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Abstract: Although linseed oil (LO) has been used in wood protection for centuries, research continues to develop new and more effective formulations and treatment approaches. In the future, growing interest in LO use could be expected due to its cost and environmental friendliness. This review summarizes recent research (from 2000 onwards) on the use of LO in wood protection, published in peer-reviewed scientific journals and included in the online publication databases Scopus or Web of Science. The studies cover surface and impregnation treatments of various wood substrates using different LO formulations, including chemically modified LO and the use of LO as a base for the development of biofinish and as a medium for thermal modification of wood, as well as research into the mechanisms behind the changes in wood properties due to treatment methods and interaction with LO formulations. Although the improvement of wood hydrophobicity and biodurability dominates, other aspects such as weathering and color stability, adhesion, and environmental safety are included in these studies. In general, almost all of the studies show a greater or lesser potency of the proposed approaches to provide benefits in wood protection; however, the level of innovation and practical feasibility varies.

Keywords: wood; linseed oil; wood protection

1. Introduction

Both wood and linseed oil (LO), each individually and in combination, have been used for a variety of applications since ancient times, although the areas and intensity of their use have transformed over time. Today, renewable materials are becoming increasingly important in light of environmental concerns and the risk of depletion of fossil resources. Together with other natural oils, LO has attracted increased interest as a raw material for various future applications, with the ability to substitute fossil-based products while providing competitive performance and cost [1,2]. LO is a triglyceride containing unconjugated polyunsaturated fatty acids produced from the seeds of the flax plant (Linum usitatissimum) by pressing. The majority of LO fatty acids consist of C₁₈-chains and contain unsaturated bonds. The main LO fatty acids are linolenic acid (35–60%), with three unconjugated double bonds; linoleic acid (17-24%), with two unconjugated double bonds; and oleic acid (12–34%), with one double bond. In addition to the unsaturated fatty acids, LO also contains a small amount of saturated fatty acids [3]. Due to its fatty acid composition, LO is characterized by a high iodine value (160-200) and is classified as a drying oil according to oil classification based on the potency to polymerize and build solid film under exposure to atmospheric oxygen via the well-studied process of autoxidation [3,4]. Although the wood industry is not the main consumer of LO and LO has lost its former position in wood protection, LO is still the predominantly used drying oil for wood in Europe [5]. Several recent reviews have addressed various aspects of the use of LO, but the use of LO for wood protection has not been the focus of these reviews [4,6-8]. The present review summarizes the findings of relatively recent research studying various aspects of the use of LO in wood protection. The time frame of these scientific papers covers the 21st century (starting from 2000 onwards), and only the articles written in English and published in peer-reviewed scientific journals included in the online publication databases Scopus or Web of Science



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Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). were considered. This review includes the studies related to the following areas of the use of LO in wood protection: surface treatment, impregnation, a base for biofinishing, a medium in the thermal modification of wood, and performance optimization by LO chemical modification (Figure 1).



Figure 1. The scope and the structure of the review.

2. Surface Treatment

The studies exploring the use of LO for wood surface protection include LO with or without functional additives and its use on both unmodified and modified wood surfaces. The most common idea behind the use of LO for wood surface treatment is related to the desire to reduce the adverse effects of water. The motivation for using LO on charred wood surfaces is also, or mainly, associated with the fixation of the crumbly surface [9]. However, the effect of surface treatment with LO cannot always be evaluated based on the reported data due to the lack of results characterizing the reference specimens without the application of LO, e.g., [10–12]. The studies addressing different aspects of LO treatment on wood surface hydrophobicity are summarized in Table 1. Different wood substrates, LO formulations, and influencing factors have been studied.

Authors	Reference	Wood Substrate	LO Specification	Hydrophobicity Evaluation
Arminger et al.	[5]	Oak, beech	LO without and with (1 wt%) dryer	Effect of drying time
Kymäläinen et al.	[9]	Norway spruce, Scots pine, silver birch, trembling aspen (sapwood, charred)	LO	Effect of contact and flame charring
Ibanez et al.	[11]	Eucalyptus (Bosisto's box), loblolly pine (charred)	LO	
Weththimuni et al.	[13]	Maple	Cooked (270 °C) LO with colophony	Ratio of components
Timar et al.	[14]	European ash, European walnut, sycamore maple	Boiled LO	Effect of aging on resistance to water
Janesch et al.	[15]	Spruce	LO emulsion (1.02 wt% oil content)	

Table 1. Studies addressing the effect of linseed oil (LO) on wood hydrophobicity.

Authors	Reference	Wood Substrate	LO Specification	Hydrophobicity Evaluation
Yaremchuk et al.	[16]	Scots pine (heart wood)	LO-based product	Resistance to water (immersion 24 h)
Šeda et al.	[17]	European beech (charred)	LO	Effect of artificial weathering (UV + water spray)
Kutnar et al.	[18]	Scots pine (densified)	Cold pressed LO	Effect of densification degree
Petrič et al.	[19]	Scots pine (sapwood, densified)	Cold pressed LO	Effect of densification degree
Ekstedt and Östberg	[20]	Norway spruce	LO paint (solvent-based)	Effect of artificial weathering (UV + water spray)

Table 1. Cont.

In general, surface treatment with LO lends the treated surface enhanced hydrophobicity and this is observed for both unmodified wood [5,13-16] and wood with modified surfaces including surface charring [17] and densification [18,19]. On the other hand, no effect on water vapor permeability was observed for unmodified and contact-charred wood with LO-treated surfaces compared with their uncoated counterparts [9]. However, an increase in moisture exclusion efficiency and a decrease in liquid water uptake were reported in the same study for the LO-treated wood. A high level of wood protection from water absorption by using LO coating was also observed for the coated wood surface subjected to long exposures (72 h) to liquid water, meeting the requirements for coatings used on wood for "stable construction" (EN 927-5) [20]. In a study exploring the treated surface repellence stability over time, it was observed that the LO-treated wood surface has a high water resistance and slightly lower resistance to ethanol and acetone which is maintained during aging in simulated indoor conditions [14]. It has also been shown that a high surface water repellency can be obtained by applying a water emulsion of only 1% LO content, which is speculated to be associated with the higher surface micro-roughness maintained when using low-load emulsions [15]. The studies show that surface hydrophobization depends on the wood substrate and the completeness of LO drying. Arminger et al. [5] found that the wood substrate can significantly influence the drying in their experiments applying a similar LO formulation to beech and oak wood, which resulted in considerably inhibited film formation on the latter. According to the authors, this may be due to the differences in the chemical composition of the two woods, with oak wood containing a higher content of extractives prolonging the induction period of the LO polymerization. In the same study, the increase in hydrophobicity with drying time is well documented. However, a decrease in contact angle due to treatment with LO was observed in one study, where the surface was dried only for 1 h after LO application [21]. However, knowing the drying peculiarities of LO with and without the drying stimulating additives, it is more likely that the polymerization process was incomplete and a solid film had not yet been fully formed due to the relatively short drying time at the moment of contact angle measurement in this study. This reasoning is also supported by the results of the studies tracing the process of LO drying [4,5,16,22–24]. The difference in the protective effectiveness of LO against water depending on the wood species was also observed for charred wood surfaces [9].

As could be expected from earlier studies, the surface treatment of unmodified wood with LO without additives provides a slight or no protective effect against wood surface photodegradation caused by exposure to UV irradiation, which has been approved by the evaluation of discoloration (CIELAB color model) and chemical changes (FTIR analyses) [14,25–27]. High lightfastness, which was determined by evaluating visual color changes and the degree of loss in glossiness, has been reported for pine wood coated with LO [16]. However, missing details about the experiment, such as information about the spectral characteristics of the used illuminant and duration of the exposure, impede the viewing of these results in an overall context of surface photostability. Interestingly, smaller

total discoloration (ΔE in the CIELAB color model) was observed for the surface treatment with an LO nanoemulsion compared to LO with the effect intensifying as the duration of the exposure increased [27]. In addition, the 10% LO nanoemulsion performed better than the 30% one, although in general both formulations only slightly reduced discoloration. On the other hand, it was observed that, despite high total discoloration characterized by ΔE , LO provides protection from greying caused by UV exposure supplemented with water spraying and condensation steps [28]. However, higher discoloration was observed for wood coated with LO in comparison with uncoated wood when exposed outdoors [29] and artificial weathering including UV irradiation and water spray [17]. In general, using additives is shown to be a more promising approach. Surface treatment with LO nanoemulsions containing nanoparticles of the well-known photo inhibitors zinc oxide or cerium oxide exhibited good UV resistance, with the former showing better results [27]. Other recently tested LO additives include pigments obtained from wood decay fungi [28,30,31]. In these studies, LO was used as a medium for blending and a carrier of dyes extracted from fungi of two genera (Chlorociboria and Scytalidium) grown on malt agar plates amended with wood. This approach could be attributed to the use of an old practice in a new way, as the use of pigments produced by fungi for wood staining has long been known [28]. A relatively high level of color coverage retained on the wood surface after an artificial weathering test (including exposure to UV radiation and water spray) was observed for wood specimens coated with LO containing a pigment dramada from *Scytalidium cuboideum* suggesting potential for its use in wood surface protection [28].

Equivocal results of the effect of surface treatment with LO on protection from weathering were observed for the modified wood. Coating with boiled LO decreased the rate of discoloration and cracking of acetylated hornbeam wood exposed outdoors while significantly increasing discoloration due to UV irradiation [26]. Both linseed oil and linseed oil varnish also increased the discoloration of ethanolamine-fumed cherry wood surfaces exposed to short-wavelength UV irradiation (radiation peak at 253.7 nm), which, however, differs considerably from the real environment [25]. A different effect of charred wood surface treatment with LO on discoloration due to outdoor weathering for one year was observed depending on the charring method (contact, flame, and flame followed by brushing) and wood species with no benefit from the LO treatment observed for pine wood independently of the charring method [29]. Also, no benefits or even impairment from the oiling of the surface for protection from discoloration was observed for beech wood with contact-charred surfaces in an artificial weathering test that included UV irradiation and water spray [17].

Other studied effects of LO use in coatings for wood include bioprotection, mechanical properties, resistance to abrasion and different liquids, and thermal stability. In the study investigating the effect of surface-charred beech wood treated with LO, a slight reduction but no protection of the wood against decay and mass loss due to exposure to white and brown rot fungi was observed for the reference and charred wood [17]. Similarly, little improvement in mold resistance was observed for the treatment of unmodified and thermally modified Turkey oak heartwood surfaces with LO [32]. On the other hand, an antiadhesive effect to some fungal strains has been reported for wood treated with LO [21]. However, as mentioned above, it is not clear how well the test surfaces can represent coated wood because of the very short drying time of the LO used in the experiments.

In the study of thermally modified Turkey oak wood finished with LO, a decrease in adhesion but an increase in hardness, abrasion, and impact resistance was observed with an increasing wood thermal modification temperature [32]. An improvement in the abrasion resistance of LO varnish was achieved by the addition of chemically modified nanofibrillated cellulose with the effect depending on the cellulose modification [33]. Enhanced hardness has also been observed for varnish prepared according to an old recipe for musical instruments using LO and colophony when a formulation with increased LO was used [13]. LO-based coating showed high resistance to temperature which did not depend on the film thickness and was higher compared to alkyd-based coating [16]. The changes in wood color due to surface treatment with LO, making the wood more yellow, is another piece of information provided directly or indirectly by several publications, but it is not the main focus of the studies and does not reveal any new aspects [14,26,27,34]. Ohshima et al. [35] studied the processes behind color changes due to the application of LO and demonstrated that the LO penetration into the wood structure results in a color change in response to structural changes in the wood causing transformations in all optical components (reflectance, transmittance, absorption). The largest total changes were observed at a wavelength of 480 nm. In addition, the authors found that the changes did not depend on the applied amount of LO.

Different performances of LO coating have been observed depending on the wood substrate when unmodified and thermally modified woods were examined. Better penetration into thermally modified aspen wood than into its unmodified counterpart was observed for an emulsion containing an alkyd and LO [36].

A study on potential emissions of aldehydes from a LO-based coating during drying showed that the use of wood as a substrate for this type of coating resulted in the lowest emissions and, consequently, the lowest threat to indoor air quality among the tested materials, which included glass, fiberboard, gypsum board, and lime mortar in addition to pine heartwood [22].

Summarizing the recently published results on LO use for wood surface protection, the main benefits of the studied treatments and formulations, apart from those associated with the ecological aspects, are related to improving surface hydrophobicity. In general, no significant groundbreaking information in this area is reported. However, the high surface hydrophobicity endowed by the technique using a low-load (1%) surfactant-free LO emulsion [15] demonstrates an innovative approach for economical and efficient wood surface protection.

3. Impregnation

Impregnation is a well-known method for enhancing wood biodurability and hydrophobicity-related properties. The use of LO as an impregnant is considered an environmentally beneficial alternative to conventional agents. An important element that determines the success of impregnation treatment is the uptake and distribution of the impregnant. LO uptake was studied at the macroscopic and microscopic levels to evaluate the introduction of LO into the wood substrate by impregnation. Near-infrared spectroscopy application for developing a model for the quantification of LO uptake has been explored [37,38]. In studies about the impregnation of Norway spruce and Scots pine with LO, the uptake differed for different wood parts with substantially more LO retention in sapwood than heartwood [37,39,40]. More detailed analyses were performed for Norway spruce, for which the influence of tissue types, the wood moisture content, and density was additionally evaluated [39,40]. Earlywood generally retained more LO than latewood. A positive correlation was observed between wood moisture content and LO uptake, suggesting that water in wood porous structures can promote LO uptake compared to wood with a low (below 30%) moisture content. The authors' proposed explanation for this phenomenon is the formation of an oil-in-water emulsion under the conditions of the experiments (60–140 $^{\circ}$ C) which penetrates the wood better than pure oil. Penetration is also explored for blends of LO with alcohols and pyrolysis oil and dependence on the used alcohol is observed [41]. The uptake also differs depending on the wood substrate [34,42,43]. A higher fraction of voids filled with LO due to impregnation is observed for the acetylated wood compared to the unmodified control [42]. Using the technique of X-ray microdensitometry allowed the tracing of the flow of the LO during the impregnation and showed a heterogeneous distribution of LO with a gradient depending on the retention levels [44]. The wood's microstructural changes due to impregnation with LO have been studied by using SEM [44–50]. The filling of cell lumens, occlusion of pits, and damages in the form of microcracks were observed, especially at higher retention levels. The latter changes

are assigned to be responsible for the impaired mechanical properties of LO-impregnated wood [44,45].

The key idea behind using LO for impregnation is to suppress the tendency of wood to absorb water by transferring the inherent hydrophobicity of LO to the wood. However, the studies differ in the wood substrates, LO-based impregnants, and impregnation methods used (Table 2).

Different results have been published about changes in wood moisture sorption and dimensional stability due to LO impregnation. An improvement in hydrophobicity and dimensional stability was achieved by impregnation with LO for unmodified and also acetylated wood [42,46,47,50–52]. A decrease in liquid water uptake into wood due to impregnation with LO has also been reported for wood that was impregnated with a copper azole preservative before impregnation with LO [53,54]. A reduction in water uptake was achieved by the substitution of the white spirit with pyrolysis oil and propanol in a LO blend used for impregnation [41]. However, a decrease in the efficacy of water repellency was observed in a long-term immersion test where the water uptake was similar for the LO-impregnated and control specimens [53]. Only lower rates of moisture and water sorption and dimensional changes with no effect on the total absorbed amount of water at the equilibrium and even increased swelling were observed for Norway spruce impregnated with LO compared with the control specimens in sorption experiments [55]. Based on these results, the authors conclude that impregnation with LO is water-repellent rather than a dimension-stabilizing treatment. In addition, only a high retention level provides a reasonable hydrophobic effect, making the material rather expensive [56].

Authors	Reference	Impregnant	Wood Substrate	Impregnation Method
Ruwoldt and Toven	[41]	Blend of raw LO, alcohol, pyrolysis oil	Scots pine	Immersion (1 h) 75 °C
Fadl and Basta	[42]	Boiled LO	Okoume, spruce (acetylated)	Vacuum, curing (1–5 h) at 70–190 $^\circ\mathrm{C}$
Liu et al.	[46]	LO	Chinese ash (sapwood)	Vacuum (0.01 MPa) 1.5 h, atmospheric pressure 1.5 h
Liu et al.	[47]	LO	Ailanthus (sapwood)	Vacuum (0.01 MPa) 1.5 h, atmospheric pressure 1.5 h
Kaya	[49]	Natural LO (100% purity)	Mediterranean cypress, field maple	Hot-cold bath 1h at 130 °C and 1h at 30 °C (followed by heat treatment at 160–240 °C)
Pelit and Arısüt	[50]	LO and synthetic thinner (1:1)	Aspen, fir	Pre-vacuum (760 mm Hg) 1 h, atmospheric pressure 24 h
Epmeier et al.	[52]	Reactive LO derivative	Scots pine (sapwood, heartwood), European beech, silver birch	Vacuum 30 min, pressure 45 min, post-vacuum 60 min
Humar and Lesar	[53]	LO (100%)	Norway spruce, European beech	Not specified
Can and Sivrikaya	[54]	LO and ethanol (1:1)	Scots pine pre-impregnated with Cu azole	Vacuum (650 mmHg), 30 min; pressure (6 bars), 1 h; hot bath (80 °C), 4 h
Fredriksson et al.	[55]	Boiled LO	Norway spruce (mature sapwood and juvenile and mature heartwood)	Pressure (1 MPa) at 100 $^\circ\mathrm{C}$
Demirel et al.	[56]	LO	Scots pine (sapwood)	Empty cell process

Table 2. Wood substrates, impregnants, and impregnation methods used in studies.

The ability to keep the moisture content below the levels required for biological deterioration is the main consideration for using wood impregnation with LO in wood protection against biodegradation [37,57]. However, studies have reported inconsistent results about the efficiency of impregnation with LO in improving wood resistance against decay fungi. Such treatment has been found both to improve performance [34,48,53,58,59] and to be rather inefficient without fungicidal additives [60,61]. The impregnation of wood with the LO formulations containing the additives of nano-CuO, nano-ZnO, and boron (trimethyl borate) showed significant improvement in resistance to both brown and white rot fungi [60,61]. Comparing the effect of both metal oxides, CuO was found to be more efficient in inhibiting fungal growth, with the efficiency of both metal oxides depending on their concentration in the formulation [61]. However, the additive of boron, which showed positive effects in laboratory tests, did not show improved biodurability in a field graveyard test [60]. In a study investigating the efficiency of impregnation with LO to improve wood decay resistance depending on the wood part, only the Scots pine heartwood, which is more durable than sapwood per se, benefited from such treatment, while no effect on mass loss was observed for the non-durable sapwood [62]. No improvement was also observed when impregnation with LO was used as the second step after impregnation with the copper azole biocide [54]. However, copper leaching was substantially reduced by postimpregnation with LO. Less leaching of boron was also observed for wood impregnated with boric acid and afterward immersed in LO [58]. In addition, such post-treatment provided significant improvement in resistance to decay in tests that included leaching before exposure to fungi.

Similarly, different results were observed in the resistance of wood impregnated with LO against termites. Fewer attacks by termites were observed in most studies both in laboratory and field tests [43,58–60,63,64]. In addition, a synergistic effect of LO and certain heartwood extracts has been demonstrated, indicating the potential of such an approach for environmentally friendly wood protection [64]. A synergetic effect was also achieved by wood impregnation with boric acid followed by immersion in LO, when leached specimens in termite tests performed much better than counterparts treated only by one of these processes [58]. However, no effect on mass loss due to field exposure to termites using the drum technique was observed for wood impregnation with LO without termiticidal additives [60].

In addition to improving hydrophobicity and biodurability, other aspects of wood impregnation with LO have also been investigated. Decreased discoloration due to artificial weathering that included cyclic UV irradiation and water spray was observed for Scots pine sapwood impregnated with LO, with better results achieved for specimens impregnated using the full cell than the empty cell process [65]. From the chemical changes analyzed by FTIR, the authors also conclude that the treatment with LO prevents the degradation of hemicelluloses but cannot prevent the degradation of lignin and cellulose. The impregnation of wood with LO is shown as a potential additional treatment of ammoniumcopper-quaternary-treated wood for reducing corrosivity, which is an important issue for the use of materials in places with high corrosion risk [66]. The immersion of wood in a hot LO bath was used as a pre-treatment before thermal modification [49]. Impregnation with LO improved mechanical properties and reduced water absorption compared to thermally modified wood without a LO pre-treatment. In the same study, the effect of impregnation with LO on wood acoustic properties was analyzed and an increase in sound absorption was caused by the introduction of LO into the wood structure, with the effect enhanced by thermal treatment.

Wood impregnation with LO has also been used as a water-repellent pre-treatment of wood followed by densification [50,67,68]. In general, pre-treatment with LO improved the properties related to water absorption while impairing strength properties and hardness. The opposite process with wood heating in an LO bath used as a post-treatment of thermally compressed wood showed that such treatment combinations complement each other, with LO treatment contributing to a reduction in water absorption and improvement in decay

resistance compared with the compressed wood without post-treatment [69]. On the other hand, strength and hardness were reduced due to LO post-treatment compared with the wood when it was only compressed. In addition, a similar decay resistance was observed for both the combined treatment and wood subjected only to the LO treatment step.

In a study investigating the biodegradability of LO among other wood preservatives, moderate biodegradability in the groundwater environment as well as relatively large abiotic degradation in a sterile environment was observed for LO, thus implying that LO as an impregnant does not pose a threat to the environment [70].

In general, although the recent publications on the use of LO impregnation in wood protection do not suggest fundamental innovations in this field, they provide more detailed knowledge about the processes involved in wood impregnation with LO, which may support optimizing the treatment.

4. Base for Biofinish

In a recently proposed wood protection approach, impregnation with LO is used to promote the formation of a functional biofinish on the wood surface. This approach is aimed at reverting the generally unwanted wood surface discoloration by wood staining fungi into a protective and decorative biofinish. The finish is a dark homogenous layer developed by microorganisms on the surface of the wood that is treated with oil-derived products, including LO. Since the biofinish is formed of living organisms, they represent Engineered Living Materials (ELM). The biofilms are purported to possess a prolonged service life with self-healing effects compared to existing coatings [71]. Wood staining fungi producing the color-giving pigment melanin of the genus *Aureobasidium* are found to dominate in several biofilms formed on wood that is treated with LO [71–75]. It is reasoned that this dominance is related to the conditions created by the oil treatment, which selectively supports the growth of *Aureobasidium* only [71]. However, the presence of more than a single dominant genus has also been observed and the species colonizing the wood are influenced by the location of the exposure site [72–75].

The synergy of LO and fungus in wood protection is explained by the prevention of liquid water by LO which, together with the fungi, protects the wood against wood-degrading microorganisms and degradation due to weathering [71,76]. However, various factors have been suggested for the explanation of the positive effect of the oil treatment [71,72]. One factor could be the availability of local liquid water on the hydrophobic surface that prevents the absorption of the water into the wood. Other potential factors include the favorable modification of surface properties for spore adhesion and the use of oil as a nutrition source by microorganisms developing the biofinish [77]. However, it was observed that on pine sapwood impregnated with stand LO, the biofilm was not formed during the test period (1.5 years) in contrast with biofilm formed on the counterparts impregnated with raw LO [72,73]. The suggested reason behind the observed differences is less energy and carbon provided by the stand compared to raw LO [78]. The wood substrate and the geographical location of the exposed wood were found to significantly influence the biofilm formation [72,73].

The results of wood coated with a commercial product containing *A. pullulans* as the main biofilm-forming component and exposed to weathering for one year showed good resistance to discoloration and a reduction in surface roughness, which is explained by two self-healing mechanisms: the filling of surface cracks by migrated LO, which subsequently polymerizes, covering the crack, and the local regrowth of damaged areas by living fungal cells present in the coating [79].

In addition, it was demonstrated that such a finishing can tolerate significant environmental fluctuations [75]. However, it is admitted that further research is needed to optimize the formulations and enhance their efficacy.

Although the use of LO for developing a biofinish on wood surfaces is an innovative and bio-based strategy for wood protection, stakeholders' awareness still needs to be estab-

lished by a comprehensive and convincing investigation of its suitability and serviceability for bringing this new concept to life.

5. Medium in Thermal Modification of Wood

One more explored use of LO is the thermal modification of wood in an oil as a heattransferring medium. LO is considered a good candidate for such a treatment due to its ease of drying after treatment and wide availability [80]. In general, similar improvements in wood properties as with other thermal modification processes are observed, although only in one study was the effect of LO analyzed by comparing it with another modification medium [81]. The role of the LO is considered to form a stable film on the outer and inner surfaces, forming a barrier to water absorption [82]. Thermal modification in LO endows wood with improved dimensional stability and resistance to biodegradation, although the improvement in the latter is species-dependent and only moderate in most studies [81–84]. However, a high biodurability was observed for all tested species when preservative additives were introduced into the LO [81]. When oil is used as the medium in the thermal modification of the wood, a considerable amount of the oil is uptaken by the wood and this is found to be wood-species-specific [81,83,84]. A decrease in water absorption was observed as the amount of oil absorbed during modification increased and the cooling time after heat treatment positively correlated with the amount of oil in the wood [84]. However, more oil uptake did not provide significant benefits for the wood's dimensional stability, indicating that a reduced cooling time would contribute to the reduction of the treatment cost by reducing the oil uptake. As with other thermal modification processes, the color of the wood is significantly altered by the thermal modification in LO [80,82,85]. In such a way, treated wood exhibited better color homogeneity between the two surfaces of the specimen compared with the control, but quite a big color variation was observed between the surface and core, with the color variation decreasing with an increasing modification temperature [82,85]. Better color stability was also observed for modified specimens in accelerated weathering tests that included UV exposure and water spraying cycles [85].

In the studies about the influence of oil aging due to heating to mimic scenarios of oil reuse, a decrease in oil uptake due to the increased viscosity and in water repellency of wood was observed for oil-heat-modified wood when oil preheated for a certain time was used [80,82]. In addition, a higher increase in viscosity was observed when LO was heated with wood specimens compared with oil heated without specimens in the oil [82].

Despite some improvement, the performed studies have not demonstrated significant benefits of the thermal modification of wood in LO as a heating medium compared with other thermal modification processes while raising concerns about the method's utility due to the rather high consumption of the LO.

6. Performance Optimization by LO Chemical Modification

A growing trend can be observed in studies on wood protection for substituting LO with modified LO. Several studies have investigated different aspects of wood protection by using epoxidized linseed oil (ELO) by itself or further modified ELO. The epoxidation converts the double bonds in triglyceride molecules into epoxy groups, improving the oil reactivity [86]. Although the main consumer of ELO is the plastic industry, which uses it as an environmentally friendly base polymer for its products worldwide, ELO has also drawn the attention of the wood protection sector due to more rapid polymerization and lower required retention levels compared to LO [24,87,88].

Similarly to LO, ELO provides hydrophobicity when impregnating the wood with the efficiency increasing with higher retention levels [56,57,89–91]. Better water exclusion efficiency and dimensional stability were observed for wood treated with ELO compared to LO [56,57,92]. A higher improvement in water repellency and dimensional stability was achieved when wood was impregnated with ELO emulsion containing 2–6% carnauba wax [91]. The enhanced dimensional stability was found to be independent of retention

and was considered to be due to the blocking of the sorption sites in wood via a reaction between wood hydroxyl groups and ELO epoxy groups [56,88,93]. However, in another study, FTIR analyses did not approve chemical reactions between ELO and wood components [91]. The discrepancies are attributed to the differences in formulation systems and oil loading. Discrepancies were also observed regarding the resistance to staining fungi. Both an improvement and impairment in resistance have been observed for the effect of ELO on resistance to surface colonization by staining fungi [57,90]. ELO did not offer an improvement regarding surface discoloration and degradation due to weathering [90]. However, surface coating with ELO, and especially ELO with the addition of a reactive UV absorber, provided, although not complete protection, a positive effect on wood protection against degradation in an artificial weathering test that included UV irradiation and water spraying steps [94]. Similarly, although ELO showed better results in decay tests, no significant improvement in wood durability was observed without biocidal additives [57,92].

Inconsistent results have been observed regarding impregnation with ELO on woods' mechanical properties. In a study by Terziev et al., increased mechanical properties were observed with a higher effect for a higher retention level [57]. On the contrary, a reduction in mechanical properties was observed by Jebrane et al., with the results depending on the method used for the introduction of the catalyst into the system [93].

Using ELO in a mixture with creosote, a high effectiveness against decay was achieved with only 30% creosote in the mixture, demonstrating the possibility of considerably reducing the use of this carcinogen [57].

ELO is used as a post-treatment for wood impregnated with a bio-oil to reduce the leachability of the latter from the wood [89,95]. The post-treatment improved the hydrophobicity and dimensional stability of wood. Ambiguous results were reported regarding the efficiency of such post-treatment on the biodurability of wood with both an increase and decrease in resistance to decay fungi and termites observed compared with specimens without additional treatment with ELO after impregnation with a bio-oil [89,95]. It should be noted that different bio-oils were used in the experiments.

Although epoxidation makes LO more reactive, the catalyst is needed to ensure the polymerization of ELO. In a study investigating the best ways of applying acetic acid as the catalyst, it was observed that a two-step impregnation process with the impregnation of the catalyst following the impregnation of ELO is a feasible and practical approach for ELO treatment of wood [88].

The possibility of developing linseed-oil-based wood coating formulations with a substantially reduced polymerization time while providing proper properties of the liquid formulation and the cured film has been demonstrated by studying different blends of ELO, fatty acid methyl esters, and cationic photoinitiators [24]. However, the potential drawbacks due to fast curing, such as insufficient penetration and a soft topcoat, were noted in the study, indicating the need to control the curing process.

Another approach tested to optimize ELO curing and coating properties is the further modification of ELO through the ring opening of the epoxy group using acrylic acid as the ring-opening agent. The effect of different hydrophobic additives and UV light intensity on the curing and film properties of the acrylated ELO was tested and it was found that none of the tested additives was the best for all properties [96]. Using different photoinitiators at various concentrations, the best coating properties, such as adhesion and scratch and solvent resistance, were observed for formulations that did not cure very fast and where the final double-bond conversion was lower than one [1,97]. In addition, a post-reaction effect after UV light exposure was observed, which allows a very short irradiation time for coatings of such type, reducing energy costs. However, the studies do not include reference coatings of LO or ELO to gain confidence in the superiority of such an approach.

Copolymerization is one more direction in the research on the use of LO in wood protection. Copolymers with methacrylates can be used for highly hydrophobic wood coating, providing wood with substantially enhanced water repellency, slowing down water

absorption [98]. ELO is also used in combination with vinyl acetate as the other component of the copolymer [86,87,99]. The copolymerization inside the wood was monitored by FTIR analyses of specimens impregnated with an emulsion containing ELO and vinyl acetate [87]. It was observed that the copolymerization depends on the degree of LO epoxidation, which corresponds to the ratio of the converted double bonds, with a higher degree providing a higher yield of the copolymer [86]. The penetration of the copolymer into wood cell walls and even middle lamella regions was observed by SEM [99]. The authors suggest that the relatively low leaching observed for the treated wood could be interpreted as covalent bonding between the copolymer and the wood components [87]. Such wood treatment rendered wood reduced hygroscopicity and substantially increased dimensional stability that positively correlated with the curing temperature and duration [87,99]. Wood impregnated with the copolymer ensured an increased decay resistance in durability tests (EN113) with the durability class depending on the weight percentage gain [86,99]. However, deteriorated mechanical performance was observed for such a treatment, which is comparable with similar treatments of LO and ELO [99]. In addition, it should be noted that the optimum curing conditions were found to be at 90 $^{\circ}$ C for a week (168 h) [87]. This could be a serious obstacle to the widespread use of such a treatment, considering its economic viability.

Another studied LO modification for wood protection is silylation with an organosilicon compound containing a vinyl group [100]. The wood impregnated with such modified LO showed high hydrophobicity and improved resistance to water, ensuring improved resistance to mold and decay fungi with the results depending on the process characteristics used for the LO modification and the product properties. However, although the study includes the results of untreated wood, the results of the reference treatment with unmodified LO are not provided, which prevents evaluation of the efficiency of the modification.

Several studies have investigated coating formulations that include LO and polyurethane (PU). A waterborne formulation of LO-based PU dispersion was developed as a potential coating for furniture finishing [2,101]. Testing of different curing methods showed that the curing process including UV exposure followed by an air-drying phase provides the best film properties [101]. Although the coating showed good film properties and good adhesion to wood and durability without crack formation in the hot-and-cold test over 10 cycles, no other important properties allowing evaluation of the coating performance on wood have been provided.

Waterborne PU dispersions synthesized from LO and different types of diisocyanates were supplemented with different metal-containing antimicrobial agents to develop an antimicrobial coating [102]. The films, regardless of the diisocyanate type and additive, showed very good adhesion on wood, while the impact and bending resistances were dependent on the diisocyanate type. Similarly, the antibacterial and antifungal activities depended on the coating base formulation and the antimicrobial agent used.

An acrylated LO prepolymer together with a reactive diluent and PU resin in various ratios was used in coating formulations to prepare UV-curable films that demonstrated good adhesion to wood [103]. Different film properties have been studied; however, the protective capacity of these formulations for wood protection has not been tested and evaluated.

The studies investigating the use of chemically modified LO in wood protection are rather sporadic and the published results are inconsistent. Although chemically modified LO has been found to outperform LO in some tests, comprehensive investigation, including sustainability aspects, is still lacking to evaluate the potential of such an approach in wood protection.

7. Conclusions

LO is considered an environmentally friendly wood protection material of increasing value and significance as part of the drive toward sustainability. LO's inherent drying capacity and hydrophobic nature are mainly exploited to enhance wood performance in

the design of various LO-based formulations and application methods for use in surface and impregnation treatments. In recent research, the effect of LO treatment on properties which are mostly, but not only, related to different aspects of wood and water interaction were evaluated and the mechanisms involved in the changes of wood were analyzed. The potential of LO use for enhancing properties was demonstrated for different wood substrates, including differently modified wood such as acetylated wood and charred and densified surfaces. In addition, the use of LO as a medium in the thermal modification of wood was evaluated; however, the results do not provide convincing arguments in favor of this approach when the advantages versus oil consumption are considered. A tendency of an increasing number of studies investigating the use of chemically modified LO in wood protection can be observed with the dominance of ELO and its derivates. In general, although almost all studies show greater or lesser potency to provide benefits in wood protection, the innovation and practical feasibility levels of the proposed approaches differ. The studies on the biofinish formation on wood impregnated with LO are among those proposing a high level of innovation that involves LO use in wood protection. However, only further studies will show the viability of this approach.

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