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REVIEW PAPER

Biomass waste as a photoprotective agent in the formulation of sunscreen preparation

Elzandra Imola Vioniken[a], and Devi Ratnasari[a]*

[a] Study Program of Pharmacy, Faculty of Health Sciences, University of Singaperbangsa Karawang, Margasari, Karawang Timur, Karawang, Indonesia

*E-mail: devi.ratnasari@fkes.ac.id

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Abstract: UVA and UVB radiation from sunlight may lead to erythema and skin cancer. Those risks can be reduced by using photoprotective agents. Sunscreen is a photoprotective agent that successfully protects the skin from ultraviolet radiation. However, the active ingredients containing synthetic chemicals can hurt the skin. This review provides knowledge regarding using lignin compounds obtained from biomass waste as photoprotective agents that have been investigated for performance. The method of study applied is a literature review of original research conducted within the last ten years. Data were searched using the keywords biomass waste, photoprotective, and sunscreen on the Pubmed and Google Scholar websites. The results showed that lignin can be found in varying levels of biomass waste, such as banana stems, coconut shells, corn stalks, rice straws, and others. Delignification of lignin from biomass can be accomplished using basic solvents, acidic solvents, organosolvents, and Deep Eutectic Solvents (DES). The delignification procedure using DES is considered more environmentally friendly and less toxic. The higher the lignin concentration in the cream, the greater the SPF value. This is because lignin contains phenolic, ketone, and other chromophore functional groups capable of absorbing UV radiation. Furthermore, converting lignin molecules into nanoparticles and modifying their chemical structure may enhance the SPF value of lignin-containing creams. The addition of phenolic hydroxyl auxochrome groups and catechol units to the lignin molecule was found to increase the SPF value of lignin. It may be concluded that lignin can be utilized as a photoprotective agent, and biomass waste can be transformed into high-value cosmetic products.

Keywords: Lignin, Photoprotective, Biomass Waste, Sunscreen, SPF

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INTRODUCTION

Indonesia is a tropical country with an Ultraviolet (UV) index ranging from high to extreme [1]. This causes a variety of skin disorders in tropical climates [2]. UV radiation is classified into three types based on its wavelength: UVA (400-315 nm), UVB (315-280 nm), and UVC (280-100nm). UVC radiation with the shortest wavelengths can be protected by the ozone layer, whereas UVB and UVA can penetrate and reach the earth's surface [3], [4]. Ultraviolet A (UVA) and Ultraviolet B (UVB) radiation, which may penetrate the ozone layer, can cause skin damage. Sunburn, a skin irritation marked by erythema or redness, can be caused by UVB exposure. UVA radiation, on the other hand, can have a more serious effect since it can penetrate deeper into the skin layer, causing DNA mutation and potentially leading to photoaging, photocarcinogenesis, and skin cancer [4], [5]. The risk of skin damage from sun exposure can be reduced by using photoprotective agents. Physical protection, such as photoprotective clothing and UV-absorbing eyewear or sunscreen, may be included among those efforts. Photoprotective clothing is designed to provide UV protection, as indicated by the Ultraviolet Protection Factor (UPF) value. However, its capacity to protect the skin from UV radiation remains limited and is determined by several factors, including fabric weight, thickness, colour choice, and composition. On the other hand, sunglasses can absorb UV radiation up to 400 nm but do not provide widespread or effective protection [6].

Sunscreen cream is a highly effective photoprotective agent that protects the skin from UV exposure, beating photoprotective clothing and UV-absorbing glasses [4][6]. Sunscreen creams are classified into two types based on how they protect against UV exposure: physical and chemical sunscreens. Physical sunscreens prevent the entry of UV radiation by reflecting them. Chemical sunscreens, on the other hand, protect the skin by absorbing and converting UV radiation into heat energy before they penetrate the skin. These two types also have different chemical ingredients. Physical sunscreens include key ingredients such as zinc oxide (ZnO) and titanium dioxide (TiO_2), whereas chemical sunscreens use avobenzone and octinoxate. The Sun Protection Factor (SPF) value determines sunscreens ability to block damaging UV radiation [4][7][8]. Sunscreen is an essential cosmetic product for everyday usage to protect against sun exposure. However, the active components in most sunscreens on the market are synthetic compounds. Long-term use of synthetic chemicals found in sunscreen cream preparations may negatively impact skin health [8].

The use of natural components such as sunscreens is becoming more popular as a way to reduce the negative effects of synthetic chemicals. Lignin is a compound that has been shown to be effective as an active component in sunscreen. This is based on the structure of lignin, which contains phenolic functional groups, ketones, and other chromophore groups that absorb UV radiation. Lignocellulose concentration in biomass waste is around 80%. Biomass waste, such as straw, stalks, stalks, and husks, is the richest source of lignin and can be used as a substitute for photoprotective agents in natural sunscreen cream formulations [9].

This article provides updated information on several lignin-containing biomasses used in sunscreen formulations and tested for SPF values. In addition to that, the review also examines several extraction methods and lignin compound modifications to increase lignin's photoprotective properties. As an outcome, this paper can serve as a reference for the future development of lignin as a photoprotective with maximum protection. This is useful for identifying the amount of lignin in various biomass wastes and the optimal extraction methods and solvents for the lignin separation process, which will be used in natural sunscreen cream formulations. It may also be a solution for more efficient and regenerative use of biomass waste. This study presents recommendations for developing lignin-active sunscreens that are low in toxicity, environmentally friendly, and meet cosmetic preparation safety standards.

MATERIALS AND METHODS

This review article is written using a literature study method sourced from original research from the past 10 years. Data collection was carried out through Pubmed and Google Scholar websites by entering keywords such as lignin, biomass waste, photoprotective, and sunscreen. Furthermore, screening was carried out on the abstract and continued screening of the article content. Data retrieved included biomass sources, methods of delignification or extraction of lignin from biomass, lignin modification, and methods of analyzing the photoprotective efficacy of lignin. Relevant data were collected and then interpreted in tabular form and re-described with a comprehensive narrative.

RESULTS AND DISCUSSION

Biomass Waste

The photosynthetic process produces organic materials as products or waste [10][11]. Agriculture, plantation, and forestry are all possible sources of biomass. Biomass waste from agriculture and plantations, such as rice straw, corn stalks, coconut shells, and oil palm empty fruit bunches, is abundant in Indonesia, but its utilization has not been optimal. According to a study conducted by Hidayati and Ekayuliana (2022), rice fields have the highest area and volume of production from agriculture and plantations in Indonesia, covering 15,994,512 ha, followed by oil palm and corn. Furthermore, Hidayati and Ekayuliana (2022) explained in their study that as many as 164,617,857 tons of agricultural products originating from Indonesia's six largest agricultural products produced biomass waste, which included rice husks and straw, corn stalks, empty oil palm bunches, coconut shells, rubber wood, and bagasse [12].

The effective management of biomass waste is required to ensure the sustainability of nature and human requirements [13]. Some are treated using the soxhlet method before being incorporated into creams [14]. Gutiérrez-Hernández *et al,* (2016) used Lignin from *Agave tequilana* pulp, and mixed it with ZnO in sunscreen formulations. Lignin from *Pinus sylvestris* and *Eucalyptus* wood, extracted from the paper industry as black liquor, was converted into nanoparticles to increase its photoprotective properties [15]. This innovation provides for more efficient utilization of biomass waste because, in addition to being an alternative to reducing biomass waste, it also has the potential to create cosmetic ingredients with fewer side effects on the skin [8].

Abundance of Lignin in Biomass Waste

Plants are primarily composed of lignocellulosic components such as cellulose, hemicellulose, and lignin. Lignocellulose is commonly found in plant cell walls [16]. Plant cellulose fibres are wrapped by hemicellulose and bonded by lignin, resulting in a stiff structure [17][18]. Biomass waste with a high lignocellulose content includes banana stems, cocoa bean shells, coconut shells, oil palm empty fruit bunches, bagasse, corn stover, and rice straw [19][20]. Table 1 shows the percentages of lignocellulose components in various biomass wastes. In lignocellulosic components, hemicellulose and lignin are connected by covalent bonds, so the structure formed becomes strong to protect cellulose fibres from damage. The process of separating lignin from lignocellulose, called delignification, requires various treatments due to the complex structure of lignocellulose [19].

Table 1. Lignocellulose Components in Biomass Waste

Biomass Waste	Hemicellulose (%)	Cellulose (%)	Lignin (%)	Reference
Banana Stems	8.8 ± 0.9	69.4 ± 0.7	15.3 ± 0.1	[21]
Corn Stalks	34.5	35.1	14.1	[22]
Coconut Shells	11	81	8	[23]
Rice Straw	21.99	37.71	16.62	[24]
Wheat Straw	24.98	36.77	23.59	[25]
Sugarcane Bagasse	26.9 ± 0.5	45.1 ± 1.5	22.2 ± 0.5	[26]
Empty Oil Palm Bunches	29.6	50.9	17.84	[27]
Empty Oil Palm Bunches	53.18	37.34	9.48	[28]
Oil Palm Leaves	21.5 ± 0.6	44.8 ± 0.2	19.8 ± 0.002	[29]

Lignin is commonly extracted with strong acids, bases, and organic solvents at high temperatures and pressures or with a specific enzyme [30]. Pulping is a common technique for extracting lignin from lignocellulosic components so that it can be utilized. Commonly used pulping methods include soda, kraft, and sulfite. Soda pulping is the process of pulping with NaOH as a solvent. [31]. Kraft pulping also uses NaOH as a solvent however it is supplemented with sodium sulfide (Na_2S) [32]. Soda pulping is considered more environmentally friendly because it does not use Na_2S, making it a better option for reducing sulfide emissions [31]. Alkaline lignin, including soda lignin and kraft lignin, is insoluble in water, whereas lignin sulfite can dissolve in water with hydrophilic groups, specifically sulfonate groups [33].

Antunes *et al*. (2023) removed lignin from sugarcane bagasse using 2% NaOH at 90°C for 30 minutes. Antunes *et al*. (2023) reported a lignin purity of 91.9 ± 1.2% after extraction. Perera *et al*. (2023) used 4% NaOH to extract lignin from oil palm empty bunches at 80 ^oC for 6 hours, yielding 11% lignin [35]. Other investigations used organosolvents as solvents for biomass waste delignification [34]. Lee *et al*. (2020) extracted lignin from rice straw biomass with 10% dioxane solution at room temperature for 24 hours. Lee *et al*. (2020) found that only 5% of the entire weight of the biomass obtained contained lignin [36]. Pasma *et al*. (2019) extracted lignin from empty oil palm bunches using 80% ethanol-water and 0.2% sulfuric acid as a catalyst, yielding 15.1 ± 1.30% [37].

Other research used ethanol/water organosolve to remove lignin from hazelnut and walnut shells. Gordobil *et al*. (2020) reported lignin yields of 19.2 ± 0.4 g and 15.3 ± 0.1 g per 100 g of extracted biomass, with lignin purity ranging between 85-95% [38]. On the other hand, Da Mata *et al*. (2022) used 90% acetic acid and 2% $MgCl_2$ as catalysts, together with HCl. Lignin production and levels of phenolic and acetyl compounds increased as the degree of acetylation increased. Adding HCL increased the yield from 21.4% to 48.8%, with acetylation degrees ranging from 0.81 to 1.58 mmol/gram [39]. Bouza *et al*. (2016) removed lignin from empty oil palm fruit bunches using acid solvents and enzymatic hydrolysis, yielding 20.4% [40]. Another study used a sequential acid/alkali treatment approach, with 1% (w/w) H_2SO_4 at 121 ^oC for 60 minutes and 2.5% NaOH at 121 ^oC for 80 minutes, yielding 28.89% lignin [41]. On the other hand, De *et al*.. (2019) tried to remove lignin from rice straw biomass using two different solvents: NaOH and organic acid. The results showed 10.75% and 14.12% yield for lignin extracted using NaOH and organic acid, respectively [42]. However, using those solvents carries several concerns, including lignin fragmentation, low lignin purity, and environmental toxicity.

Mao *et al*. (2023) investigated the efficiency of lignin extraction from cocoa bean shell waste using a variety of Deep Eutectic Solvents (DES). The most effective solvent was DES with the composition PTSA-ChCl-Gly (2:1:1), which yielded 95.5% lignin at 130 °C for 30 minutes [30]. Liu *et al*. (2019) used DES to extract lignin from wheat straw. DES was made by combining Triethylbenzyl Ammonium Chloride and Lactic Acid in various molar ratios. TEBAC: LA (1:9) was shown to be the most effective DES, removing 73.96 ± 1.26% of lignin. Liu *et al*. (2019) reported that lignin purity was 75.69 ± 1.32% after 10 hours at optimum conditions at 373 K [43]. Li *et al*. (2024) found that extracting lignin from rice straw biomass with Lactic Acid: Choline Chloride (2:1) at 60 °C for 2 hours resulted in an 81.6 ± 1.0% yield and an 88.5 ± 1.1% purity [6]. Ma *et al.* (2022) extracted rice straw biomass with Choline Chloride: Lactic Acid (1:2) at 150 °C for 6 hours, yielding 81.54% lignin and 91.33% purity [25]. Kumar *et al.* (2016) extracted lignin from rice straw using Lactic Acid: Choline Chloride (5:1) and obtained a total lignin of 60 ± 5% after 12 hours of pretreatment at 60 °C [45].

Other research concludes that DES presents numerous advantages for lignin extraction, particularly in the formulation of cosmetic products. DES may dissolve lignin effectively at relatively low temperatures. Depending on the DES composition, Lignin extraction time ranges from 30 minutes to 6 hours at temperatures ranging from 80 to 150 °C [46]. A recent study has discovered that using DES can be an option due to its low volatility, non-flammability, low toxicity, and high biodegradability, making it environmentally friendly. DES is a solvent system made up of Hydrogen Bond Acceptor (HBA) and Hydrogen Bond Donor (HBD), resulting in a new eutectic with a lower melting point [25][30][43]. Lignin extracted using DES provides a high yield with high purity and low molecular weight [47]. Lignin extracted with DES has a lower condensation rate than alkaline lignin [44]. Table 2 shows the advantages and disadvantages of DES and other solvents used for delignification.

Table 2. Advantages and Disadvantages of Solvents Used for Delignification

Solvents	Advantages	Disadvantages	References
Organosolve	• High purity • Provide high-quality lignin • Low contamination	• Low yields • Neutral pH is less effective for delignification • Requirement of catalyst • Costly	[20][37][48][49]
Acid	• Commonly used as a catalyst	• Not generally used for the main solvent • May damage the lignin structure	[20][37]
Alkali	• Commonly used • Proven to separate lignin from lignocellulose effectively • The process is easy	• Not environmentally friendly • Lack of commercialization possibilities	[20][50][51]
Deep Eutectic Solvent (DES)	• High yield • Low volatility • Non-flammable • Environmentally friendly • Low cost • Biodegradable • Low toxicity • Easy to recycle	• Low purity • Difficult to separate lignin from solvent	[20][47][52][53]

Mechanism of Lignin as Photoprotective Agent

Lignin is a complex biopolymer present commonly in plants and is safe for human health and the environment [54][55]. Recent studies have demonstrated that lignin has UV protection activity due to its functional groups, which include phenolics, ketones, and other chromophores that can absorb UV light. Lignin is formed up of phenolic units, including p-coumaryl alcohol (H unit), coniferyl alcohol (G unit), and sinapyl alcohol (S unit), connected by ether bonds (β-O-4) [56][57][58]. Lignin absorbs UV rays and converts it into heat [59]. The phenolic units contain chemical bonds conjugated to one another in the aromatic nucleus, and when exposed to sunlight, the chromophore groups in them resonate via electron transfer [60][61]. The structure is similar to that of active ingredients in organic sunscreen, such as avobenzone, oxybenzone, octinoxate, and homosalate [60]. However, these compounds have been shown to damage the aquatic environment and be potentially dangerous to aquatic organisms [62][63][64].

Therefore, lignin can be utilized as a natural broad-spectrum sunscreen, blocking the harmful effects of UVA and UVB. In addition, lignin has antioxidant properties due to the presence of phenolic groups that can inhibit free radicals, making it a good photoprotective ingredient for sunscreen [8]. Arruda *et al*. (2021) investigated the photoprotective effects of lignin from Cartaeva Tapia. They incorporated lignin into cream formulations at various numbers, including 1, 2, 5, and 10%, and then evaluated its photoprotective effectiveness with a UV spectrophotometer. The results showed that increasing the lignin concentration in the cream increased its SPF value. This study demonstrates the fascinating potential of lignin as a natural photoprotective ingredient in sunscreen compositions [14]. Other studies have found that ZnO physical filters combined with lignin provide better UV protection. Lignin is converted into nanoparticles to improve its UV light absorption. When measured using a UV-Vis spectrophotometer, lignin nanoparticles absorb more UV light than bulk lignin, which absorbs only a quarter of the UV intensity [55].

Widsten *et al*. (2020) enhanced the photoprotective effects of lignin by converting it into CATlignin, which contains phenolic hydroxyl auxochrome groups and catechol units [15]. Chromophore groups on lignin, such as quinones and aromatic rings, can be conjugated with double bonds and carbonyl groups on the side chains. Their presence is responsible for lignin's ability to absorb UVA and UVB radiation. The aromatic rings of lignin also include auxochromes such as phenolic hydroxyl and methoxyl groups, which are conjugated to their free electron pairs. Because of the chromophore and auxochrome bonding, chromophore absorption is transmitted to the UVA-UVB region [15].

Consequently, the presence of phenolic hydroxyl and methoxyl groups plays an important role in the ability of lignin to be a sunscreen agent. The auxochrome enhancement was achieved by removing aliphatic hydroxyl groups. Additionally, dimethylation and cutting of β-O-4 or alkyl-aryl ether linkages in lignin were performed to increase phenolic hydroxyl groups. Charge transfer complexes between o-quinone and phenolic groups can also enhance kraft lignin's UV-Vis absorption [15]. Widsten *et al*. (2020) also studied the effect of phenolic hydroxyl auxochrome compared to phenolic methoxyl on sunscreen performance. The results showed that hydroxyl phenolics serve a greater role than methoxyl phenolics [15].

The particle size of lignin also determines its capability as a photoprotective agent. Some researchers use this parameter to modify lignin in order to enhance its photoprotective ability. Widsten *et al*. (2020) discovered that modifying the particle size of lignin improved its potential as a sunscreen. Small lignin nanoparticles (43-95 nm) demonstrated the greatest ability, followed by lignin nanoparticles (280-450 nm), and finally, unmodified lignin, which showed a poor ability in terms of UVB Sun Protection Factor (SPF) and UV transmission parameters [15].

Piccinino *et al*. (2022) attempted to use lignin nanoparticles (LNPs) as a carrier to boost the stability of Avobenzone and Octinoxate, both Sunscreen Filters (SSFs). The method used was to encapsulate both SSFs with LNPs. The results showed increased UV-Vis absorption in the presence of bathochromic and hyperchromic transformations caused by the interaction of lignin auxochrome groups with SSF chromophore groups. The enhanced photostability of SSF agents encapsulated with LNPs is caused by their capacity to reduce the degradation process of SSFs. This transformation is done to prevent the formation of free radicals, which cause SSF toxicity after prolonged irradiation [65].

On the other hand, Gagosian *et al*. (2022) increased kraft lignin's molecular weight (MW) by an enzymatic method. The researchers evaluated the photoprotective properties of kraft lignin and its two derivatives, R1 (intermediate MW) and E60 (highest MW). The results indicated a decrease in photoprotective activity. This result was obtained from SPF measures on kraft lignin, R1, and E60 samples, with SPF values of 33.8, 22.7, and 2.1 as chemical filters, respectively. The lignin derivative with the highest MW, E60, had a very low SPF value as a chemical filter, but when evaluated for physical filter performance, it had an SPF of 22.4. The results show that the larger the lignin particles, the lower their ability to protect against UV rays. According to previous research, converting kraft lignin into lignin nanoparticles (LNPs) can improve its photoprotective properties [54].

Efficacy Testing of Lignin as Photoprotective Agent in Sunscreen Formulations

Sun Protection Factor (SPF) values determine sunscreen's effectiveness in blocking UV radiation [4][6]. To protect the skin from UVA and UVB, sunscreen's SPF should be at least 15. However, the Canadian Dermatology Association

(CDA) recommends applying sunscreens with an SPF of 30 or greater [6]. The Food and Drug Administration (FDA) divides sunscreen protection into five categories: minimal (SPF 2-4), medium (SPF 4-6), extra (SPF 4-6), maximum (SPF 8-15), and ultra (SPF ≥15) [1][66][67]. This literature review emphasizes numerous studies that have used lignin as a photoprotective agent and addresses the SPF value. Table 3 shows that each biomass's SPF values are different and altered by different treatments.

Table 3. SPF Values of Various Biomass Wastes and Solvents Used in the Delignification Process

Biomass Waste	Solvent	SPF Value	Reference
Rice Straw	Dioxane	2.2 ± 0.1	[36]
Rice Straw[c]	Dioxane	4.3 ± 0.4	[36]
Wheat Straw	Ethanol-Water	7.33 ± 1.37	[68]
Wheat Straw	Ethanol-Water	41.97 ± 7.38	[68]
Hazelnut Shell	Ethanol-Water	6.5	[38]
Walnut Shell	Ethanol-Water	4.5	[38]
Sugarcane Bagasse	NaOH	9.51 ± 2.92 (in vitro) 9.6 ± 0.8 (in vivo)	[34]
Sugarcane Bagasse	NaOH	8.65 ± 0.21	[56]
Eucalyptus Wood[a]	-	33.8 ± 0.02	[54]
Eucalyptus Wood[a]	-	6.56	[69]
Eucalyptus Wood	Ethanol-Water	14.6 ± 0.5	[70]
Eucalyptus Wood[a]	-	19.7 ± 0.1	[70]
Spruce	Ethanol-Water	18.5 ± 1.2	[70]
Spruce[a]	-	12.2 ± 0.5	[70]
Cartaeva tapia Leaves	NaOH	5.1 ± 0.14	[14]
Agave tequilana Bagasse	NaOH	5.64	[55]
Agave tequilana Bagasse	Ethanol-Acetic Acid	8.49	[55]
Bamboo	Formic Acid	4.56	[71]
Bamboo	Formic Acid-Hydrogen Peroxide	3.83	[71]
Bamboo	Acetic Acid-Hydrogen Peroxide-HCl	3.96	[71]
Pinus sylvestris Wood[a]	-	6.7	[15]
Pinus sylvestris Wood[a][b]	-	13	[15]
Pinus sylvestris Wood[a][c]	-	12.7	[15]
Pinus sylvestris Wood[a][d]	-	17.8	[15]
Pinus sylvestris Wood[a][e]	-	16.4	[15]
Pinus sylvestris Wood[a][f]	-	21.3	[15]
Eucalyptus Wood[a]	-	8.7	[15]
Eucalyptus Wood[a][b]	-	13.8	[15]
Eucalyptus Wood[a][c]	-	16.6	[15]
Eucalyptus Wood[a][d]	-	15	[15]
Eucalyptus Wood[a][e]	-	19	[15]
Eucalyptus Wood[a][f]		21.5	[15]

[a]:Kraft lignin from the paper industry. [b]: CATlignin. [c]: nanoparticles. [d]: small nanoparticles. [e]: CATlignin nanoparticles. [f]: CATlignin small nanoparticles.

According to research by Widsten *et al*. (2020), modifying lignin molecules into nanoparticles and CATlignin raises the SPF value, which is the protection strength against UV radiation. SPF measurement can performed both in vitro and in vivo. In vitro measurement involves measuring the UV transmission or absorbance of lignin-based sunscreen samples against UV light with a UV spectrophotometer [14], [34], [36], [54], [55]. In vivo testing can be done by looking at the Minimum Erythema Dose (MED), the lowest UV light dose to induce erythema on the skin after exposure for 20 ± 4 hours [34].

SPF assessment in vitro can be performed with a UV-VIS spectrophotometer by dissolving cream formulations containing lignin in ethanol

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solvent. The absorbance is evaluated at a wavelength of 290-320 nm, and the SPF value is calculated using equation (1). With CF=10 as the correction factor, Eλ is the erythemal effect spectrum, Sλ is the solar spectral irradiance, and Absλ is the UV radiation absorption [14].

$$\text{SPF in vitro} = \times \sum E\lambda \times S\lambda \times Abs\lambda \text{ .... (1)}$$

Arudda *et al*. (2021) discovered that the higher the amount of lignin in the cream, the greater the SPF value obtained. The study reported that the greatest SPF value reached 5.1 ± 0.14 at 10% lignin content [14]. Gagosian *et al*. (2022) used the same procedure to assess SPF on lignin-based sunscreen derived from *Eucalyptus* wood. The best SPF value was found in the sample that combined with LE, the lignin with the smallest molecular weight in their research [54]. The result correlates with research by Lee *et al*. (2020) findings that the smaller the lignin molecule, the greater the photoprotective property [36]

In an investigation by Lee *et al*. (2020), lignin and lignin nanoparticle creams were applied at a dose of 2 mg/cm^2 on a quartz plate and UV transmission was measured using a spectrophotometer at a wavelength of 290-400 nm. The results showed that the cream mixed with lignin nanoparticles had a higher SPF value than lignin that wasn't transformed into nanoparticles, with SPF values of 4.3 ± 0.4 and 2.2 ± 0.1, respectively [36]. Girard *et al*. (2024) also converted wheat straw lignin into nanoparticles and put it into pure cream. The cream with lignin nanoparticles had a higher SPF rating (41.97 ± 7.38) than macroparticle lignin (7.33 ± 1.37) [68].

Gordobil *et al*. (2020) also used quartz plates in SPF testing on lignin cream samples from 2 types of nut shells. It was found that the SPF value of the cream with lignin derived from hazelnut shells was higher than that of lignin derived from walnut shells with the same solvent, with SPF values of 6.5 and 4.5, respectively [38]. Gordobil *et al*. (2016) compared lignin extracted from two different biomasses, spruce and eucalyptus, using two different methods: organosolve and kraft. The results revealed that lignin extracted from two biomasses using different extraction methods resulted in varying SPF values. Organosolve spruce has a greater SPF than kraft spruce. However, organosolve eucalyptus has a lower SPF than kraft [70].

Gutiérrez-Hernández *et al*. (2016) evaluated the effects of solvent usage on lignin extraction from biomass. Commercial creams containing lignin from *Agave tequilana* delignified with two different solvents were determined using a 200-800 nm spectrophotometer. The results revealed that samples with lignin removed using organosolvents had greater SPF values than those extracted with NaOH, with SPF values of 8.49 and 5.64, respectively [55]. Li *et al*. (2019) also investigated the effect of solvents used in lignin extraction. Li *et al*., 2019 tried to extract lignin from bamboo using organic acid solvents containing various combinations of organic acids, including formic acid, formic acid-hydrogen peroxide, and formic acid-hydrogen peroxide-HCl. The extraction results provided lignin yields of 19.2%, 16.7%, and 21.3% for lignin extracted using formic acid, formic acid-hydrogen peroxide, and formic acid-hydrogen peroxide-HCl, respectively. The researchers also tested the SPF value of the cream blended with the three forms of lignin at 5%. Lignin extracted with formic acid, formic acid-hydrogen peroxide, and formic acid-hydrogen peroxide-HCl had SPF values of 4.56, 3.83, and 3.96, respectively [71]. Meanwhile, Widsten *et al*. (2020) demonstrated the influence of lignin functional group modification on its photoprotective properties. Various cream samples were analyzed using the same procedure at 290-400 nm. The results showed that modification of lignin into CATlignin and nanoparticles can increase its SPF value [15].

The source of lignin is industrial lignin taken from the pulp mill of the paper industry in the form of black liquor [15]. In the paper industry, the pulp results from fibre separation from fibrous raw materials. Pulp raw materials contain cellulose, hemicellulose, and lignin, and the pulping process separates cellulose from lignin, also known as delignification. The delignification process with the pulping method breaks down lignin into smaller molecules that dissolve in black liquor [72][73]. From the study by Widsten *et al*. (2020), the highest SPF value, which can reach >21, is owned by lignin modified into CATlignin and also nanoparticles, both from *Pinus sylvestris* wood and *Eucalyptus* wood [15]. This result is also still in line with research by Lee *et al*. (2020) and Gagosian *et al*. (2022) that the smaller the lignin molecule, the higher the SPF value.

Ratanasumarn and Chitprasert (2016) conducted in vitro tests by applying 10% lignin lotion at 1.30 mg/cm^2 to a polymethylmethacrylate plate. The UV transmittance was then measured at wavelengths 290–400 using an Ultraviolet Transmitance Analyzer. The research found that ideal lignin extraction conditions, including solvent concentration and temperature, yielded the best SPF result of 8.65 ± 0.21 [56]. Antunes *et al*. (2023) evaluated lignin-based BB cream derived from bagasse with two methods: in vitro and in vivo. An in vitro test was carried out by applying BB cream with lignin on a synthetic substrate. The UV absorbance was determined using UV spectroscopy at 290-400 nm [34]. Meanwhile, the in vivo test was carried out on 5 volunteers. The sample was applied to the skin at 2 mg/cm^2 ± 2.5%, followed by UV radiation exposure on the treated and untreated areas. The results were calculated using equation (2).

$$\text{SPF in vivo} = \frac{MEDp\ Protected\ Skin}{MEDu\ Unprotected\ Skin} \text{ .... (2)}$$

Where MEDp is the minimum dose of erythema on skin treated with BB cream and MEDu is the minimum dose of erythema on skin untreated with BB cream. MED is the lowest dose of UV light to induce erythema on the skin after exposure for 20 ± 4 hours. The SPF value of lignin-based BB cream from sugarcane bagasse from in vitro and in vivo testing showed similar results with SPF values of 9.51 ± 2.92 and 9.6 ± 0.8, respectively [34].

Lignin is modified not only to boost its SPF value but also to improve its colour appeal. Lignin's original dark brown colour will influence consumer impression if utilized as a cosmetic preparation. Zhang *et al*. (2019) attempted to eliminate the black colour of lignin through acetylation with acetic anhydride. The results revealed a brighter lignin colour than unmodified kraft lignin [69]. Lee et al., 2019 tried to minimize the dark colour of lignin by isolating it from *Miscanthus sacchariflorus* and *Pinus densiflora* biomass at room temperature and using neutral solvents. The results showed a lighter lignin colour compared to lignin extracted with organosolvent (technical lignin) [74]. It may occur because technical lignin has a lot of chromophore and auxochrome groups that contribute to its dark colour. [75][76].

When lignin is used as a photoprotectant in cosmetic preparations, it is important to assure its physical, chemical, and microbiological stability, in addition to its efficiency, which is evaluated by the SPF value. It could be a topic of future research to incorporate lignin into many forms of cosmetic preparations, particularly cream and lotion preparations, which have already been studied [34][56][77]. Researchers need to ensure that the formulations fulfil safety criteria, are physically, chemically, and microbiologically stable, and, in the future, can meet the demands of new market trends so that the commercialization of lignin-active sunscreen can be sustained. Cosmetic preparations will interact directly with the skin, so the safety of the components included inside must be assessed. Sunscreen is an essential cosmetic throughout one's life. Therefore, its long usage profile must be studied [77].

CONCLUSION

Biomass waste can be used as a photoprotective agent by applying lignin compounds to sunscreen preparations. The effectiveness of lignin in protecting the skin from the effects of UV exposure can be influenced by various factors such as the source of the biomass, the solvent used, the particle size of lignin, the modification of functional groups, and the concentration of lignin applied to the sunscreen preparation. This is shown by the different Sun Protection Factor (SPF) values. Lignin nanoparticles derived from wheat straw biomass had the best SPF value in this review. Furthermore, modifying lignin into CATlignin small nanoparticles showed excellent results regarding lignin's photoprotective properties.

Future research is expected to utilize biomass waste optimally in cosmetic products. Using DES as a delignification solvent may produce lignin with low toxicity and is environmentally friendly. In addition, sunscreen preparations based on lignin as a photoprotective agent can be formulated into various preparations such as solid sunscreen, spray, gel, or other preparations. Researchers are also expecting to assess the long-term stability in physical, chemical, and microbiological aspects of lignin-active sunscreen preparations, providing efforts to commercialize lignin-active sunscreen preparations with approved safety to continue.

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