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#### Highlights

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- · Lignin-based biopolymer bound soil composite (BSC) is a novel green material.
- · Lignin-based BSC is a sustainable alternative to concrete.
- · Lignin-based BSC is a carbon negative construction material.
- · Lignin-based BSC can be designed to meet target life cycle carbon footprint.
- · Lignin-based BSC can be phased in where lightweight concrete is currently used.

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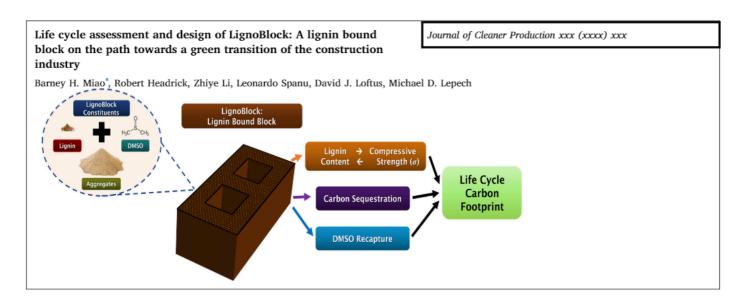


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## Life cycle assessment and design of LignoBlock: A lignin bound block on the path towards a green transition of the construction industry

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#### ABSTRACT

Lignin-based biopolymer-bound soil composites (BSCs) are a new class of sustainable construction materials that utilize a bio-based biopolymer — lignin — as a binder. Prior use of lignin suggests that lignin is a promising candidate for the development of bio-based construction materials. Inspired by these applications, lignin-based BSCs were developed using lignoboost lignin, lignoforce lignin, alkali lignin, and hydrolysis lignin. Uni-axial compressive testing of lignin-based BSC shows that the compressive strength for these BSCs range from 1.6–8.1 MPa, which makes them appropriate for low compressive strength construction applications. We performed a life cycle assessment (LCA) of lignin-based BSC, with the functional unit being a CMU-sized block ( $V = 6423cm^3$ ). The major advantage of BSC lies in the elimination of ordinary portland cement, which is common to many construction materials, including many forms of concrete. Furthermore, the use of lignin lignin-based BSC results in carbon sequestration (lignin  $\approx 60$  wt% carbon), potentially making construction materials made from lignin-based BSC carbon negative. Additionally, a design guide for estimating the life cycle carbon footprint of lignin-based BSC for a required compressive strength was developed. By utilizing the results from material tests and the LCA, designers are now able to use lignin effectively in construction applications, as they can now design lignin-based BSC for a target compressive strength with a full understanding of the life cycle carbon footprint implications.

#### 1. Introduction and motivation

Lignin is a complex organic material, known to be the second most naturally occurring biopolymer behind cellulose, and is a commonly produced as a byproduct from paper pulp and bioethanol production (Bajwa et al., 2019; Mastrolitti et al., 2021). Global production of lignin from the paper and pulp industry is currently around 50–70 million tonnes per year (Collins et al., 2019; Mankar et al., 2022). The production of lignin is projected to increase by 225 million tonnes by 2030, as a result an increase in bio-fuels production mandated by the Renewable Fuel Standard (RFS) (EPA, 2014).

The use of lignin is complicated by the fact that there are many types of lignin, defined by the industrial processes used in their manufacture. The main forms of lignin include: kraft lignin (KL), lignosulfonate lignin, alkali lignin (AL), hydrolysis lignin (HL), organosolv

lignin, and steam explosion lignin (Mastrolitti et al., 2021). Of these main technical types of lignin, kraft lignin is the most common type of lignin. Currently 85% of lignin is kraft lignin produced by purifying kraft black liquor derived from the paper and pulp industry (Mun et al., 2021; Mastrolitti et al., 2021). Currently, three different purification methods exist to precipitate lignin out of kraft black liquor which are the WestVaco, Lignoboost, and Lignoforce processes (Kouisni et al., 2016; Tomani, 2010; Kienberger et al., 2021). Target products that utilize kraft lignin include fertilizers, bio-asphalt, plywood, and other plastics/phenol applications (Mastrolitti et al., 2021; Fakhri and Norouzi, 2022; Moretti et al., 2022a; Van Vliet et al., 2016; Bourzac, 2015). By comparison, lignosulfonate lignin is produced through the older sulfite pulp process. This process is relatively detrimental to the environment in comparison with the production processes of other

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types of lignin (Aro and Fatehi, 2017). Also, by undergoing sulfonation, the solubility of the lignin increases (Mastrolitti et al., 2021). Applications of lignosulfonate lignin include use as binders, wood adhesives, bio-fuels, biopolymers, and dispersing agents (Breilly et al., 2021). Alkali lignin, like kraft lignin, is a by-product of the paper and pulp industry, but is instead generated from the alkaline pulping process (Wang et al., 2021). While alkali lignin is similar in many regards to kraft lignin, they are not identical. Alkali lignin has different molecular properties than kraft lignin, and also more arylglycerol groups and enol ether linkages (Zhao et al., 2019). Alkali lignin is used to make adhesives, sand binders, emulsifiers, and as dispersants (Mastrolitti et al., 2021). Hydrolysis lignin is typically a byproduct of bioethanol production, with the lignin being directly separated from biomass through the application of a dilute acid (Mastrolitti et al., 2021). Production of hydrolysis lignin is expected to increase dramatically in the next few years. Hydrolysis lignin has been used to make bio-asphalt, biofuels, and aviation fuels (Mastrolitti et al., 2021). Organosolv lignin and Steam Explosion lignin are implemented in similar fields as the aforementioned types, but are not as prevalent as kraft lignin and alkali lignin which make up more than 90% of the total current global production of lignin.

Previous efforts to develop BSC have been focused on alternative materials for applications in soil improvement, blocks, pavers, and other construction materials using bovine serum albumin, other blood proteins, starches, gellan gum, and xantham gum (Khatami and O'Kelly, 2013; Chang and Cho, 2014; Chang et al., 2015a; Ayeldeen et al., 2016; Qureshi et al., 2017; Dove et al., 2016; Chang et al., 2015b; Muguda et al., 2017). BSC is made by mixing soil or other aggregate, a biopolymer to bind the aggregate, and a solvent, to create a "green" material that can be formed into a desired shape. Complete desiccation results in the formation of a solid material suitable for many construction applications. Currently, the construction industry contributes approximately 11% of global CO2 emissions (Ahmed Ali et al., 2020; Huang et al., 2018). Because biologically-derived binders are used in all variants of BSC, all are attractive for reducing CO2 emissions associated with conventional construction materials, especially ordinary portland cement concrete. Lignin is an especially attractive candidate biopolymer because lignin is sourced from relatively stable waste streams, and its use leads to carbon entrapment (Obasa et al., 2022; Yang et al., 2024). Carbon sequestration occurs in construction materials that use bio-based materials during their manufacture, such as wood or agricultural residues (Lippiatt et al., 2020; Xi et al., 2016; Kuittinen et al., 2023; Pires, 2019).

## 1.1. Overview of LCAs of lignin-based construction products and types of BSC

To determine the sustainability of lignin-based BSC, a life cycle assessment (LCA) was carried out using SimaPro software using the Impact 2002+ method (Jolliet et al., 2003). This study included a life cycle assessment of the application of three types of technical lignin (kraft lignin, alkali lignin, and hydrolysis lignin) in low compressivestrength construction applications. In our analysis, we made comparisons between various types of lightweight concrete (concrete made with lightweight aggregate) and lignin-based BSC. For the life cycle assessment presented in this paper, we focused on the climate change damage category exclusively (i.e., carbon footprint). Table 1 shows previous LCA studies on various lignin-based construction products and variants of BSC. Our study is the first LCA to examine the use of asproduced lignin as a bulk material to form BSC. Also, our LCA provides an additional benefit by using the results from the life cycle assessment to develop a design guide, which helps users design lignin-based BSC to meet a desired set life cycle carbon footprint.

#### 2. Overview of lignin-based biopolymer-bound soil composites

Lignoboost lignin (LB), lignoforce lignin (LF), alkali lignin, and hydrolysis lignin were used to make BSC samples. Grade 90 sand was used as the aggregate. DMSO was used as the solvent to dissolve these four hydrophobic biopolymers. A stand mixer was used to blend the raw materials until the mixture was uniform. The resulting green material was shaped using a custom molding apparatus (13 mm inner diameter and 76 mm height) made from 316 stainless steel. A steel piston was inserted into the cylindrical mold to perform a double compaction (to 5 MPa), to ensure sample uniformity, using an MTS Criterion model electromechanical testing system. The MTS criterion model 43 was also used to extrude the samples from the mold. The samples were then desiccated in a Fischer Scientific IsoTemp oven at 60 °C until the samples were at least 80%–90% fully desiccated. The compressive strength of the samples were determined using the same MTS Criterion test system.

The compressive strength for these mixes ranged from 3.8–5.7 MPa for lignoboost lignin-based BSC, 2.9–6.0 for lignoforce lignin-based BSC, 3.6–6.8 MPa for alkali lignin-based BSC, and 1.6–8.1 MPa for hydrolysis lignin-based BSC. Fig. 1 shows the range of compressive strengths obtained using four types of lignin, in comparison to the range of results for other types of BSC. According to ASTM International (formerly known as American Society for Testing and Materials), ASTM C129 sets the minimum compressive strength for non-load bearing concrete masonry units at 3.45 MPa, while ASTM C55 specifies 17.2 MPa as the requirement for load bearing concrete masonry units. Based on our results, lignin-based BSC even at this early stage of development, satisfies the minimum compressive strength requirement for non-load bearing construction. With further development, lignin-based BSC may eventually meet the higher compressive strength standard.

#### 3. Life cycle assessment

#### 3.1. Overall considerations

This LCA will investigate the total life cycle carbon footprint incurred by manufacture of lignin-based BSC blocks ("LignoBlock") in comparison to conventional construction materials (i.e., variations of lightweight concrete). For this LCA, we chose the functional unit to be a single block with the same dimensions as a concrete masonry unit (CMU) block ( $V=6424~{\rm cm}^3$ ). Details regarding the manufacture of lignin-based BSC, production of different types of lignin, and methods for estimating the carbon footprint of lightweight concrete are provided. Information about lignin production and LignoBlock manufacture, together with equations defined later in this section, were used to estimate the total life cycle carbon footprint of lignin-based BSC and lightweight concrete.

#### 3.2. Production processes for various types of lignin

Lignin production methods vary for each type of lignin. Hence, individual process flows for the production of each type of lignin are needed to accurately estimate the carbon footprint associated with each type of LignoBlock. Furthermore, the carbon footprint that a waste product, such as lignin, should bear is also a matter to be considered. Prior LCA studies related to lignin have considered a variety of allocation approaches. The manufacture of LignoBlock, however, is the same for all types of lignin. Therefore, a common process flow, for manufacture of LignoBlock was used for all types of lignin under consideration in this study.

Table 1
Select studies covering life cycle assessment of lignin-based composites and BSCs.

Study	Scope of study	Metrics
Roedel et al. (2015)	Materials compared: Manufacture of paving units made from two types (animal blood plasma, AP920, and bovine serum albumin, BSA) of protein-bound BSC in comparison to concrete (OPCC).  Functional unit: 10,000 paving units (300 mm × 300 mm × 38 mm)  System boundary: Cradle-to-cradle (<7% of paver goes to waste)  Allocation method: Did not account for carbon sequestration from the use of protein binder in BSC.  Findings: Life cycle impact of protein-bound BSC was similar to ordinary portland cement concrete, with sufficient recycling loops of the protein binder. BSC pavers have a unique advantage of reclamation and reuse of binder at end-of-life, unlike cement, which cannot be reused after it hydrates.	• IMPACT 2002+ points
Hildebrandt et al. (2019)	Materials compared: Manufacture of wood-based fiber laminates with lignin-based phenolic resin systems (with varied lignin content). The type of lignin used in this study was organosolv lignin.  Functional unit: 1 m² of laminate board  System boundary: Cradle-to-gate (harvest wood → laminate boards).  Allocation method: Mass-based allocation method for lignin.  Findings: Partial substitution of phenolic resins with lignin-based resins may lead to significant reductions in environmental impacts of more than 80%.	Human health     Ecosystem quality     Carbon footprint     Abiotic depletion
Hermansson et al. (2020)	Materials compared: Production of kraft lignin (Lignoboost process) considering 12 different allocation methods.  Functional unit: 1 kg of extracted kraft (lignoboost) lignin.  System boundary: Cradle-to-gate (kraft lignin production).  Allocation method: 12 different methods including: mass-based, energy-based, marginal approach, etc  Findings: The life cycle impact of lignin is strongly dependent on the considered allocation method, which effects how impacts are allocated to lignin and other co-products.	Carbon footprint
Pang et al. (2021)	Materials compared: Two biorefinery processes using corncob cellulose, including current processes used at biorefineries and the proposed EXA (Ethanol, Xylose, Adhesive) biorefinery process.  Functional unit: Processing 5.25 t corncob in biorefinery.  System boundary: Cradle-to-gate (corn harvest → biorefinery).  Allocation method: Economic-based allocation method for lignin.  Findings: The carbon footprint of the EXA process (25.4 t CO₂ eq.) was lower than the carbon footprint of the reference biorefinery process (50.3 t CO₂ eq.).	Carbon footprint     Economic     (revenue)
Yadav et al. (2021)	Materials compared: Organosolv lignin from spruce bark.  Functional unit: Producing 1 kg of organosolv lignin.  System boundary: Cradle-to-gate (corn harvest → biorefinery).  Allocation method: Mass-based or main product bears all burden.  Findings: The novel lignin production method had a lower global warming potential and cost than the baseline production method.	Human health     Ecosystem quality     Carbon footprint     Abiotic depletion
Moretti et al. (2022b)	Materials compared: Assessment comparing lignin-based (kraft lignin) asphalts and conventional asphalts.  Functional unit: 1 tonne of asphalt used either as a top or base layer.  System boundary: Cradle-to-gate (lignin production) and cradle-to- grave (asphalt product).  Allocation method: Mass-based or economic-based allocation.  Findings: A 35%–70% (top) and 25%–50% (bottom) reduction of climate impact for lignin-based asphalts vs. conventional asphalts.	Human health     Ecosystem quality     Carbon footprint     Abiotic depletion
Zhou et al. (2023)	Materials compared: Lignocellosic (lignin from corncob residues) bioplastics vs. conventional plastics (PLA, PVF, PHB, etc.).  Functional unit: 1 tonne of Lignocellosic or conventional plastic.  System boundary: Cradle-to-gate (Corncob → produced bioplastic).  Allocation method: Accounted for carbon sequestration in the analysis.  Findings: The environmental impact of lignocellulosic bioplastics were lower than other plastics (PLA, PVF, PHB, etc.).	Human health     Ecosystem quality     Carbon footprint     Abiotic depletion
Our Study (2024)	Materials compared: Lignin-based BSC made from four different types of technical lignin (lignoboost lignin, lignoforce lignin, alkali lignin, and hydrolysis lignin) in comparison to various lightweight concretes.  Functional unit: 1 CMU sized block or 1 kg of lignin-based BSC.  System boundary: Cradle-to-grave (extraction → disposal at end-of-life).  Allocation method: Five different allocation methods, ranging from treating lignin as a by-product (allocated no upstream footprint) to lignin bearing the entire upstream footprint.  In this paper, we make the following contributions:  • First life cycle assessment conducted on lignin-based BSC, a novel cement-free construction material, analogous to lightweight concrete.  • Investigating the effect of solvent recapture on the environmental impacts of BSC  • Introduction of a design guide, that will help lignin-based BSC to be designed not only based on a physical parameter (compressive strength), but also on a target carbon footprint.	Carbon footprint

#### 3.2.1. Kraft lignin

Fig. 2 shows the production of kraft black liquor from the Kraft process. To obtain kraft lignin, the kraft black liquor will need to be further processed by precipitating kraft lignin out of the liquor. Currently, there are two common methods of isolating kraft lignin from kraft black liquor, which are Lignoboost and Lignoforce (Mastrolitti et al., 2021; Tomani, 2010; Kouisni et al., 2016). Kraft lignin is a byproduct of the sulfate pulping process. For this study we will use sulfate pulp as a surrogate to account for impacts associated with the production of kraft lignin. Where the input related to lignin was accounted for by the mass of sulfate pulp needed to produce the desired

amount of lignin in LignoBlock (1:3 mass of lignin to mass of sulfate pulp ratio) (Novaes et al., 2010).

#### 3.2.2. Alkali lignin

Alkali lignin is similar to kraft lignin, as it is also a by-product of the paper and pulp industry, but is notably different as alkali lignin is produced using alkaline pulping rather than sulfate pulping (Abd Latif et al., 2022; Haq et al., 2020). Fig. 3 shows the process of producing alkali lignin from birch chips. The carbon footprint of alkali lignin is likely similar to that of kraft lignin, with the only difference being an

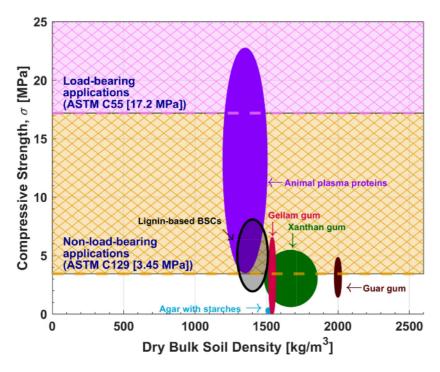


Fig. 1. Compressive strength and dry bulk soil densities of biopolymer-bound soil composites using various biopolymer-binders. The black circle represents the approximate zone (strengths and dry bulk soil densities) for lignin-based BSCs. BSC made with animal plasma proteins and BSC made with lignin show the highest compressive strength.

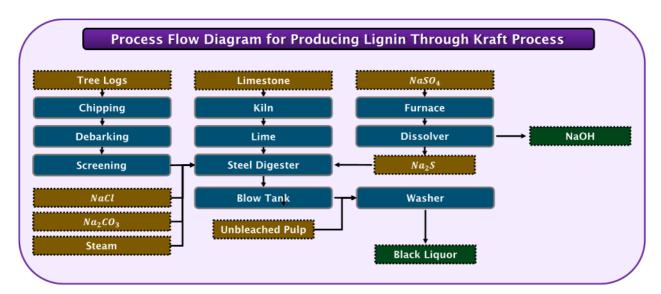


Fig. 2. Process flow diagram for the Production of kraft lignin. Lignoboost lignin and lignoforce lignin closely follow this process, with minor deviations in how lignin is extracted from kraft black liquor (Mastrolitti et al., 2021; Hermansson et al., 2020). Tan boxes represent inputs. Blue boxes represent process steps. Outputs are represented by green boxes.

additional carbon footprint associated with the drying energy of the washed alkali lignin at the end of the production process.

#### 3.2.3. Hydrolysis lignin

Unlike kraft lignin and alkali lignin, hydrolysis lignin is produced as a byproduct of the cellulose industry. Lignocellulosic biomass undergoes a fractionation process that separates out the lignin from hemicellulose and cellulose (Mastrolitti et al., 2021; Mahmood et al., 2015). Fig. 4 depicts the production of hydrolysis lignin as a waste product during the production of bio-ethanol (Balan et al., 2009). The main carbon footprint of the bio-ethanol process are related to the feedstock, chemical processing and pretreatment, refining the crude product, and, finally, distribution of the finished crude product. Typical chemicals used during the process include dilute acids such as sulfuric

acid and phosphoric acid, as they are known to be more effective on hemicellulose and lignin than on cellulose, hence making the cellulose more accessible to enzymes (Qin et al., 2014). In comparison to other forms of lignin, hydrolysis lignin has a high quantity of methoxy groups, which allows it to be used for the synthesis of a larger range of high-value chemicals (Mastrolitti et al., 2021).

#### 3.3. LignoBlock fabrication

For this study the carbon footprint impacts from three different types of lignin-based BSC was investigated. These three types of lignin cover the vast majority of lignin production, accounting for more than 90% of worldwide lignin produced, making the LCA we performed relevant to many regions across the globe. As the upstream lignin

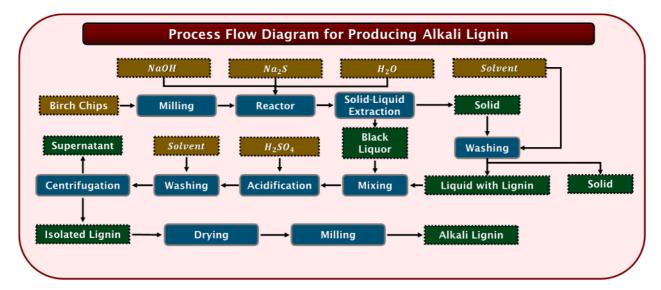


Fig. 3. Process flow diagram for the production of alkali lignin (Moreira et al., 2023; Teh et al., 2021).

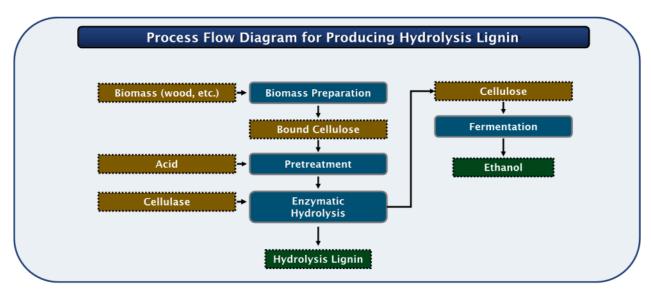


Fig. 4. Process flow diagram for the production of hydrolysis lignin (Yuan et al., 2017; Qin et al., 2014; Mastrolitti et al., 2021; Balan et al., 2009).

content and production process footprint are contributing factors to the end result, the final LCA will change if lignin with different concentrations of carbon or alternative purification processes are utilized. The solvent and fabrication method (Fig. 5) are the same for each type of lignin-based BSC.

The process flow diagram shown in Fig. 5 provides the basis for calculating the carbon footprint of lignin-based BSC. The amount of solvent needed for BSC manufacture is determined by the mix design and desired amount of lignin-based BSC. Carbon footprints associated with mixing during the manufacturing process were determined using the power consumption for a stand mixer and the time needed to blend "green" lignin-based BSC.

3.3.1. Concepts for solvent re-capture and lignin re-use relevant to the LCA DMSO re-capture can be accomplished by condensation of vaporized DMSO during desiccation of BSC and through re-use of DMSO + lignin solution produced during end of life treatment of lignin-based BSC (Fig. 6). To re-capture DMSO during the desiccation of lignin-based BSC, a cooling shaft can be constructed such that vaporized DMSO produced during the desiccation of lignin-based BSC condenses along the shaft, until reaching a collector which collects the condensed DMSO.

Depending on the design of this cooling shaft, it is likely that almost all of the DMSO used to manufacture lignin-based BSC can be re-captured. The other source of solvent use is during the end of life treatment of lignin-based BSC. During the recycling process, lignin-based BSC is crushed and mixed with DMSO, resulting in a separation of lignin from the aggregate. Fig. 6 depicts this process, where lignin covered aggregates produced from crushing lignin-based BSC are dissolved into solution, resulting in lignin being separated from the Grade 90 sand. The DMSO + lignin solution produced during recycling can be re-used completely to produce new lignin-based BSC, resulting in a circular use of the end of life DMSO. Therefore, total DMSO re-capture will be defined as the amount of DMSO that can be re-captured from oven exhaust.

#### 3.4. Life cycle assessment of blocks made from lightweight concrete

Based on its measured strength, lignin-based BSC is suitable for low compressive strength construction applications, but not for load bearing applications. For this LCA, therefore, the carbon footprint of LignoBlock was compared to comparable alternatives, such as lightweight expanded clay concrete, lightweight pumice concrete, and lightweight

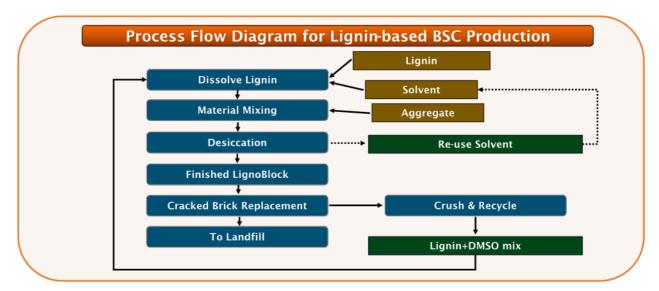


Fig. 5. Process flow diagram for the production of lignin-based BSCs. The dotted line in the figure refers to the possible recapturing of vaporized solvent during desiccation, which can be condensed and re-used to make new lignin-based BSC. When objects made of lignin-based BSC are no longer needed, they can be recycled. Recycling involves crushing the BSC and adding solvent so that new lignin-based BSC can be made. Otherwise, lignin-based BSC that is not recycled will likely be sent to the landfill.

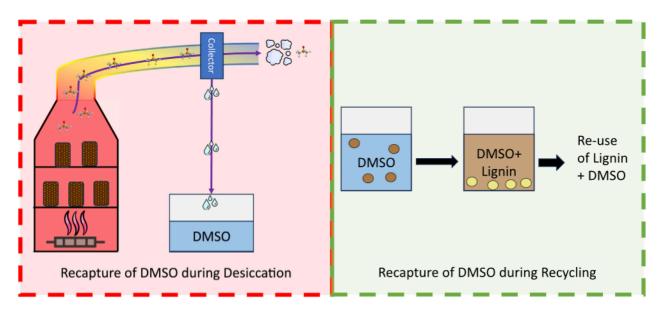


Fig. 6. During the manufacture of lignin-based BSC, an oven can be used to drive off solvent (DMSO) to achieve efficient desiccation of the material (left panel). The vaporized solvent can be re-captured by using an exhaust shaft, which permits solvent condensation. In some cases, it may make sense to re-capture lignin as well, as shown in the right-hand panel of the figure. Re-capture of both DMSO and lignin can be accomplished in this process.

perlite concrete (Ahmad et al., 2019; Sari and Pasamehmetoglu, 2005; Lanzón and García-Ruiz, 2008; Thienel et al., 2020). Fig. 7 shows the range of unit weight and compressive strength for lignin-based BSC in comparison to various types of lightweight concrete that are currently available. Lignin-based BSC falls on the upper end of the range of unit weights for the comparison materials, but is still substantially lighter than conventional concrete ( $\gamma = 2400~{\rm kg/m^3}$ ). The compressive strength of BSC is well within the range of compressive strengths of the lightweight concretes.

#### 3.5. Life cycle assessment of method

To perform the LCA for LignoBlock we examined four types of lignin-based BSC and we considered five allocation scenarios. As shown in Eq. (1) the carbon impact of LignoBlock ( $LignoBlock_{Imp.}$ ) is determined by knowing the carbon impacts of DMSO utilization, energy

consumption and the carbon footprint related to transportation calculated on a mass basis. To determine the total carbon footprint, the carbon impact associated with the allocation method and the effect of carbon entrapment must be considered where the term for entrapped carbon is subtracted from the other terms, as shown in Eq. (2). The inputs for our analysis are shown in Table 3 and are based on Eq. (1). The values shown in Tables 4–8 show the values that correspond to the five allocation scenarios considered. For this assessment, the % DMSO re-capture is assumed to be 100%. Other relevant parameters include energy efficiency,  $\eta$ , set at 90%.

$$LignoBlock_{Imp.} = DMSO_{Imp.} * (1 - wrecapture)$$

$$+ Electricity_{Imp.}(\eta) + Transport_{Imp.}$$
(1)

$$Net_{Imp.} = LignoBlock_{Imp.} + Allocation_{Imp.} - Sequestered Carbon_{Imp.}$$
 (2)

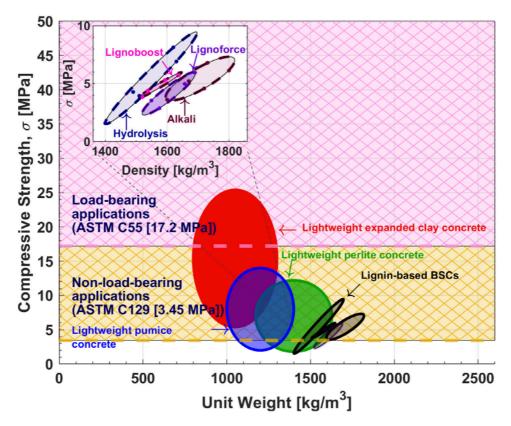


Fig. 7. Compressive strength and unit weight densities (dry density after curing/water removal) of lignin-based BSCs in comparison to lightweight concretes. The relationship between compressive strength and unit weight for lignin-based BSCs can be more easily seen in the mini-window.

By dividing the carbon footprint for each block by the mass of the block, we determined the carbon footprint on a mass-basis (as opposed to a block basis) for each type of lignin-based BSC. The electrical power needed for mixing during manufacture of LignoBlocks is found by determining the specific energy consumed per kilogram of mix as shown in Eq. (3). Where  $Power_{mixer} = \text{wattage of the mixer(W)}$  and  $T_{mix} = \text{time to blend lignin-based BSC}$ . The electrical power needed for mixing is found by finding the specific energy consumed per kg of mix as shown in Eq. (3).

$$Mixing\ Energy\ (E) = Power_{mixer} * T_{mix}$$
 (3)

The results of Eq. (3) were combined with the known mass of mix to find the mixing energy per kg (J/kg), which can be scaled up to find the energy (E) required for producing LignoBlock. Additionally, the energy needed to fully desiccate LignoBlock was found based on the amount of time it takes to fully desiccate samples in a bench-top oven operated at 60 °C. A correction for desiccation efficiency based on the load of LignoBlocks in the oven was not applied.

Finally, to calculate the transportation carbon footprint for producing LignoBlock, the distances between known paper mills, biorefineries, and chemical manufacturing plants and hypothetical use sites were taken and multiplied by the mass of each component. Eq. (4) is used to calculate the distance from each hypothetical use site to each lignin producer/chemical plant. The locations of several pulp mills and bio-refineries were identified. A factor of  $\sqrt{2}$  was applied to the calculated distances as a conservative measure to account for uncertainties in transport distances. For this study, it is assumed that the blocks are prefabricated at a separate plant at an average distance between known sources of lignin and chemical manufacturing plants. To determine distances, Eq. (4) was used, where Distance (D) is distance in kilometers,  $\Delta \phi$  is the change in latitude,  $\Delta \lambda$  is the change in longitude, and where  $K_1 = 111.13209 - 0.56605cos(2\phi_m) + 0.00120cos(4\phi_m)$ , and

 $K_2 = 111.41513cos(\phi_m) - 0.09455cos(3\phi_m) + 0.00012cos(5\phi_m)$ , and  $\Delta\phi_m$  is the mean latitude (Roedel et al., 2015).

Distance (D) = 
$$\sqrt{(K_1\Delta\phi)^2 + (K_2\Delta\lambda)^2}$$
 (4)

For this study the average transportation distance from sites of lignin production — pulp mills and bio-refineries — to hypothetical use sites was approximately 670 and 1450 km, respectively. These are conservative estimates and are much greater than is typical for transporting conventional concrete-based materials. Additionally, we used a transportation distance for DMSO of 700 km.

#### 3.6. Allocation of lignin footprint

Several different allocation methods have been implemented in allocating the footprint associated with pulping and bio-ethanol processes to waste stream lignin (Hermansson et al., 2020). For these allocation methods, however data for a specific plant/refinery is required, or are highly dependent on assumptions (e.g., economic allocation is dependent on assumptions on market price Dolezal et al., 2014). Also, as some of the carbon credits produced in the process are avoidance or sequestration, the allocation method will ultimately depend on negotiations between lignin suppliers, brick manufacturers, and end-use customers. In addition, local government regulations for carbon credit accounting will also impact this equation. Therefore, in the interest of providing a more generalizable analysis, five allocation methods independent of a specific plant, were considered. Table 2 provides details for each allocation method.

#### 3.7. Results of life cycle carbon footprint assessment

## 3.7.1. LignoBlock LCA carbon footprint according to different allocation scenarios

Table 3 shows the carbon footprint (impact, abbreviated Imp) for production of LignoBlock (LignoBlock $_{Imp}$ ), based on the mass of the

Table 2
Allocation methods applied in life cycle assessment of various LignoBlocks.

Method	Name of method	Description
1	Lignin waste stream	This conservative method assumes that lignin will take on
	bears all the burden	the entire carbon footprint associated with the production
		of pulp or ethanol, from which lignin is obtained.
2	Allocation based on	Carbon footprint is allocated proportionately based on the
	mass of co-products	mass of the co-products produced.
3	Allocation based on	Carbon footprint is allocated proportionately based on the
	energy of co-products	energy of the co-products produced.
4	Marginal approach	This method consider some impacts associated with the use
		of lignin in LignoBlocks, such as lignin extraction impacts.
5	Main product bears	Considers that lignin is a true waste product, lignin bears
	all the burden	no carbon footprint from paper pulp or ethanol production.

Table 3

Carbon footprint for production of LignoBlock without considering carbon sequestration or allocation scenarios.

Scenario	M (kg)	S (kg)	E (kWh)	T (tkm)	LignoBlock <sub>Imp</sub> (kg-CO <sub>2</sub> eq)	$LignoBlock_{Imp}$ (kg-CO <sub>2</sub> eq/kg BSC)
1 - Lignoboost	1.11	1.12	0.804	1.850	0.566	0.057
2 - Lignoforce	1.13	1.20	0.843	1.944	0.594	0.057
3 - Alkali	1.42	1.25	0.879	2.188	0.652	0.060
4 - Hydrolysis	0.92	1.33	0.694	2.646	0.696	0.072

<sup>\*</sup>M = mass of lignin, S = mass of solvent, E = energy use, and T = transport.

Table 4
LignoBlock carbon footprint with lignin bearing the footprint of the main product (Method 1).

Scenario	$LignoBlock_{Imp}$ (kg-CO <sub>2</sub> eq)	Allocated impact (kg-CO2 eq)	Sequestered CO2 (kg-CO2 eq)	Net impact (kg-CO <sub>2</sub> eq/kg BSC)
1 - Lignoboost	0.566	2.759	2.442	0.088
2 - Lignoforce	0.594	2.808	2.486	0.088
3 - Alkali	0.652	3.529	3.124	0.097
4 - Hydrolysis	0.696	2.287	2.024	0.098

Mass for LignoBlock:  $m_{LB} = 9.989 \text{ kg}, \ m_{LF} = 10.471 \text{ kg}, \ m_{AL} = 10.921 \text{ kg}, \ m_{HL} = 9.732 \text{ kg}.$ 

Table 5
LignoBlock carbon footprint using mass-based allocation (Method 2).

Scenario	$LignoBlock_{Imp}$ (kg-CO <sub>2</sub> eq)	Allocated impact (kg-CO <sub>2</sub> eq)	Sequestered CO <sub>2</sub> (kg-CO <sub>2</sub> eq)	Net impact (kg-CO <sub>2</sub> eq/kg BSC)
1 - Lignoboost	0.566	0.268	2.442	-0.161
2 - Lignoforce	0.594	0.272	2.486	-0.155
3 - Alkali	0.652	0.342	3.124	-0.195
4 - Hydrolysis	0.696	1.278	2.024	-0.005

Mass for LignoBlock:  $m_{LB} = 9.989$  kg,  $m_{LF} = 10.471$  kg,  $m_{AL} = 10.921$  kg,  $m_{HL} = 9.732$  kg.

constituents (lignin-M, and solvent-S), and based on energy consumption and transportation considerations (energy use-E, and transportation-T), without considering carbon entrapment or allocation scenarios. The higher carbon footprint for production of hydrolysis LignoBlock is a result of greater transportation distances between hypothetical use sites and currently existing bio-ethanol refineries. With the expected growth of the bio-ethanol industry, transportation distances are expected to decrease, which will no doubt reduce the carbon footprint of hydrolysis LignoBlock production to make it more similar to the other types of LignoBlock (Bajwa et al., 2019).

Table 4 shows the carbon footprint of LignoBlock production considering allocation Method 1, in which lignin bears all of the burden of its production (the most conservative allocation method) and considering the beneficial effect of carbon entrapment, via incorporation of carbonrich lignin ( $\approx\!60\%$  carbon) into the LignoBlock. While the allocation method results in substantial additional carbon footprint, the beneficial effect of carbon entrapment is also substantial and thus the net carbon footprint (net impact) is only modestly higher than the impact that is directly associated with LignoBlock production as shown in Table 3.

Table 5 shows the total carbon footprint of LignoBlock using mass-based allocation of lignin. Allocation method 1 (Table 4) is conservative as it assumes that lignin solely bares the carbon footprint associated with the production of pulp or ethanol, while the other co-products are allocated no footprint. A more balanced approach would be to perform mass-based allocation, which distributes the carbon footprint associated

with lignin production to co-products for the four different types of lignin investigated in this study. The results of this allocation method are shown in Table 5. Eq. (5) was used to calculate the allocation factor for the different types of lignin (Cherubini et al., 2011; Hermansson et al., 2020).

Allocation factor for mass product<sub>i</sub> = 
$$\frac{m_i}{\sum_{all} m}$$
 (5)

where  $m_i$  represents the mass of product $_i$  being produced, and  $\sum_{ali} m$  represents the total mass of product being produced from paper and pulp plants and from bio-ethanol plants. During the Kraft process with lignoboost extraction, four co-products — pulp, soap, heat, and lignin — are produced (Hamaguchi et al., 2012; Staffas et al., 2013). According to Culbertson et al. the outputs for a plant using the lignoboost process was 55 300 kg of pulp, 6110 kg of lignoboost lignin, 1570 kg of soap, and 70.3 MWh of heat (Hermansson et al., 2020; Culbertson et al., 2016). For this study, we are assuming that the mass-based allocation for lignoforce lignin and alkali lignin are similar to that of lignoboost lignin, as all three of these are produced from paper and pulp plants. Given the fact that the production of each liter of cellulosic ethanol during bioethanol production results in 0.5–1.5 kg of co-generated lignin (Bruijnincx et al., 2015), this type of mass-based allocation makes sense.

Similar to the mass-based allocation approach, an energy-based allocation can be performed as shown in Eq. (6) (Sandin et al., 2015).

Table 6
LignoBlock carbon footprint using energy-based allocation (Method 3).

Scenario	LignoBlock <sub>Imp</sub> (kg-CO <sub>2</sub> eq)	Allocated impact (kg-CO <sub>2</sub> eq)	Sequestered CO <sub>2</sub> (kg-CO <sub>2</sub> eq)	Net impact (kg-CO <sub>2</sub> eq/kg BSC)
1 - Lignoboost	0.566	0.119	2.442	-0.176
2 - Lignoforce	0.594	0.121	2.486	-0.169
3 - Alkali	0.652	0.153	3.124	-0.212
4 - Hydrolysis	0.696	1.135	2.024	-0.020

Mass for LignoBlock:  $m_{LB} = 9.989 \text{ kg}$ ,  $m_{LF} = 10.471 \text{ kg}$ ,  $m_{AL} = 10.921 \text{ kg}$ ,  $m_{HL} = 9.732 \text{ kg}$ .

Table 7
LignoBlock carbon footprint using marginal approach allocation (Method 4).

Scenario	$LignoBlock_{Imp}$ (kg-CO <sub>2</sub> eq)	Allocated impact (kg-CO <sub>2</sub> eq)	Sequestered CO <sub>2</sub> (kg-CO <sub>2</sub> eq)	Net impact (kg-CO <sub>2</sub> eq/kg BSC)
1 - Lignoboost	0.566	0.277	2.442	-0.160
2 - Lignoforce	0.594	0.282	2.486	-0.154
3 - Alkali	0.652	0.354	3.124	-0.193
4 - Hydrolysis	0.696	0.230	2.024	-0.114

Mass for LignoBlock:  $m_{LB} = 9.989$  kg,  $m_{LF} = 10.471$  kg,  $m_{AL} = 10.921$  kg,  $m_{HL} = 9.732$  kg.

Table 8
LignoBlock carbon footprint using main product allocation (Method 5).

Scenario	$LignoBlock_{Imp}$ (kg-CO <sub>2</sub> eq)	Allocated impact (kg-CO <sub>2</sub> eq)	Sequestered CO <sub>2</sub> (kg-CO <sub>2</sub> eq)	Net impact (kg-CO <sub>2</sub> eq/kg BSC)
1 - Lignoboost	0.566	0	2.442	-0.188
2 - Lignoforce	0.594	0	2.486	-0.181
3 - Alkali	0.652	0	3.124	-0.226
4 - Hydrolysis	0.696	0	2.024	-0.136

Mass for LignoBlock:  $m_{LB} = 9.989$  kg,  $m_{LF} = 10.471$  kg,  $m_{AL} = 10.921$  kg,  $m_{HL} = 9.732$  kg.

Allocation factor for energy product<sub>i</sub> = 
$$\frac{e_i}{\sum_{all} e}$$
 (6)

where  $e_i$  represents the energy of product, being produced, and  $\sum_{all} e$  represents the total energy of all products being produced from the paper and pulp or bio-ethanol production. Table 6 shows the total carbon footprint of LignoBlock for using energy-based allocation of lignin.

The allocation of the carbon footprint associated with the production of lignin, based on mass or energy (Tables 5 and 6) allows for a more accurate accounting of the footprint associated with lignin in comparison to allocation Method 1 (lignin bears all the burden). With allocation Method 2 or Method 3, LignoBlock has a net carbon negative footprint, because the effect of carbon entrapment is now greater than the allocated carbon footprint using these two allocation methods.

Table 7 shows the carbon footprint of LignoBlock production considering allocation Method 4, in which the allocated carbon footprint for lignin production is the sum of the energy needed to dry washed lignin and the energy needed to replace lignin that would otherwise have been used for energy generation, in existing pulp and bio-ethanol manufacturing facilities. For this study it is assumed that the average drying time and energy is approximately the same as in a bench top oven set at 60 °C. The energy needed to replace the energy value of lignin is found by comparing the amount of electrical energy produced from lignin and assuming that natural gas would be used as the substitute. The average electrical energy generated per kilogram of lignin or per kilogram of natural gas is approximately 3.3 kWh and 37 kWh, respectively (Liu and Bao, 2017; EIA, 2017). Also as 2.75 kg of CO<sub>2</sub> is emitted for every 1 kg of natural gas being burnt, the CO<sub>2</sub> impacts resulting from replacing erstwhile burned lignin needs to be considered.

Allocation Method 4 (Table 7) is based on the approach of assigning no carbon footprint to the production of lignin (Method 5), but considers the footprints associated with the energy needed to dry washed lignin and the energy needed to replace the energy value of lignin.

Table 8 shows the carbon footprint of LignoBlock production considering allocation Method 5, in which lignin is treated as a pure waste

Table 9

Analysis inputs and carbon footprints for blocks made out of lightweight concrete

Scenario	M (kg)	Total impact (kg-CO <sub>2</sub> eq)	Total impact (kg-CO <sub>2</sub> eq/kg)
5 - LWC expanded clay	6.75	1.13	0.167
6 - LWC perlite	8.99	3.05	0.339
7 - LWC pumice	7.71	2.36	0.306

\*M = mass of lightweight concrete (LWC).

product, i.e, no carbon footprint allocated to the production of lignin. This allocation method is appropriate for scenarios in which lignin would simply be discarded. This allocation method yields the most favorable net impact results for all four types of LignoBlock.

#### 3.7.2. LCA of blocks made out of lightweight concrete

Lightweight concretes were chosen as reference materials for this LCA study because their compressive strength are similar to those of lignin-based BSC. Like lignin-based BSC, lightweight concretes are primarily used for low compressive strength construction applications. To find the mass required to fabricate a block (6424 cm³) using lightweight concrete, the density of each material was used in conjunction with the required volume, resulting in the masses shown in Table 9.

#### 3.7.3. Summary of LCA results

Using the results shown in Section 3.7.1, the net impact from producing one kilogram of LignoBlock was determined for each type of lignin and each allocation method. Fig. 8 also shows the impacts of producing a kilogram of lightweight concrete blocks and lignin bound blocks.

All net carbon footprints consider the beneficial effect of carbon entrapment, since lignin is a carbon-rich biopolymer. Except for the most conservative allocation approach (Method 1), the net carbon footprint of each type of LignoBlock is negative. Even the best lightweight concretes have total carbon footprints that are significantly greater than the four LignoBlocks, even when the most conservative allocation method

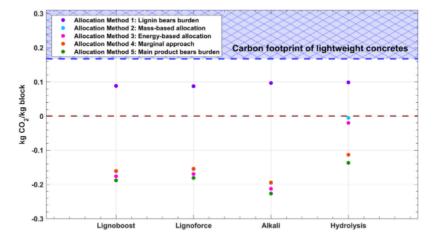


Fig. 8. Net life cycle carbon footprint for lignin-based BSC using different types of lignin. For lignoboost LignoBlock, lignoforce LignoBlock, and alkali LignoBlock, the values for allocation Method 2 and allocation Method 4 overlap.

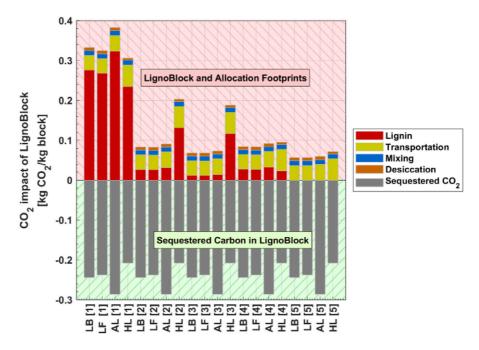


Fig. 9. Total CO<sub>2</sub> impact breakdown for producing lignin-based BSC. The top of half of the figure depicts the impacts for producing lignin-based BSC and allocated impact. The lower portion of the figure shows the associated entrapped carbon for each type of lignin-based BSC.

is used. LignoBlocks made from lignoboost lignin, lignoforce lignin, and alkali lignin exhibited similar net carbon footprints. LignoBlock made with hydrolysis lignin exhibited greater impacts, because less lignin is used to manufacture hydrolysis lignin LignoBlock, owing to its higher efficiency as a binder. As a result, carbon entrapment is less with hydrolysis LignoBlock, in comparison to the other three types of LignoBlock.

### 3.7.4. Breakdown of carbon footprint of LignoBlock according to life cycle activities

The sources of carbon footprint for each type of manufactured LignoBlock are shown in Fig. 9. The sources include carbon footprint associated with the production of lignin; carbon footprint associated with the transportation of lignin and solvent; carbon footprint associated with mechanical mixing process; carbon footprint associated with LignoBlock desiccation; and, the carbon footprint associated with carbon entrapment due to lignin sequestration into LignoBlock. This is a beneficial carbon footprint and therefore is negative (where other carbon footprint sources are positive). For allocation Method 2 (mass

basis) the carbon footprint of hydrolysis lignin is higher than that of the other three types of lignin, because hydrolysis lignin accounts for most of the mass in this production scenario. For allocation Method 3 (energy basis) the carbon footprint of hydrolysis lignin is higher than that of other types of lignin, because the production of hydrolysis lignin is not associated with significant heat generation, As expected, the carbon footprint of all types of LignoBlock is highest for allocation Method 1, because lignin production bears the entire burden for all types of lignin.

#### 3.8. Sensitivity analysis

To perform the sensitivity analysis (Fig. 10) we varied the energy efficiency from 80% to 100%. We varied the transportation distance from 90% to 110% of the nominal distance considered for this study. We investigated the effect of oven-assisted desiccation versus the scenario of desiccation without thermal assistance. Lastly, we varied the lignin content of the LignoBlocks from 90% to 110% of a mid-range mix design. We found that LignoBlock samples were significantly stronger than what is required for low compressive strength construction. Adding

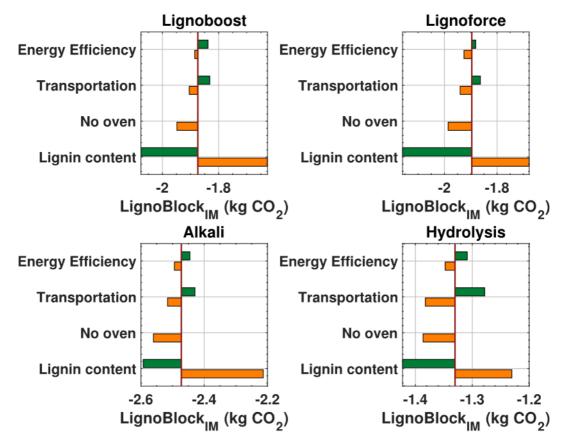


Fig. 10. Sensitivity analysis for four different types of LignoBlock. Although not explicitly shown, DMSO re-capture plays a big role in carbon footprint for all four types of LignoBlock. Lignin content has a significant impact on the carbon footprint for all four types of LignoBlock.

additional lignin may further increase the strength of LignoBlock, and may potentially increase the range of construction applications beyond standard low compressive strength applications. Furthermore, higher lignin content will result in greater carbon sequestration. The focus of the sensitivity analysis was on the impacts resulting from the production of LignoBlock (Eq. (2)) rather than on impacts associated with the production of lignin. The carbon footprints associated with the production of LignoBlock are independent of the considered allocation methods (i.e., the allocation methods only pertain to the production of LignoBlock, per se).

Based on the sensitivity analysis shown in Fig. 10, the most sensitive parameter is lignin content. Small variation in lignin content was shown to have a noticeable impact on the carbon footprint of LignoBlock, because of carbon sequestration. Carbon sequestration/entrapment is an essential benefit of LignoBlock, as it allows LignoBlock to potentially be a carbon negative construction material (depending on the considered allocation method).

#### 3.9. Developing footprint design charts

While compressive strength of a material is the design consideration that is most often emphasized, net carbon footprint can be a major design-driver for lignin-based BSC. For lignin-based BSC, lignin content is the dominant determinant of both compressive strength and carbon footprint. Smaller contributions to net carbon footprint are associated with energy use and transportation. Increased lignin content in lignin-based BSC leads to greater transportation impact and energy use impact, related to lignin production. Eq. (7) encompasses this relationship.

$$Net \ carbon \ footprint = DM \ SO_{Imp.} * (1 - \% reduction) + \\ Energy \ use_{Imp.}(\eta) + Transport_{Imp.} + Lignin \ content_{Imp.}$$

$$(7)$$

An important consideration is the degree to which DMSO is successfully re-captured during the manufacture of lignin-based BSC. For the net life cycle carbon footprint analysis presented in Fig. 8, we assumed a DMSO re-capture rate of 100%. In large-scale manufacturing operations, 100% DMSO re-capture is reasonable. However, for smaller scale manufacturing operations, DMSO re-capture systems may be cost-prohibitive. In some cases, systems with reduced DMSO re-capture capabilities may be more practical.

Fig. 11 depicts the carbon footprint design guide of LignoBlock, with carbon footprint being dependent on the degree of DMSO re-capture, compressive strength, and efficiency of energy use. The life cycle carbon footprints shown in this figure account for the impacts associated with the production of LignoBlock, but does not account for impacts related to the production of lignin. To modify these design guides to account for the carbon footprint associated with the production of lignin, a constant can be added that reflects the allocation scenario of interest.

#### 4. Discussion of results

Biopolymer-bound soil composites (BSC) are novel materials that use various biological substances to bind granular materials together to create solid materials. With compressive strength as high as 20 MPa (Rosa et al., 2020), BSC can be viewed as a concrete substitute and can be used for many applications where concrete would be used. The development of BSC is focused on the exploration of different biopolymers such as proteins, starches, and exotic substances from marine organisms, that serve to bind the granular materials.

In a very general sense, the biopolymer in BSC replaces the portland cement that is used to make conventional concrete. In conventional concrete, approximately 10%–15% portland cement is needed. Only 5%–10% biopolymer is needed for BSC. Water that is used to make

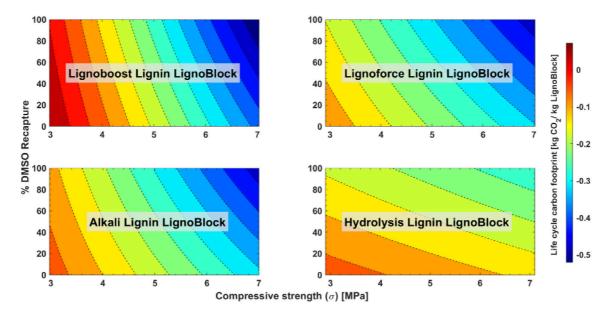


Fig. 11. For each type of lignin-based BSC, the relationships between compressive strength and life cycle carbon footprint is shown for various degrees of DMSO re-capture (dependent on the capabilities of a particular manufacturing plant). For a given degree of DMSO re-capture, the relationship between target compressive strength and life cycle carbon footprint is easily determined. All four panels assume an overall energy efficiency of 100% (i.e.  $\eta = 1.0$ ).

conventional concrete gets incorporated into the cement component by a chemical hydration process, to become permanently incorporated into the concrete. In BSC, by contrast, a solvent is needed to fully dissolve the biopolymer in order to manufacture the bio-composite, but, eventually, the solvent is removed by evaporation or other passive solvent removal methods.

The LCA reported in this manuscript concerns a newly developed variant of BSC which uses lignin as the biopolymer binding agent (lignin-based BSC). Lignin is a desirable material because it is the major component of the waste stream associated with the bio-ethanol industry. Lignin-based BSC is notable because it is the first version of BSC to use a hydrophobic biopolymer binding agent. Unlike other versions of BSC that use water as the biopolymer solvent, lignin-based BSC uses DMSO. Preliminary investigations of this material indicate that compressive strength ranges from 1.6-8.1 MPa. While this compressive strength is lower than that of other versions of BSC, it is well within the range of the compressive strength of lightweight concretes which can be used for a variety of low compressive strength construction applications. Lignin is widely available, globally, as is DMSO and other weakly hydrophobic solvents that could be used to dissolve lignin. Both lignin and DMSO are produced from processes that are independent of the fossil fuel industry.

Major aspects of the net life cycle carbon footprint of lignin-based BSC include the carbon footprint associated with the production of lignin and the carbon footprint associated with the manufacture of lignin-based BSC from lignin, granular material, and DMSO. Our analysis covered four different types of lignin — lignoboost lignin, lignoforce lignin, alkali lignin, and hydrolysis lignin — which together account for more than 90% of commercially available lignin, currently.

Although the carbon footprint varies across the four types of lignin, the results of our analysis do not vary substantially based on lignin type. With the projected growth of the bio-ethanol industry, hydrolysis lignin is expected to become more dominant and more available for use in material applications in the near future (EPA, 2014). Additionally, as the bio-ethanol industry expands, bio-ethanol production facilities will become more numerous, which will reduce the distance between lignin production sites and sites of lignin use, leading to a reduction in transportation requirements.

Notwithstanding that lignin is considered a waste product, we considered five different allocation methods in our LCA. We also computed

carbon sequestration for each type of lignin-based BSC, based on lignin content. Carbon sequestration via the use of lignin in lignin-based BSC can be considered a form of biological carbon sequestration, as carbonrich lignin is "stored" within lignin-based BSC (Farrelly et al., 2013). This is unlike geological carbon sequestration, where CO2 is injected into underground geologic formations through carbon capture and storage processes (Zhang and Song, 2014). The magnitude of carbon footprint reduction associated with carbon sequestration was large enough to substantially reduce the net carbon footprint. Considering the most conservative allocation scenario, in which lignin takes on the entire carbon footprint associated with its production, the net life cycle carbon footprint was approximately 0.1 kg CO2/kg BSC, for all four types of lignin. For the most optimistic allocation scenario, in which lignin is considered to be a true waste product, the net carbon footprints were all negative, reflecting the carbon sequestration effect, and ranged from -0.14 kg CO2/kg BSC to -0.23 kg CO2/kg BSC, for the four types of lignin. By comparison, the net carbon footprint associated with the production of lightweight concretes is substantially higher, ranging from 0.17 kg CO<sub>2</sub>/kg to 0.34 kg CO<sub>2</sub>/kg, while the carbon footprint for OPCC is 0.21 kg CO<sub>2</sub>/kg.

#### 4.1. Pathways for improvement: Future direction of lignin-based BSC

In considering the phase-in of lignin-based BSC in constructing the built environment, it is helpful to understand how lightweight concretes are used. Lightweight concrete (specifically, concrete made using lightweight aggregate) is currently used to make lightweight structural members and a variety of non-structural building elements. Examples include interior partition walls, pre-cast wall panels and other types of facade applications (Mohammed and Hamad, 2014; Thienel et al., 2020). A well-established application arena for lightweight concrete is its use as roofing tiles. All of these applications are viable paths for introducing lignin-based BSC into the construction industry. While lightweight concrete is valued for its insulating properties, thermal studies have yet to be conducted for lignin-based BSC. Future investigation of the thermal properties of lignin-based BSC would be worthwhile.

Previous life cycle assessment conducted on the manufacture of BSC made from AP920 (a mixture of blood proteins) used an aggregate impact measure (IMPACT 2002+) that accounts for four damage

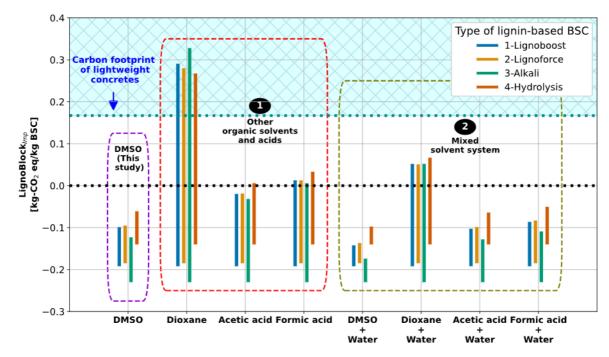


Fig. 12. Net carbon footprint (considering allocation Method 5 from Table 2) of manufacturing LignoBlock using various solvents. The bars represent the range (bottom of range = 100% solvent recapture and top of range = 0% solvent recapture) of the carbon footprint associated with manufacturing LignoBlock.

categories: human health, ecosystem quality, climate change, and resources (Roedel et al., 2015). Even though carbon footprint was not explicitly considered in the previous LCA on AP920-BSC, the use of IMPACT 2002+ points as an evaluation measure, allowed for an indirect accounting of the carbon footprint of AP920-BSC. Even without considering the beneficial effect of carbon-sequestration, the life cycle impact of AP920-BSC was comparable to that of concrete after many recycling loops of the biopolymer were carried out. While carbon sequestration takes place during the manufacture of AP920-BSC, the potential for carbon sequestration in lignin-based BSC is greater because of the higher carbon content of lignin in comparison to blood proteins (AP920). In the case of our LCA, recycling of lignin-based BSC was not considered in calculating the life cycle carbon footprint of ligninbased BSC, as there has not yet been studies of the viability of circular lignin-based BSC. Future studies should be conducted to investigate the possibility of recycling lignin-based BSC.

Fig. 12 shows the range of net carbon footprint for manufacturing LignoBlock using different solvents. For this study, DMSO was selected as the solvent, due to its effectiveness in dissolving lignin into solution. Also, lignin-based BSC made with DMSO has the lowest range of carbon footprints in comparison to other organic solvents and acids that are effective in dissolving lignin (group 1 of solvents in Fig. 12) (Sameni et al., 2017; Ma et al., 2021). While DMSO is an effective solvent for dissolving lignin, solvent removal is somewhat slower when DMSO is used as the solvent than when water is used as the solvent.

Alternative solvents (Group 1 in Fig. 12) to DMSO should be explored to consider implications for manufacturing and carbon footprint. Mixed solvent systems, such as organic solvents or acids combined with water may also be effective in dissolving lignin. Previous studies have shown that mixtures of water and organic solvent (e.g., dioxane) were more effective in dissolving lignin, than pure organic solvents (Melro et al., 2018; Evstigneev, 2010; Boeriu et al., 2014), because of the amphipathic nature of lignin. The partial replacement of organic solvent or acid with water is attractive, as the carbon footprint of water is significantly lower. Group 2 in Fig. 12 shows the potential reduction in the life cycle carbon footprint of lignin-based BSC when using mixed solvent systems. Even though the use of mixed solvents may be effective in further reducing the environmental impact of lignin-based BSC, there remains uncertainties regarding the effect of these solvents on the physical properties (e.g. compressive strength) of the finished material.

#### 5. Conclusion

Lignin-based BSC, the newest member of the BSC family, is a promising cement-free bio-composite that utilizes an agriculturallysourced biopolymer binder - lignin - to bind granular materials to form a solid material appropriate for construction. The strength and density of lignin-based BSC is comparable to many types of lightweight concrete. Our LCA showed that the manufacture of lignin-based BSC is associated with a net negative carbon footprint, except for the most conservative allocation approach. Even with the use of this approach, the net carbon footprint of lignin-based BSC is still substantially lower than the most sustainable type of lightweight concrete used as a reference material in this study (lightweight concrete made with expanded clay aggregate). Carbon sequestration — by virtue of the fact that lignin-based BSC uses a carbon-rich binder - plays a major role in offsetting the carbon footprint associated with the manufacture of lignin-based BSC. Our LCA is based on the existing distribution of lignin-production sites and anticipated sites of lignin use. Because of the Renewable Fuel Standard in the U.S, bio-ethanol production is expected to grow and therefore lignin production is expected to grow, too.

Lignin-based BSC like other members of the BSC family, is not yet commercially available, but the technology is poised to be developed for large-scale production in the near future. There is a strong potential to develop lignin-based BSC for non-load bearing construction applications, such as interior partition walls, facade applications, roofing tiles, and even pavement. Laboratory-based effort to increase the compressive strength of lignin-based BSC, if successful, may expand the range of construction applications in the future. Even at its current stage of development, lignin-based BSC is a valuable alternative to conventional concrete and other conventional building materials and shows great promise for driving a green transition in the construction industry.

#### CRediT authorship contribution statement

**Barney H. Miao:** Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Conceptualization. **Robert Headrick:** Writing – review & editing, Resources, Conceptualization. **Zhiye Li:** Writing – review &

editing, Funding acquisition. **Leonardo Spanu:** Resources. **David J. Loftus:** Writing – review & editing, Conceptualization. **Michael D. Lepech:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

#### 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

Data will be made available on request.

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