

Techno-economic and life cycle analyses of bio-adhesives production from isolated soy protein and kraft lignin

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ABSTRACT

Bioadhesives are promising alternatives to formaldehyde-based adhesives, owing to their excellent low formaldehyde emissions and sustainable raw materials. Among these, lignin-isolate soy protein bioadhesives (LISP), produced from soy protein and lignin, exhibiting significant potential applications across various wood products. This study addresses previously unidentified environmental impacts and economic feasibility of large-scale LISP bioadhesive production. This study evaluates the techno-economics and cradle-to-gate life cycle environmental impacts of a novel LISP bioadhesive production from isolated soy protein and kraft lignin, including the depolymerization of kraft lignin on a commercial scale by optimizing experimental results. Technoeconomic analysis shows that the estimated production cost of LISP bioadhesive produced at 30 MT/batch plant was \$1.32/kg and the minimum selling price was \$1.40/kg. Life cycle assessment shows that the global warming impact of LISP bioadhesive production of was 2.20 kg CO₂ eq/kg. The utilization of steam was the primary contributor to these emissions.

1. Introduction

The fossil-based chemical manufacturing industry is a major contributor to climate change (Supran et al., 2023). The use of fossil fuels totaling 36.1 billion metric tons resulted in record-high global CO₂ emissions in 2022, with current CO₂ levels at 424 ppm (Liu et al., 2023; Stein, 2023). A need for sustainable techniques to produce chemicals has also increased interest to find sustainable alternative feedstock resources to mitigate climate change (Intergovernmental Panel on Climate Change (IPCC), 2022). This can be achieved by producing chemicals from renewable resource such as biomass (Sheldon, 2014; Frihart and Hunt, 2021). Biomass feedstocks are abundant and can be a potential replacement for fossil fuels in the right circumstances (BP, 2022; El, 2023; Spengler and Chen, 2000). To lessen the environmental impact of the emission from fossil-based chemical industry, more biomass-derived materials must be produced (Jakes et al., 2016; Queneau and Han, 2022; Riazi et al., 2019; Batten et al., 2023). Under this goal, researchers have created efforts biomass conversion pathways to target bioproducts such as fuels, plastics, nanocellulose, value-added chemicals and alternative

energy sources (Hasunuma and Kondo, 2017; Kargbo et al., 2021; Mousavi-Avval et al., 2023; Rajendran et al., 2023). However, the transition from the use of petroleum-based feedstocks to biomass-based feedstock for the synthesis of chemicals has been slow due to a myriad of issues including low yield, fluctuating cost, lack of infrastructure and unknown environmental impacts negatively influences (Ahorsu et al., 2018; Huang et al., 2021; Yang et al., 2021).

Fossil-based adhesives are widely used in automotive, wood product manufacturing and general purpose construction (Marques et al., 2020). During its early history, the wood products industry used casein, starch, collage and other bioderived materials for of the manufacture of adhesives (Marques et al., 2020; Frihart and Hunt, 2021). However, development in adhesive technology increased the use of petroleum-based materials (Raydan et al., 2021). In 2020, the global market of adhesives was USD 15.8 billion with expected growth rate of 4 % per year projected to reach USD 21.9 billion in 2028 (Straits Research, 2022). Petroleum-based adhesives have advantages such as low-cost raw materials, rapid curing, high dry bond strength, and a colorless glue line. Currently, 95 % of adhesives used in wood panel industry are made of

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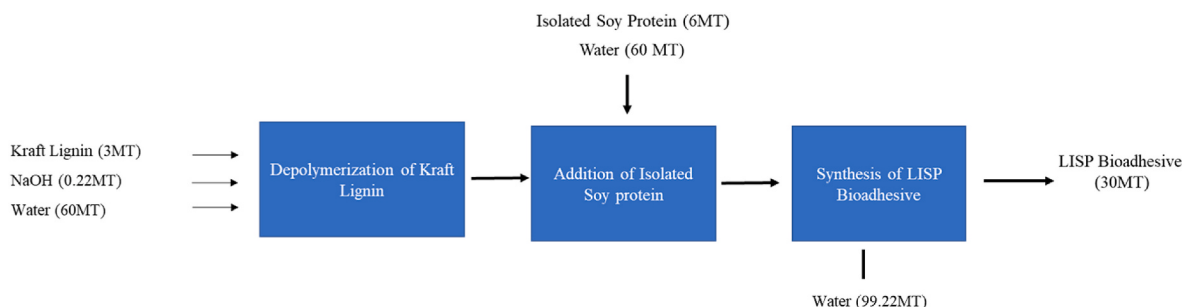


Fig. 1. Simplified overall flowchart of the lignin-isolate soy protein (LISP) production process for 30 MT/batch.

petroleum based products such as phenol-formaldehyde (PF), urea-formaldehyde (UF), melamine-formaldehyde and melamine-urea-formaldehyde (Bush et al., 2022; Carvalho et al., 2012; Mantanis et al., 2018; Raydan et al., 2021; Xu et al., 2020). One of the disadvantages of petroleum adhesives is that they can release volatile organic compounds (VOCs) and formaldehyde vapors, which have toxic and carcinogenic properties vulnerable to human health (Al-Hemoud et al., 2018; Spengler and Chen, 2000; Steinemann et al., 2017). The United States has 2 regulations, 3 product standards and 3 method standards specified for control of formaldehyde emissions (Zhang et al., 2018), which highly encourages the synthesis of formaldehyde-free adhesives.

Previous research focused on the development of soy protein-based adhesives due to the abundant supply, low cost, and safety from formaldehyde emission (Frihart and Birkeland, 2016; Vnućec et al., 2017; Zeng et al., 2021). However, currently these adhesives have limited use due to low bond strength and poor water resistance. To address these issues, researchers have explored using denaturants like urea, sodium dodecyl sulfate, and alkali to improve water resistance during the hot press process. However, these denaturants cannot meet the bond strength requirement for interior use (Luo et al., 2016). Alternatively, cross-linking agents like glycidyl methacrylate, polyamidoamine-epichlorohydrin resin, or polyethylene glycol diacrylate can effectively improve water resistance and meet the bond strength requirement but its wide utilization was limited by high cost (Gao et al., 2019; Luo et al., 2016; Wang et al., 2014). To develop sustainable and acceptable soy protein-based adhesives, they must have good water resistance, low curing temperature and proper viscosity (Kan et al., 2022; Zhang et al., 2022). There have been previous efforts to use lignin, polyethylene oxide, cellulose nanofibrils with isolated soy protein which demonstrate higher water resistance (Podlena et al., 2021).

Lignin, a byproduct of pulp industry particularly in form of kraft lignin, is widely considered a low-value material (Demuner et al., 2019). Conventionally, kraft lignin has been burned as fuel in paper mills to provide energy while recovering the processing chemicals. However, recent reports reported that kraft lignin can be utilized as renewable raw feedstock for chemicals to replace use of petrochemical resources and inherent improvement (Abdelaziz et al., 2020; Pradyawong et al., 2017). During kraft pulping process lignin is depolymerized in lignin fragments with part of it undergoing re-polymerization. The repolymerized lignin showed less reactivity. One of prevalent approach to increase reactivity of kraft lignin is base catalyzed depolymerization (Chio et al., 2019). Depolymerization of lignin produces monomers and oligomers derived from or consisting of these three basic units, p-hydroxyphenyl (H), guaiacyl (G), and syringyl (S). This method has a major advantage by generating a more reactive lignin while producing higher yields (Sun et al., 2018).

While bioadhesives have several advantages over petroleum-based alternatives, there are still challenges to their commercial production, particularly in terms of economic analysis and environmental impact (McDevitt and Grigsby, 2014; Yang and Rosentrater, 2019a). The functional properties of a product are dependent on its distinct chemical

and physical properties, which are important for commercial applications (Ahire and Bhanage, 2021). Shear strength is a key parameter used to evaluate the efficiency of an adhesive in plywood. The Lignin-isolated soy protein bioadhesive (LISP bioadhesive) meets the ASTM 12, 466-2-2007 standard requirement of greater than 1 MPa (Jiang et al., 2023; Soubam et al., 2022). It exhibits comparable performance to commercial soy protein-based adhesives, with a dry bonding strength of 1.46 MPa, compared to 2.07 MPa for the commercial adhesive. The LISP bioadhesive is sufficient to replace synthetic adhesives, as it exceeds standard requirements without using hazardous or expensive raw materials. The synthesized LISP adhesive can be used as a substitute for acrylic and Delrin-based adhesives (MasterBond, 2023).

Technoeconomic analysis (TEA) considers the costs and revenues associated with the technology, as well as the risks and uncertainties. This study is important to identify the most economically viable bioadhesive production technologies, and to make decisions about where to invest resources. Environmental impact was determined through life cycle assessment (LCA), it considers the inputs and outputs of process to determine net emission from the process system. LCA can be used to identify the better environmental performing adhesive production technologies and contribute to make decisions about how to reduce the environmental impacts of adhesive production. The objective of this study was to perform technoeconomic and life cycle assessment to evaluate the economic feasibility through unit production costs, and environmental impacts of novel LISP bioadhesive production at the commercial scale.

A comprehensive process model was formulated to investigate the environmental repercussions and production expenses linked to this process. This model was constructed utilizing information derived from both existing literature and laboratory-scale data published (Jiang et al., 2023). Sensitivity analyses were conducted to determine the most sensitive parameters in terms of the unit production cost (\$/kg). This research supports the development of a tool that helps people to decide whether to produce new bioproducts on a large scale, considering both economic and environmental factors (Wang, 2023).

2. Methods

To obtain comprehensive material and energy balance information for technoeconomic and life cycle assessment, a rigorous process model of the for producing LISP bioadhesive was simulated under SuperPro Designer v12 software (Intelligen, Inc., USA). This state-of-the-art industrial design software facilitates the modeling, evaluation, and optimization of integrated processes across a spectrum of industries. The process model for synthesis of depolymerized lignin and bioadhesive was updated by incorporating modifications to a research article published by coauthor with the resetting of energy and mass balances mentioned in supplementary information (Jiang et al., 2023). Table S5 and Fig. S2 of process model used data of experimental LISP bioadhesive preparation derived from kraft lignin and isolated soy protein in Superpro. ("Intelligen, Inc.," 2023). A cradle-to-gate LCA approach was followed based on the ISO 14040 and 14,044 standards ("ISO 14040,"

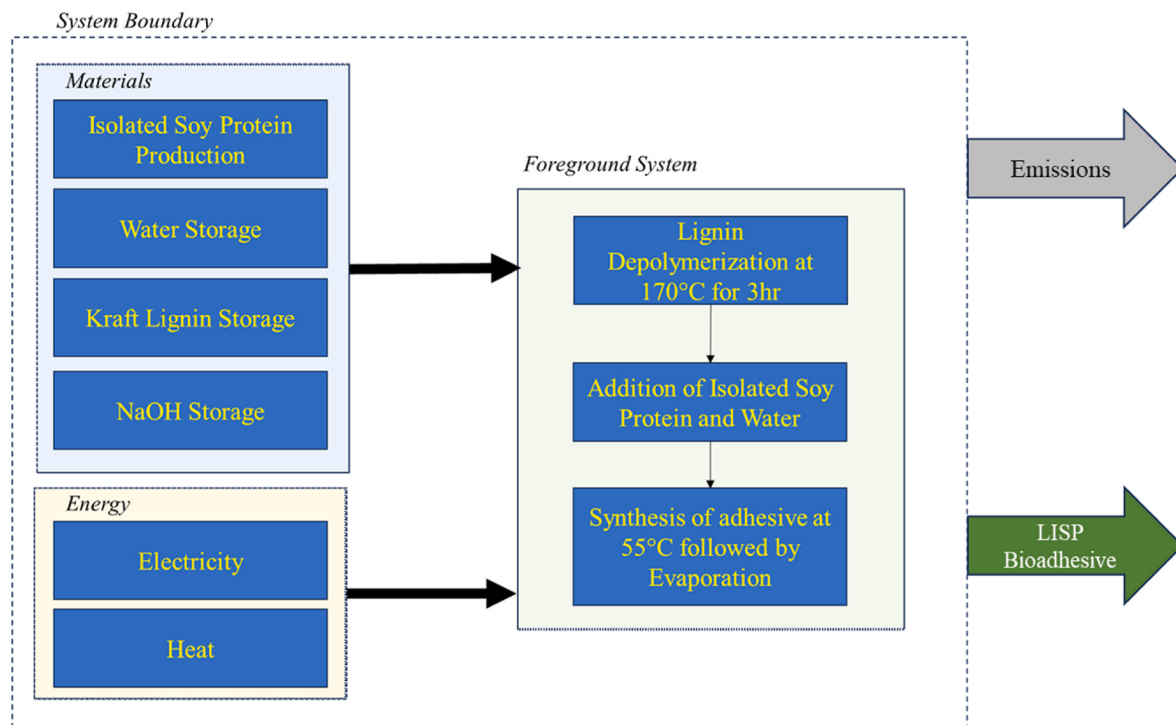


Fig. 2. System boundary for cradle-to-gate production of lignin-isolate soy protein (LISP) bioadhesive.

2014; "ISO 14044," 2014). The LCA analysis was facilitated by SimaPro 9.4 LCA modeling software ("PRé-sustainability," 2023). The LCA performed in this work used US EI and USLCI databases ("DATASmart," 2023).

2.1. Process description

The process for producing LISP bioadhesive from lignin and isolated soy protein involves two steps: synthesis of depolymerized lignin from kraft lignin, and synthesis of bioadhesive from isolated soy protein and depolymerized kraft lignin shown in Figs. 1 and 2. The study adopted a utilization rate of 330 working days per year, which mirrors the operational pattern of real-world industrial processing, operating 24 h per day. Mass and energy balance, process flow compositions and equipment compositions were carried out considering 30 MT/batch production capacity for LISP bioadhesive. It analyzed different scenarios and sensitivity analysis to evaluate economic feasibility considering input stream variations. Data from MASBio collaborative researchers, literature, and technical reports were used to evaluate costs for producing LISP bioadhesive (Jiang et al., 2023).

The following section outlined the process used for depolymerization of kraft lignin, and synthesis of bioadhesive for conducting the technoeconomic analysis and environmental impact study.

2.1.1. Depolymerization of kraft lignin

In the system boundary for the life cycle assessment of kraft lignin, it is assumed that the lignin is produced through a chlorine-free bleached process from southern pine in a mill in the southern United States (NC, 2018). The inventory of kraft lignin for life cycle assessment considered activities necessary to produce through bleached process. Initially, a lignin solution with a pH value greater than 12 is prepared from kraft lignin from storage tank (Fig. 2:P4) with 7.4 wt% NaOH (P3) (mass ratio based on lignin) and water (P1) (twenty times higher based on lignin). Water was added to the lignin and NaOH mixture to prepare a solution. The resulting solution was then mixed in a reactor for efficient depolymerization of kraft lignin. The stoichiometric reactor (P7) is used for depolymerization of kraft lignin. In the system boundary for the life

cycle assessment of kraft lignin, it is assumed that the lignin is produced through a chlorine-free bleached process from southern pine in a mill in the southern United States. (Jiang et al., 2023). Subsequently, the solution was subjected to thermal treatment at 170 °C for a residence time of 3 h. The resulting lignin solution, mainly comprised of lignin oligomers, was used as the base-catalyzed depolymerized lignin fragments, without separating the monomers. These fragments were assumed to have undergone partial degradation and were used for further analysis.

2.1.2. Bioadhesive synthesis

The isolated soy protein refers to protein from the mechanical soybean processing into soybean meal and crude soy oil ("Ecoinvent v3.0," 2020). The emission data for isolated soy protein, was used from soybean meal in DATASmart database (Arias et al., 2020). The solution of depolymerized lignin was prepared by addition of kraft lignin in alkaline water (ratio is 1:20 based on weight, pH > 12). The bioadhesive formulation was developed by mixing of isolate soy protein (ISP) and water in mass ratio (w/w) of 1:10 and the previously obtained depolymerized lignin solutions in volume ratio (v/v) of 2:1 in same reactor (Fig. 2:P7). The mixture was mixed for 1 h and then heated to 55 °C. As reported in prior research, a pre-coupling reaction occurred between soy protein and lignin occurs during the adhesive preparation process (Pradyawong et al., 2017). Multiple-effect evaporator were modeled to eliminate excess water, resulted adhesive solution has concentration of ~9 %. The concentration was controlled to maintain the desired viscosity and flowability of the adhesive, while also reducing volume for logistical concerns.

2.2. Economic analysis

For the TEA, this study used the American Association of Cost Engineers (AACE) method, which has an assumed accuracy range of ±30 % (Dysert and Christensen, 2005). The model calculated of total 33,680 labor hours, consisting hours of four operator and one supervisor would be onsite during the 330 annual operational days with hourly rate of \$33/hr and \$50/hr, respectively for one project. The purchased equipment's salvage value was assumed to be zero (Turton et al., 2008). The

project lifetime was set at 15 years with a construction period of 30 months and startup period of 4 months. The study proposed a structural bio-adhesive production plant in the North America region. The assumptions for total capital cost investment (C_{TC}) are listed in Table S4 in Supporting Information. SuperPro Designer v12 software was employed to estimate the equipment purchase costs; other assumptions were obtained from plant design and economics for chemical engineers (Kobe, 1958). The total capital cost investment (C_{TC}) was determined by adding the direct fixed capital cost (C_{DF}), working capital (C_W), and start-up costs (C_S), as shown in equation (1).

$$(C_{TC}) = C_{DF} + C_W + C_S \quad (1)$$

The annual operating cost (C_{AO}) for the structural bioadhesive production plant was categorized into utility cost (C_U), materials cost (C_M), facilities cost (C_F) and labor cost (C_L), and, with underlying economic assumptions provided in SuperPro Designer database.

$$C_{AO} = C_U + C_M + C_F + C_L \quad (2)$$

The cost of materials was obtained from retailer websites ("MOL-BASE," 2023). Electricity prices were collected from the U.S. Energy Information Administration (EIA) monthly electric power report ("EIA," 2022). Steam and water costs were gathered from the SuperPro Designer v12 software database and previous literature (Zang et al., 2020) ("Pré-sustainability," 2023). However, the costs for wastewater treatment, research and development (R&D), and distribution and marketing were not considered. Total cost investment, annual revenue, and unit production cost were evaluated. The assumptions related to total annual cost are shown in Table S4 in Supporting Information.

The gross profit, net profit, return on investment (ROI), payback time, and unit production cost were calculated using equations (3)–(7) (Yang and Rosentrater, 2019b).

$$\text{Gross profit} = \text{Revenue} - C_{AO} \quad (3)$$

$$\text{Net profit} = \text{Gross profit} - \text{Taxes} + \text{Depreciation} \quad (4)$$

$$\text{Return on investment (ROI)} = \text{Net profit} / \text{Total investment} \times 100 \quad (5)$$

$$\text{Payback time} = \text{Total investment} / \text{Net profit} \quad (6)$$

$$\text{Unit production cost} = \text{Annual total investment} / \text{Annual amount} \quad (7)$$

2.2.1. Uncertainty analysis

The economic factors influence on profitability were assessed using an analysis focused on of cost related to capital investment, lignin and scale of plant size for different scenario and sensitivity analyses was conducted. The base-case scenario considered a plant scale of 30 MT/batch, and four distinct scenario analyses were carried out as follows: 1. Adjusting the total capital investment (C_{TC}) from $\pm 30\%$. 2. Comparing the use of different lignin proportions in the bioadhesive formulation. 3. Examining the effect of kraft lignin cost from various sources on the synthesis of bioadhesive. 4. Compare the effect of different plant scale on cost of bioadhesive. For the sensitivity analysis, parameters such as materials cost and labor cost were selected based on their potential impact on product value. This enabled a comprehensive evaluation of how these factors could influence the profitability of the bio-adhesive production process. The schematic presentation of material and energy flows within the process and system boundary are showed in Figs. 1 and 2. Table S1 shows the inventory data of kraft lignin was collected from previous literature (Supporting Information) (Culbertson et al., 2016).

Table 1

Materials and energy consumption per kg of lignin-isolate soy protein (LISP) bioadhesive production in U.S. and the associated life-cycle inventory unit processes, cradle-to-gate.

Material and Energy	Unit	Quantity	LCI Unit Process
Inputs			
Material			
Kraft lignin	kg	0.1	Kraft lignin (Supporting Information)
Isolate soy protein	kg	0.2	Soybean meal, at oil mill/US-US-EI U (Arias et al., 2020)
Water	kg	4	Tap water, at user/US* US-EI U
Sodium hydroxide	kg	0.09	Sodium hydroxide, production mix, at plant/kg NREL/RNA U
Energy			
Steam	kg	7.21	Steam, for chemical processes, at plant/US-US-EI U
Electricity	kWh	0.001	Electricity, medium voltage, certified electricity, at grid/US* US-EI U
Tap water (Cooling)	kg	0.12	Proxy Water, at user NREL/US U
Outputs (material)			
LISP bioadhesive	kg	1	Final product
Water	kg	3.3	Treatment, plywood production effluent, to wastewater treatment, class 3/US* US-EI U

2.3. Life cycle assessment

2.3.1. Goal and scope definition

The goal of the present study was to quantify the life-cycle environmental impact of LISP bioadhesive production from Kraft lignin and isolated soy protein.

2.3.2. Functional unit

According to the ISO 14040 standard, the functional unit (FU) provides a reference point for inputs and outputs in an LCA study (ISO 14040:2006a). In the present study, 1 kg of LISP bioadhesive production was taken as the FU.

2.3.3. System boundary

A cradle-to-gate approach was adopted and thus all inputs (electricity, chemicals, water) and outputs (LISP bioadhesive and water vapor) were included within the system boundary (Fig. 2).

In life cycle assessment (LCA), four essential sequential steps are executed. Firstly, the goal and scope definition phase set the specific objectives, delineates the system boundaries, establishes the functional unit for comparison, and identifies the relevant life cycle stages under consideration. In the second stage, the life cycle inventory stage, we collect comprehensive data encompassing inputs and outputs associated with each life cycle phase, utilizing primary and secondary sources to construct a comprehensive inventory. Subsequently, the life cycle impact assessment phase employs characterized data and models to quantify potential environmental impacts across multiple impact categories. Lastly, the interpretation stage synthesizes the results, elucidating key contributors to environmental impacts, addressing limitations, and facilitating informed decision-making and sustainable development strategies.

The overall foreground production system is divided into two main stages: depolymerization of lignin and synthesis of adhesives. In the base-catalyzed depolymerization of kraft lignin, alkaline lignin solution is made from kraft lignin with 7.4 wt% NaOH (based on the weight of kraft lignin) and water. Subsequently, the solution was subjected to thermal treatment at 170 °C for a residence time of 3 h to produce the bioadhesive. The life cycle impact of process is evaluated through by comparing to other bioadhesive system in terms of greenhouse gas emissions, adhesion strength, comparative analysis with cost for different proportion of raw materials.

Table 2

Economic analysis results for five commercial-scale production of lignin-isolate soy protein bioadhesive, cradle-to-gate.

Scale (MT/batch)	Total capital investment (\$)	Annual operating cost (\$)	Unit Production Cost (\$/kg)
50	35,906,498	82,935,375	1.19
40	31,947,285	72,220,494	1.29
30	26,612,042	55,841,936	1.32
20	21,725,643	38,725,643	1.39
10	15,282,098	21,902,980	1.57

Life-cycle assessment was carried out using SimaPro v.9.4 to model the process based on material and energy inputs and outputs developed as part of the TEA to ensure proper integration of the TEA and LCA (Mousavi-Avval et al., 2023).

2.3.4. Life cycle impact assessment

The TRACI impact category method with US 2008 normalization was used to estimate the life-cycle environmental impacts (Bare, 2011). The impact categories were ozone depletion (kg chlorofluorocarbons-11-eq), global warming (kg CO₂-eq), smog (kg O₃-eq), acidification (kg SO₂-eq), eutrophication (kg N-eq), carcinogens (CTUh), non-carcinogens (CTUh), respiratory effects (kg PM_{2.5}-eq), ecotoxicity (CTUe), and fossil fuel depletion (MJ surplus).

3. Results and discussion

The economic and environmental impacts of producing LISP adhesive were analyzed and results presented here (see Table 1).

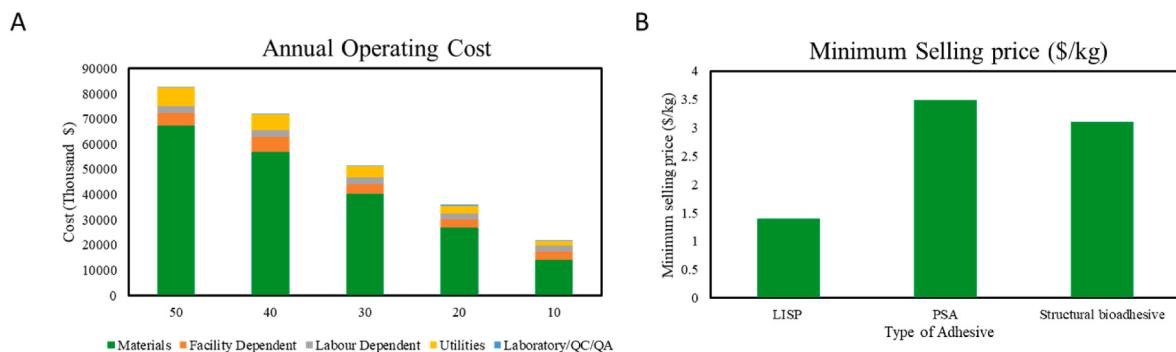


Fig. 3. (A) Annual operating cost for five different plant scale production of lignin-isolate soy protein (LISP) bioadhesive, (B) Minimum selling price of different bioadhesives. PSA: pressure sensitive adhesive; LISP (Lignin -isolated soy protein bioadhesive).

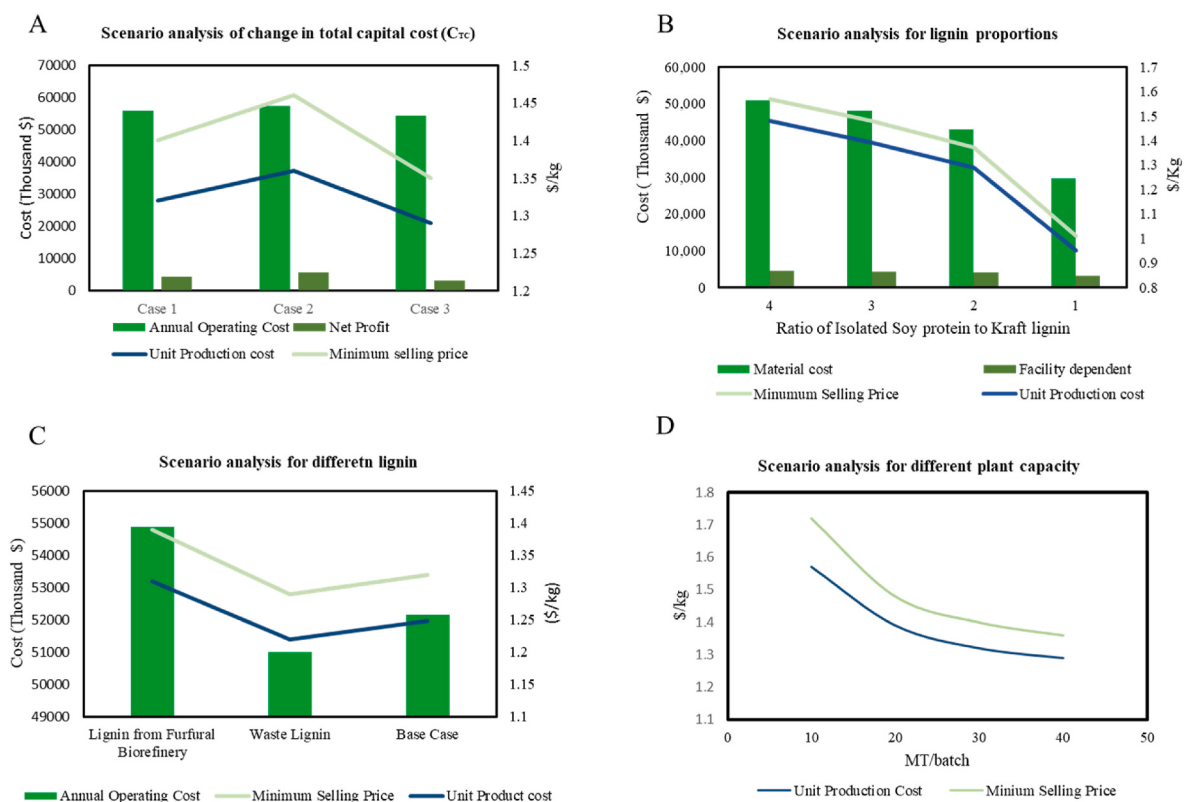


Fig. 4. Effect of unit production cost of lignin-isolate soy protein bioadhesive from changing A: Total capital cost, B: Lignin usage (%), C: Lignin source D: Plant capacity (MT/day).

Sensitivity Analysis

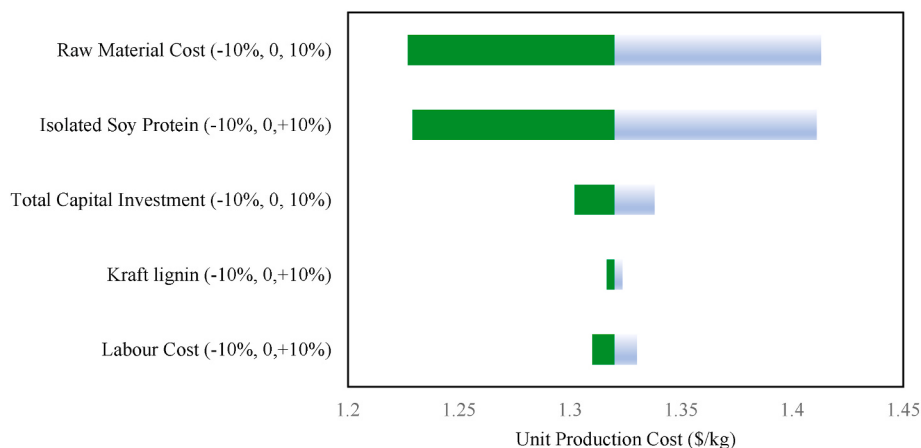


Fig. 5. Unit production cost effect on different parameters for lignin-isolate soy protein bioadhesive production plant, 30 MT/batch.

3.1. Technoeconomic assessment

The economic analysis for different production scales (Table 2), shows that the total capital investment increases from \$15.3 million for the 10 MT/batch to \$35.9 million for the 50 MT/batch plant, with direct fixed costs being highest contribution. The annual operating cost increase dramatically from the smallest plant (\$21.9 million) to the largest plant (\$82.9 million), with materials cost contributing ~80 % and facility dependent cost contributing ~7 % of the annual operating cost (Fig. 3). This indicates that the LISP production plant is driven primarily by facility dependent and materials costs. The unit production cost of LISP bioadhesive decreases from \$1.57/kg. to \$1.19/kg for plant size 10MT/batch for internal rate of return of 10 %, assumed for prediction of minimum selling price. The plant of 30 MT/batch with unit production price of \$1.32/kg was used as base case for further analysis.

The isolated soy protein has the highest materials cost, as feedstock in synthesis of LISP bioadhesive. Depolymerization of kraft lignin process requires low-cost material due and small amount required in the polymerization process. Utility costs are relatively small compared to materials costs because the LISP bioadhesive production process does not require high temperature or pressure. The minimum selling price of LISP bioadhesive of \$1.40/kg was significantly less than pressure sensitive bioadhesive (PSA) of \$3.48/kg and structural bioadhesive \$3.11/kg made from glycerol (Yang and Rosentrater, 2019a, 2019b) (Fig. 3 (B)). The cost of conventional commercial urea-formaldehyde based wood adhesive is between \$1 and 3/kg ("Alibaba," 2023). The minimum selling price of bioadhesive can up to \$1.1/kg (Fig. 4(B)).

3.2. Scenario analysis results

This study investigated the impact of uncertainty in total capital cost, usage of lignin, source of lignin and scale of plant on the unit production cost of LISP bioadhesive production.

3.2.1. Scenario analysis on total capital investments

The first analysis, shown in Fig. 4(A) demonstrates the effect of changing total capital investment (C_{TC}) on the profitability and unit production cost of bioadhesive production by changing total capital cost of investment (C_{TC}) mentioned in equation (1) on unit production price and net profit in three scenarios: Case 1 (base case using data from plant of 30 MT/batch without change in C_{TC}), Case 2 (C_{TC} increase by 30 %) and Case 3 (C_{TC} decrease by 30 %). We varied the cost of total capital investment (C_{TC}) by changing direct fixed capital cost (C_{DF}), working capital (C_W), and start-up costs (C_S) to determine the impact on annual operating cost, minimum selling price and unit production cost. In the

best-case scenario of Case 3, where C_{TC} was reduced by 30 %, the unit production cost of bio-adhesive was \$1.29/kg and minimum selling price of \$1.35/kg. In the worst-case scenario of case 2, where C_{TC} was increased by 30 %, the unit production cost was \$1.36/kg. The difference in unit production cost between the best-case scenario and the worst-case scenario was changed ~5 % shows that change in source of lignin does not affect unit product cost significantly. In terms of net profit, the change in C_{TC} shows change in net profit. Increase in C_{TC} increases net profit, while decrease in C_{TC} shows decrease in net profit.

In the second analysis, TEA on use of different proportion of kraft lignin shown in Fig. 3(B) are summarized as follows. The change in usage of kraft lignin proportion is expected to be resulted in change of production cost of bioadhesive. In scenario analysis, when the kraft lignin and isolated protein used in 1:1 and 1:2 ratio for synthesis of LISP bioadhesive, the production cost is 0.95/kg and \$1.01/kg resulting in a payback period of 6 and 6.17 years. The increase in ratio of isolated protein in kraft lignin to isolated soy protein to 1:3 and 1:4 showed an increased unit production cost of \$1.29/kg and \$1.56/kg with range of 6–6.13- year of payback period.

Kraft lignin is an important raw material in the production of LISP bioadhesive to modify its properties. Kraft lignin from a pulp biorefinery is a low-value byproduct. For our technoeconomic analysis, price of lignin was pulled from industrial data (Abbati De Assis et al., 2018). Fig. 4(C) compares three different kraft lignin sources including the price from industrial data. As observed in this figure, the unit production cost of bioadhesive when kraft lignin was used from an integrated biorefinery was \$1.31/kg. However, when kraft lignin was a waste byproduct, it showed little influence, with unit production cost of \$1.22/kg and decreased annual operating cost by ~7 %. This third scenario study (Fig. 4(C)) suggested that the source of kraft lignin production has low impact when making decisions about the production of bioadhesive. In Fig. 4(D), results show the change in unit production cost across five different LISP bioadhesive production plant scales. As expected, increase in scale of plants incurred higher capital investment and operating costs while unit production costs of LISP bioadhesive plants, showed increasing plant scales led to decreasing unit production cost as expected. The unit production cost decreased from \$1.57/kg for plant capacity of 10 MT/batch to \$1.18/kg in the 50 MT/batch LISP production plant.

3.2.2. Sensitivity analysis results

Inspecting the sensitivity analysis for different parameters, Fig. 5 shows that total direct cost investment (C_{TC}) had the highest effect on unit production cost of LISP bioadhesive. The unit production cost of LISP bioadhesive increased to \$1.34/kg after C_{TC} increased by 10 % and



Fig. 6. Life cycle assessment contributions for cradle-to-gate production of 1 kg of lignin-isolate soy protein bioadhesive.

Table 3

Impact values associated with lignin-isolate soy protein bioadhesive synthesis, cradle-to-gate. Functional unit: 1 kg LISP bioadhesive.

Impact category	Unit	Total
Ozone depletion	kg CFC-11 eq	1.27E-07
Global warming	kg CO ₂ eq	2.202
Smog	kg O ₃ eq	0.055
Acidification	kg SO ₂ eq	0.006
Eutrophication	kg N eq	0.003
Carcinogenics	CTUh	1.16E-07
Non carcinogenics	CTUh	2.52E-07
Respiratory effects	kg PM _{2.5} eq	0.0006
Ecotoxicity	CTUe	7.894
Fossil fuel depletion	MJ surplus	4.550

decreased to \$1.30/kg after C_{TC} decreased by 10 %. The isolated soy protein cost had the highest cost within the raw material cost; hence it shows same equitable changes on unit product cost. The increase of 10 % labor cost shows maximum unit production cost of LISP bioadhesive of \$1.33/kg, while a decrease of 10 %-unit production cost results in a production cost of \$1.31/kg, virtually no change.

In this study, the findings of this study are in line with the reported market price range of \$1–4/kg for bioadhesive produced via different route (Yang and Rosentrater, 2019b, 2019b). However, certain assumptions were made in this study, such as the data from process model would hold true for a commercial plant, and factors such as feedstock composition variability and fluctuations in raw material and utility costs were not considered. Therefore, the results should be viewed as estimates. When compared to other bio-based processes, the results are promising (Mousavi-Avval et al., 2023). The economic feasibility of producing bioadhesive has been found to be greatly improved in all these cases.

3.3. Life cycle assessment

In the present study, data related to bioadhesive production were

taken from SuperPro Designer process model. All mass and energy balances of this study were simulated and supported by optimizing experimental data Table 1. The background data such as cradle-to-gate production of chemicals: Isolated soy protein (Proxy: Soy meal) (Arias et al., 2020), NaOH and mix electricity were taken from US EI and USLCI databases (DATASmart, 2023). The impact categories assessed, together with the method used for their calculation, are shown in Fig. 6 and Table 3.

The data was obtained from the SuperPro designer model for a LISP plant with a production capacity of 30 MT/batch with ratio of isolate soy protein to kraft lignin of 2:1. The bioadhesive production process utilized kraft lignin, isolated soy protein, cooling water, steam. For the ten impact categories estimated, Fig. 6 shows that the use of steam contribute the highest environmental impact for most of the impact categories by contributing more than 50 % for global warming (GW), ozone depletion (OD), smog, acidification (ACD), respiratory effects (REEF) and fossil fuel depletion (FFD).

Regarding the scoring of the impact category values (Table 3), the cradle-to-gate GW impact of producing 1 kg of LISP processes was estimated to be 2.20/kg CO₂-eq. In scenario analysis (Supporting information), LCA analysis of LISP bioadhesive constituting isolated soy protein and kraft lignin in ratio of 5:1 shows lower GW impact of 1.95 kg CO₂-eq (Supplementary Information). It shows that LISP bioadhesive has a lower emission profile when ratio of isolated soy protein to kraft lignin at 5:1 compared to the lower ratio due to lower use of steam in depolymerization of kraft lignin.

For cradle-to-gate production, the GW impacts of fossil-based epoxy and glycerol resins are 8 and 6.1 kg CO₂-eq/kg (Tserpes and Tzatzadakis, 2022), respectively, while for UF resin, it ranges from 1.53 to 2.88 kgCO₂-eq/kg (Bush et al., 2022; Yang and Rosentrater, 2020).

Utilization of steam was found to be a major contributor to the GW impact (Fig. 6). Contribution from energy requirements from electricity showed less impact. Additionally, it should be noted that the use of kraft lignin and steam led to substantial impact on all categories except eutrophication (EU) where isolated soy protein contributed more than 50 % of the overall impact.

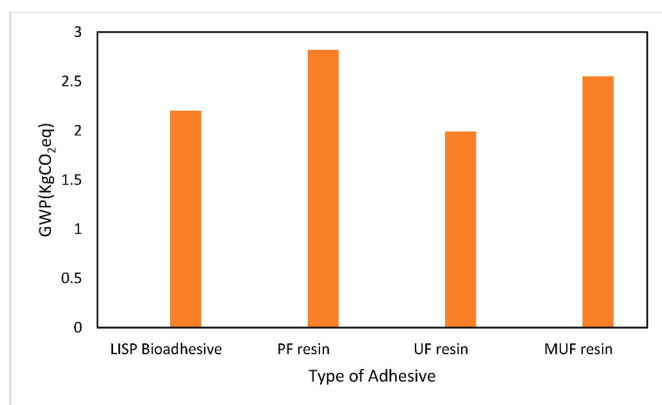


Fig. 7. Comparative analysis of GW impact of LISP bio adhesive and fossil-based adhesive with FU of 1 Kg (PF: Phenol-Formaldehyde, UF: Urea-Formaldehyde, MUF: Melamine-Urea-Formaldehyde Resin).

Regarding the scoring of the impact category values, Table 3 shows the overall value for the ten life-cycle environmental impacts. The harmful human ecotoxicity of the bioadhesive production is negligible. OD, EU, and ACD are low, reaching only 0.127 mg CFC-11-eq, 3 gm N-eq and 6 gm SO₂-eq respectively. This showed a favorable result, as the impacts on the ozone layer, eutrophication and mineral resources categories are not significant. The use of isolated soy protein shows negative potential impact in the non-carcinogenic impact category (Fan et al., 2022).

Although non-fossil resources are used as industrial feedstocks for bioadhesive synthesis, it does not imply that life-cycle environmental impacts are neutral or negative, as the process of valorizing these resources requires the use of fossil fuel-derived chemicals and energy. The supplementary materials present a detailed contribution analysis of inputs for their environmental impacts. An alternative LISP production process that reduces isolated soy protein usage could potentially improve the environmental performance of LISP produced because of high steam use in bioadhesive synthesis step.

The GW impact of bioadhesives is typically lower than the GW impact of fossil-based adhesives due to use of raw materials which made from renewable resources, such as plants, which absorb CO₂ from the atmosphere during their growth. In Fig. 7, the GW impact of a LISP bioadhesive made from soy protein and kraft lignin is showed to be 2.2 kg CO₂e per kg of adhesive. In comparison to fossil-based adhesive, the GW impact of a fossil-based resin made from melamine-urea-formaldehyde is estimated to be 2.82 kg CO₂e per kg of resin (Wilson, 2010). The urea-formaldehyde and melamine-urea-formaldehyde based resin shows slightly lower GW impact ranging from 1.5 to 2.0 and from 1.5 to 1.6 kg CO₂e per kg of resin, respectively, but emit hazardous formaldehyde (Yang and Rosentrater, 2020).

3.3.1. Uncertainty analysis

Life cycle assessment (LCA) results are used to inform decision-making, so it is crucial to quantify their uncertainty. We employed a Monte Carlo simulation to understand the influence of data uncertainty on specific impact categories. For each process, the Monte Carlo analyses involved 1000 simulations, resulting from combinations of assorted values of included life cycle inventory (LCI) parameters. We evaluated uncertainty by examining the spread of values in relation to their average. The results showed lower the standard error of mean, thus more reliable results. In Table 4, we present the key findings of our uncertainty analysis, including mean values, standard deviation (representing variability), and other pertinent metrics. The highest uncertainty was observed for non-carcinogenic impact category and carcinogenic impact category. Our results suggest that the GW impact of LISP bioadhesive varies between 1.9 and 2.5 CO₂e per kg at both ends of the confidence interval (2.5 %–97.5 %). This result shows that the uncertainty for GW impact and other impact categories considered is low, confirming the reliability of our dataset and demonstrating that variations in data selection and inherent data variability do not exert a significant influence on the examined impact categories (Giuliana et al., 2022).

4. Conclusion

This study presented economic and environmental analyses of LISP bioadhesive production using sustainable raw materials of kraft lignin and isolated soy protein. The study employed a process model designed for plant scales ranging from 10MT/batch to 50MT/batch. The unit production cost of LISP bioadhesive was \$1.32/kg at a plant scale of 30MT/batch, with the lowest achievable cost being 1.19 at 50MT/batch. Moreover, the LISP bioadhesive production process exhibited a notably low global warming impact of 2.20 kg of CO₂-eq/kg, along with a competitive unit production cost compared to alternative bioadhesive production processes. The price of isolated soy protein exerted the greatest influence on production costs. Additionally, steam usage emerged as the primary contributor to global warming impact of LISP production. The study underscores the economic viability and environmental sustainability of LISP bioadhesive production. Consequently, this research holds the potential to significantly contribute to the future advancement of sustainable bioadhesive manufacturing processes.

CRedit authorship contribution statement

J.P. Ahire: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. **S.H. Mousavi-Avval:** Validation, Writing – review & editing. **N. Rajendiran:** Formal analysis, Writing – review & editing. **R. Bergman:** Data curation, Formal analysis, Funding acquisition, Project administration, Resources, Software, Supervision, Validation, Writing – original draft, Writing – review & editing. **T.**

Table 4
Uncertainty analysis of life-cycle environmental impacts of LISP bioadhesives production.

Impact category	Unit	Mean	Median	SD	CV	2.50 %	97.50 %	SEM
Smog	kg O3 eq	0.0681	0.067	0.00775	11.40 %	0.0554	0.0865	0.000245
Respiratory effects	kg PM2.5 eq	0.00135	0.00134	9.24E-05	6.84 %	0.0012	0.00158	2.92E-06
Ozone depletion	kg CFC-11 eq	2.11E-07	1.97E-07	6.07E-08	28.80 %	1.34E-07	3.68E-07	1.92E-09
Non carcinogenics	CTUh	2.19E-07	2.10E-07	3.81E-07	174 %	−4.89E-07	9.99E-07	1.20E-08
Global warming	kg CO2 eq	2.2	2.18	0.171	7.78 %	1.9	2.55	0.0054
Fossil fuel depletion	MJ surplus	2.88	2.86	0.294	10.20 %	2.36	3.49	0.0093
Eutrophication	kg N eq	0.00456	0.00417	0.00167	36.70 %	0.00273	0.00847	5.29E-05
Ecotoxicity	CTUe	9.33	9.07	2.07	22.10 %	6.43	13.9	0.0654
Carcinogenics	CTUh	1.68E-07	1.51E-07	8.80E-08	52.20 %	7.39E-08	3.51E-07	2.78E-09
Acidification	kg SO2 eq	0.00632	0.00627	0.000527	8.34 %	0.00542	0.00748	1.67E-05

Note: SD: Standard Deviation, CV: Coefficient of Variation, CV unit is in %. SEM: Standard Error of the Mean (SEM) (standard deviation of the sample distribution of the mean).

Runge: Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Writing – review & editing. **C. Jiang:** Data curation, Writing – review & editing. **J. Hu:** Data curation, Writing – review & editing.

Declaration of competing 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.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2024.141474>.

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