WOOD STRUCTURE AND FUNCTION (A KOUBAA, SECTION EDITOR)



# Application of Liquefied Wood Products for Particleboard Manufacturing: a Meta-analysis Review

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## Abstract

**Purpose of Review** The development of eco-friendly panels is one of the major issues for the wood industry, particularly in terms of developing new adhesive systems for reducing harmful emissions and dependence from fossil fuel resources. Among the plethora of various possible solutions, liquefied wood products have demonstrated promising features to decrease the use of petroleum-based amino resins. A meta-analysis was carried out to develop a quantitative synthesis of the state of the application of liquefied wood products for particleboard manufacturing, investigating their effects on the board's properties.

**Recent Findings** The results from meta-analysis revealed that the application of liquefied wood products does not significantly deteriorate the mechanical properties of particleboards. It has also been observed that there is no negative influence on the formaldehyde content of the panel, mostly when liquefied wood products are applied in mixture with commercial resin. The most interesting observation is that the performance of the boards was not affected by the type of feedstock used for liquefaction.

**Summary** The option to apply liquefied wood products has the potential to significantly decrease the application of commercial formaldehyde-based resins without leading to a deterioration of the properties of the panels. It is however important to note that there is currently a limited number of studies on this topic. The meta-analysis showed a notable variability of the effect sizes in the trials. Further study is needed to determine the outcomes and the benefits for the use of liquefied wood products on an industrial level.

Keywords Liquefaction · Liquefied wood · Bio-adhesive · Binder · Particleboards · Formaldehyde

# Introduction

The wood industry is constantly undergoing technological developments towards innovative solutions while dealing with environmental concerns. Thus, it has taken on the challenge to recycle the industrial residues from wood processing and to convert them into value-added products. This is in line with one of the principles of circular economy, which

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<sup>2</sup> University of Lorraine, ENSTIB-LERMAB, 27 rue Philippe Séguin, BP1041, 88051, Cedex 9 Epinal, France is to maintain as much as possible the value of products and resources while minimizing the generation of waste [1]. This is particularly important taking into account the great variety of wood waste sources such as timber harvesting and wood processing residues, construction and demolition waste, wood packaging, and even more including private households or railway construction. Small-sized wood waste, including sawdust and bark, are considered particularly troublesome residues [2, 3••, 4, 5]. Moreover, some of their features, such as low bulk density and combustibility of wood sawdust, can cause concerns related to logistic and storage issues. These economic and environmental implications lead to the strong need to exploit this type of residual biomass and give it a second life. So far, the most common use of residual woody biomass is for bioenergy purposes [6–9]. However, the recent European Forest Strategy for 2030 has been promoting the sustainable use and optimization of wood-based resources in accordance with the principle of the cascading utilization of wood [10]. Cascading use is the efficient utilization of resources by using residues and recycled products for material uses while postponing bioenergy production at the end of the life cycle [11, 12].

A promising way to exploit wooden residues in accordance with a circular bioeconomy and cascade utilization approach can be obtained through liquefaction processes. Liquefaction is defined as a technology to convert biomass to liquid products via complex chemical reactions [13••]. The process has great potential to produce value-added products from available lignocellulosic biomass for many different applications including carbon materials [14], biofuels [15], coatings [16], and adhesives [17, 18].

Various factors can affect the liquefaction process itself including physical ones like temperature, reaction time, and biomass to solvent ratio and chemical ones such as liquefaction reagents-solvents relations, catalyst, and raw material type [19–22]. One of the most sought-after applications of liquefied wood products (LWP) in terms of suitability for wood-based panel technology is their application for the process of particleboard production with the aim of decreasing harmful formaldehyde release and to simultaneously become less dependent from petroleum sources. According to the current literature, there are several ways of using LWP in the manufacturing of particleboards: partial substitution of commercial resin with LWP [23], using LWP as a precursor to synthesize adhesives and applying these adhesives to completely replace the commercial resin [24], and replacing part of the feedstock used for the production of the particleboards with LWP [25].

The particleboard industry has shown substantial market development over the last two decades, with production growth of about 50% from 2000 to 2021 [26]. Particleboards are a particular type of engineered wood product, made from wood chips bonded with an adhesive. The outstanding feature is that they can be effectively produced from many different alternative feedstocks including residual lignocellulosic biomass [27–30]. However, one of the major concerns related to particleboard manufacturing is the current use of formaldehyde-based resins as binders, including urea formaldehyde (UF), melamine formaldehyde (MF), and melamine urea formaldehyde (MUF) [31, 32, 33••, 34]. These resins are produced from petrochemical feedstocks, and moreover, the presence of formaldehyde may have some negative effects on human health [35, 36].

Notwithstanding the efforts of scientific research to find alternative solutions for the reduction of pollutant emissions from formaldehyde-based resins, these are still the mostly widely used in the wood industry due to their excellent cohesion and adhesion strength as well as low cost. The application of bio-adhesives for particleboard manufacturing is still challenging at the industrial stage due to either limitations in supply such as for commercial tannin adhesives [37–43] or

some other limitations such as little bonding strength, low availability, too high viscosity, poor water resistance, a very slow hot press time, and high cost [44, 45•]. Moreover, one of the key parameters is to maintain the quality of the panels, while replacing the presently used resins with bio-adhesives.

Therefore, to go beyond the experimental phase, there is a need to find alternative manufacturing solutions, including new adhesive systems that can decrease the pollutant emissions, including formaldehyde, without compromising the mechanical properties of the particleboard and without a significant increase in their cost of production. The focus of the research lies in the transition to greener adhesives by using bio-based compounds obtained from renewable sources [33, 46, 47]. Among these, compounds from lignocellulosic materials are the most attractive since they are the most abundant and likely to be processed to value-added products. However, understanding the effect of liquefied wood products on the particleboards quality is of fundamental importance when considering their future potential in industrial application.

Therefore, the aim of this meta-analysis was to investigate the impact of the application of LWP on particleboard standard properties.

Through applying sub-group meta-analysis, the focus has been placed on the evaluation of the influence of different variables (moderators) on the mechanical properties of particleboards produced with LWP. The moderators considered were the liquefaction feedstock, the application type of LWP in the production of the panel, and the type of the industrial resin used. Furthermore, when dealing with LWP used to partially replace the commercial resin, the share of LWP used in the adhesive mixture was considered another moderator.

Taking all the above into consideration, the research hypotheses to be tested were defined as follows:

- The application of liquefied wood products for particleboard manufacturing does not significantly cause the mechanical properties of the panels to deteriorate and does not affect the formaldehyde content in the boards.
- The type of liquefied feedstock does not significantly affect the properties of the panels.

# **Materials and Methods**

### Literature Search and Criteria for Paper Selection

The literature search of relevant peer-reviewed articles was carried out using Clarivate Analytics Web of Science and Scopus databases. The publication year was not restricted; however, the search was limited only to manuscripts in English. The applied research keys were as follows: "liquef\* wood" AND "particleboard\*", "wood liquef\*" AND "particleboard\*", "liquef\* wood" AND "wood-based panel\*", "wood liquef\*" AND "wood-based panel\*", "liquef\* wood" AND "adhesive\*", "wood liquef\*" AND "adhesive\*", "wood liquef\*" AND "binder\*", "liquef\* wood" AND "binder\*", "liquef\*" AND "binder\*", "liquef\* wood" AND "binder\*", "liquef\*" AND "wood-based" AND "panel\*", "liquef\*" AND "particleboard\*", and "liquef\*" AND "bark\*".

A total of 426 papers were retrieved. This number was further refined by deleting duplicates and reading abstracts. In total, 37 papers remained after this phase. Finally, only papers fulfilling the following criteria were included in the database for the meta-analysis: (1) the paper must be focused on evaluating the quality of the particleboard produced with LWP and not on the quality of the adhesive itself; (2) the paper must report the properties of a reference control panel produced with commercial resin without the addition of a liquefied product-under the same conditions and starting from the same raw material; (3) the paper must deal with liquefied products derived from wooden material, excluding liquefaction of agricultural biomass. Considering the above, the acronym LWP (liquefied wood products) refers to the product of liquefaction derived from wooden residues, namely sawdust and bark.

#### Data Extraction and Building of the Database

From the 11 papers meeting all the selection criteria [22, 24, 25, 48–52, 53•, 54, 55], the data available in the text and tables were directly retrieved from the manuscripts, while data from graphs were extracted via WebPlotDigitizer version 4.6 software. The average, standard deviation, and

number of replications for both panels produced with liquefied wood and under the control conditions were extracted.

The considered parameters of particleboard quality were as follows: internal bond (IB, MPa) strength, modulus of rupture (MOR, MPa), modulus of elasticity (MOE, MPa), thickness swelling at 24 h (TS, %), and formaldehyde content of the panel (FC, mg/100g<sub>odb</sub> with odb meaning ovendried board). Data related to the various moderators, i.e. feedstock for liquefaction, type of application of LWP, type of resin, and share of liquefied wood products in the adhesive, were further retrieved from the publications. Data for each moderator were categorized according to the descriptors reported in Table 1.

In all the analysed manuscripts, a single paper contributed to the meta-analysis with more than one comparison between liquefied adhesive and the commercial one. Specifically, 43 comparisons were detected for IB and MOR, 41 for MOE, 35 for TSW, and 19 for FC. It was assumed that to be included in the sub-group meta-analysis, the category of a moderator should be present in at least three couple comparisons deriving from at least two different studies.

#### **Meta-analysis**

The natural log of the response ratio (Ln r =Ln [value for the panel with liquefied wood/value in the control panel]) was selected as the effect size [56, 57] and later converted into percentage of change through the equation  $(r - 1) \times 100\%$  [58, 59] to allow an immediate evaluation of the variation.

Three-level mixed effect meta-analysis was carried out to account for non-independence in the data due to fact that several comparisons in the database shared a common control [60, 61]. A weighted, multi-level linear mixed effect

Moderator	Category	Description		
Feedstock for liquefaction	Sawdust	Sawdust of coniferous and broadleaf species		
	Bark	Coniferous bark		
Application type	Partial substitution of the adhesive (PSA)	LWP are used in an adhesive mixture by replacing a certain percentage of the commercial resin		
	Full substitution of the adhesive (FSA)	LWP are used as precursor to synthesize the adhesive which is applied without any commercial resin		
	Partial substitution of the feedstock (PSF)	LWP are applied as partial substitution of wood chips		
Resin type	UF	Urea formaldehyde		
	MUF	Melamine urea formaldehyde		
	MF	Melamine formaldehyde		
	PF*	Phenol formaldehyde		
Share of LWP**	0–20%	Up to 20% of liquefied wood in the adhesive mixture		
	>20%	More than 20% of liquefied wood in the adhesive mix		

\*In the database retrieved from the analysed papers, there were not enough data to consider PF resin as a level for the moderator "resin type"; therefore, the sub-group meta-analysis was performed with only three levels for this moderator, namely UF, MUF, and MF

\*\*The moderator "share of LWP" was considered only for papers that applied LWP as PSA

model was used to calculate the overall effects using the R package "metafor", and all the estimates of the parameters were undertaken using REML (restricted maximum likelihood). The heterogeneity of effect sizes was assessed with the formal Cochran's Q test (Q). The significance of the positive and negative effects of liquefied wood on the properties of the particleboard depends on whether the effect sizes at the 95% confidence intervals (CIs) overlap with zero, with no crossover being significant.

## **Results and Discussion**

The results of sub-group three-level meta-analyses carried out for all the comparisons retrieved from the investigated papers that considered the various tested moderators are given below.

The obtained results for internal bond (IB) are reported in Fig. 1. No statistically significant difference was evident for any of the levels of all the investigated moderators apart from the case when applying LWP as a full substitution of the adhesive (FSA), with an average reduction of IB of -38.52%. Authors who applied LWP as FSA stated that the reduction in IB could probably be related to the incomplete cure of the resin during the hot pressing step and proposed a hot stacking procedure to overcome this drawback [24, 55]. On the other hand, no significant differences in IB were detected when LWP was applied as a partial substitute of the commercial resin and of the feedstock for particleboard manufacturing.

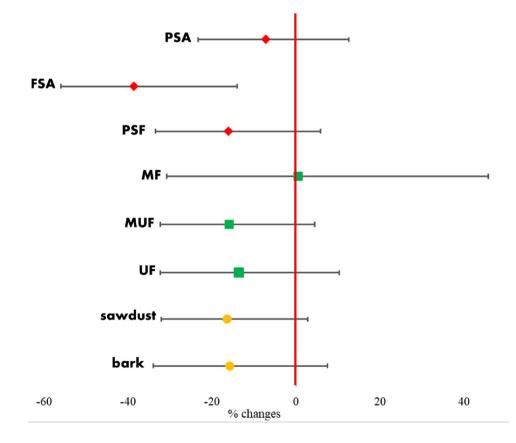
Considering the type of resin, the values of the categories for this moderator are at a comparable level, mostly between MUF (-15.94%) and UF (-13.70%), while the effect size for MF is very close to the 0 line (0.45%). For any category of this moderator, the difference with the control panel was significant according to the three-level meta-analysis; therefore, the type of resin was not an influential parameter in determining the internal bond of the panels produced with LWP.

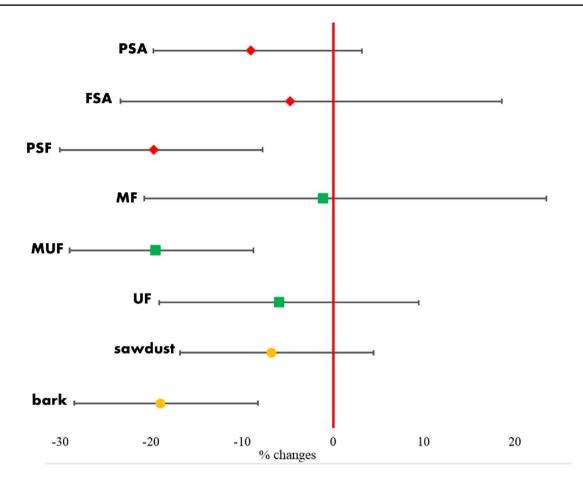
The feedstock for liquefaction did not significantly influence the IB of the particleboards. Therefore, the obtained results demonstrated that the type of application of LWP (FSA) has a significant influence on the internal bond of the panels, while other investigated moderators do not have a marked influence and did not result in significant deterioration of the panel quality in comparison to the control panel.

The type of LWP application was the most influential moderator for modulus of rupture (Fig. 2). In particular, application as partial substitution of the feedstock (PSF) led to a significant decrease in MOR (-19.68%), while PSA and FSA were not significantly different from the control values. The negative effect on the MOR parameter as a consequence of the substitution of wood chips with

comparison between internal bond (IB) of particleboards produced with LWP and control panel. The mean effect size is indicated by different symbols according to the moderator (rhombus for application type, square for type of resin in the control panel, and circle for feedstock for liquefaction). Error bars indicate the 95% CIs. CIs not crossing the 0 line (in red) indicate the presence of statistically significant differences (p < 0.05) between the values of the particleboard produced with liquefied wood and the commercial one; on the other hand, when the CIs cross the 0 line, no statistically significant difference is indicated

Fig. 1 Forest plot for the





**Fig.2** Forest plot for the comparison between modulus of rupture (MOR) of particleboards produced with LWP and control panel. The mean effect size is indicated by different symbols according to the moderator (rhombus for application type, square for type of resin in the control panel, and circle for feedstock for liquefaction). Error

LWP could be attributed to a lower density of the panels manufactured with LWP [25, 48]. The sub-group metaanalysis also revealed a significant decrease in comparison to the control panels bonded with MUF resin (-19.51%)and when applying coniferous bark (-18.97%) as feedstock for liquefaction. However, it is worth highlighting that the major part of paired comparisons between treatment vs control dealing with liquefied bark and MUF in the database are properly attributed to the application of LWP as a partial substitute of the feedstock for particleboard production. Therefore, the low performance revealed by this meta-analysis for the MOR of the panels produced with liquefied bark bonded with MUF could not be related to the application of liquefied bark or MUF themselves, but to the fact that the comparison was carried out in most cases with a specific type of application, namely PSF, which can be particularly influential for the MOR of the panel [25, 53•]. Thus, analysing the obtained results, it is possible to assert that the type of feedstock for liquefaction

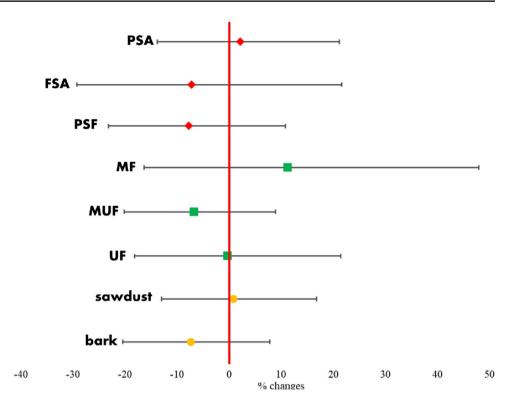
bars indicate the 95% CIs. CIs not crossing the 0 line (in red) indicate the presence of statistically significant differences (p<0.05) between the values of the particleboard produced with liquefied wood and the commercial one; on the other hand, when the CIs cross the 0 line, no statistically significant difference is indicated

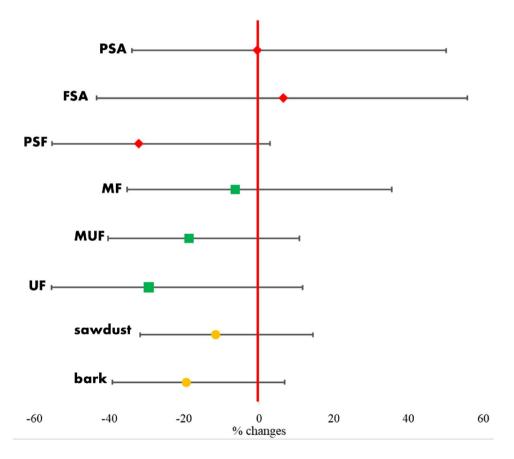
does not have a significant influence on the MOR property, especially when LWP is applied as a partial or a full substitution of the commercial resin. Moreover, it can be concluded that applying LWP as PSA the type of resin is not an influential moderator.

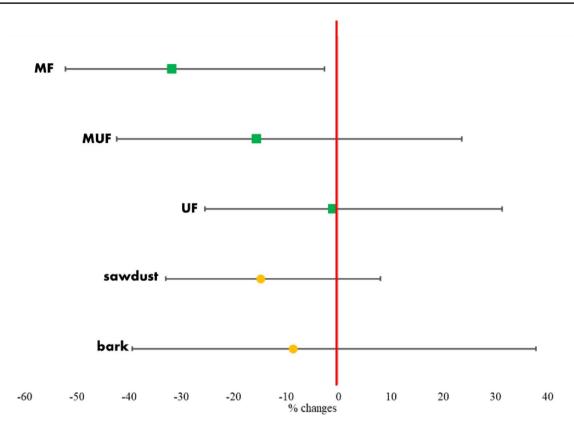
Focusing on the modulus of elasticity, the results obtained are shown in Fig. 3. For all the categories of all the moderators, no statistically significant differences were detected between the MOE of panel produced with application of LWP and that of the control panel. The results concerning the type of LWP application are particularly interesting. Indeed, in contrast to the IB and MOR results, the various methods of LWP application did not lead to a significant modification of the modulus of elasticity of the particleboards.

Promising results were obtained for thickness swelling as well (Fig. 4). No statistically significant difference was detected for this parameter in comparison to the control panel for all the categories of all the moderators. Therefore, Fig. 3 Forest plot for the comparison between modulus of elasticity (MOE) of particleboards produced with LWP and control panel. The mean effect size is indicated by different symbols according to the moderator (rhombus for application type, square for type of resin in the control panel, and circle for feedstock for liquefaction). Error bars indicate the 95% CIs. CIs not crossing the 0 line (in red) indicate the presence of statistically significant differences (p < 0.05) between the values of the particleboard produced with liquefied wood and the commercial one; on the other hand, when the CIs cross the 0 line, no statistically significant difference is indicated

Fig. 4 Forest plot for the comparison between thickness swelling (TS) of particleboards produced with LWP and control panel. The mean effect size is indicated by different symbols according to the moderator (rhombus for application type, square for type of resin in the control panel, and circle for feedstock for liquefaction). Error bars indicate the 95% CIs. CIs not crossing the 0 line (in red) indicate the presence of statistically significant differences (p < 0.05) between the values of the particleboard produced with liquefied wood and the commercial one; on the other hand, when the CIs cross the 0 line, no statistically significant difference is indicated







**Fig. 5** Forest plot for the comparison between formaldehyde content (FC) of particleboards produced with LWP and control panel. The mean effect size is indicated by different symbols according to the moderator (square for type of resin in the control panel and circle for feedstock for liquefaction). Error bars indicate the 95% CIs. CIs not

crossing the 0 line (in red) indicate the presence of statistically significant differences (p<0.05) between the values of the particleboard produced with liquefied wood and the commercial one; on the other hand, when the CIs cross the 0 line, no statistically significant difference is indicated

it can be stated that LWP application in particleboard manufacturing did not negatively affect the TS of the panel.

The results presenting formaldehyde content are given in Fig. 5. Considering that all the papers investigating formaldehyde content referred to the use LWP as a partial substitution of the adhesive, only this type of application was taken into account analysing the influence of LWP on the formaldehyde content of the panel. Referring to the type of the resin, the reduction in FC was significant only in the case of MF, while no significant difference was detected when applying it to MUF and UF. However, this aspect is probably related to the correlation among levels of different moderators. In the database for the meta-analysis, the comparisons between treatment vs control included the UF resin where only a percentage replacement lower than 20% was applied (Table 2), which probably

 Table 2 Results of the sub-group meta-analysis for the moderator

 "share of LWP" when applying LWP as a partial substitute of the commercial resin in the adhesive. \*\*Statistically significant differ

ence at p<0.01. \*Statistically significant difference at p<0.05. n.s. indicates no significant difference. *ES*, effect size; *LB95%*, lower 95% confidence interval; *UB95%*, upper 95% confidence interval

Variables	Share of LWP									
	0–20%				>20%					
	ES	LB 95%	UB 95%	<i>p</i> -value	ES	LB 95%	UB 95%	<i>p</i> -value		
FC	-2.77%	-20.28%	18.58%	n.s.	-37.51%	-52.45%	-17.89%	**		
MOE	-1.10%	-11.47%	9.26%	n.s.	6.75%	-8.96%	22.46%	n.s.		
MOR	-10.80%	-26.03%	4.43%	n.s.	-19.27%	-38.43%	-0.12%	*		
IB	-9.92%	-27.24%	7.40%	n.s.	-11.66%	-32.02%	8.70%	n.s.		
TS	-17.09%	-38.65%	4.47%	n.s.	14.33%	-6.97%	35.63%	n.s.		

influenced the results, i.e. the lack of statistically significant reduction of formaldehyde content in comparison to a pure UF resin is not related to the environmental performance of the UF resin but to the low percentage of liquefied wood in the adhesive applied in comparison to the control panels manufactured with pure UF resins. The results obtained did not show significant changes in the formaldehyde content of the panel in relation to the feedstock applied for liquefaction.

In the case where LWP was applied as a partial substitution of the adhesive, it is important to consider how much of the commercial resin can be replaced by LWP without significantly deteriorating the quality of the panels. With this aim, the share of LWP used in the adhesive mixture was considered a further moderator. The results concerning the influence of the percentage of LWP in the adhesive mixture when applying LWP as partial substitute of the commercial resin (PSA) are given in Table 2. Meta-analysis revealed a significant effect of the percentage of replacement of commercial resin by LWP on FC, with a significant decrease (-37.51%) when applying more than 20% into the adhesive mass. Replacement of commercial amino resin by LWP less than 20% results in an insignificant reduction (-2.77%). This indicates that to achieve increased environmental performance, at least 20% of the commercial resin in the adhesive has to be replaced with liquefied wood products. On the other hand, a substitution more than 20% caused a significant decrease in MOR (-19.27%); however, the upper confidence interval is very close to 0, and the difference is therefore at the border of significance. On the other hand, it was revealed that the share of liquefied wood in the adhesive did not affect MOE, IB, and thickness swelling.

# Conclusions and Future Research Perspectives

Results from the meta-analysis have confirmed the research hypotheses with some limitations. The results presented demonstrated the suitability of liquefied wood products for particleboard production. The first finding of the present meta-analysis is that the application type of LWP in the particleboard manufacturing is the determining factor on the properties of the obtained panel. The application of LWP in a mixture with commercial amino resin was a particularly promising solution both for interior type formaldehydebased UF resin and exterior type MUF resin. Three-level meta-analysis demonstrated that partially substituting commercial amino resin with LWP did not significantly deteriorate the mechanical performance of the boards. Application of LWP also reduces formaldehyde content; however, to obtain a significant difference, over 20% the LWP has to be applied in the adhesive. The other application types also reported promising results. On the other hand, a significant decrease in MOR was revealed when applying LWP to partially replace wood chips as feedstock, and a significant decrease in IB was shown when applying LWP to fully substitute the commercial resin.

What is important from the industrial point of view is that the results obtained demonstrate that there is no significant influence of liquefied raw material on final product properties; thus, the scope of possible wooden feedstock that can be subjected to liquefaction seems unlimited. Results presented here confirm the possibility of substituting part of the commercial amino resins with liquefied wood products. This substitution would allow for significantly greater utilization of wooden residues and contribute to the valorization of waste products generated by the wood processing industry in line with the cascading utilization of wood postulates.

However, further research should be focused on more-indepth study of the effects of particular moderators, including loading of LWP in the adhesive mixture, on the final product properties. Trials should be initiated which allow this process to be upscaled. Data on complex characteristics of upscaled liquefied wood products and by-products of the process should be provided with special emphasis on the optimization of parameters of technological usefulness, adequate viscosity, pH, gel time, and pot life of adhesive systems based on LWP, as these parameters are crucial for industrial manufacturing of the panels. Moreover, an environmental assessment of the liquefaction process should also be carried out to ensure sustainable production in the wood industry and support transition to the circular economy.

#### **Compliance With Ethical Standards**

**Conflict of Interest** Dominika Janiszewska-Latterini and Antonio Pizzi declare that they have no conflict of interest.

Human and Animal Rights and Informed Consent This article does not contain any studies with human or animal subjects performed by any of the authors.

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