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# Improving the lightfastness and thermal stability of black urushi (oriental lacquer) by kraft lignin oleate

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# Abstract

Urushi, an oriental lacquer, is sensitive to light and easily deteriorates upon light exposure, leading to the fading and loss of gloss of its coatings. In this study, we attempted to improve the lightfastness of black urushi by utilizing the UV-absorbing properties of kraft lignin. Oleic acid-esterified kraft lignin was prepared and added to the black urushi to improve the compatibility. No aggregation of oleic acid-esterified lignin occurred in the black urushi, and the physical properties of the films were maintained to a large extent. A possible chemical interaction between lignin and black urushi was suggested upon thermal analysis. It was also found that the addition of lignin improved the thermal dimensional stability of black urushi. Finally, the high gloss factor of the black urushi film was maintained by adding oleic acid-esterified lignin after 168 h of light irradiation. Oleic acid-esterified lignin therefore appears as a promising functional additive to improve photostability of urushi coatings.

Keywords Urushi, Lignin, Oleic acid ester, Lightfastness, Thermal stability

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# Introduction

Urushi, an oriental lacquer, has been used as a natural resin coating in East Asia for centuries. It is made by refining the sap of the urushi tree (Toxicodendron vernicifluum (Stokes) F.A. Barkley), possesses high water resistance and antibacterial properties, and gives coatings with beautiful gloss. In particular, the black urushi has been prized for its beauty as a coating agent for traditional crafts, tableware, etc. [1]. Recently, urushi coatings have received a lot of attention owing to their environmentally friendly production processes. Urushi is available without consuming energy resources like artificial polymers and polymerized without any organic solvent. However, urushi is sensitive to light and deteriorate easily upon exposure to light [2, 3]. It has been reported that both ultraviolet (UV) and visible light rays deteriorate urushi films [3]. This deterioration leads to the fading and loss of gloss of urushi films, thereby substantially decreasing the value of the products coated with them.

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This degradation phenomenon is attributed to the UVinduced cleavage of reticular bonds and photo-oxidation catalytic reaction of urushi, similar to those of other polymeric materials. The primary component of urushi, urushiol, comprises a catechol ring with two hydroxyl groups and a long unsaturated hydrocarbon side chain. In a previous report, increased oxygen content on the surface of a urushi film upon light irradiation indicated oxidation of the film surface by both fluorescent and UV lamps [3]. The oxidization of polymer materials can be effectively prevented by the addition of UV absorbers [4] and hindered phenol antioxidants [5]. In this regard, previous studies have also reported the addition of hindered amine light stabilizers such as a series of derivatives of 2,2,6,6-tetramethylpiperidine [6] and 2-(2-hydroxyphenyl)-2H-benzotriazole [7] and bromine [8] to urushi. Meanwhile, new additives of natural origin that have less impact on the environment and human health are highly desirable, because many light stabilizers are synthetic or petroleum-derived compounds that are harmful to the human body or have a large environmental impact [9].

Kraft lignin (KL) is an important industrially produced lignin derived from kraft pulping, which is presently the dominant chemical pulping process [10]. Lignin is the only biomass rich in aromatic rings in nature because of its phenylpropane group and contains UV-absorbing functional groups such as phenols, ketones, and other chromophores [11]. KL contains more phenolic hydroxyl groups than native lignin and other technical lignin due to cleavage of  $\beta$ -allyl bonds during the cooking process of wood chips in sodium hydroxide and sodium sulfide [12]. In this study, we attempted to improve the lightfastness of black urushi by utilizing the aforementioned properties of KL.

It has been reported that lignin possesses UV absorption capacity; however, upon a lignin incorporation exceeding 10 wt%, the sunscreen effect disappears owing to lignin's tendency to agglomerate easily, resulting in a short shelf life and low-level sun blockage [11]. Therefore, lignin dispersion and compatibilization with matrix materials are important for the development of novel lignin-based UV-protection composite materials. Here it is important to note that the combination of lignin and urushi properties has been recently exploited to prepare wood protective coatings based on hybrid particles [13]. Herein, oleic acid-esterified KL was prepared and added to the urushi to improve the compatibility. The thermal and physical properties of the urushi films were analyzed to evaluate the compatibility of the lignin with the urushi, and lightfastness of the prepared urushi films was measured.

# Materials and methods Materials

A black oriental lacquer (urushi; kuro-urushi, Tsutsumi asakichi urushi, Kyoto, Japan) solution, manufactured in 2019, was used to prepare the samples. The iron content in the black urushi was determined to be 0.37 wt% via elemental analysis. KL powder was provided by Nippon Paper Industries Co. Ltd. The KL powder was extracted by treating a mixture of soft wood tips and hard wood tips with Na<sub>2</sub>S and NaOH at ~ 150 °C. The total hydroxyl and phenolic hydroxyl amount of KL were 4.0-6.0 and 1.0-2.0 mmol/g, respectively, while its total sulfur content was 1.0-3.0 wt%. Oleic lignin (OL) powder, which consists of lignin oleic acid ester prepared via base-catalyzed esterification of softwood KL (BIOPIVA<sup>™</sup> 100 pine Kraft lignin (UPM, Finland), previously characterized [14]) with oleoyl chloride (Merck, 89%), was prepared as reported previously [15]. Briefly, 5.0 g of softwood KL (containing 5.94 mmol/g of total phenolic and aliphatic hydroxyl groups measured by quantitative <sup>31</sup>P NMR analysis) was dissolved for 30 min in a binary mixture of anhydrous tetrahydrofuran/dimethylformamide (30 mL/7.5 mL) containing 3.8 mL of anhydrous pyridine at 40 °C under nitrogen atmosphere. After that, oleoyl chloride (6.95 mL) was added with a syringe within 10 min. The reaction mixture was purged with nitrogen and kept for 15 h at 45 °C. After that, the reaction mixture was concentrated, dissolved in dichloromethane, and washed with brine 3 times. The organic fractions were collected, dried with MgSO4, and concentrated under vacuum to afford OL as a dark powder. The degree of esterification (DE) (DE = 50%) was calculated by direct comparison of the total amount of hydroxyl groups determined by <sup>31</sup>P NMR spectroscopy between the OL and softwood KL (Fig. 1 and Table 1).

# Preparation of urushi/lignin composite film (Fig. 2)

The preparation methodology corresponding to the urushi/lignin composite films is illustrated in Fig. 2. 1 g of urushi solution was coated onto a polyethylene terephthalate (PET) film with a size of ~15 ×8 cm using a film applicator. The samples were then thermoset in a dry oven at 90 °C for 45 h. An urushi (UR) film was obtained by peeling the PET film. The thickness of UR film was 150  $\mu$ m. Urushi/lignin films were prepared from urushi solutions with 1, 5, and 10 wt% of KL (designated UR-KL-1, UR-KL-5, UR-KL-10, respectively) and OL (designated UR-OL-1, UR-OL-5, UR-OL-10, respectively) powders following the same procedure as that of UR. Both lignin powders were dissolved in a small amount of methanol before being added to the urushi solution [16]. The thickness of each films was 146  $\mu$ m for UR-KL-1, 177  $\mu$ m for

**Table 1** Concentration of aliphatic and phenolic OH ofsoftwood KL and OL according to quantitative <sup>31</sup>P NMR

Sample	Aliphatic OH	Phenolic OH	Total OH
Softwood KL	1.89±0.04	4.05±0.01	5.94±0.09
OL	$0.61 \pm 0.05$	$2.26 \pm 0.05$	2.86±0.01

Unit mmol/g. At least three measurements were completed for each parameter. Error ranges correspond to one standard deviation from the mean value

UR-KL-5, 180 µm for UR-KL-10, 161 µm for UR-OL-1, 149 µm for UR-OL-5, and 137 µm for UR-OL-10.

# Material characterization

<sup>31</sup>P NMR spectra were recorded on a Bruker DRX 4OO NMR instrument following a previously described [15]. A 90° pulse angle, inversed gated proton decoupling, and a delay time of 10 s were used. For the analysis, 256 scans with a 6 s delay and a total runtime of 30 min were used for each sample. At least three replicated experiments were conducted, and the mean value of one standard deviation is reported.

A glossmeter (IG-331; HORIBA, Ltd., Japan) was used to measure the gloss level, and the corresponding measurement scale was calibrated from 0 to 100 gloss units. The wavelength of the light source was 890 nm and the measurement angle was 60°. Measurements were performed at 15 different points and the subsequent average was calculated.

Surface and cross-sectional observations at the micrometer scale were made via scanning electron microscopy (SEM, S-3400N, Hitachi High-Tech Science Co., Japan). The samples were coated with platinum using an ion sputter coater.

The tensile moduli, strength, and strain of the films were evaluated using a universal material testing machine (EZSX, Shimadzu Co., Kyoto, Japan) for dumbbell-shaped specimens (ISO 527-2:2012 [17], model 5B) at a crosshead speed of 1 mm/min.

Thermogravimetric (TG) and derivative TG (DTG) analyses were performed using a TG analyzer (STA7200 RV, Hitachi High-Tech Science Co., Tokyo, Japan) at a heating rate of 10 °C/min from ambient temperature to 600 °C after an initial heating at 110 °C for 20 min to dehydrate the samples in a nitrogen atmosphere.

Differential scanning calorimetry (DSC) was performed using a DSC-60APlus (Shimadzu Co., Japan) at a heating rate of 10 °C/min from ambient temperature to 300 °C. The analyses were performed in a nitrogen atmosphere at a flow rate of 60 mL/min.

The coefficient of thermal expansion (CTE) was measured via thermomechanical analysis (TMA) (TMA7100, Hitachi High-Tech Science Co., Tokyo, Japan) for 25-mm-long and 3-mm-wide samples with a 20-mm span. The measurements were performed during elongation at a heating rate of 5 °C/min in a nitrogen atmosphere at a load of 3 g. The subsequent CTE values were determined at 20–150 °C in the second run.

#### Lightfastness measurement

An exposure test was conducted for all the films using a light irradiation equipment (SUNTEST XLS+, Atlas Material Testing Technology, USA). The filter system was daylight, and the radiation wavelength was 300-800 nm with the irradiance range of 65 W/m<sup>2</sup> in 300-400 nm, 7650 W/m<sup>2</sup> in 300-800 nm, and 130 klx in Lux in order to make it more relevant to actual use. A thermostatic water circulator was connected to the sample table with a set temperature of 25 °C and water was constantly circulated during the test to maintain thermostatic conditions. The films were then exposed to light for 24, 48, 72, 96, and 168 h. It is noted that black color is achromatic and with no chromatic value. It was reported that the  $a^*$  and  $b^*$  values, the color value of CIELAB color system, of black urushi coating were approximately zero [18]. Thus, gloss is an important factor for determining the value of the black urushi coating and provides means to index its surface degradation. In this study, the gloss level was obtained for each exposure time. Attenuated total reflection Fourier transform infrared (ATR-FTIR, Spectrum One, PerkinElmer, Inc., USA) spectra were measured after 168 h of irradiation and compared with the pre-irradiation spectra. The spectra were obtained in the range of 4000–380  $\text{cm}^{-1}$  with a resolution of 4  $\text{cm}^{-1}$  for 16 scans.





Fig. 2 A schematic overview of UR film, UR-KL film, and UR-OL film

# **Results and discussion**

## **Gloss levels and SEM surface observation**

Urushi can be cured via oxidative or thermal polymerization [16, 19]. The double network structure arising from the oxidative polymerization of catechol moieties and coupling reactions of the hydrocarbon side chains take more than one month to form a tough coating. In contrast, thermal polymerization, which occurs via thermal Diels–Alder and addition reactions, is completed within several minutes [19]. Moreover, urushi coatings with ideal coloration and high water repellency are generally obtained via thermal polymerization rather than oxidative polymerization [20]. Subsequently, thermal polymerization was used in this study for the preparation of samples. The SEM micrographs of the film surfaces and their corresponding gloss levels (GLs) are shown in Fig. 3. In the case of KL, the surface appearance and GL of UR-KL-1 and UR-KL-5 was not significantly different



Fig. 3 SEM images of surface of the UR films: a UR, b UR-KL-1, c UR-KL-10, e UR-OL-1, f UR-OL-5, g UR-OL-10. "GL" means gloss level of film

from that of UR (Fig. 3a–c). However, aggregates were observed on the surface of UR-KL-10 (Fig. 3d) and GL was decreased drastically. It was suggested that the lignin aggregates in urushi result in a decrease in gloss.

In OL, the high GL either remains intact or increases slightly with the addition of 10% OL (Fig. 3g). The SEM observations show small white spots on the surface of UR-OL-5 and UR-OL-10 (Fig. 3f, g). Traditionally, plant oil has often been used as diluents to reduce the viscosity of urushi paints and to enhance the gloss of polymerized UR films [21, 22]. It is considered that the oil surface oozing from the polymerized film provides a higher gloss. A partial oil surface may have formed on the surface of the UR-OL-5 and UR-OL-10, causing an increase in GL.

#### **Thermal properties**

The results of the DSC analysis are shown in Fig. 4. The DSC signal observed in the 50–250 °C temperature range can be ascribed to free-radical chain reactions, referred to as propagation, oxygen uptake, and termination22. The endothermic peak observed in the 70-100 °C temperature range is due to water evaporation. According to the literature, the peak at ~150 °C is most likely due to the autoxidation of unsaturated alkyl chains [22, 23]. This is presumed to be due to the thermal cross-linking of uncross-linked urushiol, i.e., the exothermic value roughly corresponds to the number of the remaining double bonds in the urushiol. In our previous study, we confirmed that the thin urushi coating was fully thermally cured if it was maintained at 90 °C for more than 24 h [18]. However, in the present study, the UR film was uncured even after heating at 90 °C for 45 h. The high amount of urushi used to form the thick film may have prevent fully cured. Moreover, the calorific value increases upon the addition of lignin regardless of the presence of KL or OL (Fig. 4), indicating that the addition of lignin inhibits the cross-linking of urushiol. The degradation of GL in UR films due to light irradiation is attributed to the progressive cross-linking of urushiol [7]. The presence of lignin in the film as a component that inhibits the cross-linking of urushiol is thought to have also inhibited light degradation. This is supported by the fact that there were many uncured areas in OL-10, which had particularly high lightfastness (Fig. 4b).

The TG and DTG curves of the KL powder, OL powder, UR, UR-KL series, and UR-OL series are shown in Fig. 5. Compared to urushi, lignin has a lower pyrolysis initiation temperature, which reduced further upon modification with oleic acid due to the thermally labile ester linkages. However, after compositing with urushi, the heat resistance of urushi was maintained, and no decrease was observed in its heat resistance due to the addition of lignin. The DTG peak for urushi exhibited a shoulder and peak at 317 and 455 °C, respectively (Fig. 5b, c, Table 2). For the KL powder, the shoulder and peak were observed at 187 and 387 °C, respectively (Fig. 5b, Table 2), whereas for the OL powder, the shoulder and peak were observed at 244 and 365 °C, respectively (Fig. 5c, Table 2). The peaks at around 390, 250, and 150 °C indicate the cleavage of C-C bonds [24], release of organic substances for an alkyl [25], and dehydration [26], respectively. In both UR-KL and UR-OL, no lignin-derived degradation peaks as described above were observed (Fig. 5b, d). This suggests that a chemical bond between lignin and UR was formed. This interaction may have inhibited urushiol cross-linking and led to the improvement of lightfastness. On the other hand, the



Fig. 4 DSC curves of the UR films. **a** includes the results from films of pristine urushi and with KL and **b** includes the results from films of pristine urushi and with OL



Fig. 5 Thermogravimetric curves **a**, **c** and derivative weight change curves **b**, **d** of the UR film. **a**, **b** include the results from films of pristine urushi and with KL and **c**, **d** include the results from films of pristine urushi and with OL

Table 2 Peak terr	nperature of DTG curv	/es of the urushi	(UR) film
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Peak temperature of DTG curves/°C	
317	455
316	452
316	454
294	453
187	387
320	459
316	451
314	451
244	365
	Peak temperature of DTG curves/°C           317           316           316           294           187           320           316           314           244

first degradation peak at 294 °C was observed for UR-KL-10, which was lower than that of UR film (Fig. 5b, Table 2). The amount of KL, which is less compatible with urushi, may have been too high, resulting in insufficient bonding with the UR. For the 10% OL composite film, both the first and second peaks appear stable as UR film. It suggests that interactions between the OL and UR were formed successfully.

The thermal dimensional stability of the UR films was further examined via TMA (Table 3, Fig. 6). The UR film expanded linearly with increasing temperature, and the CTE calculated by TMA measurement was 162 ppm/K. The CTE was reduced to 109 ppm/K for the UR-KL-10

Tal	ble 3	3 Therm	al expansio	on of t	the UF	{ films
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	UR	UR-KL-1	UR-KL-5	UR-KL-10	UR-OL-1	UR-OL-5	UR-OL-10
CTE/10 <sup>-6</sup> K <sup>-1</sup>	162	123	111	109	144	138	121



Fig. 6 TMA curves of the UR films. a includes the results from films of pristine urushi and with KL and b includes the results from films of pristine urushi and with OL

and to 121 ppm/K for the UR-OL-10. Namely, the thermal stability of the UR films was successfully improved by the addition of lignin. According to the DSC results, the addition of lignin possibly inhibited the cross-linking of urushiol. Lignin upon penetrating urushiol would restrict the movement of the alkyl chains in the urushi and lower the coefficient of thermal expansion. In general, thermal expansion of composites can be controlled more effectively when there are fewer voids at the filler-matrix interface and the interface area ratio is smaller [27]. However, in this study, KL improved the thermal stability of the UR films better than OL. It is assumed that KL more effectively restricted the movement of the alkyl chains of the urushi because the molecules are less likely to move when heated than OL, which has longer alkyl chains. On the other hand, UR-KL-5 and UR-KL-10 showed almost no change in the coefficient of thermal expansion, while the coefficient of thermal expansion of UR-OL decreased as the ratio of OL increased (Table 3). This suggests that too much KL is less effective in suppressing thermal expansion due to voids at the interface with the urushi.

#### **Mechanical properties**

The tensile properties of the UR films are listed in Table 4 and Fig. 7. The elastic moduli of the UR films with KL gradually increase until 5 wt% KL (UR-KL-5). In contrast, both the tensile strength and strain decrease with the addition of KL. For UR-KL-10, the mechanical properties deteriorated significantly, resulting in a brittle film. Compatibility of lignin with the matrix components is important for lignin to act as a filler [28]. Insufficient

 Table 4
 Mechanical properties of the UR films

Sample	Elastic Modulus GPa	Tensile stress MPa	Tensile strain %
UR	1.12±0.05	40.6±3.2	6.84±0.34
UR-KL-1	1.30±0.09	$34.9 \pm 5.4$	$2.96 \pm 0.24$
UR-KL-5	1.57±0.13	$24.2 \pm 3.6$	$1.66 \pm 0.31$
UR-KL-10	0.34±0.17	$7.32 \pm 1.4$	1.18±0.25
UR-OL-1	1.13±0.09	42.2±3.1	$4.01 \pm 0.43$
UR-OL-5	$1.25 \pm 0.06$	$40.0 \pm 5.3$	$3.73 \pm 0.52$
UR-OL-10	1.50±0.09	41.9±3.7	$4.40 \pm 0.61$

The average values and standard deviations (mean  $\pm$  S.D.) were calculated using at least 5 independent specimens

compatibility causes phase separation between the filler and the matrix, leading to expansion in free volume and decreasing density, and no reinforcing effect. KL has more OH groups and thus has higher polarity than UR, and the strong cohesive force among KLs may have made uniform dispersion difficult. The compatibility between KL and UR was insufficient, resulting in agglomeration, which may have deteriorated the film's physical properties and GL before the irradiation. The results of TGA analysis also suggest that UR-KL-10 may not have sufficient chemical interaction between KL and UR.

In the case of OL, the elastic moduli improved upon increasing the amount of added lignin. The tensile strength was maintained, although the strain decreased (Table 4, Fig. 7e–g). It is likely that OL, which has a long alkyl chain similar to that of urushiol, improves compatibility of the urushi and did not cause a dramatic decrease



Fig. 7 Typical S–S curves of UR film. a UR, b UR-KL-1, c UR-KL-5, d UR-KL-10, e UR-OL-1, f UR-OL-5, g UR-OL-10

in physical properties of film when KL was added. And this high degree of compatibility probably contributed to the improvement of GL before the irradiation. Figure 8 shows the results of cross-sectional observations corresponding to tensile testing. As for the cross section of UR-KL-1 (Fig. 8b), no significant differences



Fig. 8 SEM images of cross sections of the UR films after tensile test: A Low magnification, B High magnification: a UR, b UR-KL-1, c UR-KL-5, d UR-KL-10, e UR-OL-1, f UR-OL-5, g UR-OL-10

were observed compared to UR (Fig. 8a). However, the cross section of UR-KL-5 (Fig. 8c) was rougher than UR and UR-KL-1. Then, some aggregations were observed in UR-KL-10 (Fig. 8d). These results strongly suggested to the insufficient compatibility of KL and UR.

The cross sections of UR-OL-1 (Fig. 8e) and UR-OL-5 (Fig. 8f) were also rougher than UR. However, UR-OL-10 showed a completely different morphology (Fig. 8g). Mixing urushi with oil such as linseed oil or tung oil is a common technique for enhancing rheological and mechanical properties of film [21, 22]. Since the strain of UR-OL-10 is slightly improved over UR-OL-1 and UR-OL-5, the effect of drying oil in urushi, i.e., the plasticizing effect, may have been caused by the addition of OL. It may be possible to achieve both good physical properties and lightfastness by increasing the amount of OL added or changing the DE.

#### Lightfastness

The changes in glossiness corresponded to the irradiation of the prepared UR films with visible light (Fig. 9). The GL of the UR decreased with increasing irradiation time, reaching 85% after 168 h. In KL, the glossiness is reduced by the addition of lignin before irradiation as described above, but the reduction in the GL upon light irradiation was almost suppressed. The GL of UR-KL-1 has a slightly reduced after 24 h but remained intact thereafter (Fig. 9a, red triangle). After 168 h of irradiation, the GL of UR-KL-1 and UR-KL-5 film was higher than that of the UR. The UR-KL-10 had low GL before the irradiation as described above, but did not decrease significantly beyond that after the irradiation (Fig. 9a, yellow diamond).

The GLs of UR-OL-1 and UR-OL-5 were decreased slightly after irradiation, but remained above 95% after 168 h irradiation (Fig. 9b, purple cross and blue asterisk). The UR-OL-10 showed the highest GL even after 168 h irradiation.

The degradation of urushi upon light irradiation is caused by the post-curing cross-linking of uncross-linked urushiol residues that increase the cross-link density, causing cracks, shrinkage, and whitening [7]. The presence of lignin might inhibit the cross-linking of urushi via light radiation.

Another possible mechanism is the prevention of oxidation of the unsaturated alkyl chains of urushiol by lignin. The results of the FTIR analysis before and after light irradiation showed an increase in the carbonyl groups in the absorption at  $1710 \text{ cm}^{-1}$  in all samples (Fig. 10). This indicates the progression of oxidation due to light irradiation. Urushi is oxidized by light irradiation, conforming with the previous reports [3]. The degradation of urushi is caused by the oxidation of urushiol, which induces photo-oxidation, resulting in a decrease in the gloss level. However, the oxidation of the UR films with KL and OL in the samples may be due to the oxidation of lignin. Lignin is also reacted oxidatively [29], as inferred from its structure. Meanwhile, in the samples with KL and OL, lignin is preferentially oxidized, possibly preventing the oxidation of urushiol. Moreover, the degradation of UR films with KL and OL would be prevented because lignin and its fractions can be inhibitors



Fig. 9 Gloss level of the UR films before and after irradiation with visible light. **a** includes the results from films of UR and with KL and **b** includes the results from films of UR and with OL. Error bars correspond to the standard deviation from the mean value



4000 3500 3000 2500 2000 1500 1000 500 Wavenumber (cm<sup>-1</sup>)

**Fig. 10** FTIR spectra of the UR films before and after irradiation with visible light: **a** UR, **b** UR after irradiation for 168 h, **c** UR-KL-10, **d** UR-KL-10 after irradiation for 168 h, **e** UR-OL-10, **f** UR-OL-10 after irradiation for 168 h

in free-radical auto-oxidation [30–32]. Thus, lignin, a natural material, could be added to black urushi to prevent its light exposure-induced loss of gloss and used as a stabilizer to improve its lightfastness. Furthermore, the OL was determined to be more effective in the sense that there was no decrease in glossiness before the irradiation due to its addition.

# Conclusion

Kraft lignin (KL) was evaluated as a functional additive to improve the lightfastness of black urushi UR film. The addition of KL prevented the degradation of the gloss of the black urushi film due to light irradiation, but the gloss before light irradiation decreased as the amount of KL added was increased. On the other hand, when oleated kraft lignin (OL) was added to the urushi, the gloss before light irradiation remained high even when 10 wt% OL was added, and the gloss reduction due to light irradiation was also prevented.

SEM observations and mechanical test results suggest that KL agglomerated in the UR film, and the results of TGA analysis also suggest that the chemical interaction between KL and urushi is insufficient. On the other hand, the UR films in which OL was added maintained the physical properties of the films, suggesting that lignin aggregation did not occur and that the compatibility between urushi and OL was good. The addition of KL and OL suppressed the thermal expansion of the UR film, indicating that the presence of lignin in the UR film contributes not only to the light resistance but also to the thermal dimensional stability. To further increase light stability and improve physical properties, it will be necessary to consider modifying KL by varying the DE or the length of the alkyl chain and complexing it with urushi.

#### Abbreviations

UV KL	Ultraviolet Kraft lignin
OL	Oleic lignin
DE	Degree of esterification
PET	Polyethylene terephthalate
UR	Urushi
SEM	Scanning electron microscopy
TG	Thermogravimetric
DTG	Derivative TG
DSC	Differential scanning calorimetry
CTE	Coefficient of thermal expansion
TMA	Thermomechanical analysis
ATR-FTIR	Attenuated total reflection Fourier transform infrared
GL	Gloss level

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#### Author contributions

TT contributed to methodology, investigation, and writing—review and editing. CN and KY were involved in methodology, data curation, and writing—review and editing. AM and HT were involved in investigation and writing—review and editing. MHS contributed to methodology and writing—review and editing. YO was responsible for conceptualization, methodology, validation, supervision, and writing—original draft.

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#### Availability of data and materials

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

#### Declarations

#### **Competing interests**

The authors declare that they have no competing interests.

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