradiation and steam sterilization on southern pine sapwood

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Abstract

Gamma irradiation is a commonly used method of sterilization of wood specimens prior to decay testing. As part of a larger series of studies, an investigation was made into the effects of gamma irradiation on flexural bending strength properties and its corresponding relationship to changes in lignin and hemicellulosic contents of wood after exposure to various dose rates and total dosage. Effects of irradiation were compared with effects of a steam sterilization method. Flexural strength was reduced in response to a long exposure to radiation. Klason lignin and galactan contents were also reduced by a high total dose. The other carbohydrate components of wood showed no discernable differences under the range of irradiation exposures used. The dose rate used to achieve the critical total dosage of gamma radiation required for sterilization of wood intended for laboratory decay testing was found to differentially affect both bending strength and some chemical components in irradiated pine.

Sterile specimens are required for many laboratory decay tests. Three methods for sterilizing wood specimens are steam sterilization, gas sterilization, and gamma irradiation. Each method has different effects on wood properties. Winandy and Morrell (1993) found that steam sterilization resulted in approximately 4 percent weight loss in wood with a 12 percent loss of bending strength. The arabinose and galactose components of the hemicellulose were decreased by 28 percent and 22 percent respectively. Gas sterilization seems to have little effect on mechanical properties but has been shown to leave residuals that partially inhibit decay either by changing the chemistry of the wood environment (Wallhauser 1967) or by leaving toxic residuals (Smith 1965, DaCosta and Osborne 1969).

Irradiation has been shown to have less effect on mechanical properties of wood than does steam sterilization (Hansen 1972), although at high dosage (such as 10 Mrad) effects may result in measurable weight loss (Smith and Sharman 1971). Despot et al. (2007) found that an overall radiation dosage of 3 to15 Mrad caused irreversible and permanent changes in the chemical and mechanical properties of wood. Divos and Bejo (2005) showed that loss in modulus of elasticity (MOE) in both longitudinal and transverse direction was directly related to gross irradiation dose for five different wood species, but those reported relationships were not consistent among species or properties. Aoki et al. (1977) demonstrated that irradiation caused a reduction in physical strength properties of wood. Threshold dosage values beyond which significant reduction begins (**Table 1**) were derived from data of Aoki et al. (1977) by plotting relative property against log irradiation dose, with threshold level designated as the transition point where the plot became nonlinear. Jokel and Markus (1967) suggested that irradiation caused no significant changes in the hemicellulose content of wood, although this work did not quantify changes.

Gamma irradiation is used in Europe in Standard EN113 (CEN 1996) and North America in Standard D1413–07 (ASTM 2007b) as the preferred method for the sterilization of wood and wood products. It is also used in Standard E10–07 (AWPA 2007) as a possible alternate method for the sterilization of woods. In most of these standardized uses, the total

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Table 1. — Threshold dosage at which strength property reductions begin.[a]

Strength property	Approximate threshold irradiation dose (Mrad)
Compression	56
Bending	4
Tensile	3

[a]Derived from Aoki et al. (1977).

dosage often recommended for use is 2.5 to 5.0 Mrad, based on experience and consultation (Carey 1998) and on previous work showing that 2.5 Mrad is the lowest dose required to fully sterilize wood (Hansen 1972, Schmidt and French 1979). Work by Freitag and Morrell (1998) and Pointing et al. (1998) has shown that 1.5 Mrad is sufficient to completely sterilize wood for pure cultures of a limited number of fungi.

Strength loss has been demonstrated to be a good indicator of early wood decay, with the ratio of strength loss to weight loss approximately 4:1 (Curling et al. 2002). Although strength losses may exceed 50 percent by the time 10 percent weight loss is incurred (Wilcox 1978), weight loss remains the standard measurement for decay tests for a number of reasons (such as specimen size, equipment requirements, ease of measurement).

Likewise, chemical changes during initial fungal colonization result in measurable reductions in strength before measurable weight loss (Schmidt et al. 1978, Highley 1987, Ruddick 1986, Imamura 1993, Kim et al. 1996). Previous studies (Winandy and Morrell 1993) showed a close relationship between biological degradation of hemicellulosic components, such as arabinose and galactose, and wood strength losses. Clausen and Kartal (2003) discussed a similar relationship between modulus of rupture (MOR) and galactan reduction in southern pine stakes after 4 weeks exposure to *Postia placenta*.

We felt that, because sterilization method can affect initial strength and chemical composition, the relative merits of various sterilization methods for the above-mentioned decay technique needed to be investigated.

The objective of this project was to compare the effects of gamma irradiation and steam sterilization on bending strength and chemical composition of southern pine sapwood.

Experimental method

Gamma irradiation

Seventy-two matched southern pine sapwood specimens (253 by 25 by 9.5 mm long) were exposed to a range of nine total irradiation doses (from a cobalt-60 source) applied at two different dose rates: 0.85 Mrad/h (total doses of 1.50, 2.50, and 5.00 Mrad) and 1.69 Mrad/h (total doses of 1.50, 2.00, 2.50, 3.75, 5.00, and 7.50 Mrad). Eight replicate specimens were evaluated at each combination of irradiation dose rate and total dosage.

Steam sterilization

Two more matched groups of eight replicate specimens were also used. The first was steam-sterilized at 110 °C for 1 hour and represented a steam-treated control, whereas the last group was unexposed to any sterilization treatment and represented unsterilized southern pine sapwood controls.

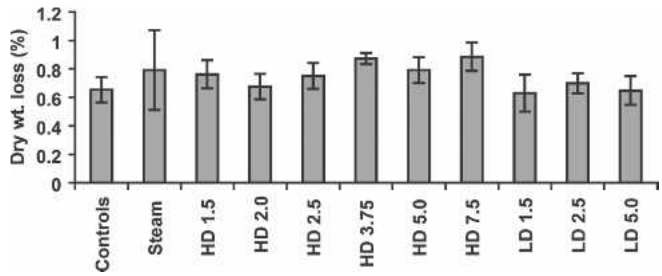

Figure 1. — Comparisons of ovendry mass loss caused by steam/irradiation, with each vertical bar indicating ±1 SD from mean weight loss value. Groups are indicated as the high dose (HD) rate of 1.69 Mrad/h or low dose (LD) rate of 0.85 Mrad/h, with the total dose indicated from 1.50 to 5.0 Mrad.

Testing, evaluation, and analysis

Eight steam-sterilized specimens, the nine groups of eight replicate specimens sterilized at each radiation dose, and a set of eight unsterilized controls were evaluated for MOR according to ASTM D 143 (ASTM 2007a), except that we used a four-point bending test configuration, rather than the simple three-point bending test normally used. The four-point loading system was selected because it produced a uniform bending stress between the two loading heads, which in turn allowed more precise assessment of the initiation of incipient decay, which initiates randomly anywhere across the decay-exposure zone. Because the three-point loading system maximizes the bending stress only directly under the center loading head, its calculated bending strength estimate can be problematic in that it can overestimate ultimate strength unless the decay initiates directly under the single-point load head, an unlikely scenario (Winandy and Morrell 1993). Realizing this problem, these authors attempted to design a decay test set-up that limited fungal exposure to the general center of the Douglas-fir test specimens and uniformly stressed the specimens over that central area.

After mechanical testing, a 25-mm long block was cut from immediately near the failure of each specimen to assess weight loss and chemical composition. Weight loss was evaluated according to ASTM D 1413 (ASTM 2007b). Carbohydrate analysis for individual carbohydrate sugars was conducted on a mixed composite (that is, equal amounts from each of the eight replicates/group) for each of the 11 groups using the methods of Davis (1998). Klason lignin was determined using the methods of Effland (1977). Statistical analysis evaluating differences in sample means used a Student's t-test (Snedecor and Cochran 1967).

Results and discussion

Weight loss and strength loss data are shown in **Figures 1** and **2**. Bending strength was calculated for each specimen, and strength loss is reported as the difference in average strength properties between each group of test specimens and the unsterilized control group.

There were no significant differences in weight loss between irradiated and steam sterilized specimens (**Fig. 1**). The strength loss data, however, showed significant ($\alpha < 0.05$) differences in strength reduction caused by irradiation and steam sterilization (**Fig. 2**). At the higher dose rate (1.69 Mrad/h), a cumulative total dose of up to 5 Mrad caused less strength reduction than did steaming. With one exception at the lowest

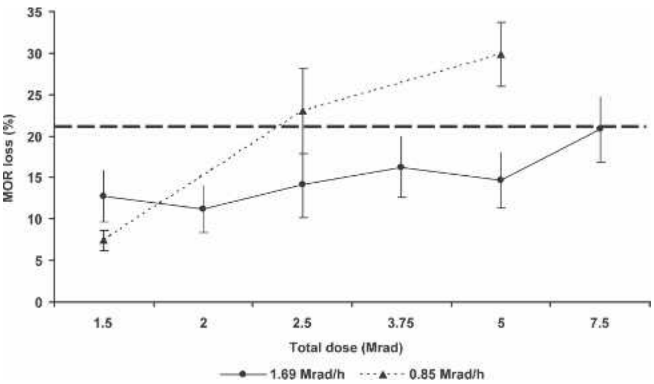

Figure 2. — Relationship between strength loss and irradiation dose, with each vertical bar indicating ±1 SD from mean MOR value. For comparison, the mean effect of steam sterilization is indicated by the dashed line.

total dose, our data clearly show that a lower dose rate (0.85 Mrad/h), which requires about twice the duration of exposure than did the higher dose rate (1.69 Mrad/h) to obtain the same total dosage of gamma radiation, caused greater strength reductions than did the higher dose rate. A clear interaction in the effect of irradiation on strength loss is clearly seen by comparing the three matched total doses at 1.5, 2.5, and 5.0 Mrad. The resultant slope of the three treatments at the lower dose rate (0.85 Mrad/h) is clearly different from that of the higher dose rate (1.69 Mrad/h). This interaction precludes a general statistical test of means and instead requires assessing the statistical significance of means by a series of direct multiple comparisons. Our analysis indicated that the lower dose rate caused significantly less ($\alpha < 0.05$) strength loss at the lowest total dose of 1.5 Mrad, than did the higher dose rate. Conversely, at a total dose of 2.5 and 5.0 Mrad, the higher dose rate of gamma irradiation caused significantly less strength loss than did the lower dose rate. Another key result for the analysis is that each dose application rate (0.85 and 1.69 Mrad/h) clearly showed no evidence of lack of linearity. This is useful in interpolating results within the ranges of our evaluation.

As the required total dose for biotic sterilization is about 2.5 to 5.0 Mrad, the higher 1.69-Mrad/h dose rate achieves that total dose between 1.5 to 3 hours This higher dose rate caused significantly less initial strength reductions than did the more commonly used steam sterilization procedures. Our data and analysis clearly show that the lower, slower dose rate of 0.85 Mrad/h caused higher strength reductions than did steam sterilization, Thus, because both steam and the slower 0.85 Mrad/h irradiation rate caused higher strength reductions than did a faster, higher irradiation dose rate, we feel that pretest sterilization using dose rates higher than 1.5 to 1.8 Mrad/h seems a reasonable recommendation for future work.

These results clearly suggest that duration of exposure may be just as critical, if not more critical, than total dose. We believe that the lower dose rate produced a greater reduction on bending strength than the higher dose rate because the kinetic energy imparted from the irradiation exceeded various physio-chemical thresholds within the wood (**Figs. 3** to **5**). Although our data showed little differential effect on wood mass between the two dose rates, various chemical constituents were measurably affected by gamma irradiation.

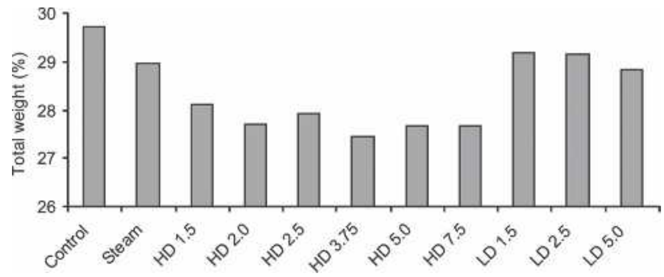

Figure 3. — Effect of various irradiation doses on Klason lignin content of pine. Groups are indicated as the high dose (HD) rate of 1.69 Mrad/h or low dose (LD) rate of 0.85 Mrad/h, with the total dose indicated from 1.50 to 5.0 Mrad.

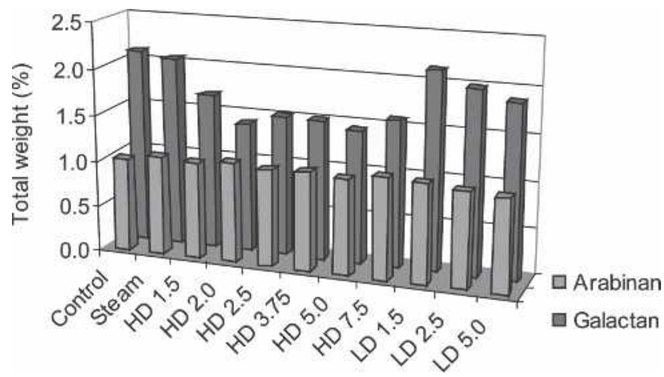

Figure 4. — Effect of various irradiation doses on arabinan and galactan content of pine. Groups are indicated as the high dose (HD) rate of 1.69 Mrad/h or low dose (LD) rate of 0.85 Mrad/h, with the total dose indicated from 1.50 to 5.0 Mrad.

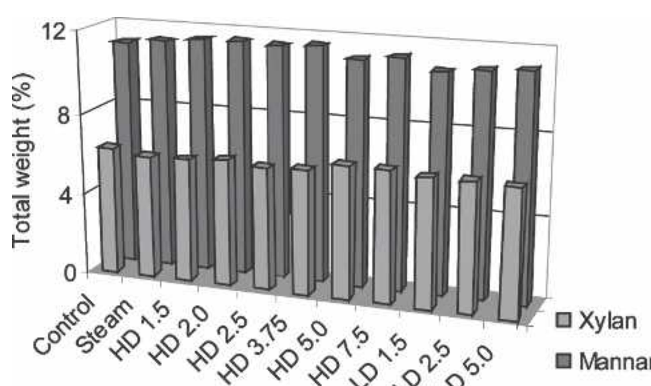

Figure 5. — Effect of varying irradiation dose on mannan and xylan content of pine. Groups are indicated as the high dose (HD) rate of 1.69 Mrad/h or low dose (LD) rate of 0.85 Mrad/h, with the total dose indicated from 1.50 to 5.0 Mrad.

The effects of gamma irradiation on various wood chemical constituents at various dose rates (1.69 and 0.85 Mrad/h) and for various durations controlled to produce set dose levels are shown in **Figures 3** to **5**. The higher 1.69 Mrad/h dose rate clearly had more negative effect on Klason lignin levels than did either the steam sterilization or the lower 0.85-Mrad/h dose rate (**Fig. 3**). However, gamma irradiation at the 1.69-Mrad/h dose rate rapidly reduced Klason lignin from 29.5 percent to 28.1 to 27.5 percent, but seemed to have little on-going effect related to dosage accumulation. A nearly identical trend was exhibited by the galactan side-chains in glactoglucomannan hemicellulose in that galactan content started at ~2 percent and was quickly reduced to about 1.5 percent by the higher 1.69-Mrad/h dose rate. However, very little additional damage to galactan occurred with increasing total dose (**Fig. 4**). At the lower dose rate (0.85 Mrad/h), the reductions in Klason lignin (**Fig. 3**) and galactan (**Fig. 4**) closely corresponded to a comparable reduction in bending strength (**Fig. 2**).

Winandy and Lebow (2001) postulated that the earliest stages of thermochemical-induced strength loss were related to degradation of hemicellulose polymers in wood. They found that the initial event of hemicellusose depolymerization involved the severing of arabinan or galactan side-chains from their primary xylan- or glucomannan-backbones, respectively,. The results shown in **Figures 4** and **5** tend to support that theory. Note how galactans, which are side-chain constituents of the hemicelluloses (**Fig. 4**), are affected sooner and to a greater degree than are the xylans or mannans that represent the primary backbone of the hemicellulose polymers (**Fig. 5**). Conversely, note that arabinan was virtually unaffected by various radiation regimes. We suspect that the β–(1$\rightarrow$3) linkage of the arabinan-xylan bond is less affected by radiation than is the β–(1$\rightarrow$6) linkage of the galactan-mannan bond.

In summary, the effect of gamma irradiation on strength loss seems directly related to a subtle radiation-induced hydrolytic breakdown of various chemicals that make up wood.

Conclusions

Dose rate and total dosage of gamma radiation differentially affected both bending strength and some chemical components in the tested pine. Density appeared unchanged by any tested level of irradiation dose or rate of exposure. The cumulative duration of the gamma-irradiation exposure period appeared to be more critical than total dose in determining overall strength loss. When total gamma-irradiation dosage was directly compared at or near the critical levels required to achieve sterilization, shorter applications using higher dose rates had less effect on strength than longer applications using lower dose rates. Klason lignin and galactan experienced a measurable, but non-progressive, initial reduction at the higher dose rate. In contrast, Klason lignin and galactan each exhibited a reduction that was lower initially but slowly progressed as irradiation continued at the lower 0.85 Mrad/h dose rate. These findings conflict with recommendations given in the two North American soil-block decay tests D1413 (ASTM 2007b) and E10 (AWPA 2007) that dose rate does not matter. This requires further study.

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