APPLIED
MATERIALS
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Research Article

From waste to value: Lapachol from teak wood waste as a green catalyst for sustainable soda cooking of *Acacia* and *Eucalyptus*

Esty Octiana Sari[1,*], Syelvia Putri Utami[1], Evelyn[1], Akiko Nakagawa-Izumi[2], Hiroshi Ohi[2]

ABSTRACT: The development of a sustainable catalyst as an alternative to synthetic anthraquinone (AQ) is urgently needed for a more efficient pulping process. This study investigates the potency of lapachol, a natural naphthoquinone isolated from teak (*Tectona grandis*) wood waste, as a catalyst in soda cooking of three industrially important hardwoods: *Acacia crassicarpa*, *Eucalyptus pellita*, and *Eucalyptus globulus*. Approximately 97.7% purity of lapachol was isolated and applied at 0.09% (on oven-dry wood). For comparison, the commercial synthetic additive, 2-Methylanthraquinone (2-MAQ) was also used at the same dosage. Cooking experiments were conducted at 160°C under varying alkali dosages (23, 27, 31%) and times (4, 5, 6 h). The result revealed that the delignification performance was species-dependent: *A. crassicarpa* (S/V=0.74) was the hardest, while *E. globulus* (S/V=3.04) was the easiest to delignify. Notably, *E. pellita* (S/V=2.04) shows the greatest selectivity index. Lapachol shows the capability of enhancing delignification across the three wood species by decreasing the residual lignin by up to 5% in *A. crassicarpa*, 5% in *E. pellita*, and 2% in *E. globulus* compared with soda cooking (control). Although the delignification is slightly lower than 2-MAQ, lapachol maintains pulp yields comparable to or higher than 2-MAQ. The selectivity index analysis confirmed that lapachol improved the balance between lignin removal and carbohydrate preservation, with the benefits most pronounced in *E. globulus*. These findings underscore lapachol as a promising sustainable pulping catalyst, offering the potential for impactful industry transformation through sustainable innovation.

Keywords: *Hardwoods, Lapachol, 2-Methylanthraquinone, Soda cooking, Green catalyst*

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Received: September 24, 2025
Revised: October 12, 2025
Accepted: October 18, 2025

How to cite: Sari, E., O., Utami, S., P., Evelyn, Nakagawa-Izumi, A., Ohi, H., (2025). From waste to value: Lapachol from teak wood waste as a green catalyst for sustainable soda cooking of *Accacia* and *Eucalyptus*. *Journal of Applied Materials and Technology*, 7(1), 22–30. https://doi.org/10.31258/Jamt.7.1.22-30.

1. INTRODUCTION

Efficient delignification is essential in the pulp and paper industry for converting lignocellulosic biomass into high-quality pulp. Soda cooking is chemical pulping method that recognized for its simplicity and sulfur-free nature, which contributes to its lower environmental impact compares to kraft cooking [1,2]. Despite this advantage, soda cooking is constrained by incomplete delignification and low selectivity, resulting increased chemical consumption and reduced pulp quality. To address this issue, the introduction of catalytic additives has been explored [3-5].

The quinone-based additives, especially anthraquinone (AQ), are well-known catalysts used in pulping processes. AQ has been employed in more than 100 paper mills worldwide since the 1990s [6]. Its synthetic derivative, 2-methylanthraquinone (2-MAQ), is widely studied as a redox catalyst in alkaline pulping [7-10]. However, nowadays, concerns about the health and safety aspects of AQ and 2-MAQ have been raised. AQ is classified as a possible carcinogen [11], and the safety of 2-MAQ remains uncertain. Some research reports 2-MAQ might be carcinogenic [12,13], while the Ames tests report no evidence of mutagenicity [7,8]. In addition, some reports highlight that the risks come from contaminants or by-products rather than the compounds themselves [14]. These debates emphasize the urgent need for alternative, safer, and more sustainable catalysts for pulping. Natural, bio-based quinones could offer a promising alternative that maintains effectiveness while improving safety and sustainability.

Lapachol, a natural compound found in teak (*Tectona grandis*), is structurally analog to anthraquinones (see Figure 1). It shows redox activity in both chemical and biological contexts [15,16]. Although most research has focused on its potential for cancer treatment suggesting the safety, its use as a pulping additive has not yet been explored. Studying lapachol as a natural pulping additive could be an eco-friendly alternative to synthetic quinones and also add value to teak wood.

Teak wood is a valuable export commodity of many tropical countries including India, Indonesia, and Myanmar. Indonesia is the second-largest teak wood producer and exporter, with about 1.47 million hectares of teak plantations in 2015. By early 2024, the forest product exports of Indonesia reached around USD 3.5 billion, and teak wood was valued about USD 3,000 per cubic meter. Besides its value, a large part of the teak wood is lost during processing, approximately 28% during felling and bucking, and approximately 41.3% lost during the production of square logs [17]. The residues from teak are often burned or thrown away, which causes environmental problems [18,19]. The utilization of teakwood waste materials to extract lapachol could help reduce environmental problems, leading to more sustainable, circular use of resources.

Our earlier study was the first to demonstrate through experiments that lapachol can act as a catalytic additive in soda pulping of *A. crassicarpa* and *E. pellita*, but only under specific cooking conditions [20]. While these initial findings confirmed that lapachol is a practical natural redox catalyst, the research only looked at two species and tested a limited set of cooking parameters. To overcome these limitations, this new study expands the research by carefully examining how cooking time and active alkali levels influence the process, and by including another key hardwood species commonly used in industry, *E. globulus*. Also, we compare the catalytic effectiveness of natural lapachol directly with that of commercial 2-MAQ, using the same experimental conditions. This broader investigation offers a more complete picture of how lapachol functions as a catalyst in soda pulping and points to its potential as a sustainable, plant-based alternative to synthetic quinones for pulp production.

Despite the importance of catalytic additives in improving soda pulping, the search for safer and more sustainable natural alternatives to synthetic quinones remains limited. This study is novel in its investigation of lapachol, a bio-based quinone from teak wood, as a soda pulping catalyst; an application of lapachol that has not been previously explored. Building on initial limited studies, this research systematically examines the effects of cooking time and alkali concentration and includes an additional hardwood species, and compares lapachol to the well-studied synthetic additive 2-MAQ. Therefore, this study offers a comprehensive evaluation of a natural, potentially safer, and more sustainable pulping catalyst to address the critical gaps in the development of eco-friendly pulp production technologies.

2. EXPERIMENTAL

2.1. Materials. Wood chips of *Acacia crassicarpa* and *Eucalyptus pellita* were obtained from two pulp mills in Indonesia: PT Riau Andalan Pulp and Paper (RAPP, Riau) and PT Tanjungenim Lestari Pulp and Paper (TELPP, South Sumatra). *Eucalyptus globulus* was obtained from a Chilean plantation and provided by a pulp mill in Japan: Hokuetsu Corporation,Tokyo. The chips (3–4 cm length) were milled and sieved to obtain wood meals of 40–80 mesh. Teak (*Tectona grandis*) sawdust was supplied by Toko Vastu Gunung Kidul (Yogyakarta, Indonesia) and similarly sieved to 40–80 mesh. Commercial 2-methylanthraquinone (2-MAQ, CAS: 84-54-8, 97% purity) was purchased from Alfa Aesar. All solvents were of special grade and obtained from Wako Pure Chemical Industries, Japan.

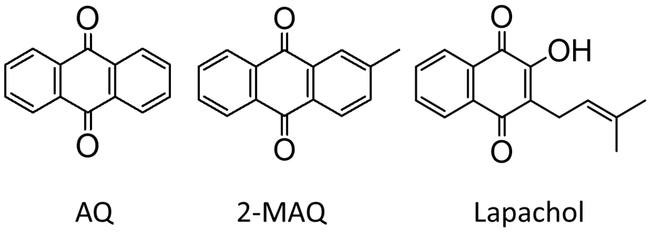

Figure 1. The chemical structure of anthraquinone (AQ), 2-methylanthraquinone (2-MAQ), and) and lapachol.

2.2. Isolation of lapachol from teak wood waste. Lapachol was extracted from teak sawdust using an acid-base extraction method. Teak sawdust (150 g) was mixed with 1 L of 1% aqueous Na_2CO_3 solution and allowed to stand for 24 h with occasional stirring. The mixture was filtered, and the filtrate was acidified by the slow addition of 6 M HCl until the solution turned from red to yellow, producing a precipitate.

The precipitate was collected by filtration, washed three times with distilled water, and dried at room temperature. Further purification was performed by silica gel column chromatography using n-hexane/ethyl acetate (7:3, v/v) as the eluent. After solvent evaporation, a yellow solid was obtained. The isolated compound was characterized using GC–MS and NMR, confirming its identity as lapachol with ~97.7% purity [20].

2.3. Soda pulping experiments. Soda pulping was carried out with three wood meals (*A. crassicarpa*, *E. pellita*, and *E. globulus*). The catalytic performance of natural lapachol (97.7% purity) isolated from teak wood waste was compared with that of commercial 2-MAQ under identical conditions. A control experiment without additives (soda cooking) was also conducted. The additive dosage was fixed at 0.09% (based on oven-dried wood meal).

For each trial, 1.0 g of oven-dried wood meal was placed in a reactor and cooked at 160°C with 8 mL of cooking liquor (liquor-to-wood ratio of 8 mL/g). The active alkali (AA) charges applied were 23%, 27%, and 31%. To investigate the effect of cooking time, additional experiments were performed at 160 °C with 31% AA for 4, 5, and 6 h. In all cases, an additional 10 min was allowed for the system to reach the target temperature. Prior to cooking, lapachol and 2-MAQ were activated by mixing with D-glucose (1:2, w/w) in 2 M NaOH, followed by heating at 80–100 °C for 1 h.

2.4. Analytical methods. Pulp yield was determined on an oven-dry weight basis. The lignin content of the pulps (residual lignin) was analyzed according to standard procedures. Acid-insoluble lignin (Klason lignin) was measured by acid hydrolysis following TAPPI T 222 om-21 (equivalent to ISO 21436:2020). A sample weight of 0.1 g was hydrolyzed with 1.5 mL of 72% sul-

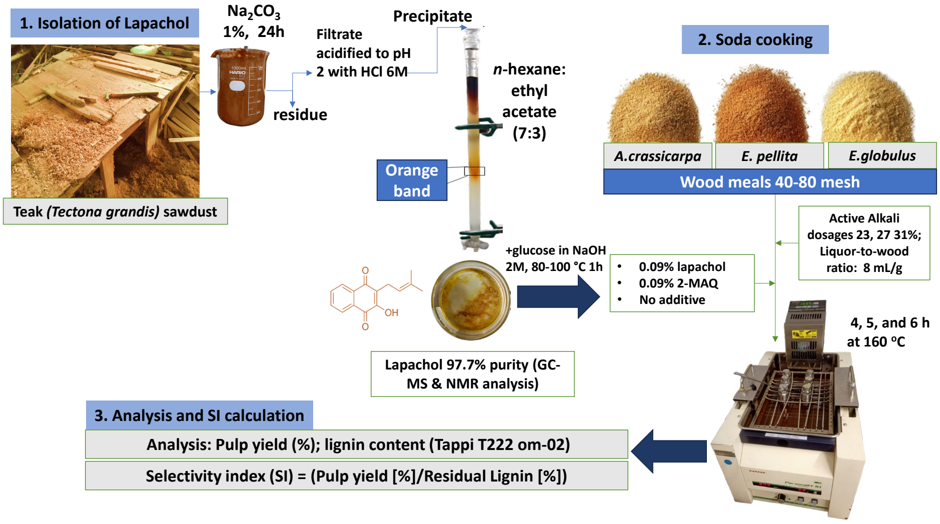

Figure 2. Schematic diagram of lapachol isolation from teak sawdust and its application in soda pulping.

furic acid room temperature with occasional stirring for 2h. The mixture was then diluted to 3% sulfuric acid using deionized water, transferred to a vial, sealed, and heated to 120°C for 1.5h. The resulting suspension was filtered, the acid-insoluble residue (Klason lignin) was determined gravimetrically. The acid-soluble lignin fraction was determined from the hydrolysate filtrate by UV–Vis spectrophotometry at 205 nm, according to TAPPI T222 om-88 procedure. The residual lignin was calculated as the sum of the acid-insoluble and acid-soluble fractions. The isolation of lapachol, soda pulping experiment and analysis are illustrated in the Figure 2. The pulping selectivity index (SI) was calculated according to the following equation:

$$\text{Selectivity index (SI)} = \frac{\text{Pulp yield (\%)}}{\text{Residual lignin (\%)}}$$

3. RESULT AND DISCUSSION

Efficient lignin removal while preserving carbohydrates is the main goal of any pulping process. Pulping performance is influenced not only by cooking conditions but also by the intrinsic characteristics of the raw material. Several studies have reported variations in delignification behavior among different hardwood species [10, 21-25].

The chemical compositions of the three studied wood species are summarized in Table 1. The percentages reported are based on oven-dry wood weight. Among them, *E. globulus* contained the highest glucan content (49.4%), compared with *E. pellita* (42.1%) and *A. crassicarpa* (41.1%). Since glucan largely represents cellulose and xylan represents hemicellulose, the glucan-to-(glucan+xylan) ratio can be used to indicate cellulose preservation potential during alkaline cooking. Glucan is more resistant to alkaline hydrolysis than hemicellulose; thus, a higher glucan ratio suggests greater cellulose retention. Interestingly, *E. pellita* exhibited the highest glucan ratio (0.839), while *E. globulus* showed the lowest (0.763). Beyond carbohydrate composition, lignin structure plays a more crucial role in delignification efficiency. Model compound studies have shown that the β-aryl ether (β-*O*-4) linkages in syringyl (S) lignin are more prone to cleavage than those of guaiacyl (G) lignin [27]. Since β-*O*-4 linkages are the predominant bonds in lignin, their reactivity strongly influences pulping performance. These linkages exist in two stereoisomeric forms, erythro (E) and threo (T). Under alkaline conditions, the E form tends to cleave over the T form. There is a positive correlation between the E/(E+T) ratio and the S/(S/V) ratio obtained by nitrobenzene oxidation, indicating that a higher proportion of S-type units enhances delignification efficiency [28,29]. The S/V ratio obtained by nitrobenzene oxidation is commonly used to estimate the S/G ratio, with a conversion factor of 0.806 [24]. In general, wood species with higher S/G ratios are easier to delignify. In this study, *A.crassicarpa* had the highest lignin content (31.4%), while *E. globulus* had the lowest (27.6%). The S/V ratio varied among species: *E.*

Table 1. Chemical composition of wood material.

	Eucalyptus pellita[a]	*Acacia crassicarpa*[a]	*Eucalyptus globulus*[b]
glucan (%)	42.1±2.39	41.1±3.9	49.4
xylan (%)	8.1±0.51	10.9±1.16	15.32
glucan ratio (%)	0.839	0.790	0.76
acid insoluble lignin (%)	28.7±0.12	29.6±0.48	22.2
acid insoluble lignin (%)	0.69±0.03	1.89±0.22	5.4
total lignin (%)	29.4	31.4	27.6
Ash (%)	0.09±0.01	0.22±0.01	0.55±0.11
Acetone extractives (%)	1.68±0.11	2.08±0.08	0.6
Acetone extractives (%)	19.0	14.2	7.1
S/V molar ratio	2.04±0.21	0.74±0.01	3.04
S dan V yield (mmol/g-lignin)	2.42±0.05	1.24±0.31	5.81

[a] [10]
[b] [26]

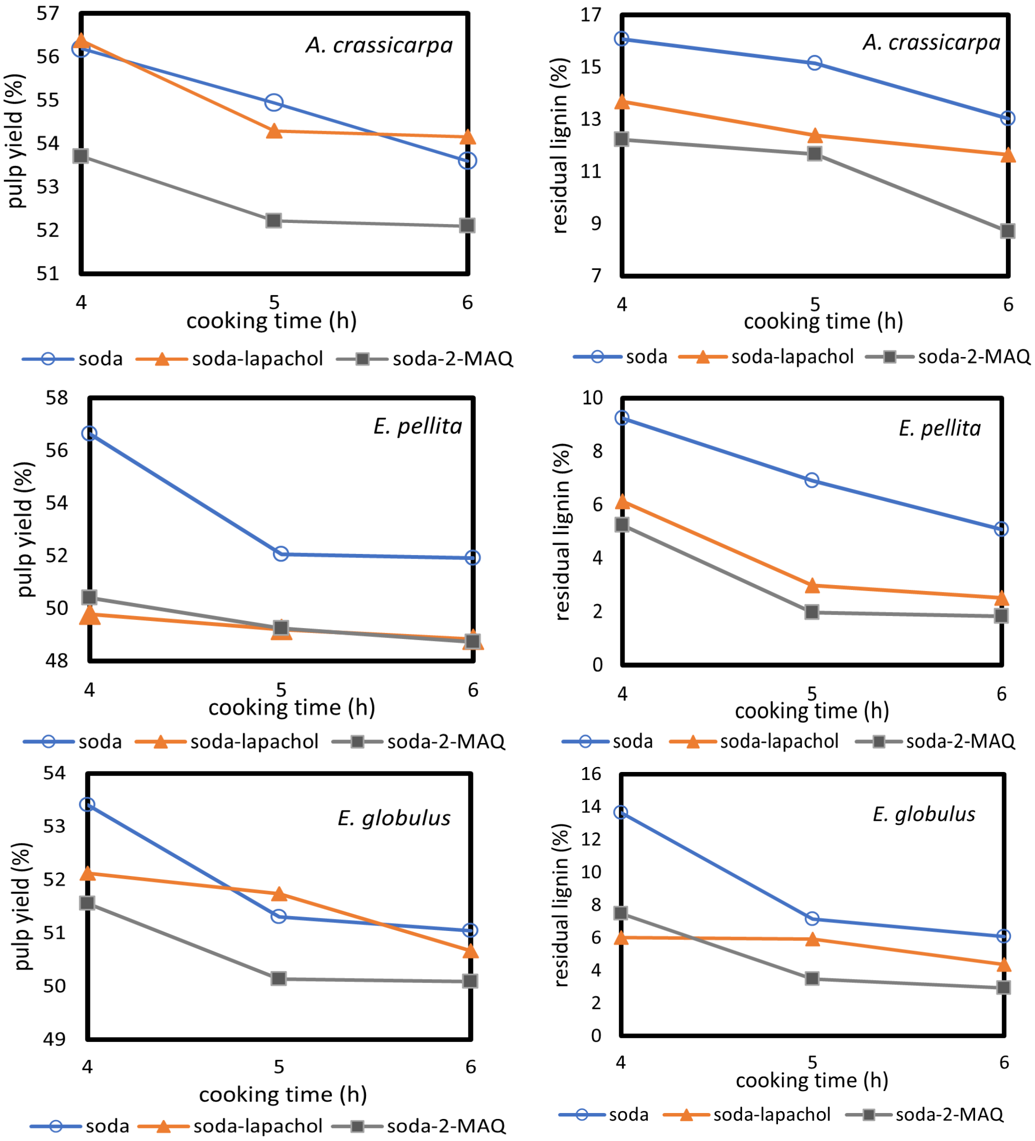

Figure 3. Effect of cooking time on pulp yield (%) and residual lignin (%) in soda cooking (AA dosage: 31%) at 160 °C, with and without additives, for *A. crassicarpa*, *E. pellita*, and *E. globulus*.

globulus showed the highest value (3.04), followed by *E. pellita* (2.04), and *A. crassicarpa* (0.74). Based on these data, *A. crassicarpa* is expected to be the most difficult species to delignify, whereas *E. globulus* is the easiest, with *E. pellita* showing intermediate delignification behavior.

3.1. Effect of cooking time on pulp yield and residual lignins. The influence of cooking time on pulp yield and residual lignin content was investigated at 160 °C and 31% active alkali for *A. crassicarpa*, *E. pellita*, and *E. globulus*. As expected, extending the cooking time from 4 to 6 h progressively decreased pulp yield across all wood species due to enhanced delignification and carbohydrate degradation (Figure 3). For example, in the soda cooking of *A. crassicarpa*, the yield decreased from 56.19% (4 h) to 53.60% (6 h), while the residual lignin content declined from 16.07% to 13.02%. A similar trend was observed for boths *E. pellita* and *E. globulus*, although the extent of lignin removal varied with wood type. The addition of lapachol consistently enhanced delignification compared to the control (soda), shown by reduced residual lignin in all wood species (Figure 2). In *A. crassicarpa*, the pulp yield across cooking times was comparable to soda, but the residual lignin content was lowered by 2-3% compared to the control. In *E. pellita*, lapachol reduced residual lignin from 6.90% (control) to 2.98% after 5 h (~4% reduction), but the pulp yield was lower than that of the control (soda). Meanwhile, in *E. globulus*, residual lignin decreased from 7.15% (control) to 6.04% (soda-lapachol) after 5 h and to 4.37% after 6 h, with stable pulp yields (~51%).

Quantitative analysis using the selectivity index (SI = yield[%]/lignin[%]), lapachol improved SI in all species (Table 2). The effect was most pronounced in *E.globulus*, where lapachol nearly tripled SI over soda at earlier cooking times and maintained the highest SI improvement at all durations, from 3.91(soda) to 8.65 (soda-lapachol) at 4 h, and from 8.41 (soda) to 11.58 (soda-lapachol) after 6 h. Although *E. pellita* exhibited higher SI values at longer cooking times, its lower pulp yields and greater variability

Table 2. Selectivity Index (SI = Pulp yield /Residual lignin) of soda, soda–lapachol, and soda–2-MAQ Cooking at 160 °C.

Wood Species	Cooking Time (h)	Selectivity Index (SI)		
		Soda	Soda–Lapachol	Soda–2-MAQ
A. crassicarpa	4	3.50	4.12	4.39
	5	3.63	4.38	4.47
	6	4.12	4.65	5.98
E. pellita	4	6.13	8.12	9.62
	5	7.55	16.50	24.85
	6	10.22	19.31	26.65
E. globulus	4	3.91	8.64	6.89
	5	7.18	8.74	14.41
	6	8.41	11.59	17.04

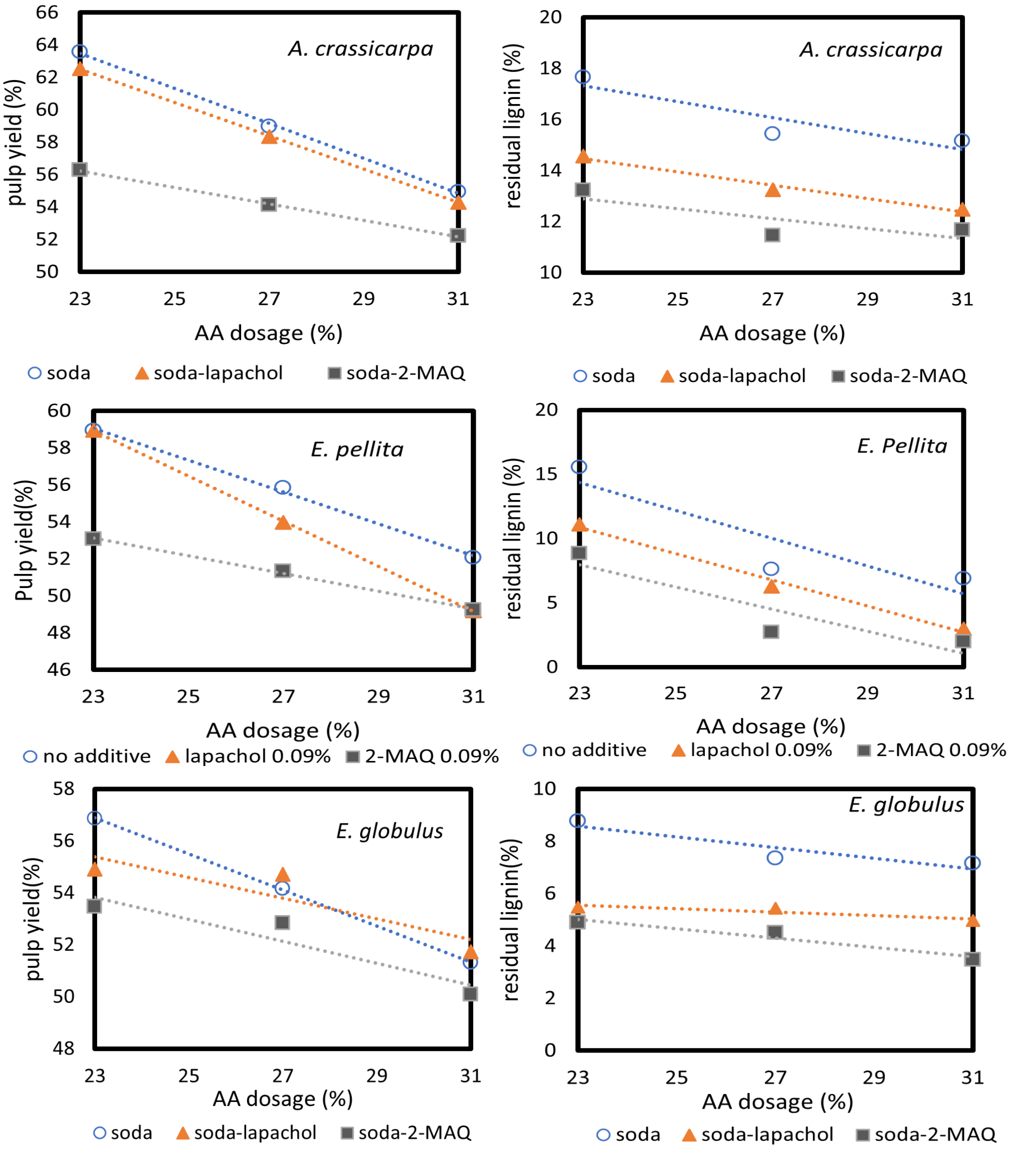

Figure 4. Effect of AA dosage on pulp yield (%) and residual lignin (%) in soda cooking at 160 °C, with and without additives, for *A. crassicarpa, E. pellita,* and *E. globulus.*

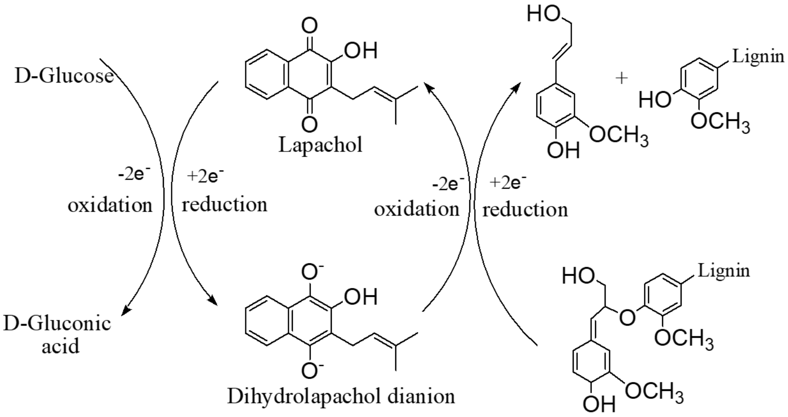

Figure 5. Hypothetical catalytic mechanism of lapachol-enhanced alkaline delignification.

in yield reduction suggest less overall process stability. In contrast, *E. globulus* maintained relatively high and stable pulp yields alongside substantial lignin removal, representing a favorable balance between delignification efficiency and carbohydrate preservation. This indicates that, in soda cooking lapachol offers its most effective balance between high pulp yield and lignin removal when applied to *E. globulus*, rather than in *E.pellita* or *A.crassicarpa*.

When compared to 2-MAQ, based on the trend in Figure 3 and Table 2, lapachol demonstrated comparable catalytic performance. The residual lignin from soda-lapachol cooking is only slightly higher than soda-2-MAQ. Meanwhile, the pulp yields are comparable to soda-2-MAQ for *E. pellita* and higher than soda-2-MAQ in *A. crassicarpa* and *E. globulus*. This finding suggests that natural lapachol can function as an effective alternative catalyst to synthetic 2-MAQ.

3.2. Effect of active alkali dosage on pulp yield and residual lignin. The effect of alkali dosage was studied at 23%, 27%, and 31% AA with 5 h cooking. Increasing alkali concentration generally lowered residual lignin but also reduced pulp yield due to more severe carbohydrate degradation (Figure 4). For example, in soda cooking of *A. crassicarpa*, yield dropped from 63.58% (23% AA) to 54.94% (31% AA), while lignin decreased from 17.65% to 15.15%. Similar patterns were observed in *E. pellita* and *E. globulus*.

Lapachol consistently enhanced delignification compared to the control at all AA dosages for three wood species. In case of *A. crassicarpa*, the total residual lignin decreased from 15.15% (control, 31% AA) to 12.48% (lapachol, 31% AA), but interestingly, the pulp yield is maintained close to the control (soda). In *E. pellita*, residual lignin decreased dramatically from 6.90% (control, 31% AA) to 1.98% (lapachol, 31% AA), an approximate 5% reduction. In *E. globulus*, lapachol lowered lignin residue from 7.15% to 5.33% with pulp yield comparable to soda. From all AA dosages in the three woods, lapachol enhances delignification without significant yield reduction, highlighting its role in promoting selective delignification.

Compared with 2-MAQ, lapachol exhibited competitive performance. In *A. crassicarpa*, residual lignin from soda-lapachol was 12.48% at 31% AA, compared to 11.67% with soda-2-MAQ. In *E. pellita*, lapachol and 2-MAQ showed comparable results (2.98% vs. 1.98%), however in *E. globulus*, lapachol was slightly less effective than 2-MAQ (5.33% vs. 3.48%). It can be notified that the yields obtained from soda-lapachol were consistently higher or comparable to those soda-2-MAQ in all AA dosages, suggesting that lapachol provides a good balance between delignification and carbohydrate preservation.

The exact mechanistic action of lapachol in enhance delignification and carbohydrate retention is remain unclear, since there were no studies about application of lapachol for pulping additive. Therefore, further study that focus on elucidating the catalytic mechanism of lapachol are needed. Hypothetically, lapachol has structure analog to the established AQ, enabling it to undergo redox cycles under alkaline pulping conditions. Lapachol is initially reduced by carbohydrate end groups to its hydroquinone form (lapachol dianion). This reduced species then donate electron to lignin-derived quinone methide intermediates regenerating the quinone form (lapachol). This redox cycling promotes cleavage of β-*O*-4 ether linkages of lignin while suppress carbohydrate peeling reactions by oxidizing the reducing end of carbohydrate (Figure 5). Additionally, the phenolic hydroxyl group at C-2 position may enhance its electron donating capacity and resonance stabilization during redox cycling, thereby increasing catalytic efficiency toward lignin depolymerization. The isoprenoid side chain may further influence interaction with lignin macromolecules, improving accessibility to hydrophobic lignin regions. However, this mechanism remains hypothetical and requires further experimental verification.

3.3. Selectivity index and catalytic efficiency. Figure 6 shows the relationship between pulp yield and residual lignin content for *A. crassicarpa*, *E. pellita*, and *E. globulus* from several cooking conditions. The trends show that soda cooking produced higher pulp yields but with very high residual lignin, indicating limited delignification. Conversely, soda-2-MAQ achieved the lowest residual lignin but at a significant yield loss. Soda-lapachol provided an intermediate result, reducing residual lignin more effectively than soda while maintaining higher pulp yields than soda-2-MAQ. This suggests that lapachol improved pulping selectivity by balancing delignification and carbohydrate preservation.

The differences in selectivity among the three wood species can be related to their chemical composition (Table 1). A. crassicarpa has the highest lignin content (31.4%) and lowest S/V ratio (0.74), making it the most difficult to delignify and resulting in

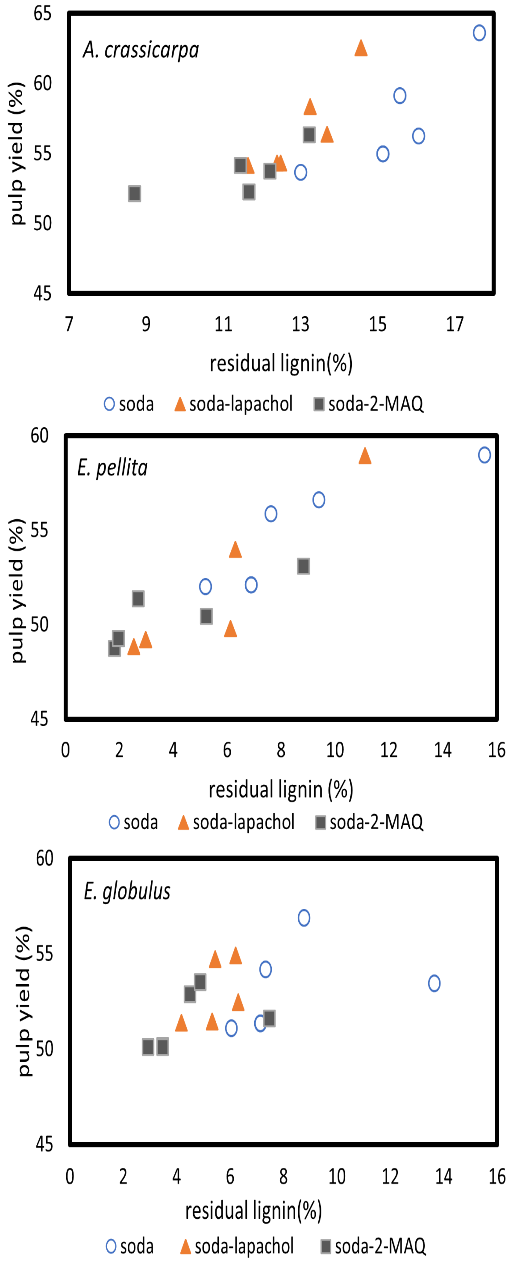

Figure 6. N_2 residual lignin (%) vs pulp yield (%) of various cooking conditions of soda cooking *A. crassicarpa*, *E. pellita*, *E. globulus*.

higher residual lignin in its pulps across all cooking conditions. *E. globullus*, with the lowest lignin content (27.6%) and highest S/V ratio (3.04), is the easiest to delignify, as shown by low residual lignin even at relatively high yields. *E. pellita* demonstrates a wide range of combination pulp yield vs residual lignin, from the high lignin residual content with high pulp yield to the lowest residual lignin for soda-lapachol and soda-2-MAQ. This could be another factor to investigate in a future study.

Overall, the plots confirm that lapachol enhances pulping performance by improving lignin removal while avoiding excessive yield losses. When correlated with wood composition, the results indicate that the lapachol benefit depends not only on cooking conditions but also on the intrinsic delignification potential of each wood species.

These trends are further confirmed in the average selectivity index plots (Figure 7). Across all species, soda cooking consistently showed the lowest selectivity, highlighting the trade-off between high yield and poor delignification. In contrast, soda-2-MAQ achieved the highest selectivity but with significant yield loss. Importantly, soda-lapachol displayed intermediate, indicating that it provides a practical compromise between soda and soda-2-MAQ. When correlated with wood composition, the results suggest that the advantage of lapachol becomes more pronounced in species with lower lignin content and higher S/V ratio, such as *E.globulus*.

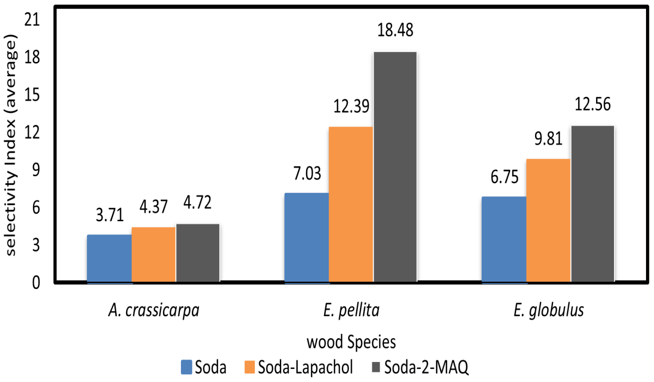

Figure 7. Average selectivity index (SI) of various cooking conditions of soda cooking *A. crassicarpa*, *E. pellita*, *E. globulus*.

The combination of yield–residual lignin relationships and selectivity indices demonstrates that lapachol can enhance pulping performance by improving lignin removal while preserving carbohydrates. Compared to 2-MAQ, lapachol may be less aggressive, but it offers a more balanced and sustainable improvement in pulping selectivity across different hardwood species.

3.4. Implications for green pulping technology. These findings underscore the significant impact of lapachol, isolated from Indonesian teak wood waste, as a green catalyst in soda pulping of *Acacia* and *Eucalyptus*. Lapachol enhanced delignification and maintained a comparable pulp yield to soda. In addition, lapachol stands out as a natural with potentially safer alternatives to synthetic AQ. The combination of sustainable pulping chemistry and valorization of forestry by-products highlights the natural lapachol in advancing circular bioeconomy strategies.

4. CONCLUSION

The wood characteristics, particularly lignin structure, strongly affect pulping behavior. *A. crassicarpa*, with the highest lignin content and lowest S/V ratio, was the most difficult to delignify, while *E. Globulus*, with the lowest lignin content and highest S/V ratio, was the easiest. Meanwhile, *E. pellita* shows the highest average selectivity index (SI) among the three wood species. Among the cooking systems and three wood species, the addition of lapachol enhances delignification comparable with soda-2-MAQ. Soda-lapachol consistently provided intermediate performance: reducing lignin more effectively than soda while maintaining higher yields than soda-2-MAQ. The most pronounce balance between delignification and carbohydrate preservation was shown when lapachol applied in soda cooking of *E. globulus*. Overall, lapachol demonstrated potential as a natural redox catalyst for alkaline pulping, suggesting promise for sustainable pulping applications

ACKNOWLEDGEMENTS

Esty Octiana Sari is grateful to the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan for providing the scholarship to conduct the current research under the MEXT Special Scholarship Program on Trans-world Professional Human Resources Management (TPHRD) for Doctoral Course.

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Esty Octiana Sari: Conceptualization, Methodology, Investigation, Data curation, Formal analysis, Writing–Original draft preparation, Writing–Reviewing and Editing. **Syelvia Putri Utami**: Investigation, Data curation, Formal analysis, Writing–Reviewing and Editing. **Evelyn:** Writing–Reviewing and Editing. **Akiko Nakagawa-Izumi**: Supervision, Methodology, Data curation, Formal analysis. **Hiroshi Ohi**: Supervision, Conceptualization, Writing–Reviewing and Editing.

DECLARATIONS

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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