



Agricultural residue lignin from novel low-temperature pretreatment as potential raw material for LPF resins

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Abstract

Appropriate digestate management is crucial for economic as well as ecologic optimization of biomethane production. Here, we present the production of a lignin-based phenol formaldehyde adhesive and its application to plywood from lignin separated after H_3PO_4 disintegration of digestate. Together with the work of Schroedter et al. (*Microorganisms* 9:1810, 2021), a full valorization of solid digestate is demonstrated. The separated sulfur-free LXP-lignin had high purity of above 90% lignin with a carbohydrate content of < 3%, an ash content below 2%, and a molecular weight of 6163 Da. The depolymerization of the LXP-lignin reduced the molecular weight to 3838 Da. The prepared lignin-based phenol formaldehyde resin has a viscosity of 350 mPa*s and a comparable tensile shear strength to the reference resin produced with the kraft lignin Indulin AT. Testing of nine-layer (500 × 500 mm) birch plywood panels showed promising results using standard resin recipes.

Keywords Digestate · Adhesive · Lignin · Biorefinery · Residues

1 Introduction

As stated in the 2018 EU Bioeconomy Strategy, Europe aims to develop a circular and sustainable economy which ensures food and nutrition security as well as reduces the dependence on non-renewable, unsustainable resources. The utilization of all available biomass (primary as well as secondary) is mandatory to reach this goal [1, 2]. As different biorefinery systems target for the same raw materials but often produce unused side streams, it is necessary to develop

integrated biorefinery systems to maximize the ecological benefit.

Anaerobic digestion is an efficient waste treatment technology and provider of sustainable energy. As by-product, it produces digestate which consists of water, un-degraded organic material, and inorganic compounds (e.g. minerals) [3]. To avoid negative impacts of intensified use of anaerobic digestion, appropriate digestate management has to be implemented. Besides ecological considerations, total process economics additionally benefits from sustainable nutrition management [4]. Solid/liquid fractionation is often the key technology for digestate processing. The separated liquid fraction can be further processed to recycle the water fraction and separate valuable ingredients for agricultural use [3, 4].

Depending on the input material and the digestion process, the composition of the un-degraded and separated solid fraction can differ significantly but carbohydrate and lignin contents between 20 and 50% and 15 and 30% are reported in literature [5–8]. For valorization of the carbohydrates and lignin present in digestate, additional treatment is mandatory. Different treatment methods as well as possible application are reported in literature [4].

One possibility is the utilization of solid digestate as feedstock within biorefinery systems. Carbohydrates can

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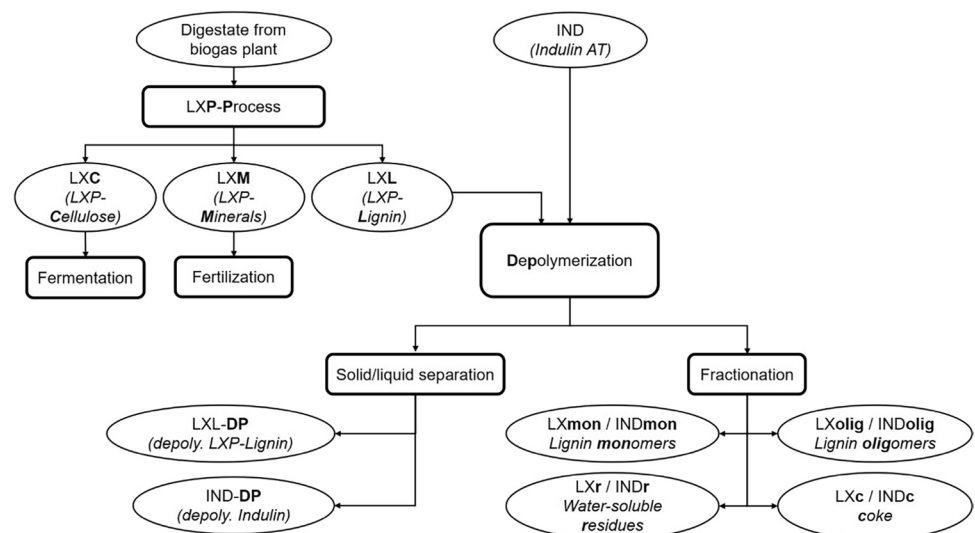
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Fig. 1 Basic flowchart of the process. Digestate from a commercial biogas plant enters the LXP process as raw material (RM). The pretreatment results in three fractions: LXP-cellulose (LXC), LXP-minerals (LXM), and LXP-lignin (LXL). LXL or untreated Indulin (IND) is depolymerized. Depolymerized lignin is then separated into a liquid fraction (IND-DP or LXL-DP) and a solid fraction, or it is fractionated into lignin monomers (INDmon or LXmon), lignin oligomers (INDolig or LXolig), water-soluble residues (INDr or LXr), and coke (INDc or LXC)



be separated for lignocellulose sugar production [5, 6, 9] or reprocessed within the biogas plant to increase the total methane yield [7]. Even though the number of studies dealing with digestate as biorefinery feedstock is limited [4], all findings and concepts for lignocellulose biorefineries can be used regarding comparable biomass composition.

Lignin with its phenolic chemical structure ensures the ability to replace phenol in the synthesis of phenol formaldehyde resin (PF). PF resins belong to a major class of condensation adhesives and are widely used as binders in the wood panel industry, mostly for the production of plywood, oriented strand board, hardboard, or glulam. These phenolic resins have excellent properties, such as high bonding strength and good resistance against moisture and temperature [10]. Because of an increasing ecological awareness, there is a need for renewable resources, such as lignin, to replace parts of the fossil components. Many studies have been published on this topic, focusing on incorporation of different types of lignin in the synthesis of lignin-based phenolic resins (LPF), including lignin after enzymatic hydrolysis, lignosulfonates, organosolv lignin, kraft lignin, and soda lignin [11]. During this intensive research endeavor, various chemical modifications have been developed to improve the reactivity of lignin toward formaldehyde, such as methylation, phenolation, or sulfonation. However, the most effective way to enhance the reactivity of lignin was achieved with thermochemical pretreatments such as depolymerization of lignin, as these processes produce degraded lignin with smaller molecular weights and more reactive sites [12, 13]. The first hybrid products with phenol content up to 50% are already available on the market (for example, by Prefere Resins (Hamina, Finland)¹;

therefore, a comparable substitution percentage was chosen for our experiments.

Within this study, we used H₃PO₄ biomass decomposition as patented by LXP Group and described in U.S. Patent No. 9969760 and No. 10144677 [14, 15]. In addition to the carbohydrate valorization reported by Schroedter et al. [5], this study focused on the possible application of separated lignin from the LXP process. In detail, the substitution of phenol in lignin-based phenol formaldehyde adhesives was investigated. As crude LXP-lignin did not show satisfactory results, we used base-catalyzed lignin depolymerization to increase the reactivity. A basic flowchart of the investigated process is presented in Fig. 1. As we reproduced the whole process in lab scale (pretreatment, depolymerization, resin preparation, and resin characterization), we were able to identify synergies and cost-saving potentials between the processes. Therefore, we investigated the application of crude lignin oligomer solution after base-catalyzed depolymerization and observed comparable performance but better processability.

2 Material and methods

2.1 Raw material and pretreatment

Digestate from anaerobic digestion of mainly maize silage was used as raw material (RM). The RM was collected from the biogas plant in Ketzin (Brandenburg/Germany) in summer of 2019. Immediately after sampling, RM was dried at room temperature until a dry matter content of at least 80%. The composition and dry matter content of RM was analyzed prior to pretreatment. The material was analyzed according to the method NREL/TP-510-42,618 (2012) [16].

RM was pretreated with phosphoric acid as described elsewhere [5] resulting in three separated fractions: one

¹ <https://prefere.com/en/company/news/biopolymers-put-to-industrial-use-at-phenolic-resin-producer-prefere-resins> (accessed Feb. 16 2023).

fraction called LXP-cellulose (LXC) containing mainly cellulose, hemicellulose, and some lignin, one containing mainly lignin and minor amounts of carbohydrates and minerals called LXP-lignin (LXL), and the third fraction containing minerals and the residual lignin and carbohydrates called LXP-minerals (LXM) (see Fig. 1). The raw lignin resulting from the LXP process was washed with water until the eluent has reached a pH of about 5 indicating the phosphoric acid levels to have dropped sufficiently to obtain the LXL as used in this study. After soft drying at 45 °C, LXL is fawn brown and can easily be ground. Just like the RM, LXL was analyzed according to the method NREL/TP-510-42,618 (2012) [16].

2.2 Lignin depolymerization

All lignin depolymerizations were performed in a Parr 4560 stainless steel autoclave. To be able to better assess the performance of the LXL, all depolymerization experiments were performed with LXL and model kraft lignin Indulin (IND). For each experiment, the lignin (IND or LXL) was dissolved in a sodium hydroxide solution (5 wt% NaOH) at a concentration of 10 wt% and put into an autoclave (PARR 4871, Parr Instrument (Deutschland) GmbH, Frankfurt a. M., Germany). The reaction mixture was heated to 250 °C for 30 min at a stirring rate of 300 rpm. The reactor was cooled to 25 °C. Subsequent treatment of the product mixture was different, depending on the final product. In case of the depolymerized lignin product (IND-DP or LXL-DP), the resulting mixture was filtered through a suction filter to remove residues of coke. To obtain the lignin oligomers (INDolig or LXolig), each product mixture from the lignin depolymerization was subjected to a fractionation process. The first step was acidification with HCl (37 wt%) to a pH of 2. The resulting solids—lignin oligomers and coke—were then separated from the suspension by a suction filter. The filtrate was extracted three times with ethyl acetate and subsequently dried by rotary evaporation and lyophilization, resulting in a fraction of lignin monomers (INDmon or LXmon). Similarly, the aqueous phase was dried to obtain a fraction of water-soluble residue (INDr or LXr). Tetrahydrofuran (THF) was used to dissolve the lignin oligomers and separate them from coke by a suction filter. The filtrate was dried again by rotary evaporation and lyophilization to obtain a dry fraction of lignin oligomers (INDolig_s or LXolig_s). Rotary evaporation was performed at 40 °C (Rotavapor® R-100, BÜCHI Labortechnik AG, Flawil, Switzerland) and lyophilization at −80 °C (ALPHA 1-2 LDplus, Martin Christ Gefriertrocknungsanlagen GmbH, Osterode am Harz, Germany). A drying cabinet (Binder Series E Classic.Line E028, Binder GmbH, Tuttlingen, Germany) was used to desiccate the coke at 105 °C (INDc or LXC). The yields of the obtained product fractions were

determined gravimetrically, referring the weight of each product fraction to the initial weight of lignin.

2.3 Lignin and depolymerization product analysis

Gel permeation chromatography (GPC) was investigated to analyze lignins (IND and LXL) and the depolymerization products (IND-DP, LXL-DP, INDolig, LXolig, INDmon, and LXmon). High-performance liquid chromatography was performed using a DIONEX ICS-5000+ (Thermo Fisher Scientific wissenschaftliche Geräte GmbH, Wien, Austria) equipped with a PSS MCX analytical 100 A + 1000 A + 100,000 A column (8 mm × 300 mm) (Thermo Fisher Scientific wissenschaftliche Geräte GmbH, Wien, Austria). The eluent was a sodium hydroxide solution (0.1 mol L⁻¹ NaOH). During analysis, the flow rate was adjusted to 0.5 mL min⁻¹ at a temperature of 30 °C. Polystyrene sulfonates with molecular weights of 891–976,000 g mol⁻¹ and vanillin were used to calibrate the system.

According to de Sousa et al. [17], the concentrations of phenolic hydroxyl groups in lignin (IND and LXL) and the depolymerization products (INDolig, LXolig, INDmon and LXmon) were determined with Folin & Ciocalteu's phenol reagent.

2.4 Resin preparation

To investigate the performance of the LXL in lignin-based phenol formaldehyde adhesives, resins were manufactured with 50% phenol substitution from four different lignin sources: LXL, LXolig, INDolig predried as powder, and IND-DP. The synthesis was conducted—according to a recipe by Metadynea (Krems, Austria) in a round flask equipped with an electronic temperature controller.

In the case of powdered lignins (LXL and LXolig), the lignins were slowly dispersed in a mixture of NaOH solution and phenol until a homogeneous dispersion was achieved and further processed into a resin in a multistage condensation process with formaldehyde. For the use of dissolved hydrothermal depolymerized lignin oligomers without predrying, the liquid lignin (IND-DP and LXL-DP) was used directly in the synthesis, whereby the time-consuming incorporation step became unnecessary. The subsequent resin condensation was conducted in a multistage process equally to the powder lignin-based resin. In addition to the LPF resins with Indulin AT, a phenol–formaldehyde adhesive was also produced in order to have an industry-oriented comparison.

2.5 Resin characterization

The resin was characterized by analyzing properties such as viscosity (after synthesis and after one week), pH, solid content, B-time, and free-formaldehyde content.

Table 1 Composition of the raw material (RM), cellulose (LXC), and lignin output of LXP process (LXL)

Composition		RM		LXC		LXL	
		Value	sd	Value	sd	Value	sd
Cellulose	%ODW	29.2%	0.1	36.2%	0.0	0.5%	0.1
Hemicellulose	%ODW	22.4%	0.2	8.5%	0.0	2.4%	0.4
Sum of carbohydrates	%ODW	51.6%	0.2	44.7%	0.1	2.9%	0.3
Acid-soluble lignin	%ODW	5.2%	0.5	2.2%	0.1	4.1%	0.4
Acid-insoluble lignin	%ODW	28.3%	0.2	36.7%	0.1	88.2%	0.9
Sum of lignin	%ODW	33.4%	0.3	38.9%	0.1	92.3%	0.5
Ash	%ODW	8.6%	0.2	8.6%	0.1	1.4%	0.6
Acetic acid	%ODW	2.2%	0.2	1.6%	0.0	1.0%	0.2
Lactic acid	%ODW	0.0%	0.0	0.0%	0.0	0.0%	0.0
5-HMF	%ODW	0.2%	0.0	0.3%	0.0	0.0%	0.0
Furfural	%ODW	1.5%	0.1	0.5%	0.0	0.2%	0.0
Fats/oils	%ODW	n.d	–	n.d	–	n.d	–
Protein	%ODW	n.d	–	n.d	–	n.d	–
Sum others	%ODW	2.5%	–	5.4%	–	2.3%	–
Sum of identified	%ODW	97.5%	–	94.6%	–	97.7%	–

The viscosity of the resins was determined at 20 °C using a cone plate system with a diameter of 50 mm and a cone angle of 1° on an MCR 302 rheometer (Anton Paar GmbH, Graz, Austria). pH was measured of a 1:1 resin to water mixture with a TitroLine® 6000/7000 pH meter (Xylem Analytics Germany Sales GmbH & Co. KG, Weilheim, Germany). Solid content was determined according to EN 827 (2006) [18] using a ventilated oven at a temperature of 135 °C for 2 h. Free-formaldehyde content was obtained with back titration (TitroLine 6000/7000; SI Analytics, Mainz, Germany) according to ISO 11402 (2004) [19].

The time required to reach the B-stage of phenolic resin curing (B-time) was measured at 130 °C, according to ISO 8987 (2006) [20] standard using a self-constructed equipment. The setup consists of a heated aluminum plate with a round depression (depth: 5 mm, diameter: 25 mm) and a temperature sensor for precise temperature control.

2.6 Development of bonding strength

Additional to the physicochemical properties, the development of the bonding strength as a function of hot-pressing time was evaluated with an automated bonding evaluation system (ABES) (Adhesive Evaluation System, Corvallis, USA). This specific setup was selected in order to allow testing according to ASTM D7998 (2015) [21] as originally described in U.S. Pat. No. 5176028 [22]. After hot pressing for 30 s to 10 min (0.5, 1, 2, 3, 5, and 10 min) at 120 °C, the tensile shear strength of the specimens was tested with a crosshead speed of 60 mm/min.

2.7 Longitudinal tensile shear strength of plywood

In order to investigate the behavior of LXL-DP in comparison to IND-DP for plywood application, nine-layer (500 × 500 mm) birch plywood panels were made from veneers 1.5 mm thick. The resins were mixed with a hardener, consisting of rye flour, chalk, and water for 2 h to reach a target viscosity of 120 s tested with a DIN 4 cup. The adhesive was then applied with a spread rate of 155 g/m² by roller, and each veneer was weighed to control the actual adhesive amount. The assembly time consisted of a 3 to 4 min open time, followed by 4 + 4 + 1 min hot-press at a pressure of 150/60/30 N/cm² at 125 °C. After pressing, the boards were preconditioned at standard climate 20 °C and 65% r.h. at least two weeks between cutting and testing.

To investigate the bonding quality of veneer plywood, shear testing occurred after sample pretreatment to evaluate plywood in class 3 (non-covered exterior) according to EN 314–1 (2005) and EN 314–2 (1997) [23, 24]. The pretreatment is conducted as follows: samples were immersed for 72 h in boiling water, followed by cooling in water at 20 °C for at least 1 h.

3 Results and discussion

3.1 Material characterization

The composition of RM, LXC, and LXL is presented in Table 1. The raw material is composed of 51.6% carbohydrates, 33.4% lignin, and <9% ash. The high amount of

Table 2 Mass distribution after fractionation of depolymerization products

Product fraction	Yield (%)	
	IND	LXL
Oligomers	82.9	63.9
Monomers	9.0	8.7
Water-soluble residues	3.7	16.7
Coke	0.5	5.4

carbohydrates in RM reveals the potential of digestate for further valorization. The lignin fraction produced by the LXP process consists of more than 90% of lignin and has only a small proportion of carbohydrates, with only 2.4% hemicellulose and 0.5% cellulose. This composition makes LXL a promising candidate for diverse lignin applications. In contrast, the LXC fraction contains 45% carbohydrates and a fairly high lignin content of 39%. The inorganic fraction, as well as the fraction of other substances such as acetic acid, is found in larger quantities in LXC than in LXL.

3.2 Characterization of products after depolymerization

The yields of the depolymerization products—oligomers, monomers, water-soluble residues, and coke—obtained from lignin (IND and LXL) are listed in Table 2.

Depolymerization of Indulin AT was optimized to maximize the amount of oligomers and monomers which can be subsequently valorized. The yield of the oligomers from LXL was 19% lower compared to that from Indulin AT. On the other hand, the yield of the side products—water-soluble residue and coke—was 18% higher. These differences might be explained by the fact that due to the lack of proper amounts of LXL the performed depolymerization process was optimized for Indulin AT. To increase the yield

of oligomers obtained from LXL, further optimization of the depolymerization parameters needs to be done. The water-soluble residues and cokes were no products of interest in this study and therefore have not been further analyzed.

The characteristic values of the molecular weight distributions as well as the phenolic hydroxyl group concentrations of lignin (IND and LXL) and the depolymerization products (INDolig, LXolig, INDmon, LXmon, IND-DP, and LXL-DP) are listed in Table 3.

In each lignin depolymerization, the obtained products had a lower molecular weight and PDI compared to the initial lignin. These results confirmed the performance of the depolymerizations, because in the course of lignin depolymerization, covalent bonds within the lignin macromolecule were cleaved, resulting in smaller molecules with a lower molecular weight. By comparison of each depolymerized lignin solution (IND-DP and LXL-DP) with the separated depolymerization products—oligomers and monomers (INDolig, INDmon, LXolig, and LXmon)—the molecular weight of each depolymerized lignin solution is between that of the monomers and oligomers. This was due to the fact that the solution of depolymerized lignin contained both oligomers and monomers. The low molecular weights of the obtained depolymerization products result in reduced steric hindrance within the lignin molecules which is favorable for the production of LPF resins [25].

By comparison of each lignin with the corresponding depolymerization products, it was found that the concentration of phenolic hydroxyl groups was about 40% higher in the obtained oligomers and 170% higher in the obtained monomers. This was due to the fact that during lignin depolymerization mostly β -O-4 ether bonds within the lignin structure were cleaved resulting in phenolic hydroxyl groups. The high concentrations of phenolic hydroxyl groups in the depolymerization products are advantageous for the synthesis of LPF resins because more positions on the lignin molecules are available for the reaction with formaldehyde [26].

Table 3 Number average molecular weight (M_n), weight average molecular weight (M_w), polydispersity index (PDI), and phenolic hydroxyl groups of different lignin fractions

	M_n (Da)		M_w (Da)		PDI		Phenolic hydroxyl groups (mmol g ⁻¹)	
	Value	sd	Value	sd	Value	sd	Value	sd
IND	1240	21	4950	10	3.99	0.06	3.16	0.04
INDolig	1102	14	2102	14	1.91	0.02	4.38	0.09
INDmon	328	3	525	2	1.60	0.01	8.65	0.06
IND-DP	857	19	2086	27	2.44	0.02	–	–
LXL	1266	5	6163	136	4.87	0.12	2.11	0.07
LXolig	1236	36	4609	30	3.52	0.03	2.97	0.05
LXmon	372	1	604	4	1.62	0.01	5.73	0.04
LXL-DP	1032	11	3838	23	3.72	0.03	–	–

Table 4 Resin properties of LPF resins using lignin with various treatments as phenol substituent

		LPF (LXolig)	LPF (LXL-DP)	LPF (INDolig)	LPF (IND-DP)
Solid content		43.3%	45.7%	42.8%	45.2%
Viscosity after synthesis	mPa*s	410	350	525	350
Viscosity after one week	mPa*s	490	1000	899	520
B-time (140 °C)	min:s	1:11	1:15	1:05	1:30
Free-formaldehyde content		0.5%	1.8%	0.2%	0.2%

3.3 LPF resin characterization

In an initial screening, four lignin-based phenolic adhesives with 50% phenol substitution, varying in lignin type and lignin pretreatment, were produced and its properties characterized. The results presented in this chapter are intended exclusively to compare the behavior of lignin sources in LPF resins and do not represent properties of optimized adhesives, ready for implementation.

During the resin synthesis, the first differences in behavior could be observed, while the lignin was added to the mixture. Powdered lignin can only be added very slowly in small amounts at a time to avoid the formation of agglomerations. During this procedure, the viscosity increases constantly, why the stirring speed of the magnetic stirrer must be adjusted. The intensity of this increase depends on the molar size of the material. LXL therefore shows a significantly higher increase due to its higher molecular weight ($M_w = 6163$ Da) than INDolig ($M_w = 2086$ Da), which was also added as powder. Using liquid lignin dispersions represents an easier and faster handling during resin synthesis and nevertheless saves the drying step after lignin pretreatment. This method allows for cost savings during the production of the adhesive in several steps.

Tables 4 shows that the viscosity after synthesis is for all resins between 350 and 525 mPa·s. These values depend on how fast and effective the resin condensation is stopped by cooling to prevent further crosslinking. After one week of storage, the viscosity was measured again and shows that the resins based on depolymerized lignins exhibit a higher increase than LXL, which suggests a higher binder reactivity. The very high value of LXL-DP (1000 mPa·s after one week) could also be explained with the very high value of free formaldehyde (1.8%), as the curing and crosslinking of phenolic resins correlate to the amount of free formaldehyde [27]. However, the general objective of a depolymerization step is the reduction of complexities of lignin molecules and increasing its chemical reactivity. With this investigation, a successful improvement could be proven and is well in line with findings of Solt et al. [13]. Observing the increase of LPF with powdered compared to dissolved INDolig, a stronger increase can be observed with powdered. This could be attributed to a possible swelling of the lignin.

Interestingly, the B-time of the resins shows similar gelation regardless of the different lignin types, pretreatments, and tested resin properties.

3.3.1 Mechanical properties of LPF resins

Figure 2 illustrates the tensile shear strength as a function of press time of all lignin-based resins with LXL and IND, with and without pretreatment and with difference consistencies. For comparison, already published shear strength values of phenol formaldehyde (PF) were added to the graph [27]. The PF reference shown here reaches a maximum strength of 6.4 N/mm² after a pressing time of 5 min, which can be seen as a leveling of the curve. The veneer specimens reached 100% wood failure, which suggests that the adhesion or adhesives exceed wood strength. In comparison, the bonding strength development of adhesives with 50% phenol substitution by lignin (of any lignin type and pretreatment) is much slower to the PF-reference resin. The slowest curing strength formation was recorded by the LXL, which reached an average strength of 2.4 N/mm² after 10 min hot pressing. The slope suggests that the final strength was not yet reached after 10 min.

With the same resin formulation, IND-DP as well as INDolig was investigated in the second step. The depolymerization reduces the M_w of the lignin and introduces more free reactive groups (amount of aromatic and aliphatic OH-groups), which are necessary for the

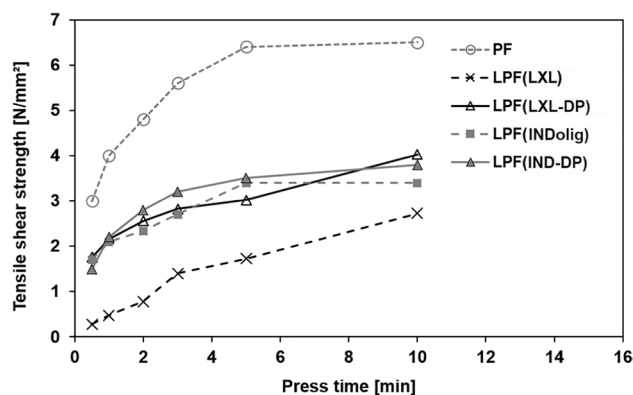
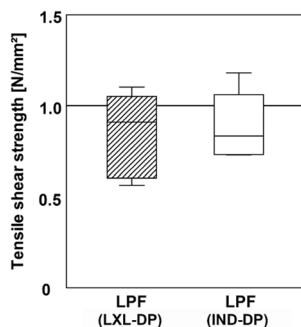


Fig. 2 Average tensile shear strength of PF and LPF bonded lap-shear samples measured after different press times

Fig. 3 Tensile shear strength of LPF resin-bonded plywoods immersed for 72 h in boiling water. The line at 1.0 N/mm² indicates the EN-314-2 (1997) standard requirements for plywood class 3 (non-covered exterior)



condensation reaction with the formaldehyde. The results show that the depolymerization resulted in a “doubling” of the strength development. The binder readily forms bonding performance immediately after contact with temperature (after 30 s 1.5–1.8 N/mm²) and increases to a final strength of ≥ 4.0 N/mm² within the 10 min pressing time. It is interesting to note that due to depolymerization LPF (LXL-DP) achieves a similar bond strength development as adhesive based on depolymerized industrial Indulin AT (LPF (IND-DP)). References describing the positive effect of lignin depolymerization in LPF resin formulations for bonding wood can be found; however, the specimens show in most cases a reduced strength compared to the PF reference [12, 13, 28]. As LXL-DP was used for the first time in LPF resins, improvements in resin quality can be assumed due to optimizations in the binder formulation. Another noteworthy point is that the consistency of the lignin (powder or liquid) during adhesive cooking has no influence on the resulting mechanical adhesive properties. This finding therefore speaks in favor of the use of dissolved/dispersed lignin in order to save energy and time-consuming drying.

In addition to the bonding evaluation, the mechanical properties of LXL-DP and IND-DP were evaluated in a nine-layer plywood board. Testing was performed exclusively according to plywood class 3 (non-covered exterior). Both adhesive types achieved similar average strength values (0.85 N/mm² for LXL-DP and 0.89 N/mm² IND-DR) with no apparent cohesive wood failure. Standard requirements for bonding class 3 could almost be achieved. Our results indicate—as the strength development results do—that thermochemical depolymerized LXL lignin leads to a comparable bond strength as the industrially produced Indulin AT in LPF resins (see Fig. 3). Both processes in combination (LXP process and depolymerization) thus make it possible to create a reactive, high-performance lignin out of lignocellulosic residues that can be successfully incorporated into phenolic resins. Of course, there is potential for optimization, especially in resin production,

to ensure good final strength of the plywood, but this example serves to illustrate a successful application.

4 Conclusions

As a strengthening of the biomethane production is assumed in the upcoming decade, appropriate digestate management as well as by-product valorization will be necessary to maximize the economic as well as the ecological performance of this technology. Disintegration of digestate with H₃PO₄ offers the possibility to maximize the methane yield or valorize plant sugars in biorefinery systems while separating parts of the lignin for further processing. We successfully demonstrated the application of separated lignin after H₃PO₄ disintegration from digestate as phenol substitute in lignin-based resins. Plywoods prepared with an un-optimized formulation almost reached standard requirements for plywood class 3. This type of lignin has never been used before in LPF resins, and therefore, further optimization of the recipe is necessary to maximize the performance of the lignin-based phenol formaldehyde adhesive. Synergies of base-catalyzed lignin depolymerization and LPF resin preparation were identified. Dissolved hydrothermal depolymerized lignin was directly used for resin preparation; no further product purification was needed.

Author contribution Leitner Viktoria: conceptualization, writing—original draft, visualization, project administration, funding acquisition. Aufischer Gottfried: methodology, investigation, resources, writing—original draft, visualization. Solt-Rindler Pia: methodology, investigation, resources, writing—original draft, visualization. Streffer Friedrich: supervision, writing—review and editing. Gabler Christoph: supervision, writing—review and editing, investigation. Noack Jakob: investigation, writing—original draft. van Herwijnen Hendrikus: supervision, writing—review and editing. Christian Paulik: supervision, writing—review and editing.

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Data availability Supplementary material if needed can be requested via the e-mail address of the corresponding author.

Declarations

Ethics approval Not applicable.

Competing interests LXP Group GmbH and Metadynea Austria GmbH are strategic research partners of Kompetenzzentrum Holz GmbH. This work was co-funded by Metadynea Austria GmbH. Laboratory experiments were performed by employees of Kompetenzzentrum Holz GmbH following principles of good scientific practice. The results were not influenced by the funding.

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