


Odor-free kraft lignin-based thermoset with remarkable mechanical properties

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Future materials should be made from renewable resources and be sustainable without compromising the mechanical properties compared to conventional products. Kraft lignin is an available renewable raw material, sourced globally as a by-product from paper pulp production, and currently burnt at a low value. Kraft lignin has been converted into thermoplastics, however the mechanical properties worsen by degree of blending. Thermosets containing kraft lignin give materials with high strength, where the lignin matrix contributes to the mechanical properties. However, pre-fractionation or multistep chemistries have been applied to give high performance materials. Herein, we have combined kraft lignin with bio-based glycerol 1,3-diglycidyl ether to give a resin with enhanced mechanical properties. This resin – LigniSet® – is odorless, which is a unique property for kraft lignin-based products. The resin is, due to its hydrophilicity, compatible with natural fibers to give strong composite materials. The material can be recycled to give new materials without reduction in performance. Life cycle assessment shows that transformation of lignin to materials instead of burning shows significant benefits with respect to environmental sustainability.

Introduction

Even though kraft pulping is focused on generating a strong cellulose-rich fiber, the process also yields crude tall oil and heat and power.¹ The recovery boiler regenerates the process chemicals by a formal reduction where sulfates are transformed into sulfides, and this process also gives heat and power that makes the pulp mill self-sufficient in energy (Fig. 1).² Most pulp mills generate a surplus of energy that sometimes is used to heat up a village in the vicinity of the pulp mill. The heat and power production in the recovery boiler is however not efficient.³ In addition, the recovery boiler is the most capital expenditure (CapEx) intensive unit in a pulp mill: thus, every pulp mill runs with the recovery boiler as the bottleneck. To increase production of pulp or produce a higher quality pulp,⁴

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Lawoko and Berglund have developed epoxy resins that have the potential to replace bisphenol A in certain applications.^{10,16} The chemistry is performed by reacting lignin with epichlorohydrin and then with a polyamine. Tunable properties could be achieved by solvent fractionation of the kraft lignin prior to transformation. Thermosets have also been reported by reacting kraft lignin with adipic acid and an anhydride.¹⁷ These technologies demonstrate the possibility of valorizing kraft lignin. However, most of the technologies suffer from either tedious pre-treatments, such as solvent fractionations, or multistep reactions to increase the reactivity of the lignin. A major showstopper for commercialization is the inherent smell of lignin in the resulting materials. Even when volatiles, such as C1–C4 carboxylic acids, guaiacol and sulfur containing compounds such as mercaptans and alkyl sulfides/sulfoxides, have been removed by different techniques, the smell returns after aging or in the presence of humidity, and this prevents any indoor application.¹⁸

Herein, we report on a thermoset resin with a 50% unmodified kraft lignin content – LigniSet®. The material has remarkable mechanical properties and is completely odor-free after curing (Fig. 2).

Materials and methods

Starting materials

Kraft lignin was obtained *via* the Lignoboost® process from a mixture of softwood and hardwood black liquor supplied by Södra Skogsägarna's Mörrum paper mill. The kraft lignin was received with a dry content of 65%, dried to above 95% dry content in an oven at 60 °C. Diepoxyethers: ethylene glycol diglycidyl ether (EGDE, TCI), glycerol diglycidyl ether (GDE, Nanjung Yolatech), neopentyl glycol diglycidyl ether (NGDE, Sigma-Aldrich), bisphenol A diglycidyl ether (BADE, TCI), epoxidized linseed oil (ELO, Traquisa) were used as received. Resorcinol diglycidyl ether (RDE) was obtained by reacting a solution of resorcinol (20.00 g, 1 eq.) in epichlorohydrin (45.08 g, 4 eq.), stirring at 80 °C and a solution of sodium hydroxide (9.987 g, 2.05 eq.) in water (10 ml) was added over 4 min. The reaction

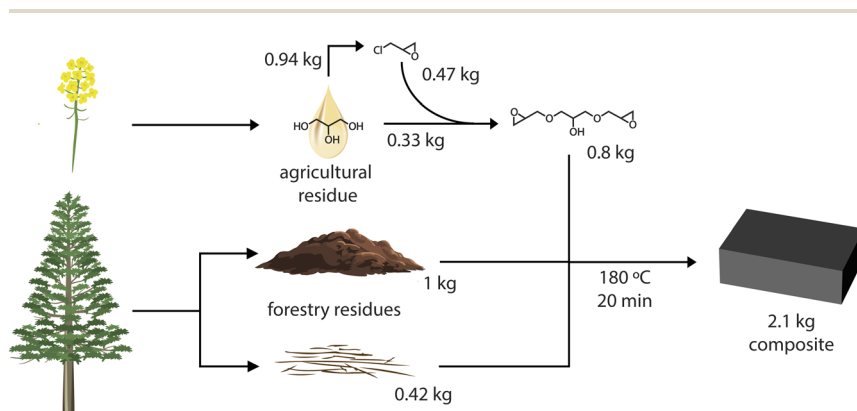


Fig. 2 Residues from agriculture, glycerol from fatty acid methyl ester production, are used to produce glycerol diglycidyl ether, which is combined with the precipitated kraft lignin and wood fibers that generate a composite material that can substitute bisphenol A.



was continued for 80 min. After cooling to room temperature, water (40 ml) and diethyl ether (100 ml) were added. After extraction, the aqueous phase was discarded. The organic phase was washed with aqueous sodium hydroxide solution (50 ml, 10%), water (2×50 ml) and finally dried over sodium sulfate. After filtration, solvent was removed on a rotovapor to afford 33.57 g yellowish oil (83.2%). The product was used without further purification. Catalysts: 2,4,6-tris(dimethylaminomethyl)phenol (TDAP, Sigma-Aldrich), imidazole (Im, Sigma-Aldrich), 4-(dimethylamino)pyridine (DMAP, Sigma-Aldrich), tripotassium phosphate (K_3PO_4 , Sigma-Aldrich) and potassium hydroxide (KOH, Sigma-Aldrich) were used as received. Bentonite clay was obtained from cat litter purchased at a local retail store, which was milled (Retsch GM 300, 4000 rpm, 1 min), fractionated through a 250 μ m sieve and dried in an oven at 100 °C for 3 h. Fibers: recycled cellulose (iCell) were milled (Retsch GM 200, 10 000 rpm, 20 s) before use. Wood fibers (Hunton) were used as received. Bamboo fibers were provided by NPSP.

General procedure for resin preparation

Diepoxyether, catalyst (when used) and dry kraft lignin amounting to a total of 20 g were manually mixed, first at room temperature and then at 140 °C for 5–6 minutes. The resulting dough was then portioned into 5 pieces and placed in a Teflon mold, where each rectangular slot has the following dimensions: 43 \times 17 \times 3.2 mm. The curing took place in a home-built hot press consisting of two aluminum blocks equipped with 700 W heating elements.

General procedure for composite preparation

Diepoxyether, catalyst and dry kraft lignin amounting to a total of 40 g were manually mixed, first at room temperature and then at 140 °C for 5–6 minutes. The resulting dough was loaded into a measuring mixer 50 EHT equipped with Banbury blades and mounted on a MetaStation 4E (Brabender). The mixing was carried out at a speed of 100 rpm and a set temperature of 90 °C, until the internal temperature reached 90 °C. The resulting composites doughs were placed into Teflon molds and cured as previously described.

Flexural properties

Flexural properties were evaluated with a 3-point bending setup using a Sauter TVS 5000N240 test stand equipped with a FH500 500N cell, LD length measuring device and a 3-point adapter AD9315 (30 mm spacing). The measurements were carried out at a speed of 2 mm min^{-1} on 5 specimens per sample.

Composite preparation

GDE, TDAP and dry kraft lignin were manually mixed initially, followed by mixing in a Maxima planetary mixer at room temperature. After uniform mixing of the ingredients for 10 minutes, fibres are added to this dough and mixed for another 10 minutes for uniform mixing. This dough is then taken out and pressed in a hot press using compression molding technique at 150 °C (Fig. 2).



Mechanical properties

Flexural properties were evaluated using a 3-point bending method using an Instron 50kN tensile testing machine. The testing is carried out according to ISO 14125. Impact strength was evaluated using a 1J hammer impact tester using ISO 179 testing standards.

End of life

The End-of-Life (EOL) research is carried out by mechanical recycling of the plates produced during compression moulding. The plates are shredded and milled to particle size less than 100 microns. These particles are then used as fillers in the newer composites made as shown in Fig. 2.

To demonstrate the EOL of products, they are divided into two categories:

(1) Post industrial waste products: these are products with defects which cannot be sent to the customers, hence considered waste.

(2) Accelerated weathered/aged products: to simulate real life ageing of a product in the real world, products were placed in a QUV chamber with cycles of UV-B rays at 0.89 W m^{-2} and condensation. This causes accelerated weathering of products and 1 month in the chamber can be approximately considered as 2 years in the real-world environment.

These products were shredded and milled to smaller particles less than 100 microns and used as fillers in a new batch of composite preparation, *vide supra*.

Life cycle assessment (LCA)

To evaluate environmental sustainability the ISO 14040/14044 standard was followed, comprising the four phases: (1) goal and scope definition; (2) inventory analysis; (3) impact assessment; (4) interpretation. In this study, an attributional gate-to-grave study including the effect of carbon storage was performed. Ecoinvent 3.11 datasets, cut-off by classification where the polluter pays was used. SimaPro (9.6.0.1) software, Environmental Footprint 3.1 (adapted) impact assessment, and the ILCD handbook was followed for carbon storage calculations.

Results and discussion

The dried lignin was mixed with a range of diepoxyethers in a 2 : 1 ratio at $140 \text{ }^\circ\text{C}$ for 5–6 minutes without catalyst. Ethylene glycol diglycidyl ether (EGDE), glycerol diglycidyl ether (GDE), neopentyl glycol diglycidyl ether (NGDE), resorcinol diglycidyl ether (RDE), bisphenol A diglycidyl ether (BADE) and epoxidized linseed oil (ELO) were compared. The resulting dough was added to a mold and cured at $180 \text{ }^\circ\text{C}$ for 120 minutes. The cured thermoset was taken out from the oven and cooled down before removal from the mold. Flexural strength was evaluated at room temperature (Table 1) and it was found that BADE and EGDE ranged close to 55 MPa whereas GDE gave 46 MPa. Nevertheless, GDE was chosen because glycerol can be sustainably sourced and used both in the preparation of epichlorohydrin¹⁹ and in the production of GDE, and when they are produced at the same plant, toxic epichlorohydrin would be handled as an intermediate.

The ratio of lignin to GDE was evaluated using 2,4,6-tris(dimethylamino-methyl)phenol (TDAP) as a catalyst (0.5 wt%), where an increase in epoxide



Table 1 Comparison of diepoxyethers mixed with kraft lignin in a 1 : 2 ratio at 140 °C: flexural strength after 120 min curing at 180 °C

Type of diepoxyether	Flexural strength (MPa)
EGDE	55 ± 20
BADE	54 ± 1
GDE	46 ± 8
RDE	31 ± 8
NGDE	22 ± 9
ELO	15 ± 3

Table 2 Influence of GDE to lignin ratio: flexural strength after 20 min curing at 180 °C, with TDAP as a catalyst

GDE content (wt%)	Flexural strength (MPa)
33	23 ± 8
40	34 ± 14
50	31 ± 6

content to 40 wt% gave higher flexural strength than 33 wt% (Table 2). However, the increase did not motivate higher loadings of GDE, as GDE is a cost driver in the formulation. Surprisingly and gratifyingly, the thermoset had no detectable odor. Even grinding the material and placing it in a container for a month did not release any detectable odor.

A catalyst screening was performed, including both organic and inorganic bases, at 200 °C. The following catalysts were tested: 2,4,6-tris(dimethylamino-methyl)phenol (TDAP), imidazole (Im), 4-(dimethylamino)pyridine (DMAP), tri-potassium phosphate (K_3PO_4) and potassium hydroxide (KOH). To study the effect of catalysts, the curing time was reduced from 120 min to as little as 10 min (Table 3). The flexural strength was affected by the choice of catalyst. DMAP and TDAP gave flexural strengths above 40 MPa, that is, similar to the results running the curing for 120 min without a catalyst. DMAP reached this performance after only 10 min curing, while TDAP required 30 minutes. Although this proves DMAP

Table 3 Influence of catalyst: flexural strength of a 2 : 1 kraft lignin : EGDE resin cured at 200 °C using different catalysts and curing times

Catalyst type	Curing time (min)	Flexural strength (MPa)
DMAP	10	48 ± 1
TDAP	10	8 ± 4
TDAP	20	22 ± 9
TDAP	30	40 ± 4
Im	20	21 ± 6
K_3PO_4	10	14 ± 6
KOH	10	14 ± 3



Table 4 Addition of bentonite clay as filler: flexural strength of a 2 : 1 kraft lignin : GDE resin cured at 180 °C for 20 min, catalyzed by a mixture of TDAP and K₃PO₄

Clay type	Clay (wt%)	K ₃ PO ₄ (wt%)	Total filler (wt%)	Flexural strength (MPa)
Na-bentonite	9	9	18	37 ± 3
Na-bentonite	20	8	28	46 ± 5
Ca-bentonite	20	8	28	39 ± 3
Ca-bentonite	30	7	37	34 ± 3

is a more effective catalyst, TDAP was chosen because it is an established catalyst for curing epoxides; it has a high boiling point, is not toxic and can easily be bound in the resin. Imidazole performed similarly to TDAP after 20 min curing but, similarly to DMAP, poses toxicity issues. Inexpensive inorganic bases can be used in larger amounts in order to also act as filler material, however they were found to substantially increase the samples' water sensitivity and can only be considered for niche applications. The material was still completely odorless.

The effect of filler material was further assessed through the addition of a mixture of Na- or Ca-bentonite clay and K₃PO₄ to a TDAP-catalyzed resin. The results are summarized in Table 4. Addition of 20 wt% Na-bentonite clay and K₃PO₄ had negligible effects on the flexural strength and gave material with above 40 MPa. Other fillers such as biochar and coffee grounds were also tested and showed no reduction in mechanical properties. Increasing the loading to 30 wt% with Ca-bentonite slightly reduced the mechanical properties compared to 20 wt% loading, showing a wide tolerance of additives to the recipe. A benefit of using bentonite was a visual improvement of the surface properties, which became smoother.

Two types of commercial cellulosic fibers were also tested to produce a composite material with either recycled paper or wood fibers (20 wt%). The results are shown in Table 5 and demonstrate good reinforcement capabilities for cellulosic fibres, which potentially implies good fiber–matrix compatibility.

Production of a composite comprising kraft lignin, GDE and fibers was scaled-up to 1 kg. The mixing was performed in a Maxima planetary mixer. The resulting dough was placed in a mold and cured using compression molding hot press at

Table 5 Fiber reinforcement: flexural strength of a 2 : 1 lignin : GDE resin after 20 min curing at 180 °C, with TDAP as a catalyst. Fiber content is 20%

Fiber type	Flexural strength (MPa)
None	11 ± 6
Recycled cellulose	36 ± 7
Wood fiber	47 ± 3



150 °C for 10 min. The material was cooled before removal from the mold. Two different pressures were tested (70 and 90 bars) and both samples gave similar flexural strength (42 and 42.5 MPa). No odor was detected in the finished product. The material was subjected to enhanced aging tests modelling 2 years of outdoor use in central Europe. This did not affect the mechanical properties either, however, some minor defects on the surface could be detected.

To study the recyclability of the material, the resulting material was shredded and milled to particle size of less than 100 microns (Fig. 3). This part of the material was used as filler (10 wt%) in a new batch of composite made in a similar way. Interestingly, the material showed no loss in mechanical properties, and still no odor was detected. This is very promising for environmental sustainability aspects of the material.

Prompt by the good mechanical properties of the material, the fact that it can be recycled and has no detectable odor, we wanted to benchmark using lignin as LigniSet® instead of burning lignin from an environmental sustainability perspective. If the proposed material was introduced, one candidate that would be substituted is bisphenol based bisphenol A diglycidyl ether (DGEBA).²⁰ DGEBA is one of the most used epoxy resins, and has excellent mechanical properties, for instance flexural strength of around 100 MPa.²¹ However, this compound is banned due to its toxicity. Thus, there are incentives to find alternatives that are not toxic and preferentially environmentally more sustainable. It has been demonstrated in several studies that bisphenols made from lignin containing methoxy groups are non-toxic.^{22,23}

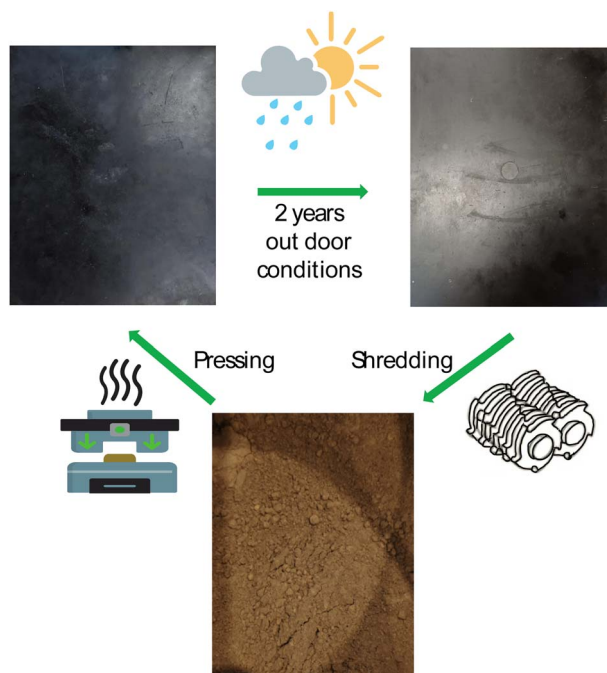


Fig. 3 LigniSet® plates were aged in a QUV chamber for 1 month (equal to 2 years outdoor conditions). The weathered product was shredded to a size less than 100 microns, and then used as a filler in a new batch.



The environmental sustainability of the value chain of producing the composite material was compared to burning the precipitated lignin in a cogeneration plant; an LCA-based study was performed. The Joint Research Centre recommends feedstock based functional units when a non-dedicated feedstock is used in prospective LCA studies.²⁴ This applies to lignin as it is generated as a by-product when pulp is produced. Furthermore, the standard recommends to use a gate-to-grave study when using the same feedstock and producing different products.²⁵ Thus, a gate-to-grave study was performed to study and compare the two scenarios: burning the lignin or valorization to composite material. In addition to study the emissions by these transformations, effect of carbon storage was also included. In our study, we assume that the pulp mill is being debottlenecked, that is the pulp mill removes lignin to increase the pulp production.⁵ The production of the composite comprised 1 kg of lignin, 0.67 kg of GDE, 0.42 kg of wood fiber (Table 6). GDE was not available in the database. In this study, GDE has been manufactured from glycerol in both formation of epichlorohydrin and then GDE, and an inventory was built up using mass balances in a simulated process.²⁶ As the mechanical properties were 47% compared to DGEBA, only the corresponding amount was substituted, in the system expansion, *vide infra*.

To compare the environmental sustainability of incineration of 1 kg of kraft lignin to valorizing the lignin to a composite comprising 1 kg of lignin, the

Table 6 Inventory analysis for production of composite

Process materials and energy	Amount	Unit	Comment
Drying			
<i>Inputs</i>			
Precipitated lignin (66% DS)	1.5	kg	Measured in lab
Electricity	0.5	kW h	Measured in lab
<i>Outputs</i>			
Dry precipitated lignin	1	kg	Measured in lab
Deionized water	0.1	kg	Calculated
Mixing			
<i>Inputs</i>			
Dry precipitated lignin	1	kg	Measured in lab
DGE ^a	0.67	kg	Measured in lab
Wood fiber	0.42	kg	Measured in lab
Catalyst ^b	0.01	kg	Measured in lab
Electricity	0.04	kW h	Measured in lab
<i>Outputs</i>			
Dough	2.1	kg	Measured in lab
Curing			
<i>Inputs</i>			
Dough	2.1	kg	Measured in lab
Electricity	0.16	kW h	Measured in lab
<i>Outputs</i>			
Cured composite product	2.1	kg	Measured in lab

^a An inventory for producing 1 kg epichlorohydrin from glycerol was developed using: 1.4 kg glycerol, 1.07 kg of HCl, 0.58 kg of NaOH; 0.7 kg of epichlorohydrin was reacted with 0.330 kg of glycerol using 0.55 kg of NaOH. ^b EDTA was used as a proxy as TDAP was not available.



comparison is: 1 kg of kraft lignin incineration vs. 2.1 kg of composite (comprising 1 kg of lignin). The carbon content in lignin is 60% (0.6 kg in 1 kg of lignin) and this equals 2.2 kg of CO₂ equiv. In the case of the composite, burdens and benefits of the additional GDE and wood fiber is added and the resulting processing. However, in the carbon storage calculations, only the lignin is included.

Incineration of lignin in a cogeneration plant was modelled according to Marson *et al.*³ where the energy produced (10.62 MJ heat and 1.53 kW h electricity) substitutes the marginal energy mix in Sweden. The benefits of incineration of lignin are marginal, yet negative (−0.044 kg CO₂ equiv.). The reason is mainly that the energy mix in Sweden is relatively green relying on hydro, wind and nuclear power. If incineration was performed in Germany with an energy mix that is heavily dependent on natural gas, the benefit is higher (−0.72 kg CO₂ equiv.). It should be noted that incineration does not give any storage benefits and the stored 2.2 kg of biogenic CO₂ is released immediately.

Valorization of kraft lignin to composite comprises the addition of fiber and DGE as well as the processing and gives an impact of 0.6 kg CO₂ equiv. As in the case of incineration, where the heat and power substitute the energy mix in Sweden or Germany, *vide supra*, the composite material would substitute products on the market. Given the mechanical properties of the composite, we propose that DGEBA could be substituted. It is important to note that DGEBA has a flexural strain of around 100 MPa, and the LigniSet® based composite has a mechanical strength of 47 MPa. Thus, in the system expansion only 47% of 2.1, is used in substitution of DGEBA. Crediting LigniSet® with 0.9 kg of DGEBA gives total impact of an impressive −3.4 kg CO₂ equiv., that is a net-negative effect climate change. We also performed the same calculations substituting acrylonitrile butadiene styrene (ABS), and found a similar score (−3.5 kg CO₂ equiv.). Taking the carbon storage into account according to the ILCD guidelines,²⁷ and the fact that lignin has a high carbon content (60 wt%), the correction flow for delayed emissions would subtract another 0.022 kg CO₂ equiv. per year stored for only the lignin, and as is demonstrated, the resin can be recycled without losing mechanical properties. In addition, the end of life scenario is most likely incineration, where an additional 0.044 kg CO₂ equiv. would be subtracted from the score, in addition to the stored 2.2 kg of biogenic CO₂. Besides climate change, potential burdens in other impact categories: particulate matter, land use, resource use (both fossil and non-fossil) were studied. Besides land use, where the DGE contributed to impact in the case of LigniSet®, there are significant benefits in producing a material of the kraft lignin instead of incineration in the 4 other impact categories (Fig. 4).

The technoeconomics of producing composites from lignin involve both the costs of precipitating the lignin as well as the cost converting the lignin to a marketable product.² The revenues are found in both the increased pulp production, coined “marginal tonnage” as well as product from lignin, in this case LigniSet®. All costs are scale-dependent and in the following example a 50 000 ton lignin precipitation plant is modelled. The total invest cost for the precipitation plant, such as a LignoBoost®, is around 50 MEuro. Using an annuity factor of 0.1 (10 years depreciation), the resulting CapEx is around 100 USD per ton lignin precipitated or 80 USD per ton pulp produced (1 ton of pulp gives 0.8 ton of lignin). The investment cost of mixers and hot presses are estimated to be



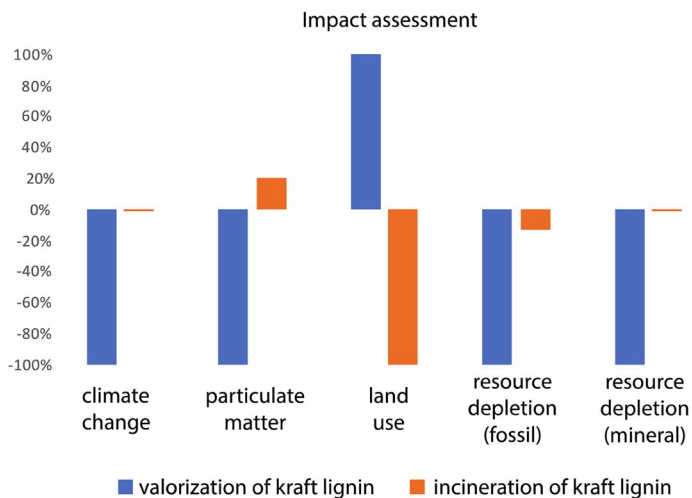


Fig. 4 Impact assessment results comparing valorization of kraft lignin (blue) and incineration (orange).

20 MUSD to convert 50 000 ton of lignin. The total CapEx is thus 120 USD per ton of lignin. Operating expenditure (OpEx) of precipitating lignin is the associated cost of chemicals (CO_2 , H_2SO_4 and NaOH) and has currently been fluctuating between 40–160 MUSD; the costs for manufacturing of the composite is estimated to be 1000 USD per ton of lignin transformed where the DGE is the major cost driver. The OpEx of running both facilities, using a 3 × 3 shift-rotation (three operators 3, 3 × 8 hour shifts) would have a total cost of 800 000 USD per year, and give an additional 16 USD OpEx per ton of transformed lignin. Thus, CapEx and OpEx costs per ton lignin are 1280–1400 USD per ton of transformed lignin, noteworthy, each ton of lignin will generate 2.1 tons of composite material.

The revenue of the marginal tonnage pulp is around 300 USD. The estimated market price for a green bisphenol A substitute is around 2000 USD per ton of composite. Thus, 4200 USD in the case of LigniSet® (1 kg of lignin gives 2.1 kg of composite). Thus, the gross revenue taking both pulp and composite material is 4500 USD. The net revenue is thus 3100–3220 USD per ton of lignin transformed.

Conclusions

Herein we report a procedure to obtain an odorless composite material – LigniSet® – from kraft lignin showing impressive mechanical properties. The material is 100% bio-based and uses residues from forestry (lignin and wood fiber) and agriculture (glycerol). An LCA shows significant environmental benefits, especially in climate change, of transforming the lignin to a material that substitutes either bisphenol based DGEBA or ABS. Technoeconomic assessment shows potential in both revenues from increased pulp production as well as product from lignin. This study demonstrates that lignin outtake is both economically and environmentally sustainable, if thermoset resin LigniSet® is produced and used for the manufacture of composite.



Data availability

The data in this publication is included in the tables and in the text. We don't have any other data or spectra. All data has been measured. For LCA, the inventory is given in the table. The Ecoinvent databases have been used.

Conflicts of interest

JSMS is professor at Stockholm University and founder of LigniCore, a company that has the rights to LigniSet®.

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