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Solvent selection enables sustainable and affordable lignin biocomposite for cement-free construction

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ABSTRACT

Cement-free construction materials are essential to reduce global carbon emissions, yet scalable alternatives remain limited. We report the development of a lignin-based biopolymer-bound soil composite (BSC), a novel cement-free material with mechanical properties comparable to lightweight concrete. To advance its scalability and environmental performance, we also used a systematic framework for solvent selection in lignin biocomposite fabrication. Applying this approach, we identified an acetic acid–water solvent system that speeds up manufacturing and enhances material quality. BSCs fabricated with this system exhibit increased strength (5.4 MPa vs. 3.7 MPa), attributed to acetylation of lignin. In addition, the acetic acid–water system dramatically reduces drying time compared with the alternative solvent, dimethyl sulfoxide (2 days vs. 14 days), enabling more efficient production. Life cycle assessment reveals additional CO_2 sequestration and a 70 % reduction in material cost (US$122–237/m^3 vs. US$409–933/m^3) relative to lignin biocomposite made using DMSO as the solvent. These improvements stem from solvent-induced modifications in lignin chemistry that enhance composite performance. This work demonstrates how both material design and rational solvent selection can pave the way for adoption of lignin-based composites as scalable, affordable, and low-carbon alternatives for the built environment.

1. Introduction and motivation

The construction industry is one of the main sources of greenhouse gases. It accounts for 11% of global carbon emissions (Huang et al., 2018). Cement production, in particular, represents a significant portion of these emissions, contributing 5%–8% of annual global anthropogenic carbon emissions (Cheng et al., 2023). Cement production is an energy-intensive process, primarily due to the calcination of limestone and the combustion of fuels (Barbhuiya et al., 2024). In the building industry, attention is now focused on cleaner materials, with less reliance on cement, the development of which promises to improve resource efficiency and reduce the embodied carbon of the built environment (Forbes and Ahmed, 2010; Marjaba and Chidiac, 2016; Sizirici et al., 2021; Chen et al., 2023; Labaran et al., 2022; Li et al., 2025).

Biopolymer-bound soil composite (BSC) is a novel class of construction materials on the path toward a green transition in the construction industry (Miao et al., 2024c). Unlike conventional construction materials, such as ordinary portland cement concrete, BSC uses biologically derived polymers to bind granular material, forming a material as strong as concrete (Rosa et al., 2020). Furthermore, as many of these biopolymers – blood proteins and lignin – are by-products of major industries (Hu et al., 2018; Fernández-Rodríguez et al., 2017; Gupta et al., 2024), they are readily available in large quantities to be incorporated into these materials on a large scale. BSC materials benefit the environment in two ways. First, they avoid the use of portland cement, and second, they sequester carbon through the incorporation of carbon-rich biopolymers (i.e., the biogenic carbon originally fixed from the atmosphere by plants is stored long term in lignin biocomposite), resulting in a negative carbon footprint.

Lignin-based BSC is the newest member of the BSC family, with compressive strength similar to that of lightweight concrete (Miao et al., 2024b,c, 2025a). In addition, lignin-based BSC can be recycled, to recover the biopolymer, so that the material can be re-manufactured (Miao et al., 2025b,a). The main contributors to the carbon footprint of lignin-based BSC are the energy required for desiccation and the solvent used to manufacture the biocomposite (Miao et al., 2024c). Based upon the compressive strength of this material that we have been able to

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Table 1
Overview of biopolymer-bound soil composites (BSC) and their applications.

Study	Biopolymer	Applications	Density (kg/m^3)	Compressive strength (MPa)
Dove et al. (2016)	• Alginates (polysaccharides with salts)	Masonry and bricks	1800–2100	0.8–3.0
Qureshi et al. (2017)	• Xanthan gum	Soil improvement	1630–1670	0.7–1.7
Chang et al. (2018)	• Casein	Soil improvement	———	2.7–5.6
Rosa et al. (2020)	• Animal plasma AP920 (proteins)	Soil improvement, structural construction, pavers	1330–1720	1.6–8.1
Roberts et al. (2021)	• Spider silk • Bovine serum albumin	Extraterrestrial construction and 3D printing	———	6.6–37.1
Miao et al. (2024b)	• Hydrolysis lignin • Alkali lignin	Soil improvement, non-structural construction, pavers	1280–1500	1.6–8.1
Miao et al. (2025b)	• Lignoforce lignin • Lignoboost lignin	Soil improvement, non-structural construction, pavers	1730–1910	2.9–6.0
This study	• Lignoforce lignin with acetic acid water solvent	Pavers, pre-cast wall panels, interior partition walls, roofing tiles	1460–1740	0.1–5.4

achieve thus far, the applications of lignin-based BSC include pavers, pre-cast wall panels, interior partition walls, roofing tiles, and building facades (Miao et al., 2024c). Table 1 summarizes how lignin-based biopolymer-bound soil composite compares to other biopolymer-bound soil composites in terms of biopolymer type, applications, and physical properties (i.e., compressive strength and density).

Lignin is a complex aromatic biopolymer and the second most abundant natural polymer after cellulose. Commercially, it is primarily produced as a byproduct of the paper and pulp industry and from bioethanol production (Bajwa et al., 2019; Mastrolitti et al., 2021). Industrially, lignin exists in several forms depending on the extraction process, including kraft lignin, lignosulfonate lignin, alkali lignin, hydrolysis lignin, organosolv lignin, and steam explosion lignin (Mastrolitti et al., 2021). Kraft lignin, obtained from the purification of kraft black liquor, is the most common type and represents the majority of global lignin production. Lignosulfonates are produced *via* sulfite pulping and are highly water-soluble, while alkali lignin comes from alkaline pulping and differs structurally from kraft lignin. Hydrolysis lignin is derived from biomass during bioethanol production.

For many biopolymers used in the manufacture of BSC, water is the preferred solvent, with or without pH modification. For lignin, however, a wide range of solvent options can be considered. Lignin is a complex hydrocarbon, the solubility of which is influenced by abundant aromatic rings, aryl-ether linkages, and numerous hydroxyl groups that are part of a large molecule with complex branching (Abolore et al., 2025; Cazier et al., 2024). The various forms of lignin that are created during the isolation processes, from paper and pulp production and bio-ethanol industries, have different solubilities (Mastrolitti et al., 2021).

This study was motivated by a desire to seek an alternative to dimethyl sulfoxide (DMSO) as the solvent for lignin-based BSC. DMSO is a relatively safe organic solvent. In mammalian model systems, for instance, ingestion of a few grams of DMSO per kg is required to see significant toxic effects, with LD50s reported in the range of 14-28 g/kg in rodents (Brobyn, 1975). By contrast, other solvents used in the manufacture of construction materials can have LD50s that are order of magnitudes lower (i.e., they are more toxic). DMSO is also considered relatively benign when exposure occurs by inhalation (Fishman et al., 1969).

The use of DMSO, however, in the manufacture of lignin-based materials presents several challenges. For example, DMSO evaporates more slowly than does water. In laboratory settings, BSC specimens made with water typically dry in 2–3 days, while those made with DMSO require approximately two weeks to fully desiccate (Biggerstaff et al., 2022; Miao et al., 2024b). This is consistent with the difference in vapor pressure between DMSO and water. Other solvents, such as acetic acid and formic acid, are attractive as they are more volatile than DMSO, while also being effective in dissolving lignin (Khare et al., 1999). The use of undiluted organic acids, however, instead of DMSO, results in a higher carbon footprint for lignin biocomposite (Miao et al., 2024c). Thus, to strike a balance between low environmental impact and speed of desiccation, a mixed solvent system (organic acid and water) may be ideal. Previous studies have shown that mixtures of water and an organic solvent (e.g. dioxane) were more effective in dissolving lignin (Melro et al., 2018; Evstigneev, 2010; Boeriu et al., 2014), due to the amphipathic nature of the lignin molecule (Sun et al., 2022).

To help address these challenges, we introduce a solvent-based approach for optimizing the design and manufacture of lignin biocomposite. Our method incorporates the Hansen approach to examine various candidate solvents for lignin, forming the foundation for designing our mixed solvent system. Acetic acid is an effective solvent for lignin, enabling the development of a mixed solvent system consisting of acetic acid and water. This approach aims to fine-tune the manufacturing process while minimizing the environmental impacts and economic costs of BSC. In addition, the acetic acid–water system can significantly reduce the time and energy required for desiccation (Miao et al., 2024c). More rapid dry-out also enables additive manufacturing approaches, such as 3D printing.

Our study contributes to the development of cleaner materials by advancing a cement-free, bio-based composite that combines reduced process emissions through lower energy demand with long-term storage of biogenic carbon.

2. Methods

2.1. Materials

To manufacture lignin biocomposite, feldspathic grade 90 sand (# 90 Silver Sand; grain shape — sub-angular; hardness — 6.5 Mohs; moisture content — < 0.1 %; bulk density — 1409 kg/m^3; soil particle density — 2.60 g/cm^3; uniformity coefficient 1.75-1.95) from P.W. Gillibrand, Simi Valley, California was used. According to Rosa et al. the maximum dry bulk density of this material ($\rho^{max}_{dry,bulksoil}$) was 1.48 g/cm^3 (Rosa et al., 2020). Lignoforce lignin was obtained from West Fraser Timber Company, Ltd (Vancouver, Canada), from their Hinton Plant, located in Alberta, Canada. The acetic acid (glacial) was obtained from Fischer Scientific.

Table 2
Hansen solubility parameters for candidate solvents.

Pure solvents	Dispersion, δ_d ($MPa^{1/2}$)	Polarity, δ_p ($MPa^{1/2}$)	Hydrogen, δ_h ($MPa^{1/2}$)	Relative energy difference (RED)
Water	15.6	16.0	42.3	2.27
Dimethyl sulfoxide	18.4	16.4	10.2	0.22
Acetic acid	14.5	8.0	13.5	0.92
Dioxane	19.0	1.8	7.4	1.29
Pyridine	18.8	4.9	9.2	1.03
Formic acid	14.3	11.9	16.6	0.78

Table 3
Hansen solubility parameters of acetic acid–water mixtures.

Composition of mixed solvent	Dispersion, δ_d ($MPa^{1/2}$)	Polarity, δ_p ($MPa^{1/2}$)	Hydrogen, δ_h ($MPa^{1/2}$)	Relative energy difference (RED)
100% Water	15.6	16.0	42.3	2.27
20% acetic acid & 80% water	15.4	14.4	36.5	1.87
40% acetic acid & 60% water	15.2	12.8	30.8	1.50
60% acetic acid & 40% water	14.9	11.2	25.0	1.18
80% acetic acid & 20% water	14.7	9.6	19.3	0.96
100% acetic acid	14.5	8.0	13.5	0.92

2.2. Estimation of solubility by the Hansen approach

The Hansen solubility approach was used to identify candidate solvents, in their pure form, that are effective in dissolving kraft lignin. The use of a mixed solvent composed of acetic acid and water may help lower the carbon footprint of lignin biocomposites (Miao et al., 2024c). Again, the Hansen solubility approach was used to make initial predictions regarding the effectiveness of all the mixed solvents investigated.

To determine the interaction between a solute and solvent, the Relative Energy Difference (RED) parameter was computed, with RED being formulated as the ratio between the radius of interaction of a solvent (R_a) to that of the solute (R_0). The Hansen approach takes into account dipole–dipole interactions (δ_p), London dispersion forces (δ_d), and hydrogen bonding (δ_h) between the solute and the solvent. Eq. (1) shows the calculation of the radius of interaction of the solvent (R_a) and RED ratio.

$$(R_a)^2 = 4(\delta_{d2} - \delta_{d1})^2 + (\delta_{p2} - \delta_{p1})^2 + (\delta_{h2} - \delta_{h1})^2$$
$$RED = \frac{R_a}{R_0} \quad (1)$$

Where if the $RED < 1$ the solute will dissolve in solvent, if $RED = 1$ the solute will partially dissolve into the solvent, and if $RED > 1$ solute will not dissolve into the solvent. To determine viable solvents to dissolve kraft lignin, the Hansen solubility parameters will be used to plot the Hansen space for kraft lignin (Fig. 1).

Additionally, Eq. (1) can be used to find the RED ratio, which measures the effectiveness of each solvent in dissolving lignin. The Hansen solubility parameters for candidate solvents and various acetic acid–water mixtures considered in this study are summarized in Tables 2 and 3, respectively (Dogenski et al., 2020; Burke, 1984). For this study we used the Hansen solubility parameters for eucalyptus urograndis kraft lignin (δ_d = 17.8 $MPa^{1/2}$, δ_d = 18.4 $MPa^{1/2}$, δ_d = 12.1 $MPa^{1/2}$, and R_0 = 13.5 $MPa^{1/2}$) to approximate the Hansen solubility parameters for kraft lignin obtained through the LignoForce system (Ribeiro et al., 2020). The relative energy difference values in Table 2 and Table 3 were computed by finding the ratio between the radius of interaction of the solvent (R_a) and the radius of interaction of the solute (Kraft lignin with R_0 = 13.5 $MPa^{1/2}$) according to Eq. (1).

2.3. Fabrication of lignin biocomposite

To allow lignin to effectively bind to granular material, a suitable solvent is used to first dissolve the biopolymer with the aid of mechanical tools, such as a stand mixer. The biopolymer and granular materials are mixed for approximately 5–10 min, producing a wet mix with similar consistency to cookie dough (Miao et al., 2024b). In the wet state, lignin-based BSC is easily moldable, and can be formed into a variety of shapes depending on the desired application (i.e., tiles, bricks, roofing tiles, pavement, etc.). For laboratory testing, however, the wet BSC mix is formed into cylindrical test articles (height = 52.0 ± 2.5 mm, & diameter = 25.4 ± 0.5 mm) through a molding apparatus composed of 316 stainless steel. The test articles are compacted via a steel piston which is inserted into the cylindrical mold. A total of 8 specimens were manufactured for each batch (i.e., for a given acetic acid–water mixture and desiccation condition, 8 samples were produced). A double 5 MPa compaction method was performed on all test articles by an MTS Criterion Model 43 electromechanical universal testing machine (compaction occurs at 0.2 mm/s) (Miao et al., 2024b; Rosa et al., 2020). The compacted test articles were mechanically extruded from the mold using the steel piston. In the wet state, measurements of the dimensions (height and diameter) and weight of BSC specimens were taken to calculate the fresh density and fresh bulk soil densities of the composites.

Solvent removal occurs in BSC by evaporation of the solvent from the biocomposite. The evaporation process can be accelerated through energy enhanced methods (i.e., a laboratory oven at 40 °C), but otherwise can be fully accomplished at ambient conditions. Strategies for monitoring solvent removal include conventional tracking through mass-loss measurements and more recent non-destructive methods using vibration based measurements (Rosa et al., 2020; Miao et al., 2024a,b). For this study, however, we monitored solvent removal on a mass-loss basis using a Fischer scientific Iso-4000 measuring scale.

The finished specimens (after desiccation is performed) were again measured, in order to compute the unit weight and dry bulk soil density of the specimens in their dried state. All tested specimens had roughly the same heights (±1mm) and diameters (±0.50mm). Specimens that did not fall within these dimensions were not considered in this study (Miao et al., 2024b). A uni-axial compressive test was conducted on the finished specimens (using the MTS Criterion Model 43 testing machine) at a rate of 0.02 mm/s, to obtain the compressive strength of lignin-based BSC with varying acetic acid and water mixtures.

2.4. Fourier transform infrared spectroscopy

Fourier Transform Infrared Spectroscopy (FTIR) was performed on desiccated lignin samples combined with 100% DMSO and on samples combined with 100% acetic acid using a Nicolet iS50 FT/IR Spectrometer. Scans were collected in absorbance mode at a resolution of 2 cm^{-1} over a spectral range of 400 to 4000 cm^{-1}. Each sample was dried prior

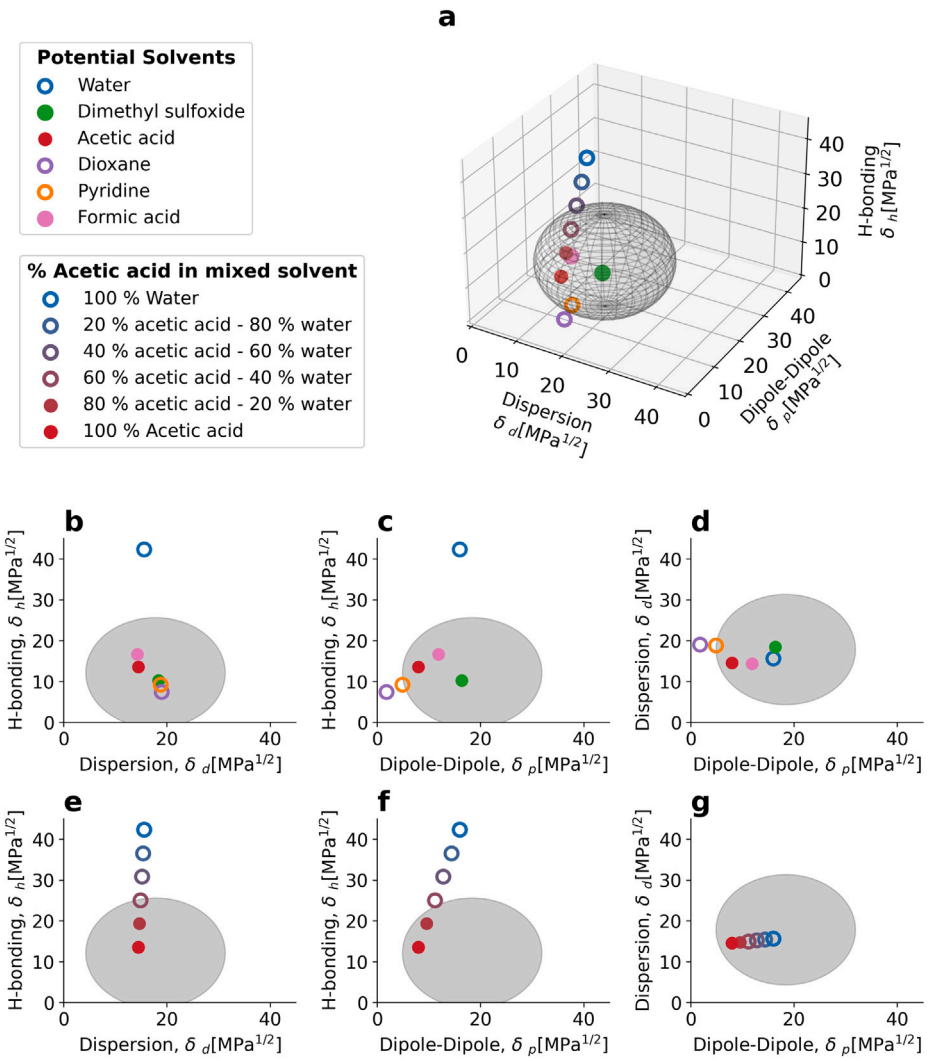

Fig. 1. The Hansen method provides a rational approach for solvent selection. The solid colored dots represent candidate solvents and candidate solvent mixtures for dissolving kraft lignin (**a**). Our solvent-based optimization method uses the Hansen approach to initially identify candidate solvents for dissolving lignin (**b-d**). Depending on various factors – such as solvent cost, volatility of the solvent, and environmental impact – a mixed solvent, typically a solvent-water mixture, is selected. In this study, we investigated the effect of using an acetic acid–water mixture on the manufacture of this material (**e-g**). For each panel, a solid dot represents an effective solvent (Relative Energy Difference, RED < 1) and an un-filled circle represents an ineffective solvent (Relative Energy Difference, RED > 1), as summarized in Tables 2 and 3.

to analysis: for lignin mixed with DMSO, desiccation corresponded to near-complete solvent removal, whereas for lignin mixed with acetic acid, drying reached a plateau at approximately 80% solvent removal. A total of five independent samples were analyzed for each solvent condition.

Before analysis, a background spectrum (either of the empty sample holder or of the pure solvent) was collected and subtracted from the sample spectrum to isolate the sample's absorbance data. The OMNIC software processes this by ratioing the sample and background spectra and applying a logarithmic calculation according to $A = \log_{10}(I_0/I)$, where I_0 is the incident light intensity (background) and I is the transmitted light intensity (sample). Baseline correction was then applied using the same software, and the absorbance spectra were normalized such that the highest peak in each sample was set to 1. This scaling preserves the shape of the spectra while enabling direct comparison of relative peak intensities across samples. The resulting spectra represent the mean absorbance profiles, with shaded regions indicating the full range of variation observed among replicates.

2.5. Scanning electron microscopy

Scanning Electron Microscopy (SEM) was performed on lignin biocomposite samples using a Thermo Fisher Scientific Apreo S LoVac scanning electron microscope. The biocomposite was ground into particulates consisting of clusters of aggregates bound together by the lignin biopolymer. These clusters were affixed to mounts using a black epoxy adhesive and coated with an Au/Pd layer (60:40 ratio). The samples were visualized using a working distance (WD) of 10.0 mm, an accelerating voltage (HV) of 5.00 kV, a beam current of 50 pA, dwell time of 200 ns, using an average of 4 filters, resolution of 768 × 512, using "Optiplan" mode with an Everhart–Thornley Detector (ETD), and in high vaccuum conditions (i.e., below 9×10^{-5} mbar).

2.6. Life cycle assessment

A cradle-to-gate life cycle assessment (LCA) was carried out using SimaPro software and the Impact 2002+ method. One of the primary reasons for selecting the Impact 2002+ method over others is

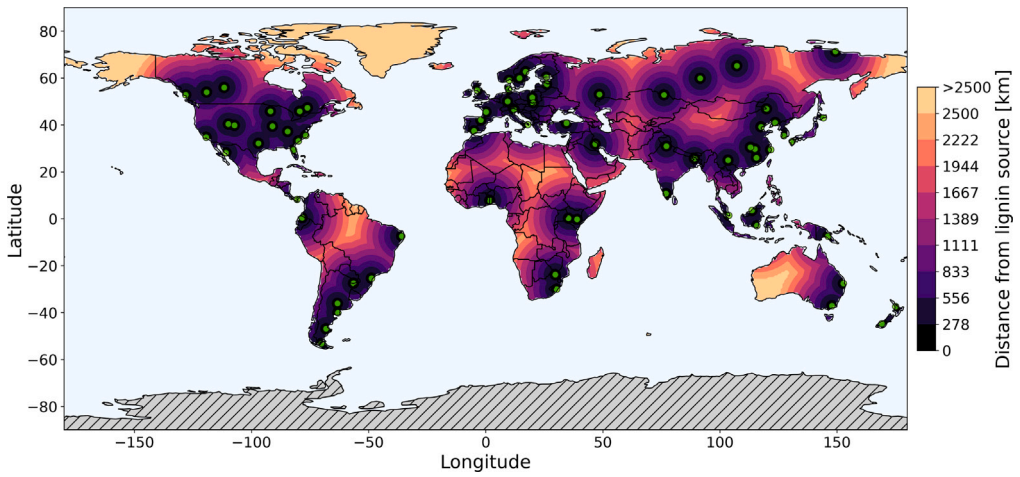

Fig. 2. Locations where lignin can be sourced. To estimate the carbon footprint of manufacturing lignin biocomposites globally, potential lignin sourcing locations were identified. Regions closer to lignin sources exhibited a lower carbon footprint, while those farther away tended to have a higher footprint. The locations of lignin production facilities are essential for calculating the distance from lignin plants to the manufacturing sites of lignin-based BSC.

its established use in performing LCAs on other soil-based building materials (Roedel et al., 2015; Miao et al., 2024c). The Impact 2002+ method reports environmental impacts across 14 different characterization categories (Jolliet et al., 2003), covering human health, ecosystem quality, climate change, and resource depletion.

Eq. (2) was used to calculate the environmental impacts of manufacturing lignin biocomposite. The total impact of lignin-based BSC can be attributed to four main activities: transportation requirements, electricity use, solvent use, and lignin production. For this LCA, we do not consider the additional footprint incurred by transporting the manufactured lignin biocomposite to the application site (i.e., the footprints calculated in this study were done using a cradle-to-gate approach). The LCA was conducted using the Impact 2002+ method in SimaPro. Eq. (3) shows the general form used to calculate the environmental impacts of lignin-based BSC, where f is the impact per unit, m is in kilograms (kg), E is in kilowatt-hours (kWh), and T is in tonne-kilometres (tkm).

$$Total\ Impact = Transportation_{Imp.} + Electricity_{Imp.} + Solvent_{Imp.} + Lignin_{Impact} \quad (2)$$

The impact of transportation, $Transportation_{Imp.}$ (Eq. (3)), accounts for the transportation of the constituents (solvent, lignin, and aggregate) of the lignin biocomposite to the manufacturing site. To calculate this impact, distances between the manufacturing site and the locations where lignin can be sourced (Fig. 2) were obtained and multiplied by the mass of lignin required to produce 1 kg of lignin-based BSC (Peng et al., 2022). The transportation distance for the solvents was estimated based on an average distance between chemical plants and manufacturing sites and was then multiplied by the mass of solvent used (DMSO or acetic acid). For this life cycle assessment, it is assumed that the water used in the mixed solvent is sourced directly from the manufacturing site.

$$Transportation_{Imp.} = f_{transportation} * (m_{DMSO} * T_{DMSO} + m_{acid} * T_{acid} + m_{lignin} * T_{lignin}) \quad (3)$$

The impact of energy use, $Electricity_{Imp.}$ (Eq. (4)), accounts for the electricity consumption associated with the fabrication of lignin-based BSC—that is, the physical mixing of constituents and the energy required to desiccate the wet mix. During fabrication, the lignin is first dissolved in solution using an electrically powered stand mixer for 5-7 min and mixed with the aggregate to form a wet mix of lignin-based BSC. An electrically powered oven (at 40 °C) is used to accelerate the desiccation of the composite. Fig. 3 visualizes the common sources of electricity production around the world, while Fig. 4 provides a more detailed breakdown of electricity generation for each country (Ritchie and Rosado, 2020; International Energy Agency (IEA), 2023). Using the energy mix for a given country, an average impact factor for electricity ($f_{electricity}$) is calculated and multiplied by the sum of the energy used to power the stand mixer ($E_{Fabrication}$) and the energy used to desiccate the composite ($E_{desiccation}$). For this LCA, an electrical grid efficiency (η) of 90% is assumed.

$$Electricity_{Imp.} = f_{electricity} * \left(\frac{E_{fabrication} + E_{desiccation}}{\eta}\right) \quad (4)$$

The impact of solvent use, $Solvent_{Imp.}$ (Eq. (5)), is the solvent used to initially dissolve lignin during the manufacture of the lignin biocomposite. Eq. (3) shows the general form for calculating the impact of the solvent, which considers the environmental impact of lignin-based BSC when either DMSO or a mixture of acetic acid and water is used as a solvent. However, the impact of solvent can be significantly reduced – or even avoided – through solvent recapture, represented by the percentage of recapture ($\%recapture$). During desiccation, vaporized solvent can be re-captured by installing a long duct that allows the solvent vapor to condense on its surface so that the solvent can be collected. With an optimized duct design, nearly all of the off-gassed solvent potentially could be recovered (Miao et al., 2024c).

$$Solvent_{Imp.} = (m_{water} * f_{water} + m_{DMSO} * f_{DMSO} + m_{acid} * f_{acid}) * (1 - \%\ recapture) \quad (5)$$

The impact from lignin use, $Lignin_{Imp.}$ (Eq. (6)), accounts for both the environmental impact of producing kraft lignin and the additional benefit of carbon sequestration in lignin-based BSC. The footprint from lignin production ($f_{lignin} \cdot m_{Lignin}$) is multiplied by an allocation factor for lignin. While lignin is often considered a waste product and thus may not be allocated any environmental burden, its potential large-scale production – as a co-product or even a main product driven by commercial adoption of lignin-based materials – necessitates the inclusion of its production impact in this LCA. To accommodate a range of allocation scenarios, an allocation factor, L_{fac}, is introduced in Eq. (3). For this study, two allocation methods were considered: "Lignin waste stream bears all the burden" and "Main product bears all the burden," both represented by L_{fac} in Eq. (6). Carbon sequestration is one of the key environmental benefits of BSC, as the incorporation of carbon-rich biopolymers in the composite enables biological carbon storage (Miao et al., 2024c). Accordingly, when accounting for the carbon footprint associated with lignin production, an additional term, $Lignin_{sequestered\ CO_2}$, is included in the equation.

$$Lignin_{Imp.} = f_{lignin} * m_{Lignin} * L_{fac} - Lignin_{sequestered\ CO_2} \quad (6)$$

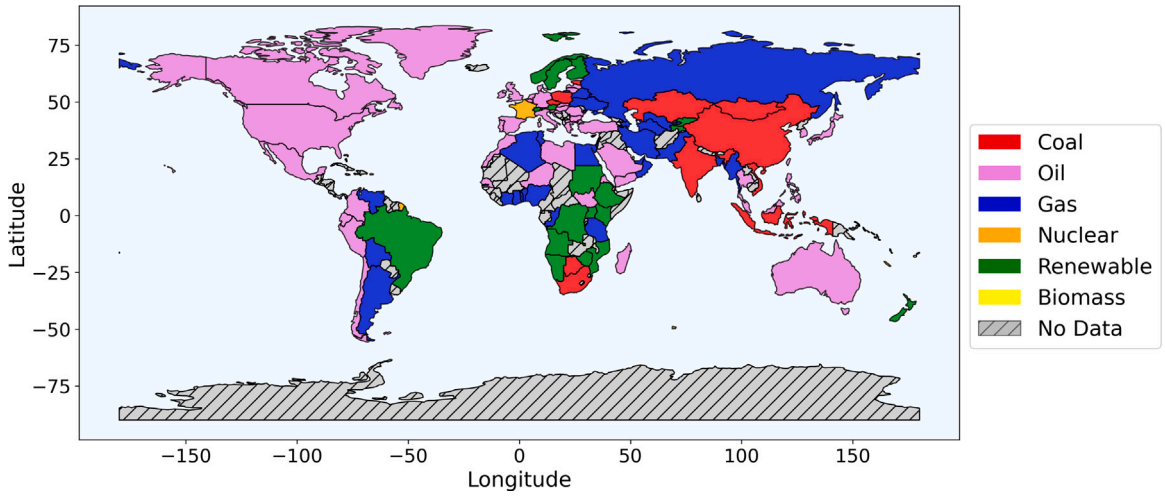

Fig. 3. Predominant source for electrical energy use in each country. The source of electrical energy is a critical factor in assessing the carbon footprint of lignin-based BSC production, as regions with cleaner electricity grids typically result in lower associated emissions.

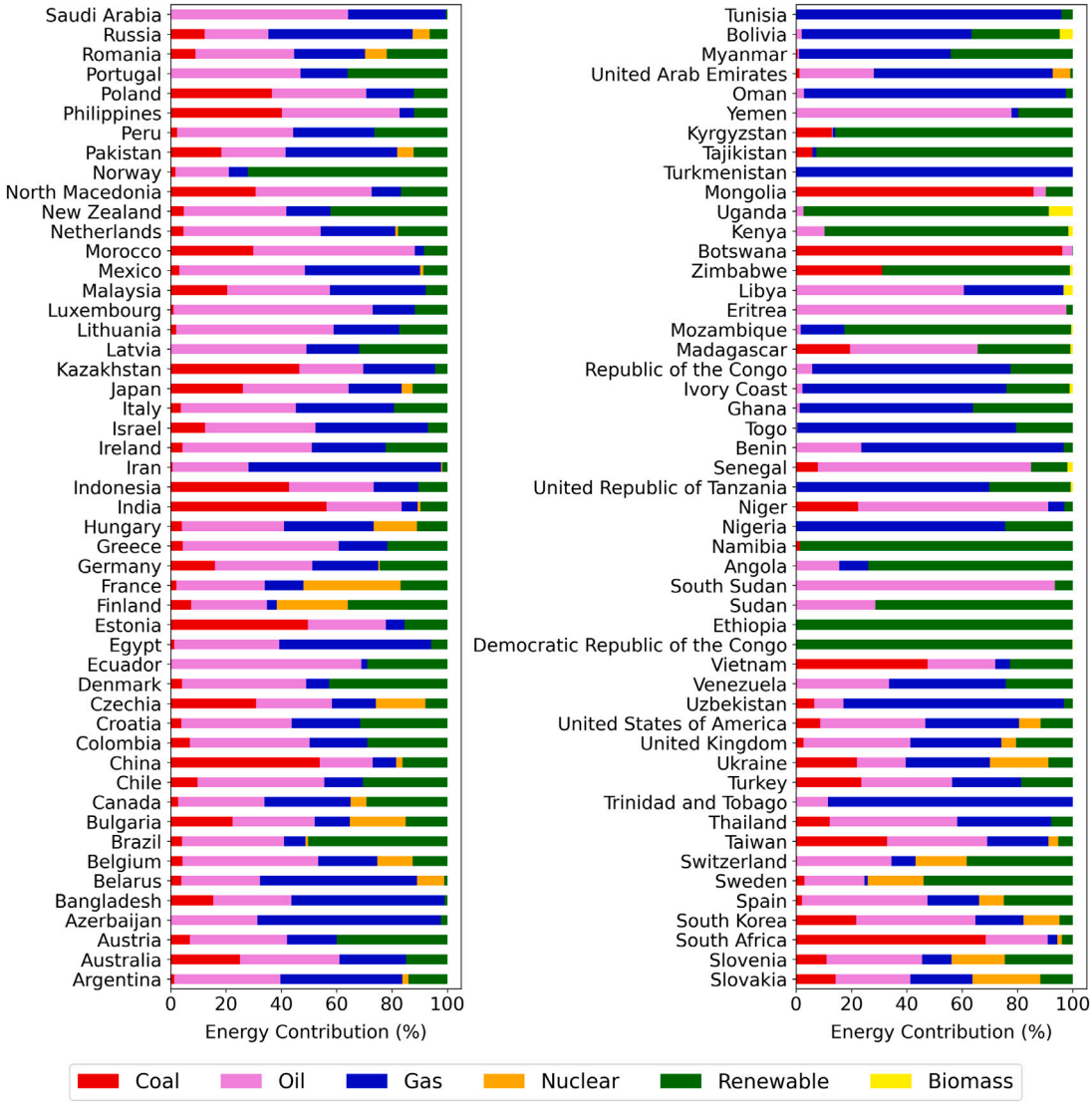

Fig. 4. Sources of electrical energy per country. To calculate the carbon footprint associated with the manufacture of lignin biocomposites globally, the source of electrical energy for each country must be considered. Based on the national breakdown of electricity generation sources, we calculated an $f_{\text{electricity}}$ value for each country. This value represents a proportional weighting that reflects the country's reliance on coal, natural gas, nuclear, renewable, and biomass energy sources for electricity production.

2.7. Techno-economic assessment

A cradle-to-gate approach was adopted to estimate the economic costs associated with the production of the lignin-based biocomposite. This approach encompasses all costs incurred from the extraction of raw materials up to the point where the final product leaves the manufacturing facility. Specifically, the calculated cost of the biocomposite includes expenses related to the solvent used (either dimethyl sulfoxide (DMSO) or an acetic acid–water mixture), the lignin feedstock, aggregate materials, and the electricity required for processing steps such as mixing and desiccation.

The cost of each solvent was determined using publicly available online sources, where the average price per kilogram was calculated based on bulk purchasing rates rather than small-scale laboratory prices. Similarly, the costs for lignin and aggregate materials were derived from the average bulk prices in the relevant country, reflecting industrial-scale procurement rather than research-grade pricing. Electricity costs were country-specific and calculated based on the average price per kilowatt-hour (kWh) for industrial usage.

The price of lightweight concrete, with an assumed density of 1640 kg/m^3, was obtained from publicly available online sources. The collected data represent typical price ranges for concrete in each country considered. This information was included to provide a reference point for assessing the economic viability of the lignin biocomposite, particularly when produced using the acetic acid–water mixture.

3. Results

3.1. Physical properties of lignin biocomposite using aqueous acetic acid

For this study, we used the Hansen solubility approach to consider various candidate solvents. In comparison to DMSO, both acetic acid and formic acid look attractive in terms of ability to dissolve lignin, as shown in Fig. 1 a-d. We chose acetic acid because it has a lower carbon footprint and is more volatile compared to formic acid (Miao et al., 2024c). A mixed solvent composed of 80% acetic acid is well within the sphere of solubility, while a 60% acetic acid–water mixture is predicted to be marginally effective at dissolving lignin, Fig. 1 e-g (Ribeiro et al., 2020; Dogenski et al., 2020).

Cylindrical specimens of lignin biocomposite (25.4 mm diameter, 52.0 mm height) were manufactured as described in Section 2.3, "Fabrication of lignin biocomposite." Specimens were desiccated either in the open under ambient conditions or in an oven at elevated temperatures. For oven-assisted desiccation, specimens made using DMSO were desiccated at 60 °C, while specimens made using acetic acid were desiccated at 40 °C, to stay below the flash point of acetic acid (Eidman, 2001). The degree of desiccation was monitored through gravimetric means. Complete desiccation is defined as no further change in weight of the specimen over the course of serial measurements, i.e., the weight plateaus.

Fig. 5a shows the impact of a mixed solvent composed of water and acetic acid on the time course of desiccation of the lignin-based composite. For all specimens studied, a consistent mix design was used, maintaining a total composition of 76.4% w_{soil}, 11.8% w_{BP}, and 11.8% w_{Solv}, resulting in a biopolymer-to-soil ratio of 15.5%. The solvent consisted of a mixture of acetic acid and water at a solvent concentration of 15.5%, corresponding to a biopolymer solution concentration of 50.0% $(w/w)_{BP}$. The ratio of biopolymer solution to minimum void volume ($V_{BP\ sol.}/V_v^{min}$) ranged from approximately 0.89 to 0.93.

Lignin-based BSC made with 100% DMSO took more than 2 weeks to desiccate (completely remove all solvent) in a 60 °C oven. The time course for desiccation, however, can be rapidly accelerated when a more volatile solvent such as acetic acid or water is used. Lignin biocomposite specimens made using an acetic acid–water mixture take only 2 days to desiccate completely in an oven. This is not surprising, as acetic acid (vapor pressure approximately 35.18 mmHg at 40 °C) and water (vapor pressure approximately 55.4 mmHg at 40 °C) are much more volatile than DMSO (vapor pressure approximately 5.75 mmHg at 60 °C), respectively (Jakli and Van Hook, 1972; Wexler, 1976; MacDougall, 1936).

As shown in Fig. 5a, it appears that a substantial proportion of solvent (acetic acid) was retained in the biocomposite (comparing the blue trace to the gray trace), even after a prolonged period of desiccation. This can best be explained by a permanent incorporation of the solvent into the biocomposite, a result of an esterification reaction between acetic acid and the lignin molecule. Acetylation of lignin hydroxyl groups in the presence of acetic acid, even at ambient temperature, has been well described in the literature (de Oliveira et al., 2020; Johansson et al., 2023; Dehne et al., 2016; Zhao et al., 2017; Shakoor Shar et al., 2023). Lignin that undergoes esterification has been shown to have higher tensile strength, higher impact strength, greater thermal stability, and increased moisture repellence than unacetylated lignin Johansson et al. (2023), Dehne et al. (2017). When an oven was used to aid in the desiccation process, a further increase in solvent incorporation into the biocomposite was observed, as shown by the comparison between the orange and blue traces. When no oven is used, 80.4% of the solvent is removed from the composite, however, when an oven is used only 77.2% of the solvent is removed. This small difference appears to be significant, because the error envelopes for these two traces are non-overlapping as shown in Fig. 5a. Its likely that acetylation is going on in both cases, but when an oven is used to aid in desiccation, acetylation is enhanced such that less acetic acid can be removed from the biocomposite. Some acetylation likely occurs during the desiccation process even when an oven is not used. Elevated temperatures are known to enhance esterification reactions (Ding et al., 2012).

When lignin biocomposite is manufactured with higher percentages of acetic acid in the solvent, higher compressive strength is achieved (Fig. 5b). Higher solubility of lignin, as acetic acid percentage is increased, is likely the dominant explanation for this phenomenon. Although our mix design called for 50% solubility of lignin, we did not achieve this degree of solubility as acetic acid percentage was reduced. Determination of the solubility of lignin is challenging, because lignin is dark in color and it is difficult to reliably identify precipitated lignin. From simple laboratory-based estimates of solubility (involving lignin and solvent only), it appears that 50% solutions of lignin can be achieved in 100% acetic acid and nearly achieved in 80% acetic acid. Solubility tests consisting of mechanical mixing of lignin and solvent in test tubes were conducted to estimate the solubility limit. In aqueous solvent containing 60% acetic acid, lignin is clearly not well dissolved at the level required for our mix design at ambient temperatures. With acetic acid content 40% or less, lignin is not well dissolved at ambient temperatures. When an oven is used to aid in the desiccation process, a significant increase in compressive strength is seen at all concentrations of acetic acid 40% or higher. The observed increase in compressive strength may be associated with greater lignin solubility at higher temperatures and enhanced acetylation, as suggested by FTIR analysis and solubility tests, although direct quantification of these effects was not performed in this study. Acetylation modifies the lignin matrix itself by making it more hydrophobic. Enhanced hydrophobicity of the lignin chains may increase lignin–lignin association, and may improve the adhesion to the aggregate particles. The most dramatic increase in compressive strength was seen in specimens manufactured with 40% acetic acid, for which we suspect the dominant factor is an increase in the solubility of lignin at 40 °C, compared to ambient conditions.

In Fig. 5c, the density (unit weight) of the lignin biocomposite also increases with the percentage of acetic acid in the solvent. Unlike compressive strength, the unit weight of the lignin biocomposite is not significantly affected by the temperature of desiccation, as the unit weights of lignin biocomposite were statistically equivalent for

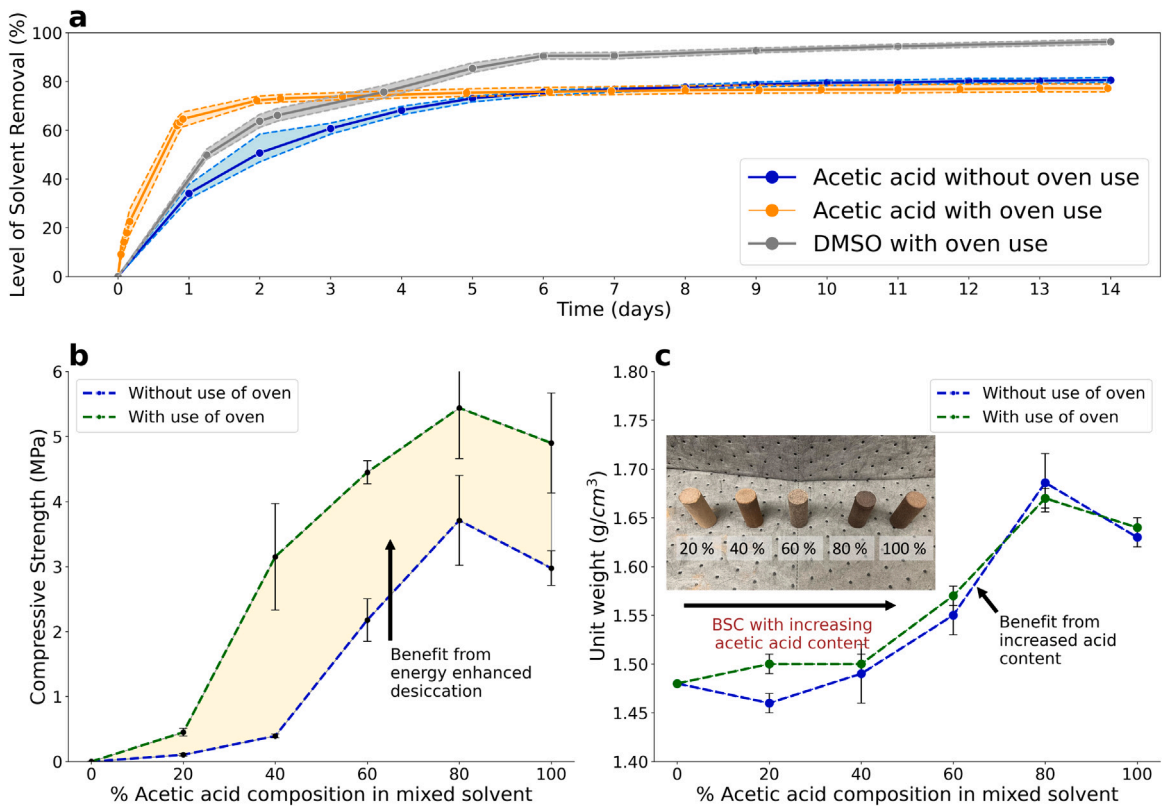

Fig. 5. An acetic acid–water mixture is an alternative to DMSO in the manufacture of lignin biocomposite. **a**, Desiccation occurs more rapidly with a volatile solvent such as acetic acid as compared to DMSO. In addition, the final level of desiccation for lignin-based BSC made using aqueous acetic acid plateaus at approximately 80%, indicating that some of the solvent molecules likely become incorporated into the material, possibly as a result of acetylation of lignin. **b** & **c**, There is a strong dependence of compressive strength and density (unit weight) on acetic acid content in the solvent mixture. Low acetic acid concentrations are likely insufficient to fully dissolve the lignin, as shown in Table 4. This is expected, as low acetic acid content results in a high Relative Energy Difference, which would predict insolubility. In addition, these panels show a notable benefit from oven desiccation. The application of heat during the desiccation process likely enhances the acetylation of lignin in the presence of acetic acid.

Table 4
Results of lignin-based BSC using Mixed Solvent System.

Mixed Solvent System	Fresh unit weight γ_{BSC}^{fresh} (g/cm^3)	Fresh bulk soil density $\rho_{bulksoil}^{fresh}$ (g/cm^3)	Unit weight γ_{BSC} (g/cm^3)	Dry bulk soil density $\rho_{bulksoil}^{dry}$ (g/cm^3)	Compressive strength σ_c (MPa)
Mixed solvent BSC without oven use					
20% acetic acid and 80% water	1.63 ± 0.02	1.24	1.46 ± 0.01	1.26	0.10 ± 0.02
40% acetic acid and 60% water	1.64 ± 0.03	1.25	1.49 ± 0.03	1.29	0.39 ± 0.03
60% acetic acid and 40% water	1.70 ± 0.01	1.30	1.55 ± 0.02	1.34	2.18 ± 0.33
80% acetic acid and 20% water	1.85 ± 0.02	1.41	1.74 ± 0.15	1.50	3.71 ± 0.69
100% acetic acid	1.81 ± 0.09	1.38	1.63 ± 0.01	1.42	2.975 ± 0.27
Mixed solvent BSC with oven use					
20% acetic acid and 80% water	1.68 ± 0.02	1.28	1.50 ± 0.01	1.30	0.45 ± 0.06
40% acetic acid and 60% water	1.67 ± 0.02	1.27	1.50 ± 0.01	1.29	3.15 ± 0.82
60% acetic acid and 40% water	1.74 ± 0.02	1.33	1.57 ± 0.01	1.36	4.45 ± 0.18
80% acetic acid and 20% water	1.83 ± 0.02	1.40	1.67 ± 0.01	1.45	5.44 ± 0.78
100% acetic acid	1.77 ± 0.01	1.36	1.64 ± 0.01	1.42	4.90 ± 0.77

all acetic acid–water mixtures used in this study. The increase in both compressive strength and unit weight is likely due to higher acetic acid content having a greater ability to dissolve the lignin biopolymer, resulting in the formation of more lignin cross-links that pull the aggregate particles together. Notably, the compressive strength and density values achieved for the lignin-based BSC are comparable to those of typical lightweight concrete, indicating that the material exhibits sufficient mechanical performance for potential non-structural building applications (Mohammed and Hamad, 2014; Thienel et al., 2020).

3.2. Observing acetylation of lignin through fourier transform infrared spectroscopy

To confirm that acetylation of lignin is taking place, we performed Fourier transform infrared spectroscopy (FTIR) of lignin combined with pure acetic acid in comparison to lignin combined with pure DMSO (Fig. 6). FTIR analysis was conducted on specimens that had undergone desiccation: for lignin mixed with DMSO, this corresponded to near-complete solvent removal, while for lignin mixed with acetic acid, desiccation reached a plateau at approximately 80% solvent removal.

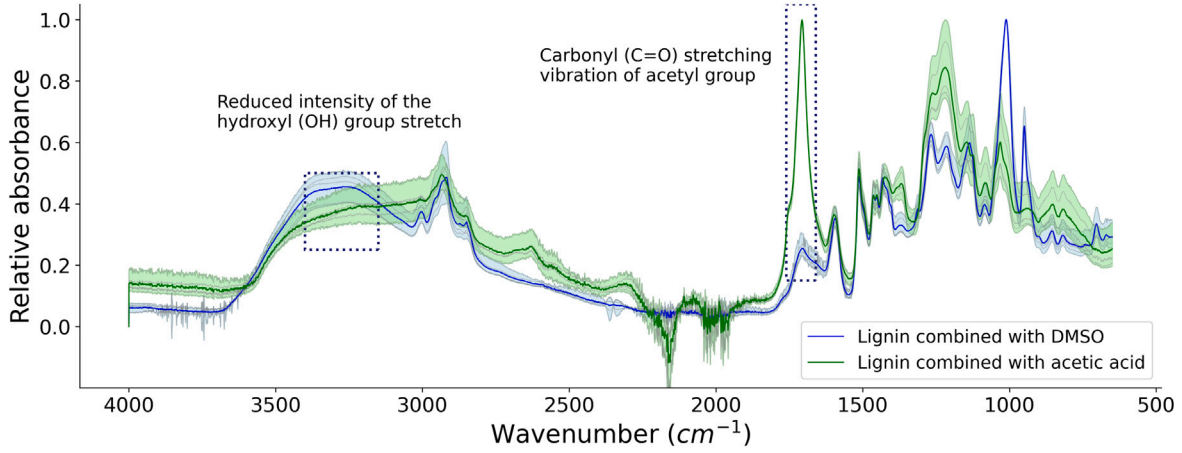

Fig. 6. Fourier Transform Infrared Spectroscopy (FTIR) analysis of lignin combined with pure acetic acid and of lignin combined with DMSO. To investigate the possibility of lignin acetylation (esterification), we combined lignin with pure acetic acid and then removed unreacted acetic acid by thorough drying in an oven. The FTIR spectrum after exposure of lignin to acetic acid revealed a prominent peak of absorbance at approximately 1730 cm^{-1}, which corresponds to carbonyl stretching. By comparison, lignin exposed only to DMSO showed no significant peak at this wavenumber. In addition, a reduction in intensity of the hydroxyl group stretch region of the spectrum, comparing acetic acid treatment to DMSO treatment, was observed. Both of these findings are consistent with acetylation of the lignin molecule in the presence of acetic acid.

A set of five independent samples was analyzed for each condition. presents the mean absorbance profiles along with an envelope representing the full range of absorbance values observed across the replicates. Relative absorbance was plotted to highlight characteristic spectral features, including changes in the hydroxyl group stretching region and the carbonyl group stretching region, the latter indicating the acetylation of lignin.

The FTIR spectrum of lignin mixed with pure acetic acid, after exhaustive desiccation to remove unreacted acetic acid, shows clear evidence of ester formation, indicated by the emergence of a new absorbance peak at approximately 1730 cm^{-1}, corresponding to carbonyl (C=O) stretching. Additionally, a reduction in the intensity of hydroxyl group peaks further supports the occurrence of esterification (Rashid et al., 2016).

3.3. Scanning electron microscopy for visualizing microstructure of lignin biocomposite

Scanning electron microscopy (SEM) analysis (Fig. 7) was used to examine the microstructure of lignin-based BSC. Panel **a** shows the raw aggregate used in fabrication, while panel **b** highlights the lignin-based composite. The SEM images reveal that the lignin biopolymer plays a dual role in composite formation: it forms inter-particle bridges between aggregates, which contribute to a tightly connected and solidified structure, and also coats individual aggregate particles. This coating may enhance recyclability by preventing permanent bonding. Notably, shrinkage cracks observed in panel **b** exhibit internal bridging, suggesting that lignin contributes to structural cohesion even at micro-crack levels. However, visible cracks at the interfaces and within biopolymer bridges indicate potential points of mechanical weakness.

3.4. Environmental impacts of lignin biocomposite using an acetic acid–water mixture

A cradle-to-gate life cycle assessment (LCA) was conducted to evaluate the environmental impacts of using an acetic acid–water mixture as the solvent in the fabrication of lignin-based BSC. We chose 1 kg of lignin biocomposite as the functional unit of our investigation using a mixture of 80% acetic acid and 20% water as the solvent. Fig. 8 shows the scope of the LCA, which covers all activities from the cradle (i.e., impacts associated with sourcing the "raw" constituents of lignin biocomposite) to gate (i.e., impacts associated with the manufacture of lignin biocomposite) for the manufacture of lignin-based BSC.

Lignoforce lignin is isolated from kraft black liquor using the LignoForce system (Kouisni et al., 2012). Kraft lignin is a by-product of the sulfate pulping process. Black liquor, the residual stream from this process, contains a mixture of lignin, hemicellulose, inorganic chemicals, and degraded extractives. In the LignoForce system, lignin is extracted from black liquor through a two-step process involving oxidation with oxygen followed by acidification, which precipitates the lignin for recovery. In this study, sulfate pulp is used as a surrogate to represent the environmental impacts associated with kraft lignin production. Accordingly, the input related to lignin is based on the amount of pulp required to produce the desired quantity of lignin in the lignin-based BSC, using a 1:3 mass ratio of lignin to sulfate pulp (Novaes et al., 2010).

Substituting acetic acid–water mixtures in the place of DMSO does not change the overall manufacturing process of lignin-based BSC (Miao et al., 2024c). The acetic acid–water mixture results in a shorter desiccation time (2 days instead of 14 days). The process flow diagram detailed in Fig. 8 provides the basis for calculating the carbon footprint of lignin-based BSC. Environmental impacts covering impacts related to human health, ecosystem quality, and climate change (carbon footprint) are investigated in this LCA, according to Eq. (2).

Fig. 9 shows the carbon footprint associated with the manufacture of 1 kg of lignin biocomposite using an acetic acid–water mixture, compared to the use of DMSO, across different geographic regions, where distance from sources of lignin to manufacturing sites plays a strong role in the environmental impact of this novel material. The use of a mixed solvent (80% acetic acid and 20% water) has the potential to reduce the carbon footprint of lignin-based BSC in more remote regions, as only the acetic acid needs to be transported to the site since water should be readily available. Additionally, the use of a more volatile solvent to manufacture the composite further reduces the carbon footprint, as less energy is needed to complete the dry-out.

These benefits, however, are outweighed by the higher carbon footprint of the manufacture of acetic acid, resulting in lignin biocomposite made with a mixed solvent having a greater carbon footprint than that made with pure DMSO when no solvent re-capture is performed (Fig. 9a and Fig. 9b). Solvent re-capture can be achieved by condensing the vaporized solvent back into liquid form for reuse in subsequent BSC production (Miao et al., 2024c). Figs. 9c and 9d show the carbon footprint of lignin-based BSC under full solvent re-capture conditions.

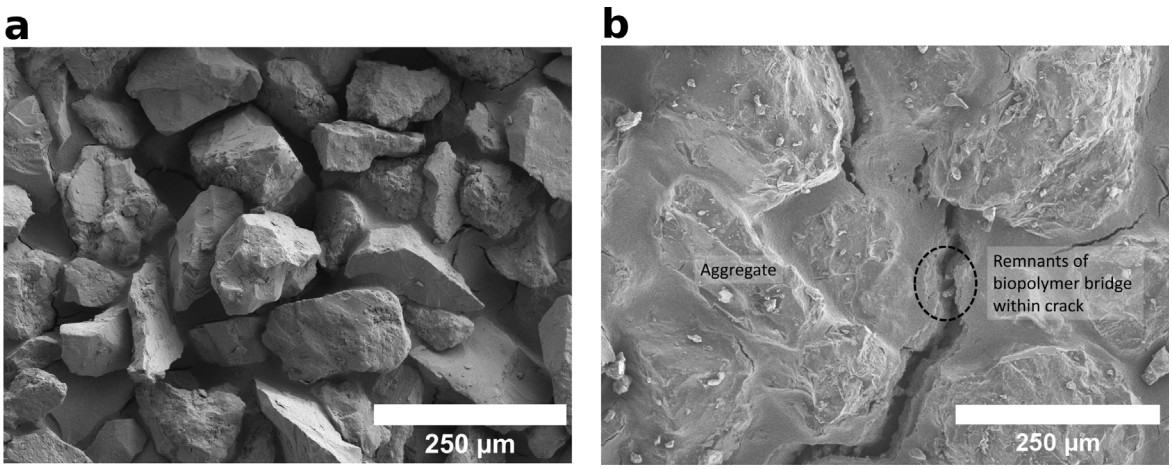

Fig. 7. Lignin biopolymer helps bind together granular material to form lignin biocomposite. **a**, We used a fine-grain aggregate, Grade 90 silver sand, to manufacture the biocomposites. **b**, These aggregates are bound together by a lignin biopolymer, which forms a matrix that connects the granular material to create a composite (made using a mixture of 80% acetic acid and 20% water). The image shown here depicts fragments of the biocomposite after failure in compression. Failure occurs when cracks develop at the interface between the biopolymer bridge and an aggregate, then propagate toward the bridge, eventually linking with other cracks in the material and resulting in overall failure. In addition, remnants of biopolymer bridges are visible within the crack (the small granules bridging the crack).

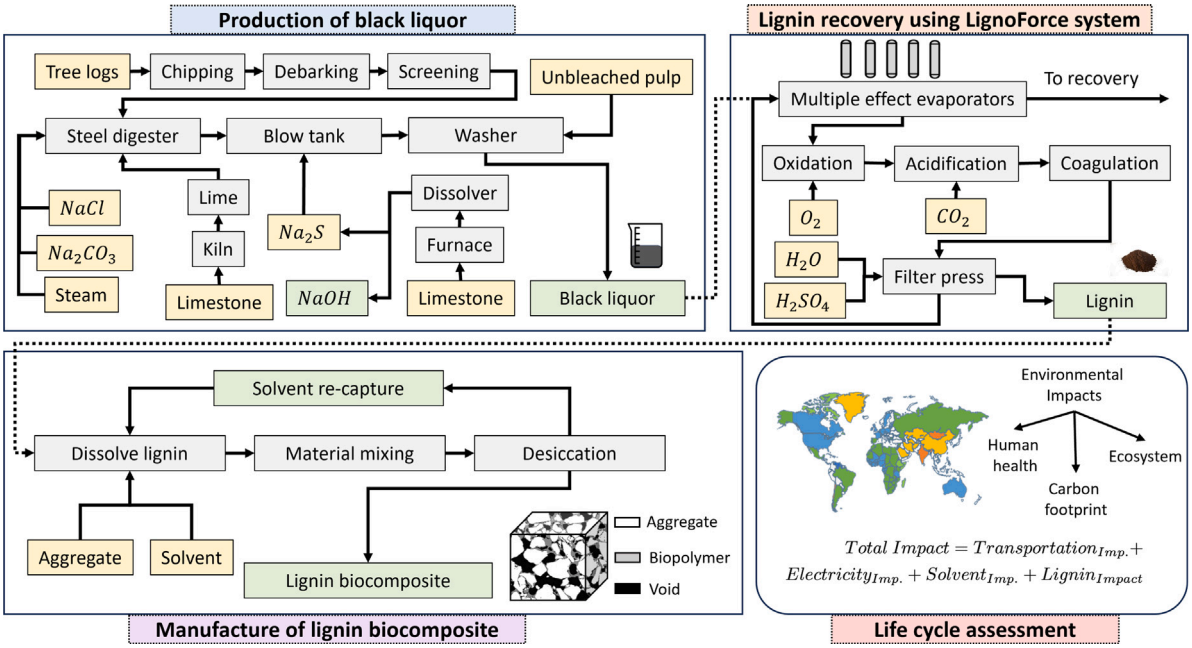

Fig. 8. For this investigation, we selected Lignoforce lignin as the biopolymer for manufacturing lignin-based biocomposite structural concrete (BSC). Lignoforce lignin is a variant of kraft lignin, produced using the LignoForce system to isolate lignin from kraft black liquor (Kouisni et al., 2012; Hubbe et al., 2019; Mastrolitti et al., 2021). Although kraft lignin (e.g., Lignoboost lignin and Lignoforce lignin) is typically considered a by-product of the sulfate pulping process, to account for scenarios where lignin is produced as the main product, we used sulfate pulp as a surrogate for lignin. To manufacture the lignin-based BSC, we used an electrically powered mixer and oven to mix and desiccate the composite, respectively. In the BSC, the biopolymer coats the aggregate particles and forms interparticle connections through a biopolymer-bridging matrix, resulting in a three-phase material composed of aggregates, biopolymer, and voids (Miao et al., 2025b). To assess the environmental impacts associated with manufacturing the lignin biocomposite, a cradle-to-gate life cycle assessment (LCA) was conducted. The functional unit for this assessment was 1 kg of lignin biocomposite. Our analysis evaluated multiple environmental metrics, including human health, climate change (carbon footprint), and ecological quality.

In this case, the carbon footprint of BSC made with the mixed solvent is lower across all regions of the world, as compared to BSC made with pure DMSO.

In most regions of the world, the manufacture of lignin biocomposites is less carbon-intensive than the production of many conventional construction materials. For instance, producing 1 kg of lightweight concrete results in at least 0.17 kg of carbon emissions (Miao et al., 2024c). As a result, even in cases where solvent re-capture is not performed, the carbon footprint of lignin-based BSC generally remains lower than that of traditional alternatives, ranging from 0.24 kg to −0.09 kg CO_2 per kg of BSC. In the world maps shown in Fig. 9, lignin is assumed to be the main product of its production process (i.e., the lignin waste stream bears the full environmental burden). This is a conservative assumption, as lignin is typically treated as a waste product from the paper and pulp industry or bioethanol production (Miao et al., 2024c). However, future large-scale manufacturing of lignin biocomposites may

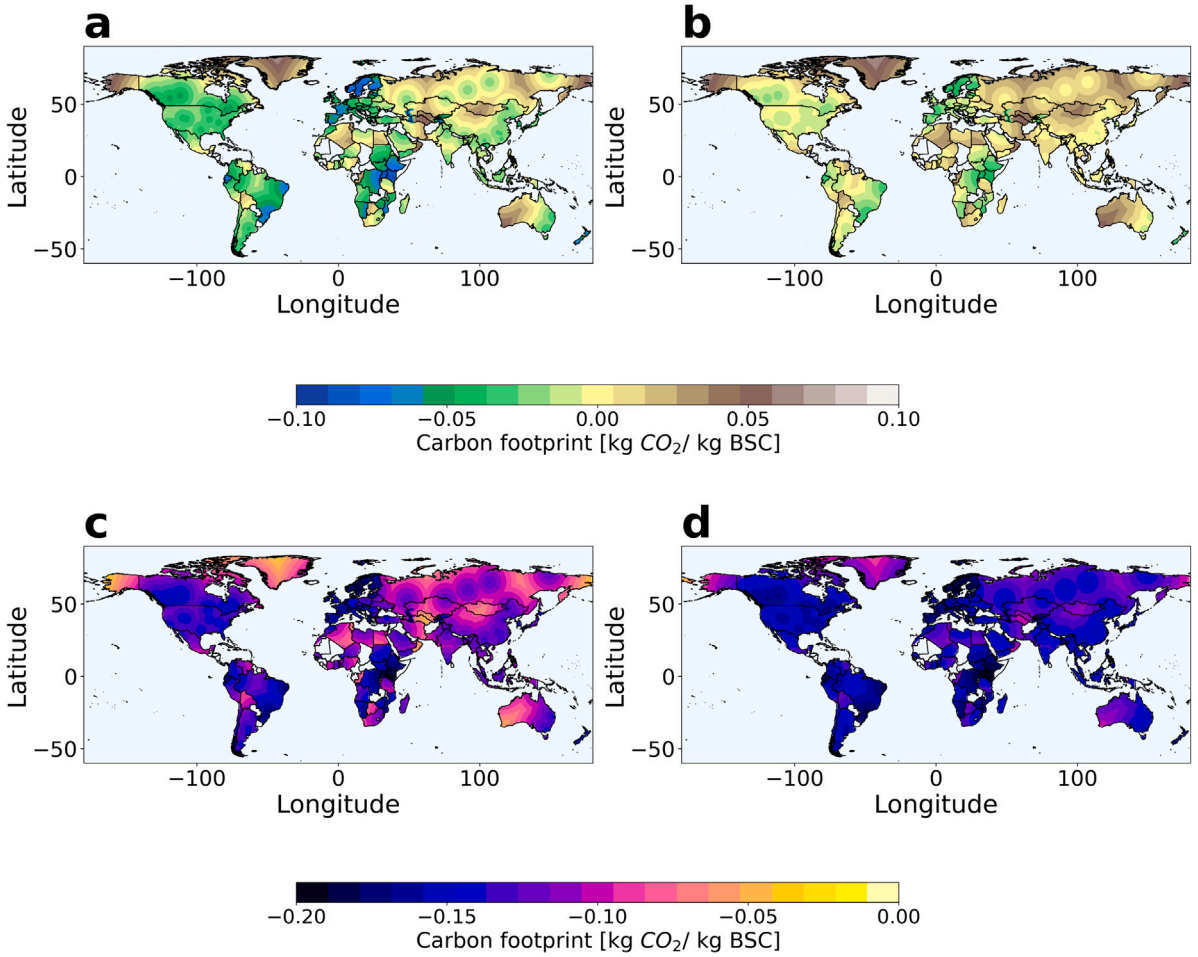

Fig. 9. Worldwide carbon footprint for manufacturing 1 kg of lignin biocomposite using aqueous acetic acid in comparison to using DMSO. **a**, Nature-based biocomposites made using lignin with pure DMSO as a solvent are net carbon-negative in most regions of the world, even when no solvent recapture is performed. This is especially true in areas where lignin is locally available. **b**, The use of an acetic acid–water mixture (80% acetic acid and 20% water), however, for manufacturing lignin biocomposites results in a higher carbon footprint when the solvent is not recaptured. If solvent recapture is performed, lignin biocomposites made using a mixed solvent (**d**) exhibit a lower carbon footprint than those made with pure DMSO (**c**). This reduction is primarily due to the decreased energy demand for the desiccation process when using more volatile solvents such as acetic acid and water, as compared to DMSO, thereby enhancing the carbon-negative potential of the material.

involve more intentional lignin production, where lignin becomes a co-product or even the primary product.

Fig. 11 shows the estimated environmental impacts associated with the production of 1 kg of lignin-based BSC. Using the known locations of potential lignin sources (Fig. 2), we calculated transportation distances on a global scale (Fig. 10), and found that the "most likely" transportation distance for lignin during the manufacture of the biocomposite was approximately 1009 km. Similar to Figs. 11c and 11d, we accounted for solvent re-capture during the manufacture of the material.

The environmental midpoint impacts of lignin biocomposite are shown by the blue bars – representing its manufacture using DMSO—and the green bars – representing its manufacture using an acetic acid–water mixture. If lignin is considered a true waste product, the lower bars for both solvents are more representative, as they correspond to scenarios where lignin "bears no burden." Conversely, if lignin is produced as the main product, it will "bear all the burden" of its production. The upper bars represent this case. While lignin is currently a waste product of the pulping and bioethanol industries, the potential for large-scale manufacture of lignin-based products may shift its status, requiring it to bear a portion of the production burden. For this LCA, the environmental impact associated with lignin is accounted for based on its production via the pulping process (Miao et al., 2024c).

The range between the upper and lower bars represents the range in environmental impacts under different allocation scenarios. For example, if lignin is no longer considered a pure waste product, several allocation methods can be applied, such as allocation based on the mass of co-products, allocation based on the energy content of co-products, the marginal approach, among others (Hermansson et al., 2020). The environmental impacts of lignin biocomposites manufactured using DMSO and an acetic acid–water mixture are also compared to those associated with the production of various types of lightweight concrete (e.g., expanded clay, pumice, and perlite), as indicated by the colored dots in the LWC column.

In general, the manufacture of lignin biocomposites results in lower impacts on human health (Fig. 11a-e), ecosystem quality (Fig. 11f-k), and climate change (Fig. 11l) compared to the production of conventional building materials such as lightweight concrete. Although the impacts of lignin biocomposites may be higher than the lightweight concretes in some scenarios (as indicated by the upper bars), greater emphasis should be placed on the impacts represented by the lower end of the range, as lignin, currently, is treated as a waste product and is allocated minimal footprint.

In terms of human health impacts, the manufacture of lignin biocomposites results in lower emissions of carcinogenic substances in comparison to lightweight concrete and other established building

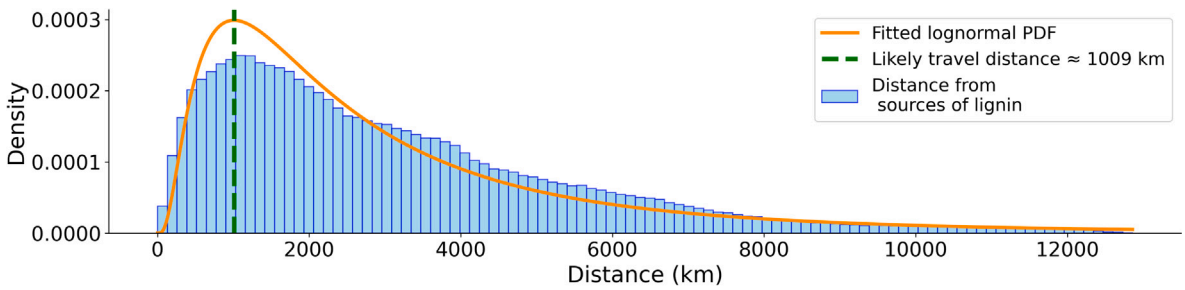

Fig. 10. Distance from sites of manufacture to potential sources of lignin. To estimate the most likely environmental impacts associated with the manufacture of the lignin biocomposite, we fit a lognormal distribution to the histogram representing typical transport distances for lignin. From the resulting probability density function, the mode – or most probable travel distance – was determined to be approximately 1009 km. This value was used as the representative transport distance in the life cycle assessment to calculate the environmental impacts shown in Fig. 13.

Table 5
Environmental impact from manufacturing lignin biocomposite using DMSO and an acetic acid–water mixture.

Impact Category	Lignin-based BSC		Lightweight Concrete (LWC)		
	DMSO	AA-H_2O	Clay	Perlite	Pumice
Carcinogens (kg C_2H_3Cl)	5.0E−4 – 5.4E−3	3.3E−4 – 5.3E−3	3.5E−4	2.00E−2	−6.5E−4
Non-carcinogens (kg C_2H_3Cl)	2.2E−3 – 1.4E−2	2.0E−3 – 1.4E−2	0.013	0.031	0.0039
Respiratory inorganics (kg PM2.5)	6.0E−5 – 4.2E−4	4.9E−5 – 4.1E−4	2.1E−4	2.5E−4	4.4E−4
Ozone depletion (10^{-7} kg CFC-11)	2.7E−10 – 8.2E−9	1.8E−10 – 8.1E−9	4.8E−9	9.2E−8	6.9E−9
Respiratory organics (kg C_2H_4)	1.8E−5 – 1.2E−4	1.6E−5 – 1.1E−4	1.2E−4	3.7E−4	9.8E−5
Aquatic ecotoxicity (kg TEG water)	14 – 57	14 – 57	89	230	15
Terrestrial ecotoxicity (kg TEG soil)	2.3E−2 – 15	1.5E−2 – 15	31	81	3.9
Terrestrial acidity (kg SO_2)	1.9E−3 – 6.0E−3	1.6E−3 – 5.8E−3	5.7E−3	0.013	5.4E−3
Land occupation (m^2 arable)	2.0E−6 – 0.15	1.4E−6 – 0.15	0.096	0.24	0.019
Aquatic acidity (kg SO_2)	5.0E−4 – 1.5E−3	3.9E−4 – 1.4E−3	9.1E−4	0.0023	0.0011
Aquatic eutroph. (kg PO_4 P-lim)	3.1E−7 – 10.1E−5	2.7E−7 – 10.09E−5	5.4E−5	1.2E−4	3.1E−5
Global warming (kg CO_2)	−0.23 – −0.14	−0.24 – −0.15	0.17	0.34	0.31

materials. For instance, the production of clay-fired bricks has been linked to significant health issues, particularly in developing countries, where it contributes to respiratory problems—especially among children under the age of five (Brooks et al., 2023). Therefore, the manufacture of lignin-based BSC not only supports the decarbonization of the construction industry but also helps mitigate the release of harmful air pollutants that negatively impact the health of surrounding communities.

In terms of life cycle activities (Fig. 12), the proportion each process contributes to the environmental impacts of producing lignin biocomposite depends on the allocation scenario considered for lignin production. For instance, in scenarios where lignin is considered a waste product (as represented by the lower bars in Fig. 11), most of the environmental footprint is attributed to the transportation of ingredients to manufacturing sites and the energy required for material processing (e.g., mixing). In scenarios where lignin is considered the main product (as represented by the upper bars in Fig. 11), however, the production of lignin contributes the majority of the environmental impacts associated with manufacturing this novel material.

Therefore, in the near term, improvements to the manufacturing of lignin biocomposites should focus on the use of locally sourced materials and enhancing the energy systems that power production. However, in the long run, attention should shift toward improving the lignin production process itself, as it becomes the primary contributor to the environmental impacts associated with producing the lignin biocomposite.

3.5. Techno-economic assessment of lignin biocomposite using an acetic acid–water mixture

We conducted a techno-economic assessment to determine if lignin biocomposite is an affordable material. Fig. 13 illustrates the economic costs associated with the manufacture of lignin biocomposites, with expenses attributed to individual raw materials and production processes. Although lignin-based BSC produced using DMSO is a carbon-negative building material, it is not economically competitive compared to conventional construction materials such as concrete. This is primarily due to the relatively high cost of DMSO compared to more affordable alternatives. Moreover, the DMSO-based process requires substantially more energy, as it involves 14 days of desiccation, whereas the use of an acetic acid and water solvent system reduces desiccation to just 2 days. The acetic acid and water mixture, being both volatile and inexpensive, lowers energy requirements and raw material costs, thereby improving the economic feasibility of the biocomposite.

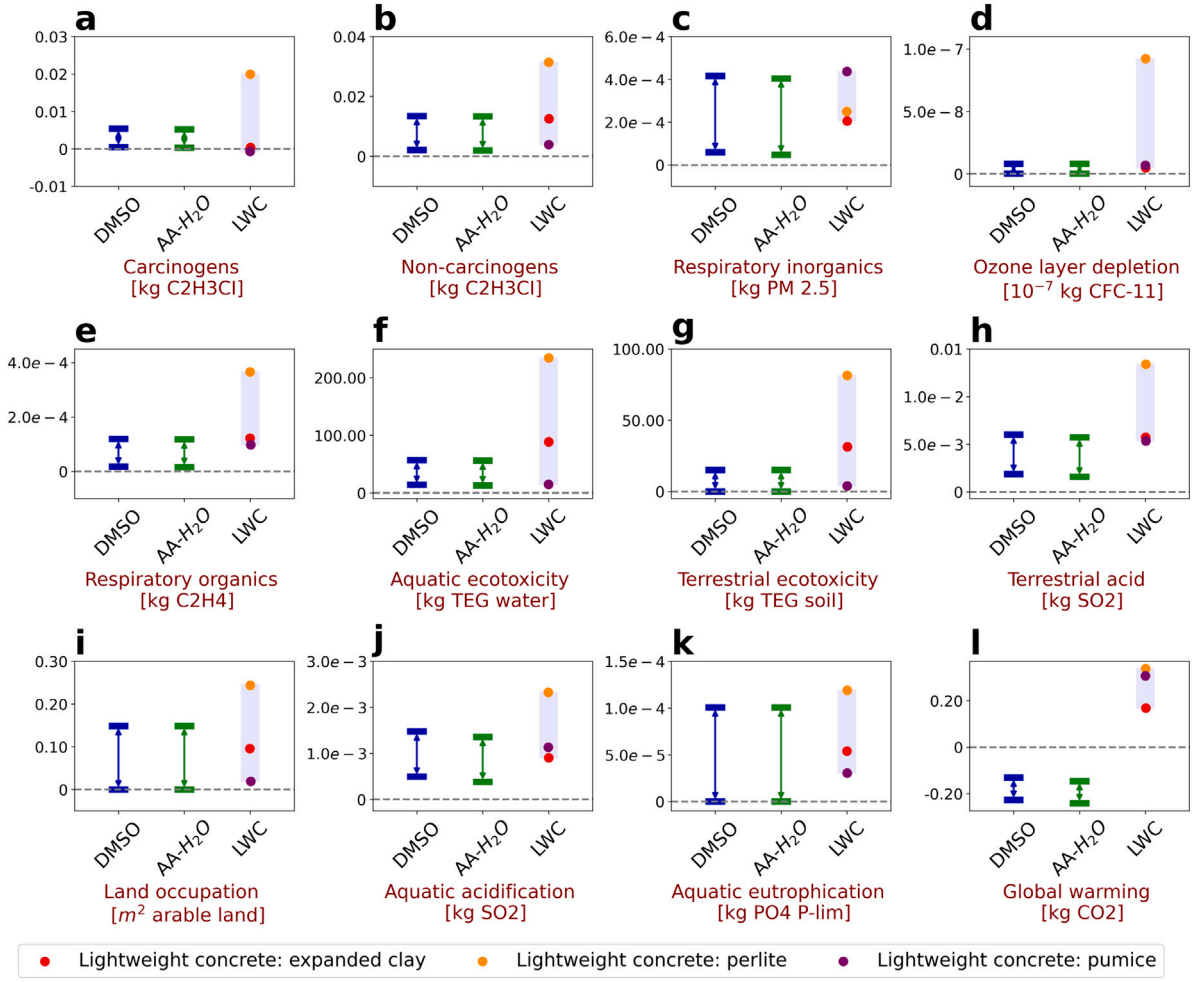

Fig. 11. Environmental impact characterization associated with the fabrication of 1 kg of lignin biocomposite. Environmental impacts from lignin biocomposite manufacturing were assessed using the Impact 2002+ method (Jolliet et al., 2003). We compared biocomposite produced using 100% DMSO (blue bars, "DMSO") and biocomposite produced using an acetic acid–water mixture (green bars, "AA-H_2O"). Panels **a–e** show *"human health"* impacts, panels **f–k** show *"ecosystem quality"*, and panel **l** shows *"climate change"* impact. For each panel, the upper bars (for lignin-based BSC made with 100% DMSO or an acetic acid–water mixture) represent the environmental impact for allocation scenarios where lignin bears the entire burden of their production (lignin becomes the main product), while the bottom bars represent scenarios where lignin is considered a waste product (i.e., it bears no burden). The range between the upper and lower bars represents the impacts of intermediate allocation scenarios (e.g., allocation based on the mass or energy content of co-products). Additionally, the environmental impacts of various lightweight concretes – including expanded clay, pumice, and perlite types – are shown. Compared to lightweight concrete, the manufacture of lignin biocomposites results in lower environmental impacts. This environmental benefit can be further enhanced by using a more volatile solvent (i.e., a mixture of acetic acid and water). The reduction in impact is indicated in each panel by the ranges and is summarized in more detail in Table 5.

Lignin biocomposites produced with the acetic acid–water system are significantly cheaper than those produced using DMSO, with an estimated cost range of US$122/m^3 to US$237/m^3, compared to US$409/m^3 to US$933/m^3 for the DMSO-based counterpart. In most of the countries considered, the cost of lignin biocomposites using the acetic acid–water process is now comparable to, or even lower than, several commercially available concrete options. These findings highlight the critical role of solvent selection in enabling lignin biocomposite to become a cost-effective material for decarbonizing the built environment.

4. Discussion

Solubility of the biopolymer is the first consideration in designing novel nature-based biocomposites, as solubility is essential to enable the biopolymer to be used as an effective binder. If the biopolymer is not fully soluble at the concentration required for a desired mix, the result will be that undissolved biopolymer merely functions as a bystander in the material and does not become bonded to the aggregate. If the goal is to optimize for mechanical strength or environmental impact, additional experimental testing is required to account for these other influencing factors.

The Hansen solubility approach can serve as a preliminary screening tool for identifying viable candidate solvents and providing rough estimates of solvent compositions likely to dissolve lignin effectively. For instance, the Hansen solubility approach predicts that lignin would be borderline soluble in solvents containing 60% or less acetic acid. We confirmed the marginal solubility of lignin in 60% acetic acid using simple solubility tests performed in the laboratory. To determine the "optimum" solvent composition, however, experimental tests are necessary to evaluate the physical properties of the lignin biocomposite.

Through our solvent optimization approach, we arrived at 80% acetic acid in water as being an effective compromise. The organic acid, better than water by itself, helps to dissolve the somewhat hydrophobic lignin biopolymer, and avoiding DMSO helps to speed up the desiccation process, since both acetic acid and water are significantly more volatile. The desiccation time of the lignin biocomposite was reduced

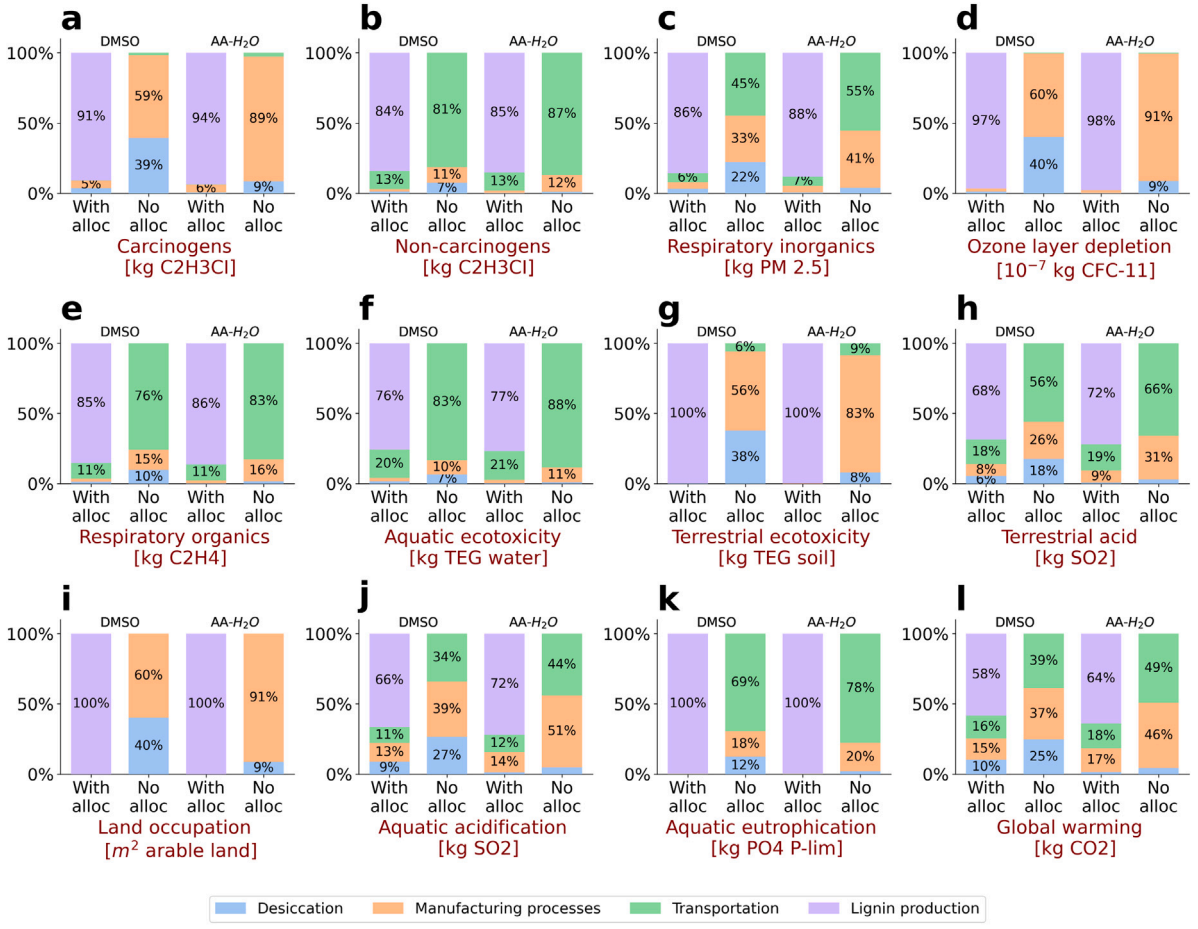

Fig. 12. Environmental impacts associated with production processes. The impacts shown in the figure can be broken down by each process involved in the manufacture of the lignin biocomposite. "With allocation" represents the most conservative approach, where lignin bears the entire burden of its production (i.e., lignin is treated as the main product), and is illustrated by the upper bars in Fig. 5. "No allocation" represents the current state of lignin production, where lignin is considered a pure by-product, and is depicted by the lower bars in Fig. 7. Under the current production model, the main contributors to environmental impacts – namely human health, ecosystem quality, and climate change – are associated with activities related to the manufacture of the lignin biocomposite (e.g., transportation of raw ingredients to the manufacturing site, energy required for desiccation, and energy used for mixing). In the event of a scale-up in lignin biocomposite production, however, the majority of environmental impacts would shift to being associated with lignin production itself.

from 14 days to as little as 2 days, simply by substituting a more volatile mixed solvent for DMSO. This substantial reduction in desiccation time broadens the potential applications of lignin biocomposite. For instance, with this faster drying capability, the material can now be considered for time-sensitive applications such as *in situ* construction, where rapid desiccation is often desirable, if not essential.

By incorporating acetic acid into the solvent for lignin, we observed an unexpected result, namely the acetylation of the lignin molecule, which was confirmed by infrared spectroscopy. The incorporation of acetate into the biocomposite, at concentrations of acetic acid that were sufficient to dissolve the biopolymer, resulted in an enhancement of compressive strength of the material. We can attribute the observed strength improvement from acetylation primarily to changes within the biopolymer phase of the material. It remains unclear, however, whether this benefit stems from enhanced strength of the biopolymer bridges themselves — the biopolymer connections between adjacent aggregate particles. Alternatively, acetylation may improve the strength of the interfacial bond between the biopolymer and the aggregate. It is worth noting that preliminary experiments suggest that the dominant mode of failure in BSCs occurs through fracture of the biopolymer bridge, rather than at the interface. Further experimentation, particularly tensile strength testing of the biopolymer phase by itself, will be necessary to tease out the specific role of acetylation in enhancing mechanical performance of the material.

Additionally, our solvent optimization approach contributed to reducing the environmental impact associated with the production of lignin biocomposites, primarily by shortening the oven desiccation time. As such, the solvent-based optimization strategy presented in this study not only improves solvent selection and design for biocomposite development but also enhances the overall environmental performance of the material. Beyond environmental benefits, acetic acid–water provides practical advantages, including reduced energy consumption and accelerated strength development of the final material. Most importantly, this solvent-based optimization helps make lignin biocomposites a financially viable alternative to conventional building materials. By enabling the use of low-cost, volatile solvents and minimizing processing times, our approach establishes a pathway for adoption of cement-free carbon-negative construction materials that are both affordable and accessible. This represents a critical advance in the development of sustainable materials for the built environment, with the potential to transform how we design and construct climate-resilient infrastructure.

5. Conclusions

In this paper, we introduce an approach for solvent selection in the manufacture of a novel nature-based biocomposite. Our method incorporates the Hansen solubility approach to evaluate various candidate

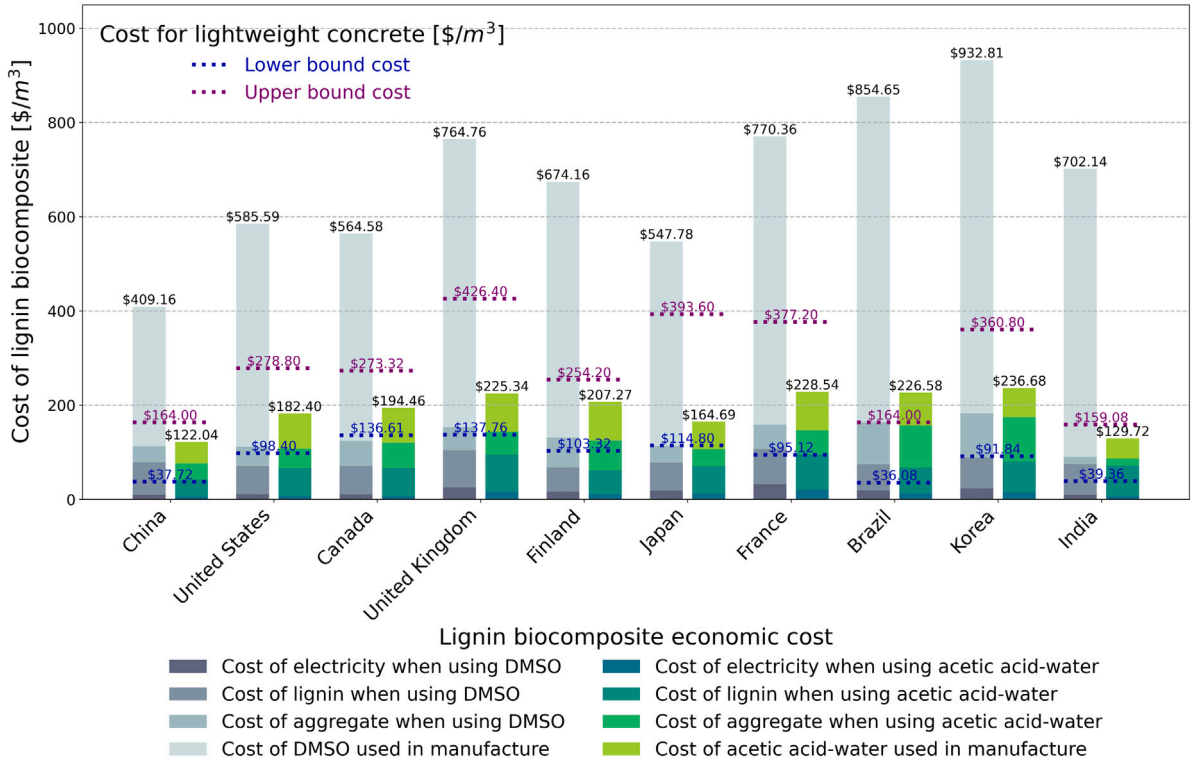

Fig. 13. Economic cost associated with the manufacture of lignin biocomposite in comparison to lightweight concrete. The production costs of lignin biocomposite made using DMSO versus acetic acid–water are broken down by manufacturing processes and raw materials. Dotted blue lines and dotted purple lines indicate the lower and upper bounds of typical concrete prices in the countries studied. Using acetic acid–water as a solvent lowers both material and energy costs, making production more economically viable. In most countries analyzed, lignin biocomposites are comparable in cost to lightweight concrete, and in some cases even cheaper. The shift away from DMSO makes lignin biocomposite a practical and affordable low-carbon alternative for the built environment.

solvents for dissolving lignin. By using this approach, we considered two organic acids, formic acid and acetic acid, as alternatives to DMSO, a solvent we have used in previous studies. We chose to explore acetic acid, because the acetyl group is more likely to interact favorably with the hydrophobic lignin molecule, as compared to the more hydrophilic formate group. A solvent mixture of 80% to 100% acetic acid was able to completely dissolve the lignin at high concentrations, resulting in a material with sufficient strength.

Somewhat unexpectedly, we observed that acetic acid becomes permanently incorporated into the biocomposite during the desiccation process *via* acetylation of the lignin molecule. Incorporation of acetic acid into the biocomposite is associated with an increase in the compressive strength of the material. We attribute this enhancement in compressive strength to either an increase in the biopolymer bridge strength or an increase in the interfacial bond strength between the biopolymer and the aggregate. In addition to the strength enhancement of the material, our optimization approach offers practical advantages to the overall manufacture of lignin biocomposite. For instance, the use of a more volatile solvent helps reduce the time needed for desiccation from 14 days to as little as 2 days. From an environmental perspective, the manufacture of lignin biocomposite generally results in lower impacts on climate, ecosystems, and human health, as compared to conventional building materials.

CRediT authorship contribution statement

Barney H. Miao: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Daniel Woo:** Writing – review & editing, Investigation. **Andrew C. Lesh:** Writing – review & editing, Investigation. **David J. Loftus:** Writing – review & editing, Supervision. **Michael D. Lepech:** Writing – review & editing, Project administration, Funding acquisition.

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.

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Data availability

Data will be made available on request.

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