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ORIGINAL ARTICLE

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The effects of within-species and between-species variation in wood density on the photodegradation depth profiles of sugi (*Cryptomeria japonica*) and hinoki (*Chamaecyparis obtusa*)

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Abstract Low density wood is more rapidly eroded than denser wood when exposed to the weather, possibly because it is more susceptible to photodegradation. Fourier transform infrared microscopy was used to examine: (1) the depth of photodegradation in earlywood and latewood of sugi (Japanese cedar) and earlywood of hinoki (Japanese cypress) exposed for up to 1500h to artificial sunlight emitted by a xenon lamp (375 W/m^2 within the 300 to 700nm spectral range); and (2) the relationship between the density of wood tissues and depth of photodegradation. The depth of photodegradation varied between species (sugi and hinoki) as well as within a growth ring (sugi earlywood and latewood), and there was an inversely proportional relationship between depth of photodegradation and wood density. These findings may explain why low density earlywood is more rapidly eroded than latewood during weathering, and more generally, why there is an inverse relationship between the density of wood species and their rate of erosion during artificial and natural weathering.

Key words Density · Depth profile · FT-IR · Photodegradation · Wood

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Introduction

Wood exposed to the weather undergoes slow surface erosion.[1,2] The depth of such erosion, which has often been used as an indicator of the susceptibility of different wood species to weathering and the effectiveness of chemical treatments in restricting photodegradation, varies with wood density.[3–5] Borgin[6] noted that earlywood is eroded first when wood is exposed outdoors producing the characteristic corrugated surface appearance of weathered wood. Both accelerated and natural weathering studies have confirmed that there is a negative relationship between wood density and erosion.[1,3,7–9] Accordingly, the erosion rate of softwoods exposed vertically and facing south in the northern hemisphere was estimated to be 5–10mm per century, whereas comparable figures for denser hardwoods were 2–5mm.[3] Much larger differences in erosion rate, however, can be seen within species in growth rings composed of thin-walled earlywood and denser latewood tissues. Sell and Feist[3] reported that the erosion rate of earlywood was 1.0 to 4.4 times larger than that of latewood, based on the artificial weathering of six softwood species. Williams et al.[8,9] found that during the first several years of natural weathering, earlywood eroded much more quickly than latewood until the latewood bands became isolated when their rate of erosion increased, albeit, not to the level of earlywood.

There are currently two different interpretations of the relationship between wood density and erosion during weathering. Sell and Feist[3] concluded that the negative relationship between density and erosion is "approximately linear within a density range of 0.3 to 1.0g/cm^3." Williams et al.[8] believe that there is an inverse relationship between density and erosion. More recently, Evans et al.[10] reexamined the erosion data of various wood species and concluded that "the relationship is inverse, but not linear, within the density range of 0.3 to 1.2g/cm^3."

It is reasonable to assume that the erosion of wood during weathering depends on photodegradation of wood and removal of degraded constituents by water or wind-blown particulates. The depth of erosion of wood may therefore be related to depth to which wood is photodegraded. There have been relatively few studies, however, that have examined the relationship between wood density and depth of photodegradation. Horn et al.[11] used Fourier transform infrared (FT-IR) spectroscopy to examine the weakening of western redcedar (*Thuja plicata* D. Don) exposed to artificial weathering. They found that earlywood was photodegraded faster than latewood, but they did not measure the depth of photodegradation in latewood. Yata and Tamura[12] used histochemical techniques to examine depth of photodegradation in three softwoods, including sugi (*Cryptomeria japonica* D. Don) and hinoki (*Chamaecyparis obtusa* Endl.), following natural weathering. They reported that earlywood was degraded to a greater depth than latewood. Park et al.[13] also used histochemical techniques to compare the depth of photodegradation in hinoki earlywood and latewood following ultraviolet (UV) irradiation. They measured the number of cell walls in which degradation of lignin could be detected. Photodegradation was detected up to five and two cell walls deep in earlywood and latewood, respectively. The findings of these authors[11–13] are consistent with observations that earlywood is more susceptible than latewood to erosion during weathering.

In this article, we report on photodegradation depth profiles in sugi earlywood, sugi latewood, and hinoki earlywood, and corresponding variation in density within and between species. By combining both types of measurements, we aim to precisely assess the relationship between the density of the different wood types and the depth of photodegradation, and more generally to help to resolve the discrepancy that exists in the literature regarding the relationship between the density of wood species and their rate of erosion during weathering. It is important to clarify this because of the large within-species and between-species variations in wood density and the important effects of photodegradation on the surface properties of wood.

Experimental

Wood samples

Wood specimens were obtained from air-dried sugi (Japanese cedar) and hinoki (Japanese cypress) sapwood. For IR depth profile studies, wood blocks measuring 5 (L) × 5 (R) × 2 (T) cm were used (Fig. 1a). For soft X-ray densitometry studies, crosscut wood sections of 2.00-mm average thickness were prepared from the same wood specimens used for depth profile analyses (Fig. 1b).

Artificial solar radiation

The radial faces of wood blocks were exposed to artificial sunlight from a xenon light source (>290 nm) at 375 W/m^2 (measured in the 300–700 nm range) for up to 1500 h at 60°C (black panel) and 30% relative humidity in a commercial fade-meter chamber (Suga FAL-25AXC-HC). The total

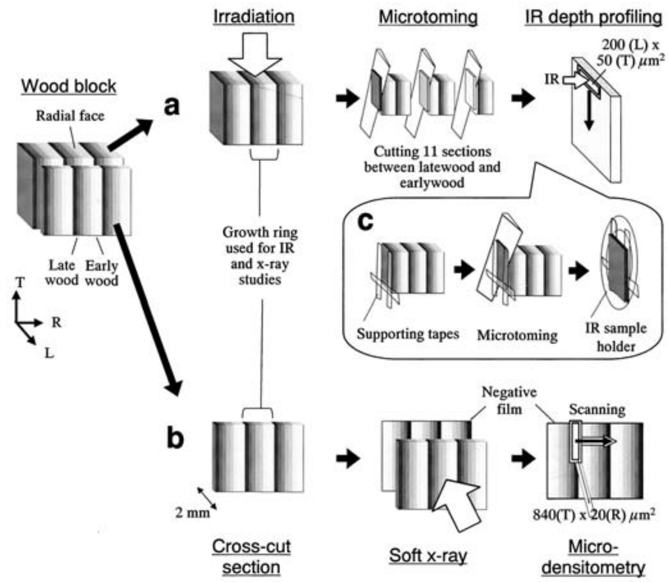

Fig. 1. Methods used to prepare and section wood for infrared (IR) depth profile analyses (*a* and *c*) and soft X-ray densitometry measurements (*b*)

energy received by specimens was roughly equivalent to that of 10–11 months of natural exposure in a horizontal plane in Tokyo. After each exposure period of 50, 100, 300, 600, and 1500 h, tangentially orientated wood sections (10–30 μm thick) were cut vertically from exposed surfaces, and were used for microscopic FT-IR measurements, as shown in Fig. 1a, and described previously.[14,15] For within-ring analyses of photodegradation depth profile, sugi specimens exposed for 1500 h were used.

FT-IR depth profile analyses

Figure 1a shows the method of obtaining within-ring FT-IR depth profile spectra for sugi earlywood and latewood at intervals of approximately 100 μm. To check the accuracy of IR analyses, spectra were also obtained from areas adjacent to each sampling position. Sugi latewood was difficult to section and its high density reduced the transmission of IR radiation, as mentioned previously.[15] These difficulties were overcome by supporting wood blocks during sectioning, which made it possible to cut dense sugi latewood into 10-μm-thick sections that transmit sufficient IR radiation (Fig. 1c). Thus, the thickness of wood section samples varied from 10 (latewood) to 30 μm (earlywood), depending on the density of the tissues. Each sampling position (the distance from the beginning of the growth ring) was measured using a calibrated digital microscope (Keyence VH 7000) at a magnification of 50 to 500 times. Thus, it was possible to precisely match the positions of IR and density measurements.

Spectra were obtained using a Nicolet Magna 860 spectrometer coupled to a Nicplan microscopic unit, which focussed IR radiation on small areas of 200 (L) × 50 (T) μm of wood tissues, as shown in Fig. 1a, and described previously.[14,15] All spectra were the averages of 64 scans at 4 cm^{-1} resolution. Changes in absorption intensity at 1730 cm^{-1} were monitored to assess the effect of light on the chemical composition of wood. For all intensity measurements, the height of the band at 1370 cm^{-1} due to CH deformation in polysaccharides was used as an internal standard in accordance with the method of Tolvaj and Faix.[16]

Soft X-ray densitometry

X-ray density profiles[17] were obtained for the same annual ring used for IR depth profile analyses. Crosscut wood sections were conditioned for 2 days at 20°C and 65% relative humidity and were then exposed to soft X-rays generated by SOFTEX inspection apparatus (20 kVp, 14 mA, 4 min). X-ray images of the wood sections were recorded on negative films (Fuji IX-FR), and the images were enlarged 25 times using a projector. The intensity of light transmitted through the negative images was measured using a photosensor and converted into density data using an image analysis program (Walesch Electronic Dendro 2003) and a calibrated optical step-wedge. The dimension of the photosensor was 21 (T) × 0.5 (R) mm on the enlarged images and equivalent to 840 (T) × 20 (R) μm on the negatives. The photosensor scanned along the wood in the radial direction between earlywood and latewood and density data was collected at 10-μm intervals.

Results and discussion

Photodegradation depth profiles in sugi and hinoki

Figure 2 shows FT-IR depth profile spectra for sugi earlywood (a), sugi latewood (b), and hinoki earlywood (c) with densities of 0.24, 0.95, and 0.35 g/cm^3, respectively, subjected to 1500 h of irradiation. The weakening of the IR absorption peak at 1510 cm^{-1} (due to photodegradation of lignin), and the strengthening of the peak at 1730 cm^{-1} (formation of carbonyl groups), are notable features of the spectra.

The depth of photodegradation was greater in low-density sugi earlywood (a) than in hinoki earlywood (c) or sugi latewood (b). The maximum depth at which wood was photodegraded was determined by plotting changes in peak height at 1730 cm^{-1} (carbonyl) against the reference peak at 1370 cm^{-1} as shown in Fig. 3, and described previously.[14,15] The filled circles (sugi earlywood), squares (sugi latewood), and triangles (hinoki earlywood) in Fig. 3 indicate statistically significant changes in intensity, and provide a measure of the maximum depth of photodegradation (thresholds for statistical significance were different between wood types). Significant changes in the carbonyl peak were detected at depths of up to 650, 200, and 450 μm in sugi earlywood, sugi latewood, and hinoki earlywood, respectively.

The increased depth of photodegradation with decreasing wood density may explain the negative relationship between wood density and erosion. Horn et al.[11] noted that "a more dense structure would make it more difficult for penetration of light and water." Such effects of wood density on the penetration of light into wood and depth of photodegradation will be described and discussed later.

Rates of penetration of photoinduced chemical changes into sugi and hinoki earlywood

We reported previously that there was a logarithmic increase in the depth of photodegradation in sugi earlywood as a function of irradiation time[14] and suggested that this

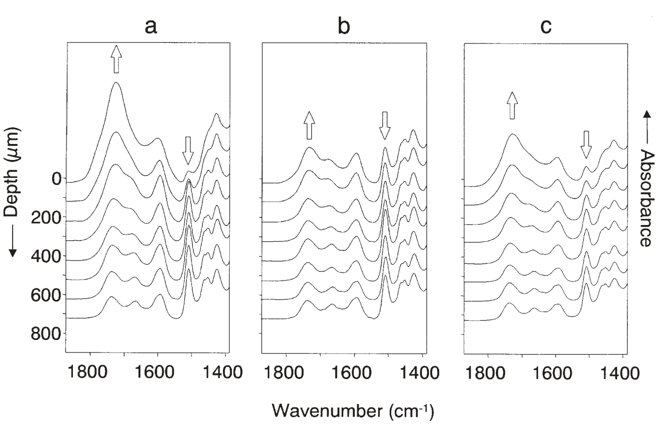

Fig. 2. Fourier transform infrared (FT-IR) depth profile spectra in the range 1800 to 1400 cm^{-1} of sugi earlywood (**a**), sugi latewood (**b**), and hinoki earlywood (**c**) after 1500 h of exposure to light. Not all the spectra are shown for clarity. *Arrows* indicate where pronounced changes in peak heights occurred

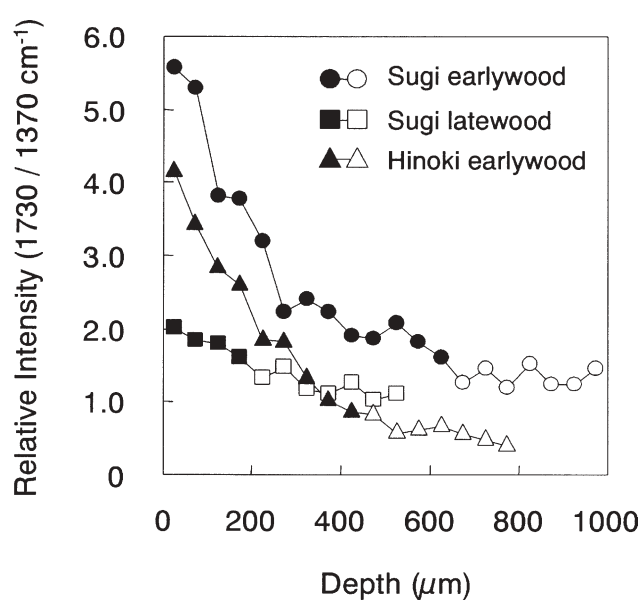

Fig. 3. Changes in absorption at 1730 cm^{-1} as a function of depth obtained from **a**, **b**, and **c** in Fig. 2. *Filled circles* (sugi earlywood), *squares* (sugi latewood), and *triangles* (hinoki earlywood) indicate that significant changes in peaks occurred as a result of exposure of wood to light (difference between unexposed controls and exposed specimens assessed using Smirnov-Grubbs test at the 5% significance level)

Table 1. Maximum depth of photodegradation and wood density values measured at 11 matching positions in the same sugi growth ring

Sample number	Density (g/cm^3)	Photodegradation depth (μm)	Distance from the beginning of growth ring (μm)	Tissue type
1	0.24	650	237	Earlywood
2	0.25	650	347	Earlywood
3	0.24	650	446	Earlywood
4	0.25	550	548	Earlywood
5	0.29	500	649	Earlywood
6	0.34	400	749	Earlywood
7	0.44	350	853	Earlywood
8	0.53	300	954	Transition
9	0.76	200	1053	Latewood
10	0.83	250	1154	Latewood
11	0.95	200	1256	Latewood

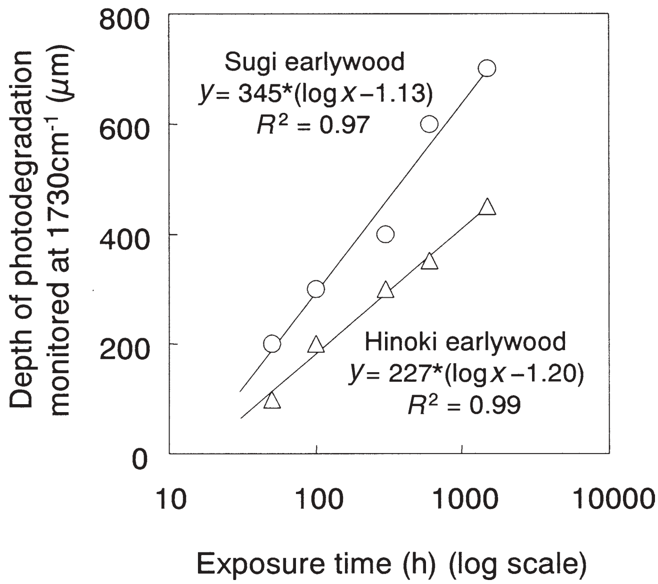

Fig. 4. Increases in depth of photodegradation in sugi and hinoki earlywood as a function of irradiation time, assessed by monitoring changes in IR absorptions at $1730\,cm^{-1}$. The data are plotted using a half-logarithmic scale

could be related to the exponential attenuation of light that occurs as it passes through wood.[15] Figure 4 plots the rate of the increase in depth of photodegradation in hinoki earlywood ($0.35–0.36\,g/cm^3$, $0.36\,g/cm^3$ on average) compared with that of sugi earlywood ($0.21–0.23\,g/cm^3$, $0.22\,g/cm^3$ on average). It is clear that there was a logarithmic increase in depth of photodegradation in hinoki (earlywood) as a function of irradiation time, although the rate of increase (a slope of linear fit) was smaller than that of sugi earlywood. The regression lines plotted in Fig. 4 indicate that at every exposure period the depth of photodegradation in hinoki earlywood was approximately 0.67 times that of sugi earlywood.

The differences observed in the photodegradation depth profiles of the two wood species may be related to differences in their density, because the average density of sugi exposed here was 0.61 times that of hinoki. Browning[18] reported that earlywood generally contains more lignin and less cellulose than latewood, although Ifju and Labosky[19] found "slight variations" in loblolly pine (*Pinus taeda* L.). Therefore, the possibility that differences in the chemical composition between wood species could have affected the results cannot be discounted, even though we used sapwood samples. In order to minimize the effect of between-species variation in wood chemical composition on photodegradation depth profiles, we compared the depth of photodegradation in serially cut sections obtained from earlywood and latewood.

Within-growth ring variation in photodegradation depth for sugi earlywood and latewood

The results of photodegradation depth profile analyses at 11 positions within a sugi growth ring, starting at the first-formed earlywood and finishing at the last-formed latewood at intervals of approximately $100\,\mu m$, are shown in Table 1, together with the corresponding density data obtained by X-ray densitometry.

The increased depth of photodegradation in sugi earlywood compared with latewood accords with previous observations of the susceptibility of these tissue types to photodegradation,[11–13] although Park et al.[13] observed that the depth of photodegradation in hinoki earlywood was 2.5 times that of latewood compared with the ratio of 3.25 observed here for sugi. This discrepancy may be due in part to differences in the ratio of density of earlywood and latewood between wood species, and the methods used to assess the depth of photodegradation in the two studies. Park et al.[13] did not measure the density of hinoki earlywood and latewood. Therefore we measured the density ratio of these tissue types in ten hinoki growth rings. The density of hinoki latewood was up to 2.78 times greater than that of earlywood whereas the comparable ratio for the sugi exposed here was 3.96 (Table 1).

Sell and Feist[3] obtained results that more closely match those obtained here. They observed that western red cedar earlywood and latewood with densities estimated to be 0.25 and $0.95\,g/cm^3$, were eroded to depths of up to 630 and $150\,\mu m$, respectively, following 2400 h of artificial weathering. The densities of sugi earlywood and latewood exposed to artificial sunlight here were 0.24 and 0.95 g/cm^3, respectively, and the depth of photodegradation in the two tissue types were 650 and 200 μm (Table 1). The similarity in erosion depth in western red cedar noted by Feist and Sell[3] and our observations of photodegradation depth profile in sugi suggests that depth of photodegradation and erosion are linked.

It can be seen in Table 1 that the density of sugi increased from 0.24 g/cm^3 in earlywood to 0.95 g/cm^3 in latewood, although the rate of increase was not linear across the growth ring. The variation in density observed between sugi earlywood and latewood here is representative of the extremes in density that occur between species. For example, Sell and Feist[3] examined the erosion rate of a range of species whose densities varied from 0.3 to 1.0 g/cm^3. Therefore, examination of photodegradation depth in earlywood and latewood exposed to light may provide useful insights into previous studies of the relationship between wood density and erosion.

Relationship between wood density and photodegradation depth

Figure 5 shows the changes in depth of photodegradation in sugi as a function of the reciprocal of wood density (volume per unit mass, cm^3/g) obtained from Table 1. A linear fit indicates that there is a precise inversely proportional relationship between wood density and maximum depth of photodegradation. This also explains our findings that the rate of the increase in depth of photodegraded hinoki was 0.67 times that of sugi when the density of sugi was 0.61 times that of hinoki (Fig. 4). Moreover, if erosion rate is plotted against the reciprocal of wood density using figures in the literature,[1,3,5] a similar relationship is obtained (Fig. 6). These findings further suggest that the depth to which light can penetrate and degrade wood tissues and the susceptibility of wood to erosion are linked. Clearly the relationship between wood density and erosion during weathering is inverse, but not linear. Sell and Feist[3] and Horn et al.[11] concluded that the most obvious reason for the variation in the weathering of wood is differences in wood density, rather than chemical differences (including extractives). Findings here support this conclusion.

Some understanding of the physics of the penetration of light into wood is useful in explaining the precise relationship between the inverse of wood density and photodegradation depth (Fig. 5) and erosion of wood exposed to UV light (Fig. 6). It has been established that the intensity of light transmitted by wood falls off exponentially with depth, as predicted by the Beer-Lambert equation:[20]

$$\log_{10}(I_0/I) = \varepsilon\beta\chi \tag{1}$$

where I_0 is the intensity of incident light, I is the intensity of transmitted light, ε is the wavelength-dependent absorptivity coefficient, β is the path length (or wood thickness), and χ is the concentration of the material (or wood density).

In a previous article[15] we showed that Eq. 1 could be used to predict the attenuation of light transmitted by thin sugi wood sections (within a range of a few hundred micrometers); however, it should be noted that such a prediction may become less accurate with increasing wood thickness because of the scattering of light that occurs in wood. The Beer-Lambert equation predicts that when the path length (β) and the concentration of the material (χ) are varied inversely, the same absorbance [($\log_{10}(I_0/I)$)] is obtained.

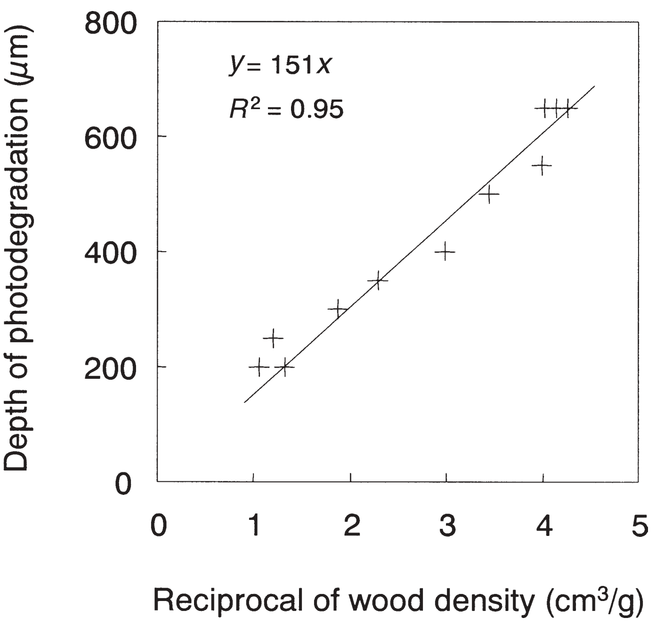

Fig. 5. Relationship between the reciprocal of wood density within a single sugi growth ring and maximum depth of photodegradation

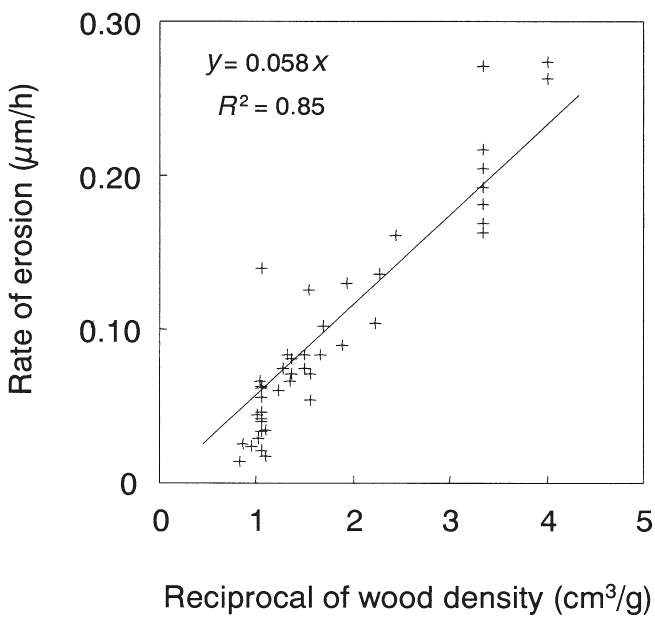

Fig. 6. Relationship between the reciprocal of wood density and erosion rate. Values for earlywood and latewood of softwoods and various hardwoods available in the literature[1,3,5] are plotted

This indicates that the depth (β) at which a certain percentage of the initial light is present in wood (i.e., I_0/I is constant) should decrease inversely with increases in wood density (χ). Accordingly, our finding that there is an inversely proportional relationship between the depth of photodegradation and the density of wood may be explained by the attenuation of light that occurs in wood surface layers exposed to solar radiation.

Conclusions

The photodegradation depth profiles that develop in sugi and hinoki during exposure to artificial sunlight largely depend on wood density. The finding that there is an inversely proportional relationship between the maximum depth of photodegradation and wood density explains why low density earlywood erodes more rapidly than denser latewood, and more generally why there is a similar inversely proportional relationship between the density of wood species and their rate of erosion during artificial and natural weathering. Future studies should examine the effect of other factors, such as wavelength of light and presence of chemical additives, which may affect the photodegradation depth profile of wood. It is hoped that such studies will allow the development of more effective photoprotective treatments that target the wavelength dependency of the phenomenon.

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