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Sustainable production of densified wood as advanced structural materials through oxygen delignification

This work demonstrates scalable oxygen delignification (O_2-delig) of dimensional wood at 90–110 °C for transforming natural wood into high-performance structural materials through subsequent densification at 2 MPa. The resultant wood achieved bending modulus of rupture over 220 MPa and Brinell hardness over 80 MPa. Life cycle assessment showed that densified O_2-delig wood has substantially low environmental impacts compared with high recycling content metals.

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Sustainable production of densified wood as advanced structural materials through oxygen delignification

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This study demonstrated a sustainable and commercially scalable low temperature (~100 °C) oxygen delignification (O_2-delig) technology for delignifying pristine wood boards to produce advanced super-strong wood structural materials through subsequent densification. Sustainable dimensional wood delignification is the prerequisite first step with a significant barrier to commercialization due to great economic and environmental costs, though the groundbreaking concept of substantial wood densification through delignification has been well established and demonstrated. O_2-delig was conducted at low temperatures of 90–110 °C under semi-gas–solid phase reaction conditions with a lower base chemical charge of 28–56 g kg^{-1}, achieving approximately 20% lignin removal. Subsequent densification of the delignified basswood at a low pressure of 2 MPa for only 20 min resulted in a densified wood with double the density, a bending modulus of rupture over 220 MPa, and a Brinell hardness over 80 MPa. This study for the first time revealed that gains in wood strength by wood densification need to be greater than the "mathematical enhancing factor" (MEF) to avoid actual loss in wood load-bearing capacity due to the loss in wood thickness by densification. Life cycle assessment showed that densified O_2-delig wood has substantially low environmental impacts compared with metals even with high recycling content.

Green foundation

1. This study demonstrated sustainable and commercially scalable delignification of wood boards using oxygen at low temperatures for producing super-strong wood through densification to replace nonrenewable and high carbon footprint ore-based materials. Using oxygen eliminates the energy, water, and capital-intensive chemical recovery with lower environmental impacts than metal production supported by LCA.
2. O_2-delig was conducted at 90–110 °C with lower alkali charge 28–56 g kg^{-1} than alkaline pulping and achieved approximately 20% delignification. Subsequent densification at only 2 MPa for 20 min achieved a densified wood with a bending modulus of rupture of 220 MPa and a Brinell hardness of 80 MPa.
3. Further research will include using potassium- and ammonia-based alkalis, *e.g.*, KOH, K_2CO_3, and urea, for impregnation so that the spent liquor can be used as a fertilizer, evaluating the performance of this fertilizer, and characterizing the dissolved lignin for high value valorization.

Introduction

Wood has long been used as a structural material. Its inherent renewability and ability to store carbon attracted great interest as an alternative to ore- or mineral-based materials. Wood has good mechanical strength with lower density and thermal conductivity than metals and glasses, ideal for Leadership in Energy and Environmental Design (LEED) building (https://www.usgbc.org/leed). However, substantial improvements in wood mechanical and other properties are required for certain large structural applications where ore- or mineral-based materials are preferred currently. Existing dimensional wood treatments, such as thermal,[1,2] chemical,[3,4] preservative,[5] impregnation,[6–8] and densification[9–12] are not sufficient to meet the required performance. Recently, groundbreaking research successfully demonstrated that wood can be transformed into advanced structural materials with superior properties, such as outstanding strength,[13] moldability,[14,15] thermally radiating and insulative properties,[16] and optical transparency,[17–20] through chemical delignification with sub-

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sequent engineering of wood cell walls such as wood densification. These studies provided opportunities for advancing wood as a sustainable structural solution through product and market innovations, which not only can sequestrate carbon but also promote sustainable healthy forest management and rural economic development by upcycling low grade forest thinning materials. However, effective and sustainable delignification of dimensional wood as the first prerequisite step is the key to achieving commercial success.

Several chemical delignification approaches have been attempted to remove lignin in dimensional wood.[21] For example, hydrogen peroxide with acetic acid was initially introduced to improve densification of resin-impregnated wood at low pressures.[22] Alkaline sulfite solution was used in the pioneering work of producing super strong densified wood.[13] NaOH with anthraquinone (AQ) was also applied for delignifying and densifying pine wood.[23] Recently, carbonate and/or pulping spent liquor were also proven effective for wood delignification to produce strong densified wood.[24] However, peroxide is cost prohibitive for removing a substantial amount of lignin because of the high chemical dosages required. Sulfite delignification is known to have difficulties in sulfite recovery in addition to the serious environmental concern of SO_2 air emission. AQ has carcinogenic potential.[25] Oxidative delignification using chlorinated compounds produces dioxin – a carcinogen well known in the pulp and paper industry.[26] Sodium hydroxide[27] and carbonate[24] delignification seemed logical choices because mature alkali chemical recovery systems are commercially implemented in commercial wood pulping for fiber production. However, the complex alkali recovery system involving several energy- and capital-intensive processes such as calcination, causticization, and black liquor concentration and combustion constitutes a significant cost barrier to commercializing densified wood at much smaller scales than wood pulping. Co-location of wood delignification and densification operations at a pulp mill is not a viable option either because most pulp mills are bottle-necked by their chemical recovery operation in addition to the need for a very different material handling and reactor system.

The existing delignification technologies discussed above were developed for wood pulping and fiber bleaching, very different from those for dimensional wood treatment to produce structural wood materials. The former aims to achieve a complete separation of cellulosic fibers using small and thin wood chips, whereas the latter requires keeping the physical structure of dimensional wood (boards or panels) intact to preserve its structural mechanical strength. Removing lignin from dimensional wood while maintaining its physical integrity is very difficult because lignin inherently serves as the "glue" to glue wood cell wall components together, and because of the limited mass transfer or chemical accessibility to lignin in large-dimension wood. Fortunately, a near-complete removal of lignin is not necessary for many applications such as producing translucent wood panels and high strength or moldable structural materials through densification.[13,14] Oxygen has been used in the pulp and paper industry as an economically and environmentally sustainable[28,29] approach for partial removal of a very small amount of residue lignin in chemical pulp fibers. Due to the low oxygen solubility in water, mass transfer or lignin accessibility to oxygen in aqueous fiber systems is a major rate limiting step.[30] Although oxygen delignification (O_2-delig) has been proven effective, but only for removing a very small amount (~3%) of residue lignin in chemical wood fibers,[28,31] its applicability for removing a large amount of bulk wood lignin, especially in solid dimensional wood, has never been hypothesized and evaluated. Using O_2-delig for *in situ* delignification of dimensional wood presented in this study is therefore novel. It also represents a major technological breakthrough for its great commercialization potential in terms of process scalability, technological readiness, capital and operational costs, and environmental impact, as well as its ability to advancing wood as a sustainable alternative low-density structural material to energy-intensive ore-based and other materials for many applications.

We hypothesize that direct application of O_2-delig to pristine dimensional wood could achieve significant bulk wood delignification for subsequent effective and desired wood densification.[32] This is because not only lignin in natural wood is highly reactive unlike the highly condensed residue lignin in chemical pulp fibers with low reactivity, but also a higher wood solid content in alkali-impregnated wood boards/panels of approximately 35% results in a thinner water film on wood fiber surface and greatly improves oxygen mass transfer (a rate controlling process), compared with fibers at 15% solids limited by pumping fiber suspensions in commercial chemical pulp bleaching operations.[28] Oxygen partially delignifies wood *via* depolymerization and oxidation[29,33] (Fig. 1). Depolymerization decreases the size of lignin polymer, whereas oxidation introduces hydrophilic (such as carboxylic) groups onto lignin (Scheme 1). Both reactions promote lignin dissolution in aqueous alkaline solutions. The partial removal of lignin softens the wood structure and facilitates wood densification by hot-press. The introduction of carboxyl groups by O_2-delig can increase inter-fiber bonding through densification. The moderate temperatures of O_2-delig not only save energy but also prevent carbohydrates (cellulose and hemicelluloses) from undesirable depolymerization and loss of wood mass. Furthermore, any base chemicals such as Na_2CO_3, $Ca(OH)_2$, KOH, NH_4OH, or urea[32,34] can be used in O_2-delig in addition to the commonly used NaOH in commercial pulping operations because their role is merely to buffer the proper pH for effective O_2-delig,[29] very different from that in alkaline delignification. The wide selection of compatible base chemicals can decrease chemical cost. The residual spent liquor from these alternative alkaline systems could contain suitable nutrients for agricultural applications without the need for chemical recovery,[35] which removes the key and significant barrier to commercialization due to the very expensive capital cost for alkali recovery. Moreover, because the oxidation reaction occurs at relatively low temperatures without sulfur-containing chemicals, the dissolved lignin may offer opportunities for future valorization compared with highly condensed

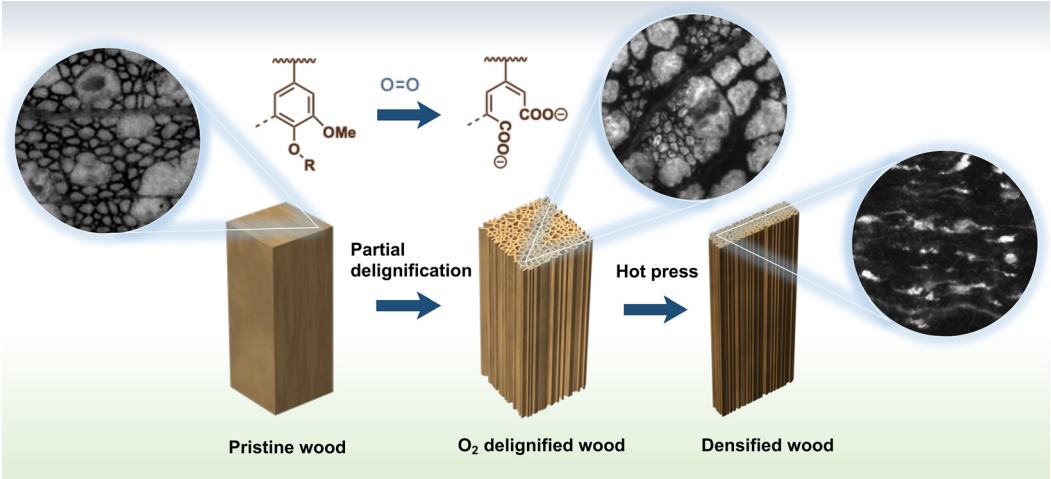
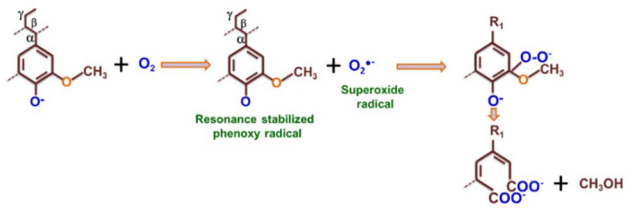


Fig. 1 A schematic illustrating the oxygen delignification mechanism and subsequent wood densification.



Scheme 1 Oxidation and carboxylation of lignin by oxygen.

sulfur-containing lignin from conventional alkaline/kraft delignification.[36,37] However, evaluation of nutrient recovery for fertilizer use would require further compositional and agronomic assessment and was not the scope of the present work.

To evaluate the potential environmental benefits of densified wood through O_2-delig compared with traditional building structural materials, we performed a comparative life cycle assessment (LCA) for wood-based and ore-based materials including Al-alloy and steel. Both ore-based materials were studied at low and high levels of recycled material content (LRC and HRC), with the objective to illustrate that the O_2-delig method is sustainable. The LCA results concluded that O_2-delig has minimal to no adverse effects on the environment and the properties of resultant wood materials, which reveals great potential in applying high strength[13] and moldable wood[14,15] for sustainable economic development and carbon sequestration.

Materials and methods

O_2-delig and densification of wood were carried out according to the experimental flow schematic illustrated in Fig. 1. Wood is impregnated with an alkali solution, *e.g.* KOH, NaOH, Na_2CO_3, or urea, and then delignified using O_2 typically at 0.75 MPa and 110 °C for one hour. As will be discussed later, the alkali loading is substantially lower than that for alkaline delignification. Furthermore, using low temperatures (85–115 °C) is a significant advantage in terms of energy savings over alkaline delignification, typically operated between 150 °C and 170 °C.[24,27] As shown in Scheme 1, the primary and initial O_2 attack on lignin results in oxygen gaining an electron from phenolic lignin to form a superoxide anion radical ($O_2^{\bullet-}$) and a resonance stabilized phenoxy radical.[29,33] The superoxide radical ($O_2^{\bullet-}$) is a weak oxidizing agent and does not react with carbohydrates but continues to react with phenoxy radicals and, therefore, is a highly selective oxidant for wood delignification. There are many other parallel reactions including carbohydrate degradation reactions caused by unselective hydroxyl radicals and oxyl anion radicals formed through the conversion of superoxide anion radicals. By properly controlling the pH around 11 along with increasing oxygen pressure in O_2-delig, carbohydrate degradation can be minimized.[29]

Materials

Pristine basswood boards, with dimensions of 45.21 cm × 17.53 cm × 2.29 cm (17.8 in × 6.9 in × 0.9 in), were sourced commercially and complimentarily provided by Inventwood, Inc. (Frederick, MD, USA). The moisture content of the boards was measured at 4.16%. Compressed oxygen with a purity of 99.0% was obtained from Badger Welding Supplies, Inc. (Madison, WI, USA). ACS reagent grade magnesium sulfate and sodium hydroxide were purchased from Sigma-Aldrich (St Louis, MO, USA).

O_2-delig and densification of wood

Eleven pristine basswood boards labeled as A through K were weighed individually and used for producing densified wood boards through O_2-delig under different conditions with board K being the control without O_2-delig. As shown in Fig. 2, the wood boards were first soaked in NaOH solution for alkaline impregnation to buffer O_2-delig reactions. NaOH impregnation was conducted either at a room temperature of 25 °C for a

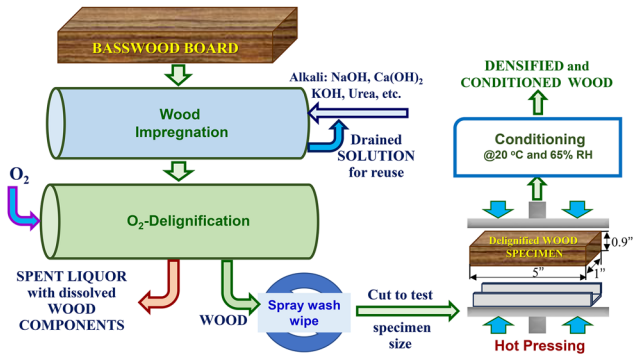


Fig. 2 A schematic flow diagram showing aqueous wood impregnation, gas phase O_2-delig, densification, and conditioning.

week (long) or at 70 °C for only 2 hours (short). $MgSO_4$ at a loading of 5 g kg^{-1} wood was supplemented into the NaOH solution to prevent cellulose degradation during O_2-delig. Applying $MgSO_4$ in O_2-delig is a common practice in commercial wood fiber production with a wealth of literature. The main mechanisms are preventing the generation of unselective hydroxyl radicals by absorbing transition metals to form a complex[38] and the synergistic effect between $MgSO_4$ and phenol to behave as a protector system for carbohydrates and improve delignification selectivity.[39] O_2-delig was carried out at a temperature of 90 °C and 110 °C and an oxygen pressure of 0.62 MPa (90 psi) and 0.76 MPa (110 psi) for 1 h except for one run for 0.5 h. NaOH concentration varied from 15 to 50 g L^{-1} (Table S1). The actual NaOH charge on wood ranged between 28 and 98 g kg^{-1} (Table S1) based on the measured amount of NaOH solution uptake (Table S2), which is substantially lower than approximately 200 g kg^{-1} in alkaline wood pulping. The specific impregnation and delignification conditions are listed in Table S1. The run conditions were abbreviated as *t*A*zz*T*xx*O*yy* with *t* and *zz* representing NaOH soaking time ($t = h = 2$ hours at 70 °C, $t = w = 1$ week at 25 °C) and concentration in g L^{-1}, respectively, and *xx* and *yy* representing O_2-delig *T* in °C and O_2 pressure in psi, respectively. For the O_2-delig run of 0.5 h, the abbreviation was amended as *w*A30T125O110-*t*05. Impregnation was conducted in a vessel of 1.7 m internal diameter and 3.6 m length with an effective volume of approximately 8 m^3. The wood package was loaded into the impregnation vessel. After alkaline impregnation, each wood board was removed from the solution, drained of excess liquid, and wiped with paper towels. Each board was weighed and subsequently loaded into a vertical 21 L wood pulping digester heated *via* a steam jacket. Steam was injected into the reactor to heat the wood boards to the desired O_2-delig temperature. Oxygen was then introduced to pressurize the vessel to the target pressure (Table S1).

The O_2-delig of wood boards was maintained at the desired temperature and O_2 pressure for a specified reaction duration using several short bursts of direct steam and O_2 injection during treatment. At the end of the reaction, the wet wood boards were weighed, and the mass changes due to impregnation and O_2 delignification were recorded (Table S1). The pretreated boards were then left to air dry under ambient room conditions.

After O_2-delig and air drying, the wood boards were cut into several smaller size test specimens with dimensions $L \times W \times H$ of 127 cm × 25 cm × 23 cm. The specimens were placed in a U-shaped steel restraining channel and densified (Fig. 2) using a Carver laboratory heated-platen 12 cm × 12 cm hydraulic press (Platen 2, Sterling, Inc., Menomonee Falls, WI, USA) at 2.0 MPa and 150 °C for only 20 min. This low pressure and short time densification process was employed to decrease densification cost as a long occupying time of a capital-intensive high-pressure press can be costly. Compared with over 12 h pressing time reported in the literature,[13] our process intensity increased over 36 times. After densification, the wood samples were conditioned in a climate-controlled chamber at 22 °C and a relative humidity (RH) of 65% for two weeks, which resulted in an equilibrium wood specimen moisture content of 12%.

Wood yield and chemical composition determination

A strip of wood sample was cut from each O_2-deliged basswood board. The dimensions of the strip were recorded. By comparing the dimensions of the sample with the dimensions of the corresponding board, the oven-dry weight of each corresponding untreated wood (pristine wood) strip was determined assuming that the wood density was uniform throughout the entire board. The strip was then Wiley milled to 30 mesh. All milled samples were then washed. The solid content in the oven-dry weight of the washed sample was determined. The solid yield from O_2-delig for each board was determined by comparing the oven-dry weight of milled samples to the corresponding oven-dry weight of the pristine wood strip.

The washed and milled samples were used for chemical composition analyses. Chemical compositions of wood samples were determined by the Analytical Chemistry and Microscopy Lab at the Forest Products Laboratory using the conventional two-step acid hydrolysis method following a procedure developed by the National Laboratory of the Rockies (NLR).[40] Briefly, carbohydrates in wood samples were solubilized into monomeric sugars in two steps using aqueous sulfuric acid solution first at 72% (v/v) and 30 °C, and then at 3.6% (v/v) and 120 °C. The hydrolysate was analyzed using an ion chromatographer (Dionex ICS-5000, Thermo Fisher Scientific, Sunnyvale, California, USA) equipped with an integrated amperometric detector and a CarboPac PA1 column (4 × 250 mm) at 30 °C. Klason lignin was quantified using the gravimetric method with the ash content determined through combustion in a muffle furnace at 575 °C for 12 h.

Wood characterization

The bending modulus of rupture (MOR) and bending modulus of elasticity (MOE) of the densified basswood boards were measured by three-point static bending tests according to a modified standard procedure described in ASTM D4761-19.[41] Wood specimens were cut into dimensions of approximately

127 mm length by 25.4 mm width by 8 mm thickness and tested with a span of 114 mm (short) and 356 mm (long for pristine wood only) at a crosshead speed of 5 mm min^{-1} using a universal mechanical testing machine (Instron 5869, Grove, PA, USA). Mean and standard deviations of MOR and MOE of each wood sample were calculated from the data of 10 replicate specimens based on the following equations:

$$\mathrm{MOR} = \frac{3 \times F_{\max} \times l}{2 \times b \times t^2} \quad (1)$$

$$\mathrm{MOE} = \frac{l^3}{4 \times b \times t^3} K \quad (2)$$

where $F_{\max}$ is the peak (or failure) load, l is the span of supports, b is the sample width, t is the sample thickness, and K is the slope of the load *vs.* deflection curve between 20% and 40% at the maximum load obtained from linear regression.

Wood board hardness was measured by the Brinell hardness test according to European Standard EN 1534[42] with minor modifications, similar to that carried out in the literature.[43,44] Specimens with dimensions 20 mm (length) by 20 mm (tangential) by 8 mm (radial) were tested using the same universal mechanical testing machine for MOR with a ball diameter of D = 3.18 mm. During testing, the ball was penetrated to a fixed depth h = 1.60 mm. At least six replicate samples were tested for each wood board sample to obtain the mean and standard deviation of the Brinell hardness (H_B) in N mm^{-1} according to the below equation:

$$H_B = \frac{F}{\pi \times D \times h} \quad (3)$$

where F is the applied load and D is the diameter of the indenter ball.

Life cycle assessment of densified O_2-delig wood production

Calculations for mass and energy balances were accomplished using a Microsoft Excel spreadsheet (Microsoft, Redmond, Washington, USA). A copy of this spreadsheet can be obtained from the authors. The assumptions for the O_2 treatment and the results of mass balance calculations can be found in Tables S3 and S4. The densified wood was produced in this study using O_2-delig. The life cycle inventory for O_2-delig is presented in Table S5.

Cradle-to-gate system boundary

The functional unit of 1 kg of building material was selected and subsequently changed to MPa cm^3 g^{-1} based on the specific strength. The LCA was conducted using openLCA software and the TRACI (Tool for the Reduction and Assessment of Chemicals and Other Environmental Impacts) methodology developed by the U.S. Environmental Protection Agency (EPA).

Wood production was referenced from hardwood lumber production in the Northeast and North Central United States.[45] For this inventory, a selection of allocation methods was required. Mass allocation was used as the base for the calculations as suggested by the authors.[45] The transportation distance to the processing facility was assumed to be 300 km upon harvest.

Al-alloy production, which includes some additives such as 0.3% magnesium and 1.3% titanium, is referenced from Ecoinvent 3.10 documentation.[46] Two levels of recycled content were studied: LRC is defined as 20% scrap aluminum and 80% aluminum ingots and HRC for Al-alloy is defined at 20% aluminum ingots and 80% aluminum scrap (The Aluminum Association. https://www.aluminum.org/Recyucling, last accessed 05/15/2026).

In the same way, steel production was referenced from Ecoinvent 3.10.[46] Two levels of recycled content were defined: LRC at 19% iron scrap and 81% pig iron and HRC at 93% iron scrap and 7% pig iron (https://www.aisc.org/media/c00penl2/more-than-recycled-content.pdf, last accessed 05/15/2026).

In every process/inventory, the electricity source was changed to U.S. production (market group for electricity, medium voltage | electricity, medium voltage | Cutoff, U – US),[46] which sources its electricity from coal (20%), natural gas (39%), nuclear (18%), hydro (6%), solar (4%), and wind (10%) principally (International Energy Agency, 2024. Energy Statistics Data Browser – Data Tools, IEA. https://www.iea.org/data-and-statistics/data-tools/energy-statistics-data-browser?country=USA&fuel=Energy%20supply&indicator=ElecGenByFuel, last accessed 08/04/2024). Steam production for the advanced wood materials was modeled as being produced 50% from natural gas, 30% from oil, and 20% from coal.[46]

Chemical production was taken from the Ecoinvent database. The methods used to produce NaOH and O_2 are chloralkali electrolysis and cryogenic air separation. Chemicals were assumed to be transported 100 km by truck.

For comparative purposes, densified wood and ore-based materials were evaluated based not only on mass but also on specific strength. In this comparison, the MOR is used to represent the strength of wood, as it is the standard measure of flexural failure in brittle and quasi-brittle materials such as timber. For the metallic materials, the ultimate tensile strength (UTS) is used as the functionally comparable parameter, because metals typically undergo significant plastic deformation prior to fracture and do not fail abruptly in bending as wood does. UTS therefore better reflects the ultimate load-carrying capacity of ductile metals under tensile or flexural stresses. Accordingly, using MOR for wood and UTS for metals allows a consistent assessment of their ultimate strength-to-weight performance on a specific strength basis.

The first part of the analysis consisted of a cradle-to-gate system boundary. The inventory starts from the reception of raw materials and chemicals and continues until the finishing of the building materials at the facility gate.

Cradle-to-grave system boundary

The second part of the analysis consisted of adding end-of-life emissions. However, at this point, emissions during the use stage were omitted due to a lack of better data and the sensitivity of consumer practices during this stage.

End-of-life factors were based on the Waste Reduction Model (WARM) developed by the EPA.[47] Two possible end-of-life fates for wood-based materials were combustion and landfilling. Conversely, recycling was selected as the end-of-life of steel and Al-alloys. The possibility of recycling wood materials is limited due to the chemicals, glues, and additives added during the use stage, which can hinder their recyclability potential.

As suggested by the WARM documentation, wood materials were modeled using dimensional lumber, whereas construction and demolition steel and Al-alloy were modeled using aluminum ingot and steel cans (Table S6). Negative values in Table S6 represent the CO_2 credits taken by the end-of-life decisions in each case. Combustion of wood materials displaces fossil fuels, whereas landfilling of wood materials creates temporal carbon sequestration. Comparatively, recycling steel and Al-alloys avoids the production of their respective virgin materials.

Results and discussion

O_2-delig of dimensional wood

The chemical compositions of pristine wood and O_2-delig wood were analyzed (Table 1). Wood cell wall component mass losses were calculated based on the analyzed composition and wood solid yield. The extent of delignification based on acid insoluble lignin (AIL) or Klason lignin (KL) varied from 10% to 30% depending on the reaction conditions (Table S1). Cellulose and xylan losses were less than 10% and between 10 and 22%, respectively, except for the run impregnated with NaOH of 50 g L^{-1} or 98 g kg^{-1} wood. In general, the O_2-delig effectively delignified the cell walls with minimum loss of cellulose and moderate removal of xylan (the major hemicellulose). It is noticed that the variations in lignin removal and carbohydrate degradation between the regions of the wood board surface and center are in general small for the short alkali impregnation O_2-delig runs (A, B, and C in Table 1). It is expected that this variation is negligible for the week-long impregnation runs as verified by the uniformity in the color of the cross-sectional images of the wood boards (Fig. 3A). Except for the highest alkali loading 98 g kg^{-1} wood impregnation run, the reaction temperature and time played a minor role in affecting lignin removal and cellulose dissolution while week-long alkaline impregnation had the most noticeable effect for the range of O_2-delig conditions carried out.

The dissolved lignin could be separated in principle from the spent liquor for utilization and the remaining liquor could potentially be reused for subsequent impregnation steps depending on the base chemistry employed. Preliminary characterization of the physical properties of the dissolved lignin collected from all the O_2-delig runs with week-long impregnation was carried out in terms of solubility, molecular weight by GPC, visual color, and potential for sunblock application. While a further study is needed, the preliminary ana-

Table 1 Chemical compositions of O_2-delig basswood along with wood component mass removals (values in the parentheses)

Run	Run abbreviation	Wood yield (%)	AIL (%)	ASL (%)	Arabinan (%)	Galactan (%)	Glucan (%)	Xylan (%)	Mannan (%)
A (outside)			22.53	5.11	0.50	0.51	51.64	14.23	1.10
A (whole)	hA30T110O90	83.38	23.26 (10.3)	5.46 (9.7)	0.50 (16.6)	0.52 (32.3)	46.65 (8.7)	13.83 (21.5)	1.22 (64.4)
B (outside)			21.83	5.65	0.37	0.34	52.84	12.82	1.34
B (whole)	hA30T110O110	82.70	22.26 (14.9)	4.87 (20.1)	0.42 (30.5)	0.40 (48.3)	52.62 (−2.2)	14.18 (20.1)	1.56 (54.9)
C (outside)			20.64	5.76	0.49	0.52	50.31	13.23	0.84
C (center)	hA30T90O90	84.92	22.24	5.40	0.50	0.58	44.69	14.38	1.18
C (whole)			21.46 (15.7)	5.61 (5.5)	0.50 (15.1)	0.56 (25.7)	48.25 (3.8)	14.05 (18.7)	1.04 (69.1)
D	wA30T110O90	80.70	20.17 (24.7)	5.36 (14.2)	0.46 (25.8)	0.46 (42.0)	51.08 (3.2)	13.57 (25.4)	0.62 (82.5)
E	wA30T110O110	81.72	21.95 (17.0)	5.22 (15.4)	0.48 (21.6)	0.48 (38.7)	47.93 (8.1)	15.41 (14.2)	1.41 (59.7)
F	wA30T90O90	83.17	20.43 (21.4)	5.04 (16.8)	0.49 (18.5)	0.44 (42.8)	49.71 (3.0)	15.91 (9.9)	2.32 (32.5)
G	wA30T125O110-t05	77.28	22.52 (19.5)	5.53 (15.2)	0.44 (32.0)	0.37 (55.3)	51.29 (7.0)	14.24 (25.0)	0.80 (78.4)
H	wA30T125O110	78.47	20.95 (24.0)	5.35 (16.7)	0.45 (29.4)	0.45 (44.8)	51.48 (5.2)	14.63 (21.8)	0.65 (82.2)
I	wA50T110O110	69.18	22.15 (29.1)	4.47 (38.6)	0.41 (43.3)	0.31 (66.5)	54.17 (12.0)	11.60 (45.3)	0.92 (77.8)
J	wA15T110O110	79.84	21.81 (19.5)	5.32 (15.7)	0.49 (21.8)	0.43 (48.4)	51.20 (4.0)	14.62 (20.5)	1.80 (49.8)
K	Control	100.00	21.62	5.04	0.50	0.64	42.60	14.68	2.86

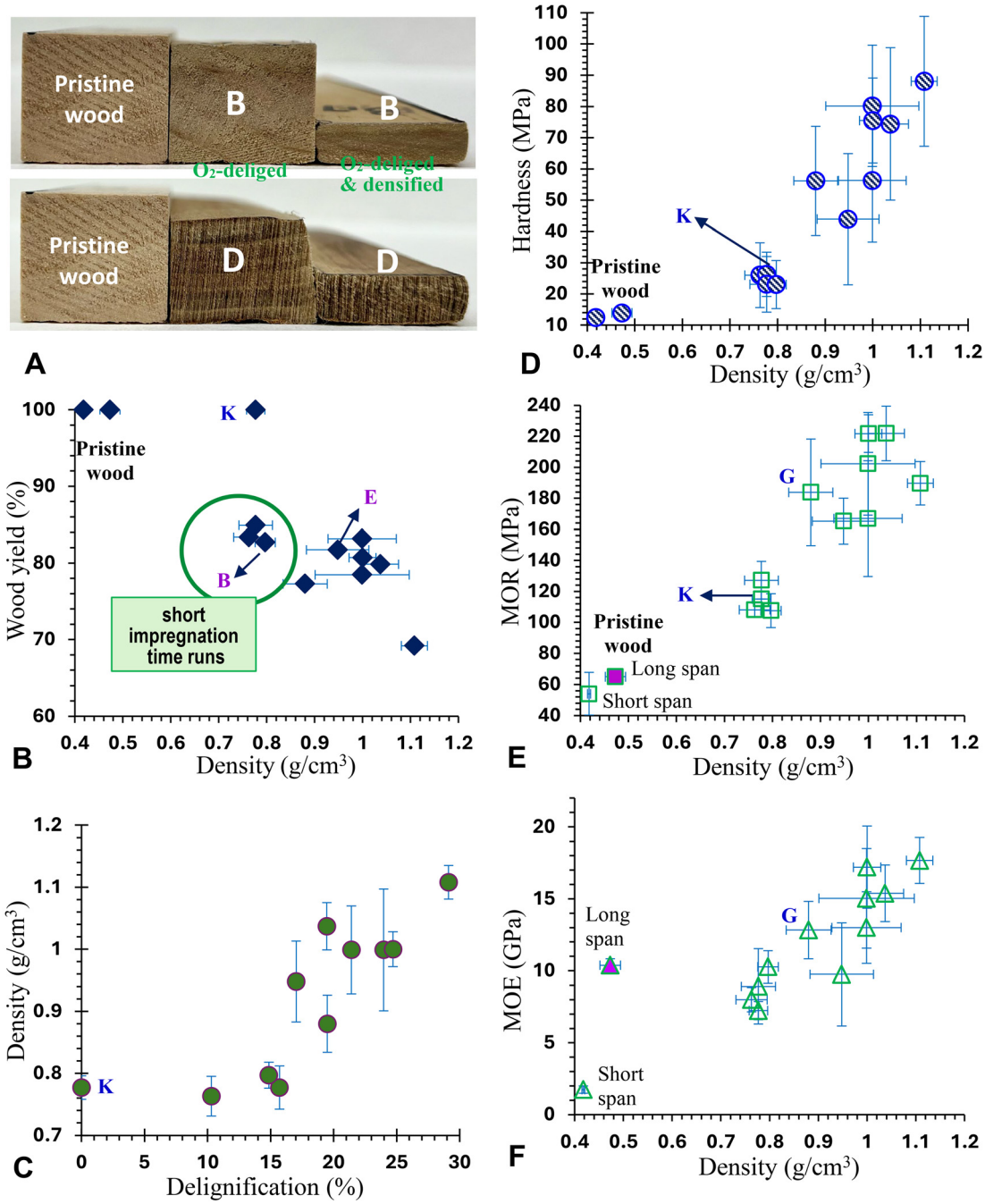


Fig. 3 Results from O_2-delig under various conditions and subsequent wood densification. (A) Images of oxygen delignified and delignified and densified wood from the two runs with short (2 h, Run B) and long (7 days, Run D) alkali impregnation times under the same O_2-delig conditions. (B) Relation between O_2-delig wood yield and density of the resultant densified wood. (C) Effect of the extent of delignification on the density of the densified wood. (D–F) Effect of wood delignification and subsequent densification on wood hardness (D), bending strength (MOR) (E) and bending modulus (MOE) (F).

lyses show interesting lignin properties such as light color and great anti-UV ability for sunscreens (Fig. S1).

Mechanical properties of densified wood

Oxygen delignification with subsequent densification achieved a substantial increase in wood density to 1.1 g cm^{-3} or by 265% in Run I (wA50T110O110, Tables S1, S2, and 1) at the highest NaOH charge of 98.5 g kg^{-1} wood, as shown in Fig. 3B and Table 2. The extent of wood densification under the given densification conditions (150 °C for 20 min under 2 MPa) was somewhat dependent on wood yield, suggesting that increasing wood loss or the extent of wood structural loosening facili-

Table 2 Density and mechanical strength of the densified basswood

Board labelling	Run abbreviation	Wood solid yield (%)	Thickness (cm)	Density (g cm^{-3})	Hardness (MPa)	MOR (MPa)	MOE (GPa)
Pristine wood	Short span	100	2.2349 ± 0.0023	0.418 ± 0.003	12.4 ± 0.9	54.0 ± 13.7	1.74 ± 0.26
Pristine wood	Long span	100	2.2342 ± 0.0066	0.470 ± 0.021	13.9 ± 2.0	65.0 ± 3.0	10.38 ± 0.45
A	hA30T110O90	83.38	0.9589 ± 0.0653	0.763 ± 0.032	26.0 ± 10.4	108.1 ± 4.5	8.00 ± 0.85
B	hA30T110O110	82.7	0.8480 ± 0.0375	0.797 ± 0.021	23.0 ± 7.7	107.6 ± 11.0	10.27 ± 1.13
C	hA30T90O90	84.92	0.9735 ± 0.0446	0.777 ± 0.035	23.1 ± 8.9	127.2 ± 12.2	8.92 ± 2.61
D	wA30T110O90	80.70	1.0330 ± 0.0633	1.000 ± 0.028	75.5 ± 13.6	221.8 ± 12.2	17.20 ± 2.85
E	wA30T110O110	81.72	0.9705 ± 0.0952	0.948 ± 0.065	43.9 ± 21.0	165.3 ± 14.8	9.75 ± 3.58
F	wA30T90O90	83.17	0.9098 ± 0.0645	0.999 ± 0.071	56.3 ± 19.7	167.0 ± 37.3	13.00 ± 2.49
G	wA30T125O110-t05	77.28	1.0859 ± 0.0444	0.88 ± 0.046	56.2 ± 17.5	183.9 ± 34.4	12.83 ± 2.00
H	wA30T125O110	78.47	1.0370 ± 0.1339	0.999 ± 0.098	80.2 ± 19.4	202.3 ± 33.1	15.03 ± 3.47
I	wA50T110O110	69.18	1.0166 ± 0.0830	1.108 ± 0.027	88.1 ± 20.8	189.7 ± 14.1	17.67 ± 1.60
J	wA15T110O110	79.84	0.9154 ± 0.0213	1.037 ± 0.038	74.4 ± 24.4	221.9 ± 17.6	15.38 ± 1.97
K	Control	100	1.0349 ± 0.0279	0.777 ± 0.019	26.3 ± 7.1	115.1 ± 4.6	7.24 ± 0.63

tated wood densification. However, large variations in wood densities from 0.75 to 1.10 g cm^{-3} were observed among runs with wood solid yields around 80%. The variations appear to be caused by the difference in the extent of wood alkali impregnation (Fig. 3B, comparing Run B with E under the same O_2-delig conditions but different impregnation times). It is generally understood that room temperature alkaline impregnation may swell cellulose and degrade some fast reaction hemicelluloses including acetyl groups,[48] but not delignify wood. Aqueous NaOH (40 g L^{-1}) soaking at 145 °C for 10, 20, and 30 min of juniper, hawthorn, and birchwood did not result in noticeable changes in the chemical compositions of the NaOH treated wood for all these species.[49] The slight increase in cellulose content is simply due to the enrichment as some acetyl groups were dissolved. Based on the general rule of reaction kinetics, the 20 min reaction at 145 °C is equivalent to 1365 h (or 56 days) at 25 °C or 43 h at 75 °C, much longer than the 7 days at 25 °C and 2 h at 70 °C applied in the present study. In other words, the NaOH impregnation in the present study is much milder than that in this reported work.[48] Unfortunately, this work[48] did not report wood solid yield after NaOH soaking, so, the exact component mass losses such as delignification and carbohydrate dissolutions were not available.

The above analysis based on literature data from wood[48] suggests that the observed difference in delignification under two different sets of impregnation conditions was not due to delignification during impregnation, but rather can be attributed to limited alkali diffusion in wood using a short impregnation time. Statistical analysis using ANOVA indicates that the lower mean density of 0.78 g cm^{-3} for the densified wood group (K and A − C) with no or a lower extent of delignification (short impregnation time runs) compared to the mean density of 1.00 g cm^{-3} of the densified wood group (D–J) with greater extent of delignification (a week long impregnation) is significant with a probability of over 99.9% (Table S7). Plotting the wood density against delignification clearly indicates that densification was improved with increasing delignification (Fig. 3C). Densification of the pristine wood under the given pressure of 2 MPa at 150 °C for 20 min only achieved a wood density of 0.78 g cm^{-3} (Run K), while runs with 20% or more delignification resulted in a wood density of approximately 0.90 g cm^{-3} or greater under the same densification conditions. This effect of wood impregnation on wood densification can also be seen from the light microscopy images of the wood cell wall anatomic microstructures (Fig. S2). The wood sample from Run B with a short impregnation time (2 h) had an almost identical wood cell anatomical microstructure to the pristine wood Run K; in comparison, the wood sample from Run E with a long impregnation time (1 week) showed deformation in the vessel elements.

Densification substantially improved wood mechanical properties. The improvement is dictated by the increase in wood density. The pristine wood had a Brinell hardness of 12 MPa, as shown in Fig. 3D. Densification of the pristine wood without delignification (Run K) with 86% density increase improved hardness to 26 MPa, or increased by 112%, similar to those runs with 10–15% delignification with a short alkali impregnation time (Table 2). When the wood density was increased to 0.90 g cm^{-3} or greater (>115% increase) with 20% or more delignification, the Brinell hardness increased to over 40 MPa, or increased by 254% or more. When the wood density was increased to 1.11 g cm^{-3} or increased by 166%, the Brinell hardness increased to 88 MPa or by 691%. ANOVA of the mean hardness data between each two groups among the three groups: *i.e.*, (1) the pristine wood, (2) the densified pristine wood and densified low extent delignified wood (K and A − C) using a short impregnation time, and (3) the densified greater extent delignified wood (D–J) using a week long impregnation, shows that the differences are statistically significant with probability all over 99.7% (Table S8).

A similar trend was also observed in the bending strength. The bending modulus of rupture (MOR) was around 50 MPa for the pristine wood (both from the short and long span tests), as shown in Fig. 3E. Densification of the pristine wood and the wood samples with less than 15% delignification resulted in nearly doubling the MOR of the densified wood. Further increasing densification to a density of 0.90 g cm^{-3} or greater, with 20% or more delignification, resulted in approximately tripling or more wood MOR. Similarly, ANOVA of the

mean MOR data between each two groups among the three groups: the pristine wood, the densified pristine wood and densified low extent delignified wood (K and A − C), and the densified greater extent delignified wood (D–J), shows that the differences are statistically significant with probability all over 99.8% (Table S7). The non-linear trend in the Brinell hardness and bending MOR data (Fig. 3D and E) suggests that densification to approximately 0.90 g cm^{-3} or more is required to achieve substantial improvement in wood mechanical strength.

The bending modulus of elasticity (MOE) also increased with wood densification. Because MOE measurements depend on bending span, a long span test defined by standard ASTM D4761-19 was carried out for the pristine board as a reference, as shown in Fig. 3F. The pristine board was cut to dimensions of 8 mm by 127 mm by 356 mm for the short and long span tests, respectively, with span-to-depth ratios of 16 and 45, both meeting ASTM D143. ANOVA of the short span MOE data indicates that the difference between the mean MOE of the group of the densified pristine wood and densified delignified wood (K and A − C) with lower extents of delignification using a short impregnation time, and the group of the densified wood with greater extents of delignification using a week long impregnation (D–J) is statistically significant with probability all over 99.6% (Table S9).

Mathematical wood mechanical strength enhancing factor

Stress is often used to express wood strength, *e.g.*, tensile strength is measured by tensile stress calculated from the measured tensile failure force divided by the wood cross-sectional area. Densification decreases wood cross-sectional thickness, which results in a mathematical enhancement (increase) in tensile stress. We can define a term "mathematical enhancing factor (MEF)" to quantitatively determine this strength increase that is purely due to mathematics and has nothing to do with the actual wood strength improvement. Using the definition of tensile stress (σ), MOR (eqn (1)), and MOE (eqn (2)), the MEF for these three strength measures can be expressed as below:

$$\mathrm{MEF}_{\sigma} = \frac{\sigma_{\Delta t}}{\sigma} = \frac{t}{t-\Delta t} = \frac{1}{1-\delta_{\mathrm{t}}} \tag{4}$$

$$\mathrm{MEF}_{\mathrm{MOR}} = \frac{\mathrm{MOR}_{\Delta t}}{\mathrm{MOR}} = \frac{t^2}{(t-\Delta t)^2} = \frac{1}{(1-\delta_{\mathrm{t}})^2} \tag{5}$$

$$\mathrm{MEF}_{\mathrm{MOE}} = \frac{\mathrm{MOE}_{\Delta t}}{\mathrm{MOE}} = \frac{t^3}{(1-\Delta t)^3} = \frac{1}{(1-\delta_{\mathrm{t}})^3} \tag{6}$$

where t and Δt are the respective wood cross-sectional area thickness and thickness reduction by densification, and $\delta_{\mathrm{t}} = \frac{\Delta t}{t}$ is the relative thickness reduction. MEFs are only dependent on the wood cross-sectional area thickness functionality in the wood property measure equations such as eqn (1) and (2), and independent of wood species and wood delignification and densification processing conditions.

Wood strength enhancement by densification needs to be greater than the MEF to achieve actual gain in wood mechanical strength in terms of total load-bearing capacity. For example, densifying pristine wood (Run K) increased the wood density by 86% while decreasing the wood thickness by 46% (Table 2). This 46% decrease in wood thickness should have resulted in an increase in MOR by a factor of 3.4 according to eqn (5), substantially greater than the 2.1 increase from measurements (Table 2). This suggests that densifying pristine wood, under the present low pressure pressing conditions, resulted in a net loss in bending load-bearing capacity. It appears that a breakeven point for MOR was nearly achieved for Run D with MOR reaching 222 MPa (Table 2) when densification resulted in a wood density of 1.00 g cm^{-3} and a thickness of t = 1.03 cm.

The concept of MEF is critically important to practical applications because it is the total load, or the failure force, not the failure stress, that determines the amount of material needed for an application. The amount of material needed dictates the material cost. Unfortunately, this issue has never been discussed in the existing literature on densified wood. In fact, existing commercial products of densified wood are produced by densifying pristine wood, which results in a strength enhancement below MEF. This indicates that more timber (higher cost) is needed using densified pristine wood than using pristine wood itself for the same application. Densification does allow using more wood for increasing loading baeraing without occupying useful space. The experimentally measured gain in MOE for Run D was approximately 10 (MEF also approximates to 10) based on the data in Table 2, so Run D achieved the breakeven MOE.

The data obtained here (Table 2) indicate that a 20% delignification was needed to achieve the desired wood density of around 1.00 g cm^{-3} for basswood, or the breakeven wood density, under the relatively low densification pressure of 2 MPa. Identifying this lower delignification requirement of 20% under a lower densification pressure than the 45% delignification identified by the earlier study[13] that focused on achieving maximal strength using much higher pressures, provided significant flexibility for wood delignification to significantly decrease product cost and facilitate commercializing wood densification technology because delignification is the most expensive step. A lower delignification of 20% also improves wood yield, which is important for the final product cost as well as LCA to be discussed later. The lower densification with a maximum density of 1.11 g cm^{-3} reported in this study, lower than the 1.30 g cm^{-3} reported in the literature,[13] was mainly attributed to the low densification pressure of 2 MPa applied and can be improved by using a higher densification pressure of 5 MPa.[13]

The effect of O_2-delig alkali charge in wood densification

The O_2-delig alkali also had some effects on the mechanical properties of the densified wood, although the extent of densification and the extent of wood alkali impregnation played major roles. It is known that proper control of the pH in O_2-

delig of chemical pulp fibers is important for effective delignification.[29] The appropriate pH for O_2-delig of dimensional wood has never been studied or established. Here, we varied the alkali charge to verify the pH effect on O_2-delig of dimensional wood due to practical difficulties in *in situ* pH measurements in semi-gas phase reaction systems. Due to the low temperatures used in the present study (Table S1), the alkali delignification mechanism should have limited contribution. The observed effects discussed below should be attributable to the pH buffering effect in O_2-delig. The highest alkali charge (~100 g kg^{-1} wood) run (I) resulted in a significantly greater amount of delignification of 30% (Table 2) compared with the remaining runs with approximately 20% delignification at lower alkali charges (~28–60 g kg^{-1}).

The wood anatomical microstructures using light microscopy imaging clearly show the vessel elements and tracheid cells in the control sample (no delignification, pristine wood, K2). O_2-delig slightly modified the wood anatomical microstructure after drying (left column of Fig. 4), as can be seen by comparing samples from the lowest with the highest alkali charge runs (J7 *vs.* I3), and samples from the highest alkali charge run with the control (I3 *vs.* K2). The removal of lignin loosened the wood structure to result in shrunk wood with some extent of cell collapsing after drying induced hornification or hydrogen bonding. This is especially obvious for sample I3 with the greatest delignification of 30% (Table 1) under the highest NaOH loading (Table S1).

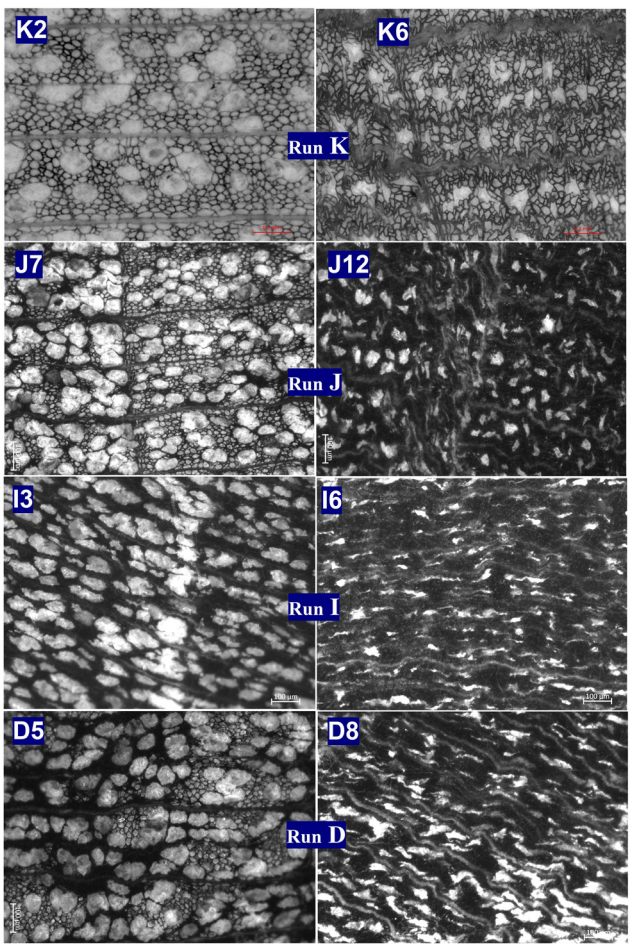


Fig. 4 Light microscopy images showing wood anatomical microstructures before and after densification. Left column: after O_2-delig before densification. Right column: after densification. Run K is the control without O_2-delig. O_2-delig conditions for Runs J, I, and D are listed in Table S1.

The extent of wood structure loosening also affected subsequent wood densification as discussed earlier and shown in the right column of Fig. 4. The fiber lumens remained widely open with minor deformation in the control sample K6 without O_2-delig but were not clearly observed in the samples with delignification (J12, I6, and D8). The voids of the vessel elements were also substantially deformed and decreased in these three O_2-delig samples, especially for sample I6 with the greatest delignification and densification. Interestingly, the MOR of the densified wood from Run I with the greatest density of 1.1 g cm^{-3} was 190 MPa (Table 2), lower than the densified wood from Runs J and D of 222 MPa with a slightly lower extent of densification of density 1.0 g cm^{-3}. ANOVA indicates that the observed differences in MOR between Runs I and J, and I and D are statistically significant both with the probability of 98.7% (Table S8). Perhaps, the greatest O_2-delig severity at the highest NaOH loading of 100 g kg^{-1} in Run I resulted in severe cellulose degradation and depolymerization as indicated by the greatest carbohydrate loss (Table 1) to negatively affect wood bending strength. Careful examination of the data in Table S8 indicates that a lower severity Run J at a NaOH loading of 28 g kg^{-1} achieved an overall great wood mechanical performance of hardness 74 MPa, bending MOR 222 MPa, and MOE 15.4 GPa, similar to those achieved from Run D of a moderate NaOH loading of 61 g kg^{-1}. This was supported by ANOVA with a probability of 98.9% for MOR and 90.9% for Brinell hardness (Table S8). This suggests that low alkali loading may be preferred.

Life cycle assessment (LCA) of densified wood

For the chosen boundaries used in LCA, densified wood from O_2-delig showed (Fig. 5) a low carbon footprint (CF) of 0.61 kg CO_2 eq. per kg, with the principal hotspot being wood sourcing (72%) and energy (18%). Comparatively, the primary hotspots for metal-based materials are the extraction of virgin metal (69%) for materials with LRC, and chemical production (41–64%) for those with HRC. Regarding CF, densified wood produces 3.2% of the emissions from producing Al-alloy with LRC and 6.5% of the emissions from producing Al-alloy with HRC. In better light, the densified wood produced 67.0% of the emissions released by producing steel with HRC and 26.5% of the emissions released by producing steel with LRC. LCA results show that using densified wood can decrease CF by at least 49% compared with metal-based materials.

In addition to global warming potential, nine other environmental impacts were studied (Fig. 5 and Table S9a). Al-alloys with LRC and HRC presented higher emissions on all impact categories when compared with densified wood. In the case of

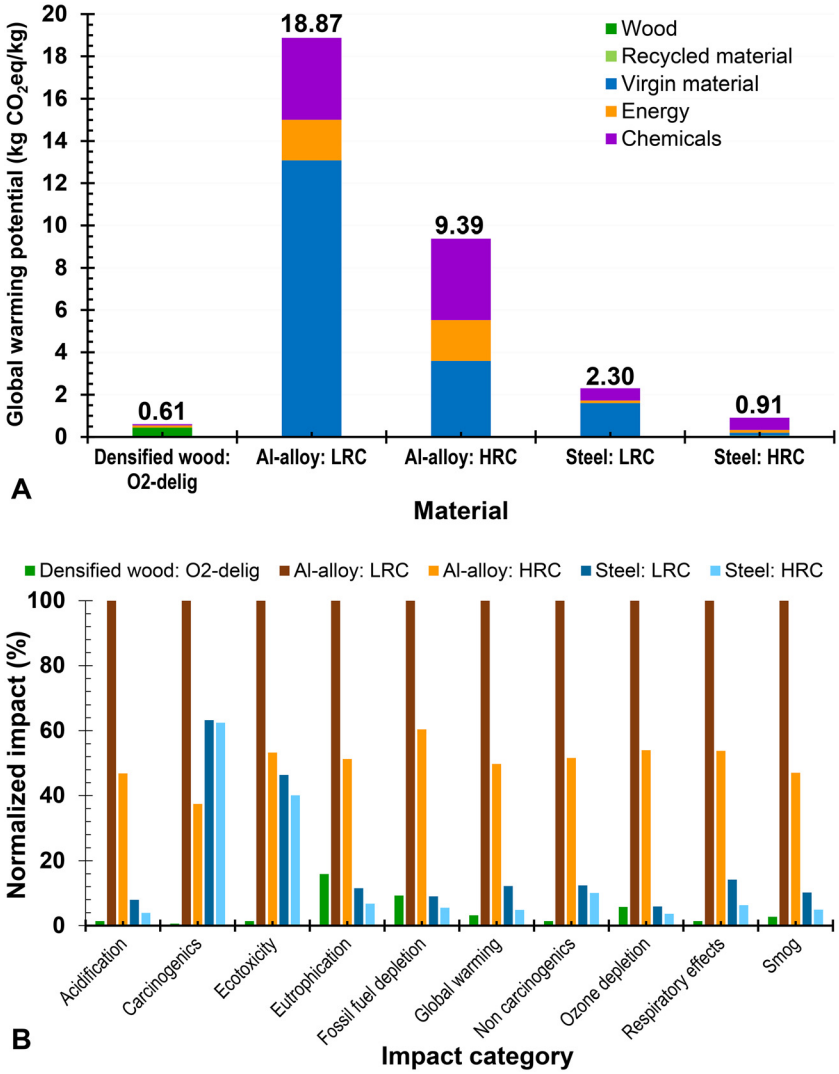


Fig. 5 Cradle-to-gate environmental impacts to produce 1 kg of building materials. (A) Carbon footprint. (B) Normalized TRACI environmental impacts.

steel, the results were more diverse. LRC and HRC steel presented higher values in eight out of the ten impacts, with the other two lower impacts being eutrophication and fossil depletion (18% and 5% lower, respectively). A higher concentration of organics in the wood-treatment effluents explains the greater eutrophication. Regarding fossil fuel depletion, steel production modeling bases its energy production only on natural gas, whereas the steam generated for wood heating comes from different sources, including oil and coal, which have lower high heating values, resulting in higher fossil fuel requirements.

When the functional unit is changed and is based on material specific strength, the environmental impacts follow the same trend.[14] The use of densified wood instead of Al-alloy with LRC means a reduction of at least 88% in all environmental impacts and particularly 97% in CF (Table S9b). Comparatively, when HRC steel is used as a baseline, the CF of wood is 51% that of steel. Additionally, eight out of the ten environmental impacts are lower for wood than for HRC steel (Fig. S3). In this sense, densified wood represented 49% reduction in the CF compared with the best-case metal-based building material (HRC steel). Using the material specific strength for comparison benefits the densified wood because more Al-alloy and steel are required to supply the same amount of strength as the densified wood.[13]

The environmental relevance of the alkali loading should be interpreted together with the overall process inventory rather than considering the alkali dose alone. Based on the life-cycle inventory used in this study (Table S5), the O_2-delig requires approximately 0.00389 kg of NaOH and 0.12 kg of water per kg of bleached wood, corresponding to about 3.9 kg of NaOH and 0.12 m^3 of water per metric ton of treated wood. These values indicate relatively low chemical make-up and limited liquid throughput for the pretreatment stage. For com-

parison, industrial wood delignification processes such as kraft pulping typically operate with substantially larger aqueous process streams and chemical inputs. Modern pulp mills operating under Best Available Techniques report wastewater discharges on the order of approximately 30–50 m^3 per ton of pulp after treatment, and active alkali charges commonly range from roughly 15 to 35 kg NaOH-equivalent per ton of pulp depending on wood species and operating conditions.[50] Although the objectives and system boundaries of pulp production and the present densification process differ, this comparison highlights that the O_2-delig operates with substantially smaller liquid volumes and lower alkali consumption. Nevertheless, such comparisons should be interpreted cautiously because industrial pulping processes involve extensive fiber processing, chemical recovery systems, and integrated wastewater treatment infrastructure that are outside the scope of the present laboratory-scale study.

To further contextualize the process intensity of the proposed O_2-delig process, a quantitative comparison with representative densified wood pretreatment strategies reported in the literature was performed (Table S10). These approaches typically rely on peroxide-, sulfite-, or strongly alkaline delignification chemistries, often involving several weight percent of reactive chemicals (*e.g.*, H_2O_2, Na_2SO_3, or NaOH-based liquors), elevated temperatures (100–170 °C), and extended treatment times (up to 12 h). Depending on liquor-to-wood ratios, such conditions can correspond to chemical inputs on the order of tens to more than 100 kg of reactive species per ton of wood. In contrast, the O_2-delig process developed in this work operates at moderate temperature (90–110 °C) and a short reaction time (1 h), with a substantially lower alkali requirement (≈3.9 kg NaOH per ton of wood, Table S5) and molecular oxygen as the primary oxidant. Despite this lower chemical intensity, the resulting densified materials exhibit mechanical performance comparable to previously reported "superwood" systems (MOR ≈200–300 MPa). This comparison indicates that similar structural performance can be achieved with significantly reduced chemical input, supporting the favorable environmental profile observed in the LCA. It should be noted that these comparisons are approximate due to differences in wood species, sample dimensions, and processing conditions across studies.

The economic benefits of the proposed O_2-delig process with lower intensity can be estimated by examining the costs of chemicals and energy inputs for delignification as it is the most expensive step in densified wood production. As detailed in the SI, the costs of chemicals are roughly estimated at \$0.020 per kg densified wood for O_2-delig compared with \$0.025–0.092 per kg for conventional NaOH delignification. The costs of heat input are roughly estimated at 0.59–0.93 MJ kg^{-1} densified wood for O_2-delig at 90–110 °C compared with 1.46–2.14 MJ kg^{-1} densified wood for conventional NaOH delignification at higher temperatures of 150–170 °C. These comparisons demonstrate up to a total of 4.4-fold cost reduction in chemicals and heat, highlighting the potential economic benefit for the O_2-delig process.

Additionally, a cradle-to-grave analysis was performed to study the influence of the end-of-life conditions. The use stage for building materials was not considered in this analysis because the variability in consumer practices is wide. The densified wood was analyzed using two possible end-of-life conditions: landfilling and combustion. Recycling is restrictive in this case because if adhesives or preservatives have been added, recyclability of the material would be hindered.[51] Comparatively, metal-based materials were assessed using recycling (Table S6).

Densified wood has more benefits from landfilling considerations with a CF of −0.31 kg CO_2 eq. per kg, as shown in Fig. 6. The negative sign illustrates that the carbon sequestration in landfills for more than 100 years is a carbon sink.

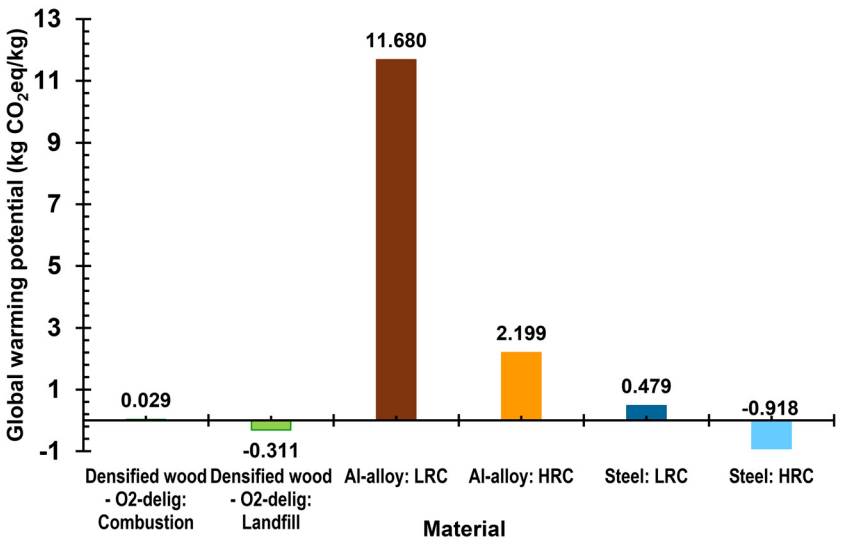


Fig. 6 Cradle-to-grave environmental impacts to produce 1 kg of building materials.

Combustion, comparatively, creates an almost neutral material with a CF of 0.03 kg CO_2 eq. per kg. In this sense, the combustion of wood materials displaces fossil-based fuel for energy generation, creating a useful output and reducing pressure on landfills.

The CF of combusted densified wood products represents less than 0.2% of the emissions by LRC Al-alloy and 6% of the emissions by LRC steel. HRC steel has the lowest CF, with −0.92 kg CO_2 eq. per kg, meaning that recycling importantly favors end-of-life options in this opportunity due to avoiding virgin material (pig iron) extraction. The modification of the functional unit to a specific strength basis does not modify the conclusion, with the combusted wood having 1.2% and 16.1% of the emissions from LRC Al-alloy and LRC steel, respectively (Table S11).

Conclusions

This study demonstrated a major technological breakthrough using low temperature (~100 °C) oxygen delignification (O_2-delig) to address a key barrier but prerequisite step – sustainable and scalable delignification of dimensional wood for advancing densified wood as a renewable alternative to energy-intensive ore or mineral-based nonrenewable structural materials. O_2-delig was conducted at low temperatures of 90–100 °C with a minimal alkali charge of 28–56 g kg^{-1} wood and achieved approximately 20% lignin removal with low carbohydrate loss or great wood yield of approximately 80%. Subsequent wood densification of the O_2-deliged wood at a low pressure of 2 MPa for only 20 min nearly doubled the wood density and substantially improved the wood mechanical strength with a bending modules of rupture over 220 MPa and a Brinell hardness over 80 MPa, suitable for many structural applications currently not possible using pristine wood. The technological advantages of O_2-delig are not only in terms of energy consumption but also in terms of process scalability, technology readiness, and eliminating the expensive chemical recovery operation as oxygen was used. Life cycle assessment showed that densified wood through O_2-delig has substantial benefits in terms of environmental impact compared with metals even with high recycling content – key to sustainable LEED building structural materials. This study also proposed the concept of "mathematical enhancing factor" (MEF), and for the first time revealed that when gains in wood strength measures are below MEF, such as the current commercial wood densification of pristine wood without delignification, wood densification results in actual loss in wood load-bearing capacity for a given wood mass due to the reduction in wood thickness.

Author contributions

Forfora and Zhang as co-first authors respectively contributed to LCA and experimental work. Forfora and Gonzales conducted LCA and drafted the LCA part of the manuscript. Zhang and Gleisner conducted the oxygen delignification. Zhang conducted wood densification and some data analysis, and drafted the experimental part of the manuscript. Houtman carried out engineering process modelling for LCA. Kitin conducted LM imaging. Begel and Fishwild conducted wood mechanical testing. Pan contributed to the technical issues of oxygen delignification. Kim conducted 2D NMR lignin chemical structure analysis. Zhu conceptualized and initiated the entire work, coordinated and supervised the study, and revised and finalized the manuscript.

Conflicts of interest

Gleisner and Zhu are co-inventors of a U.S. patent on oxygen delignification for wood densification to produce superstrong structural wood materials.

Data availability

All relevant data are included in the article and supplementary information (SI). Some data that support the findings of this study are available from the corresponding authors upon request. Supplementary information is available. See DOI: https://doi.org/10.1039/d6gc00841k.

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