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Fabrication and design of wood based high-performance composites

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Cover Letter

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28.5.18

Dear Mr. Werth,

We hereby submit "**Fabrication and design of wood based recyclable high-performance composites**", by Marion Frey, Meri Zirkelbach, Clemens Dransfeld, Eric Faude, Etienne Trachsel, Mikael Hannus, Ingo Burgert and Tobias Keplinger, for consideration as a Video Publication to Jove.

Natural fibers are increasingly considered as reinforcement of composites because of their recyclability and low energy consumption for production combined with high specific stiffness and strength. However, certain disadvantages such as poor fiber-matrix adhesion or variability in mechanical properties of fibers still have to be addressed in order to use natural fibers as a reliable replacement in structural applications. Recent progress in delignification of bulk wood, while retaining its structural hierarchy (Song et al. (2018) Nature; Frey at al. (2018) Applied Materials and Interfaces), represent an alternative top-down approach to obtain load-bearing cellulose scaffolds. After densification, these scaffolds possess superior mechanical properties to natural fiber reinforced composites such as flax- or sisal-fiber reinforced polymers and represent a promising alternative approach for the development of sustainable bio-based materials.

The manufacturing of the novel high-performance wood-based material is conducted in two subsequent steps, delignification and densification, but the shaping into 3D parts is rather challenging until now. Formability prerequisites free water in between cells. But in this state, delignified wood can be hardly densified with standard processing techniques. Water in between fibers creates a counter pressure, which leads to reduced densification, cracks and distortions in the fiber alignment.

In this manuscript, we demonstrate our novel open-molding process to manufacture cellulose composites based on wood in a scalable way by vacuum forming in open-molds and compare this method to standard densification in closed molds. Our new approach allows to combine densification and shaping in a one-step process and will facilitate new design strategies for manifold application areas as automotive industry or aviation. This research should appeal to the broad readership of Jove and specialists alike and is thus appropriate for publication here. We thank you for your consideration of this manuscript.

We hope that you find this work interesting for Jove and look forward to hearing from you.

Kind regards,

Tobias Keplinger

Marion Frey and Tobias Keplinger, on behalf of the authors

TITLE:

Fabrication and Design of Wood-Based High-Performance Composites

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KEYWORDS:
Delignified densified wood, bio-based, recyclable, natural fiber composite, cellulose scaffold, wet shaping, vacuum forming, design

SUMMARY:
Delignified densified wood represents a new promising lightweight, high-performance and bio-based material with great potential to partially substitute natural fiber reinforced- or glass fiber reinforced composites in the future. We here present two versatile fabrication routes and demonstrate the possibility to create complex composite parts.

ABSTRACT:
Delignified densified wood is a new promising and sustainable material that possesses the potential to replace synthetic materials, such as glass fiber reinforced composites, due to its excellent mechanical properties. Delignified wood, however, is rather fragile in a wet state, which makes handling and shaping challenging. Here we present two fabrication processes, closed-mold densification and vacuum densification, to produce high-performance cellulose composites

based on delignified wood, including an assessment of their advantages and limitations. Further, we suggest strategies for how the composites can be re-used or decomposed at the end-of-life cycle. Closed-mold densification has the advantage that no elaborate lab equipment is needed. Simple screw clamps or a press can be used for densification. We recommend this method for small parts with simple geometries and large radii of curvature. Vacuum densification in an open-mold process is suitable for larger objects and complex geometries, including small radii of curvature. Compared to the closed-mold process, the open-mold vacuum approach only needs the manufacture of a single mold cavity.

INTRODUCTION:

The development of novel natural fiber (NF) based composites equipped with superior mechanical properties represents one of the main tasks in materials science, as they can be sustainable alternatives for current synthetic systems such as glass fiber composites[1-3]. Besides traditional NF composites (flax, hemp, kenaf, etc)[4,5], the densification of wood after partial or complete removal of matrix components has received increasing attention in recent years[6-11]. The top-down fabrication route, based on delignification of bulk wood followed by densification, is conceptually contrary to rather complex bottom-up processes for pulp and slurry based products.[12] In pulp and slurry based products, the beneficial wood fiber alignment is not retained as fibers are separated in the process. In contrast, structure-retaining delignified wood, which is obtained in a top-down process, transfers the sophisticated architecture with aligned cellulose fibers into the new material. To achieve densification of delignified wood without fiber alignment distortions, new processing routes must be developed.

Direct densification of water-saturated delignified wood samples leads to a limited densification degree, cracks, and fiber alignment distortions due to the wet-sample-inherent free water that creates a counter pressure during densification. Current solutions to avoid structural integrity loss upon densification includes utilization of partially delignified wood followed by high-temperature densification[9] or pre-drying of delignified wood prior densification[6]. Both methods enhance connectivity between neighboring cells, either due to the remaining lignin that acts as glue or free water removal between cells.

In both cases, reduced formability occurs, which limits design applications; the required sample pre-conditioning also leads to longer processing times. Therefore, a fast and scalable process that combines shaping and densification in a single step is necessary.

In this regard, we present here open/closed-mold densification and vacuum processing of delignified wood as methods to combine shaping, densification, and drying in a simple and scalable approach. **Figure 1** shows delignified densified wood-composite parts, which were obtained by using the techniques described in this work.

[Place **Figure 1** here]

PROTOCOL:

1. Delignification of wood veneers

NOTE: This delignification protocol is based on our previous works, published by Frey et al. 2018[6] and Segmehl et al. 2018[13].

1.1. Mount a stainless-steel sample holder in a crystallizing dish or in a beaker and place a magnetic stir bar below the sample holder. Stack wood veneers on top of the holder and separate them by metal meshes or metal mesh stripes (**Figure 2A**). Here, we use radial cut spruce veneers with a thickness of 1.5 mm. Wood species and type (tangential, radial, rotary cut veneer) as well as the thickness of veneers can be varied.

1.2. Prepare a 1:1 volume mixture of hydrogen peroxide (30 wt%) and glacial acetic acid and pour the mixture into the crystallizing dish until the veneers are fully covered. Use glass dishes (e.g. Petri dish) to keep the veneers in the solution. Soak samples in the solution at room temperature (RT) overnight while stirring at 150 rpm.

1.3. Heat the solution to 80 ˚C and run the reaction for 6 h for full delignification. Adjust the delignification time depending on the sample thickness.

1.4. After delignification, pour the delignification solution into an empty beaker and let it cool down before disposal. Gently rinse the delignified veneers multiple times with deionized water. Then, continue washing the veneers without stirring by filling the crystallizing dish (beaker) with deionized water. Replace the water twice a day until a pH value of the washing water of above 5 is reached (**Figure 2B**).

1.5. Handle wet delignified wood veneers with care, as the cellulose scaffold is rather fragile. Use a metal mesh as support for transportation and draping (**Figure 4**).

[Place **Figure 2** here]

2. Storage and "cellulose prepreg" production

2.1. Consider processing the wet delignified wood samples within 2–3 weeks. Alternatively, preserve the material for long-term storage in ethanol (EtOH) or dry the sheets between metal meshes.

2.2. Store the dry, flat cellulose sheets ("cellulose prepregs") below 65% relative humidity (RH). Rewet the sheets in water prior to further shaping and processing.

3. Densification and forming of delignified wood in closed molds

3.1. Use closed molds made out of an open-porous material (e.g. ceramic molds, porous 3D printed polymer molds) to enable water removal and sufficient drying. Pore sizes should be below 2 mm, especially towards the surface, to obtain a smooth surface of the final composite part.

3.2. Condition the delignified wood at desired RH. For curvature radii in the cm range or plane structures, use samples that are conditioned at 95% RH at 20 ˚C. For smaller curvature radii, drape the veneer in water-saturated state, pre-dry the draped material in an open mold at 95% RH, or pre-dry the material in an oven (65 ˚C) for 5–30 min (the time is depends on the sample thickness). Curvature considerations are made in relation to veneer thickness (here 1.5 mm).

3.3. Densify the material in the closed mold either by using screw clamps or in a press. Readjust the pressure if needed to compensate for shrinkage. The drying process can be speed up by placing the mold into an oven at 65 ˚C or by increasing the temperature of the press.

NOTE: A relatively low pressure in the range of a few MPa is sufficient to densify wet delignified wood. The final thickness can be controlled by using spacers with the targeted thickness between the mold surfaces rather than by controlling the pressure.

3.4. After full drying, demold the composite part and reuse the mold for a new run.

4. Vacuum shaping and densification of delignified wood in open molds

4.1. Use a porous open mold as described in 3.1. Alternatively, use non-porous molds with a porous layer (e.g. mesh, textile, breather) on top of the mold or on top of the delignified wood to enable drying (**Figure 3A**).

4.2. Use a textile layer (e.g. peel-ply) to protect the mold from contamination. Drape a water-saturated delignified veneer on top of the textile (**Figure 3B**) and cover it with a second textile layer and flow mesh.

NOTE: To obtain a smooth surface finish, we recommend using porous closed-mold processing. For this, replace the flow mesh with the porous top part of the mold. However, if surface patterning with e.g. a mesh is desired, the open-mold process is a good alternative.

4.3. Place the mold on top of a stainless-steel plate, apply sealing tape and vacuum tubing, and wrap the mold (open or closed) with a vacuum bag. Use flow mesh to enable water flow to the vacuum tubing. Optionally, place additional mesh layers below the mold to enhance the drying process and to avoid local vacuum pressure drops, especially for bigger parts (**Figure 3C**).

4.4. Apply a vacuum for drying and simultaneous densification of the composite. For accelerated drying, place the setup into an oven at elevated temperature (e.g. 65 ˚C).

NOTE: Make sure to use cold traps to avoid water entering the vacuum pump. We here use an oil pump in a pressure range of 10^{-2} bar. However, it is also possible to use a membrane pump but trade-offs regarding densification degree might need to be taken into account.

4.5. After drying, demold the dry composite and reuse the mold and vacuum setup for a new composite part (**Figure 3D**).

[Place **Figure 3** here]

5. Manufacturing of laminated composite parts

5.1. Manufacture thick multi-layer composite parts by lay-up techniques and choose the fiber orientation angle of the layers (e.g. [0˚], [0˚/90˚], [0˚/-45˚/90˚/+45˚]$_S$) as in traditional composite manufacturing.

NOTE: The number of layers can be chosen depending on the targeted thickness of the final part. However, the vacuum time strongly depends on the size and thickness of the part and ranges from 2 h (single layer, 1.5 mm thick) up to 2 days for an 8-ply part.

5.2. Increase bonding between delignified wood layers by applying adhesive between layers during the draping process. Use a water based adhesive (e.g. starch) which allows combined drying and curing of the adhesive.

NOTE: We apply 0.04 g/cm^2 of a 16.5 wt% starch solution between the layers. However, other water-based glues could be used alternatively.

5.3. Demold the composite part and machine finish by hand or with standard wood tooling (**Figure 6E,F**).

6. Re-use and recycling of composite parts

6.1. Place delignified non-glued wood composites in water until the part regains formability. Then, either reshape the material to obtain a new product (see Frey et al. 2019[7]) or reduce it to small pieces.

6.2. Reuse the small pieces of delignified wood to create new products inspired by standard pulp techniques (e.g. pulp molding) and finally let the material biodegrade after end of life.

REPRESENTATIVE RESULTS:

Delignification and handling of wood veneers.

Complete delignification leads to a mass reduction of around 40% and a volume reduction of around 20% after drying at 65% RH[6]. Besides lignin, a fraction of hemicelluloses gets removed too. Removal of these components results in a fragile cellulose material (see **Figure 4**). Using metal meshes as supports eases handling and draping.

[Place **Figure 4** here]

Densification and forming of delignified wood in closed molds.

Densification of water-saturated delignified wood (**Figure 5A–C**) is demanding, as free water in the scaffold creates a counter pressure upon densification and allows the material to flow during processing. This causes fiber deviations and cracks in the final material (**Figure 5B,C**). One possibility to bypass these limitations is to use moist pre-conditioned (95% RH and 20 ˚C), delignified wood. In this condition, delignified wood is still reasonably shapeable and its densification does not lead to fiber alignment distortions and defects.

Pre-conditioned material, however, is more rigid compared to the water-saturated state, which makes it difficult to obtain small curvature radii without material damage. For small curvature radii, wet draping followed by conditioning in an already shaped state prior densification can be used. However, conditioning is rather time consuming and therefore not recommended for large-scale applications.

[Place **Figure 5** here]

Vacuum shaping and densification of a laminated part in an open mold.

Exemplarily for vacuum shaping, we manufactured a helmet in a self-made gypsum mold using an open-mold process (**Figure 6A,B**). As lay-up, we draped 2 layers of hexagon-flakes for surface texturing followed by 4 layers of delignified wood veneer in a [0˚/90˚] lay-up (**Figure 6C**). The flakes provide an attractive surface design, whereas the unidimensional (UD) layers add strength and stiffness to the composite. We applied 16.5 wt% starch as adhesive between layers to prevent delamination[14].

Vacuum densification (**Figure 6D**) leads to full drying of the part within 48 h and densification down to a thickness of 3 mm (1/3rd of the initial thickness). After the vacuum processing, the composite part is demolded (**Figure 6E**) and the edges are trimmed with a cutter (**Figure 6F**).

The maximum layup thickness that could be densified and fully dried with the open-molding approach was an 8-layer layup (8 x 1.5 mm veneer) with an end thickness of this part of 2.5 mm, which corresponds to a densification down to approximately one quarter of the initial thickness of dry delignified wood, taking into account the layer shrinkage upon delignification and drying. To obtain such high densification degrees, a low vacuum in the range of 10^{-2} bar is needed.

Delignified wood composites that are densified to around one quarter of their initial thickness typically achieve elastic moduli values around 25 GPa and strength values in the range of 150-180 MPa, as shown in our previous work (**Table 1**)[7].

[Place **Table 1** here]

[Place **Figure 6** here]

Utilizing flow meshes typically results in a mesh-imprint in the sample. This can either be considered as a process-inherent design strategy or can be prevented by placing an additional thicker textile layer between delignified wood and flow mesh.

Alternatively, closed molds combined with vacuum processing as described in protocol step 4.2 can be used. Regular patterning can be obtained by placing small pieces of delignified veneers in a defined order, as shown before for our example with the hexagon patterning on the helmet.

Problems that can arise during vacuum processing include warpages in the composite part, which are caused by incomplete drying and the occurrence of cracks (**Figure 7**). Cracks mainly result in delignified wood that was stored in EtOH prior composite fabrication. Therefore, after EtOH storage, we recommend to carefully soak delignified wood in water before further processing. Additionally, careful draping followed by slight densification by hand to remove some free water reduces the risk of cracking.

[Place **Figure 7** here]

Re-use or decomposition of composite parts.

Our cellulose-starch composite is all bio-based and can disintegrate in water. On the one hand, the hydrophilicity of the material is a disadvantage, as it leads to reduced mechanical performance when in contact with water. A simple method to protect the composite from liquid water comprises hydrophobic coatings, as we have shown in Frey et al. 2019[7]. On the other hand, a hydrophilic behavior of the material can also be beneficial when it comes to end-of life use and recycling aspects. The sample can simply be disintegrated in water to smaller pieces and the fibrous slurry can further be used for the production of new fiber-based products as shown in **Figure 8**. Furthermore, the fibrous material is fully biodegradable, as shown in **Figure 9**.

[Place **Figure 8** here]

[Place **Figure 9** here]

FIGURE AND TABLE LEGENDS:

Figure 1: Examples of delignified densified wood composite parts. (**A**) Door panel, (**B**) side mirror, (**C**) door handle of a car, (**D**) orthosis, (**E**) cut open helmet, and (**F**) tachometer cover of a car.

Figure 2: Delignification setup. (**A**) Crystallizing dish with metal mesh sample holder and wood veneers stacked on top of the sample holder. Metal mesh stripes separate the individual veneers from each other. (**B**) Delignfied veneers covered by water during the washing process.

Figure 3: Schematic illustration of the open-mold process. (**A**) Porous mold with smaller pores towards the surface. (**B**) Delignified wood draped on top of the porous mold (grey) and optional textile layer for mold protection (green). (**C**) Textile, flow mesh and vacuum bag placed on top of

delignified wood. Pressure is applied through the vacuum bag and leads to densification and drying of the material. (**D**) Final composite after demolding.

Figure 4: Handling of delignified wood in wet state. (**A**) Fragile delignified wood in its wet state. (**B**) Handling of the material is eased by using a metal mesh for transportation or (**C**) for draping the material to a mold. (**D**) Delignified wood draped on top of a porous 3D-printed mold.

Figure 5: Closed-mold densification of delignified wood in a wet and moist state. (**A**) Densification of the water-saturated cellulose material leads to (**B,C**) cracks and fiber misalignment. (**D–F**) Densification of moist material, conditioned at 95% RH results in a better preservation of fiber alignment and less defects.

Figure 6: Manufacturing of a helmet by open-mold processing. (**A,B**) Molding of the original helmet using a gypsum mold. (**C**) Draping of two outer layers with hexagon flakes followed by draping the inner 4-layers in a [0/90] layup. (**D**) Densification and drying of the part by vacuum. (**E**) Demolding of the dry part and (**F**) finish using a cutter.

Figure 7: Possible problems arising in fabrication of complex geometries. (**A**) Back view and (**B**) side view of the manufactured helmet. (**C,D**) Small cracks due to shrinkage of the material during processing.

Figure 8: Re-use of delignified wood fibers. (**A–C**) Reduction of delignified wood veneers into small pieces by dispersing the material in water. (**D–F**) Re-use of the fiber slurry for producing the lining of a helmet. (**D**) Reveting of a silicon mold with fiber slurry. (**E**) Final lining of the helmet. (**F**) Lining made out of disintegrated delignified wood inside of the hard shell of the helmet.

Figure 9: Degradation of delignified wood fibers. (**A**) Petri dish filled with soil. (**B**) Placing the fiber slurry on top of the soil and (**C**) filling it with water. (**D**) Bio-degradation after one day, (**E**) after eight days, and (**F**) after 26 days.

Table 1: Literature values for tensile elastic modulus and tensile strength of densified delignified wood. The vacuum processing results in a densification down to 1/4th of the initial thickness, which corresponds to an FVC of 66%.

DISCUSSION:

We present versatile fabrication techniques to obtain high-performance delignified wood–based composites and suggest possible re-use and recycling strategies. Closed-mold processing prerequisites pre-conditioning of the material, as it cannot be processed in water-saturated state. Utilizing a closed-mold process, however, could be the method of choice especially if e.g. there is no vacuum setup available or if a nice (smooth) surface finish on both sides is desired.

Open-mold vacuum processing of delignified wood allows for combining shaping, densification, and drying of water-saturated samples in a simple and scalable approach. The technique is applicable for the production of complex geometries and offers a scalable alternative for closed-mold processes. We have manufactured composites by stacking delignified wood veneers using starch as adhesive between layers. Densification down to one quarter of the initial thickness resulted in a final thickness of 2.5 mm of the 8-layer thick composite part. For obtaining a smoother surface finish in the vacuum process, the use of a closed porous mold could be an appropriate alternative.

For both processing methods, we recommend the use of an adhesive system in between delignified wood layers in order to decrease the risk of delamination. For the given example, we choose starch, as it is a well-known bio-based glue for pulp and paper products, such as paper bags, and is water based. Future works will focus on the fabrication of thicker laminates to resolve current limitations in terms of drying and fiber flow deviations.

In general, vacuum processing of delignified wood has the potential for a easy and fast production of large-scale densified cellulose fiber composites. After addressing the material's durability issue by applying proper coatings, water-stable adhesive systems or chemical modification, possible industrial applications may include automotive components such as door panels, floors, and dashboards. Our material could replace metals or fiber reinforced composites in order to reduce weight for better fuel efficiency and to improve recyclability.

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DISCLOSURES:
The authors have nothing to disclose.

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Figure

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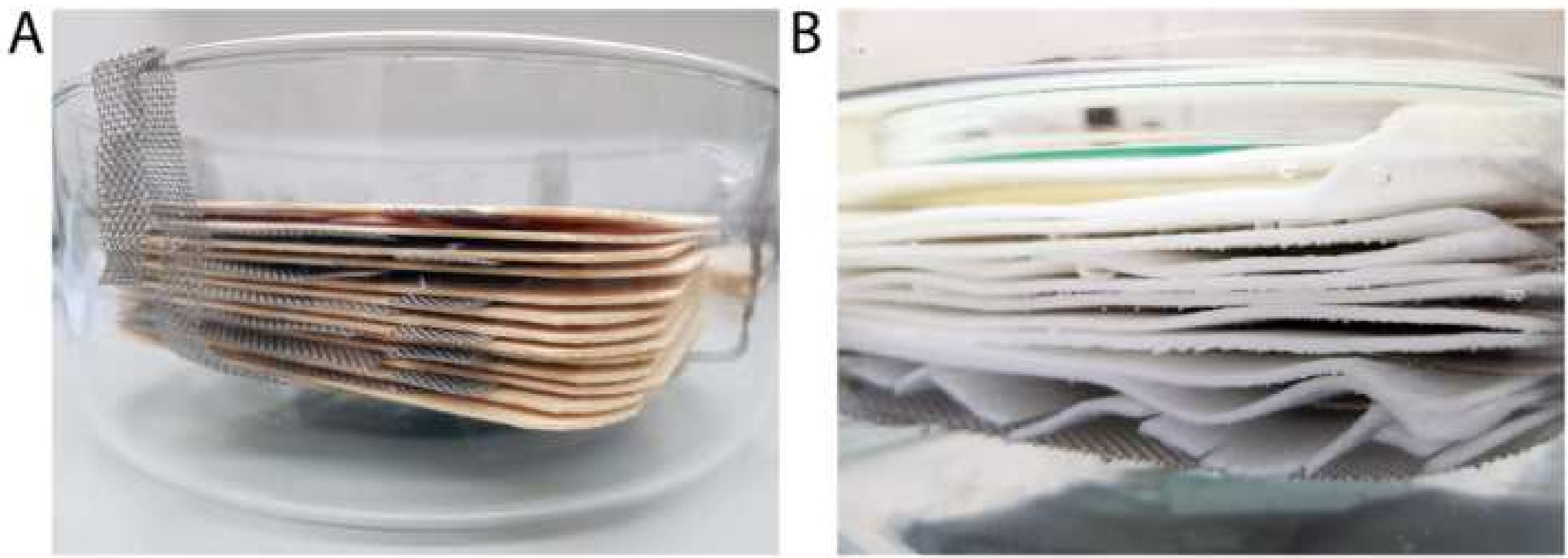

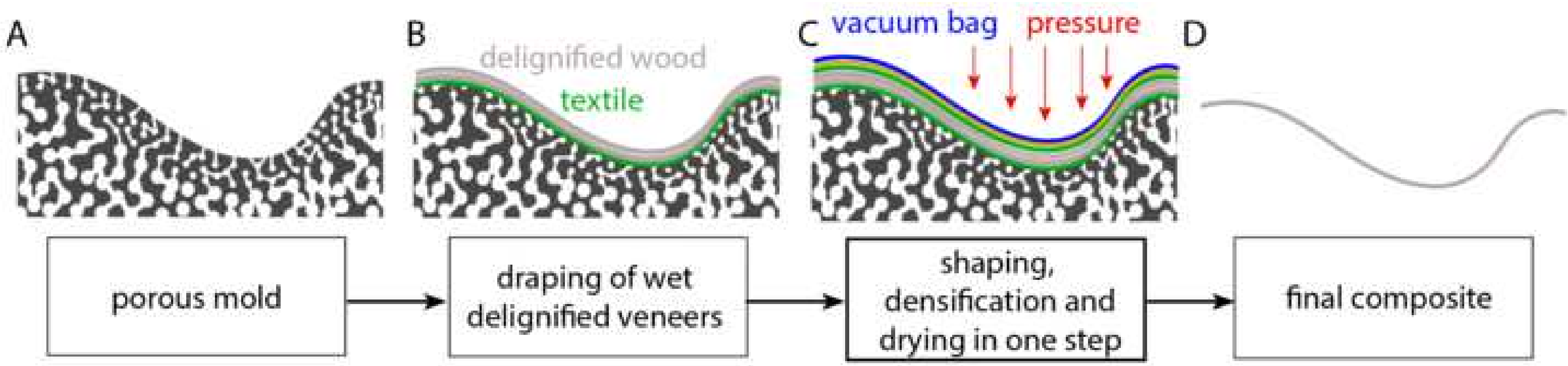
A
B
delignified wood
textile
C
vacuum bag
pressure
D
porous mold
draping of wet delignified veneers
shaping, densification and drying in one step
final composite

Figure

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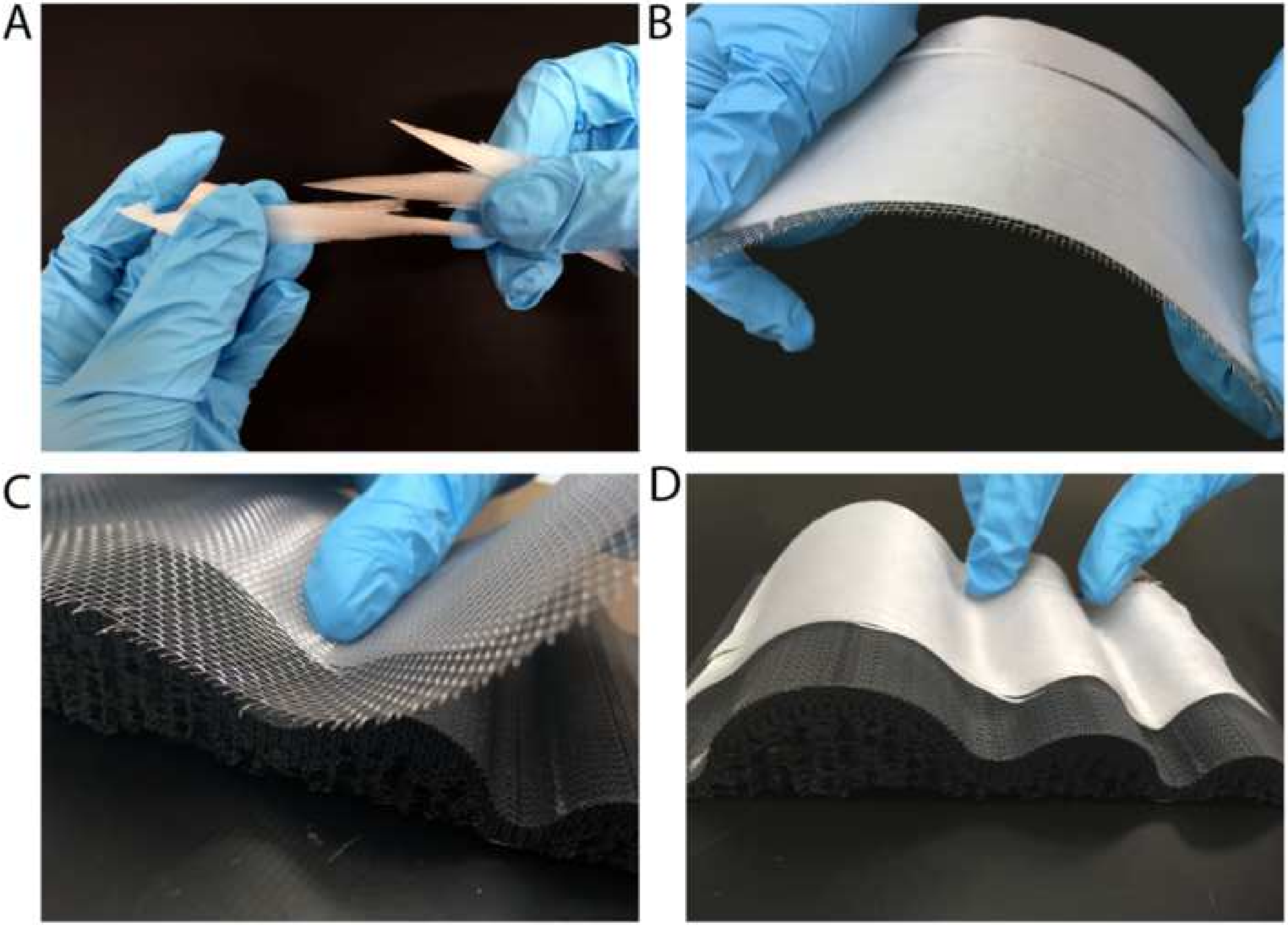

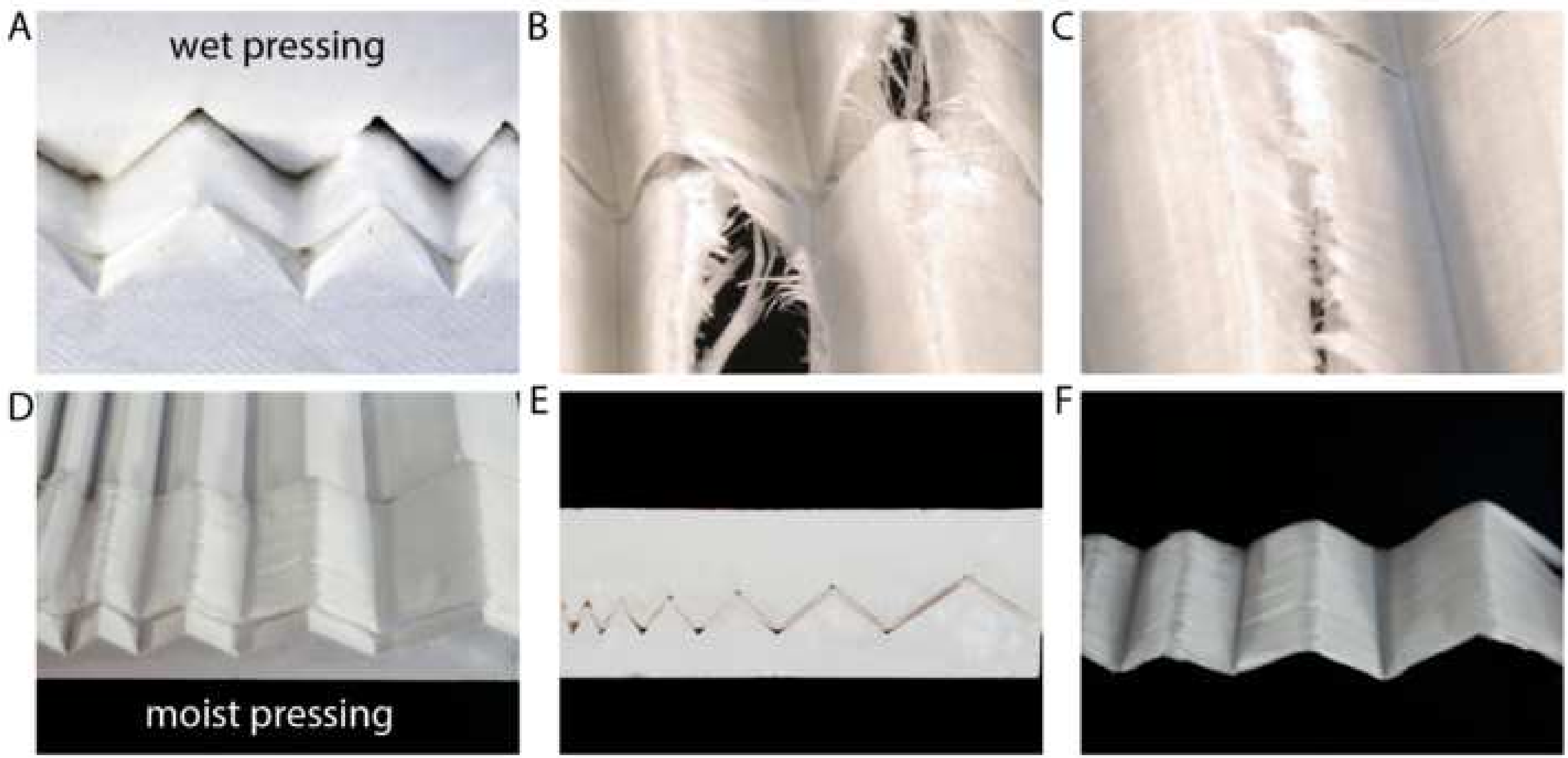
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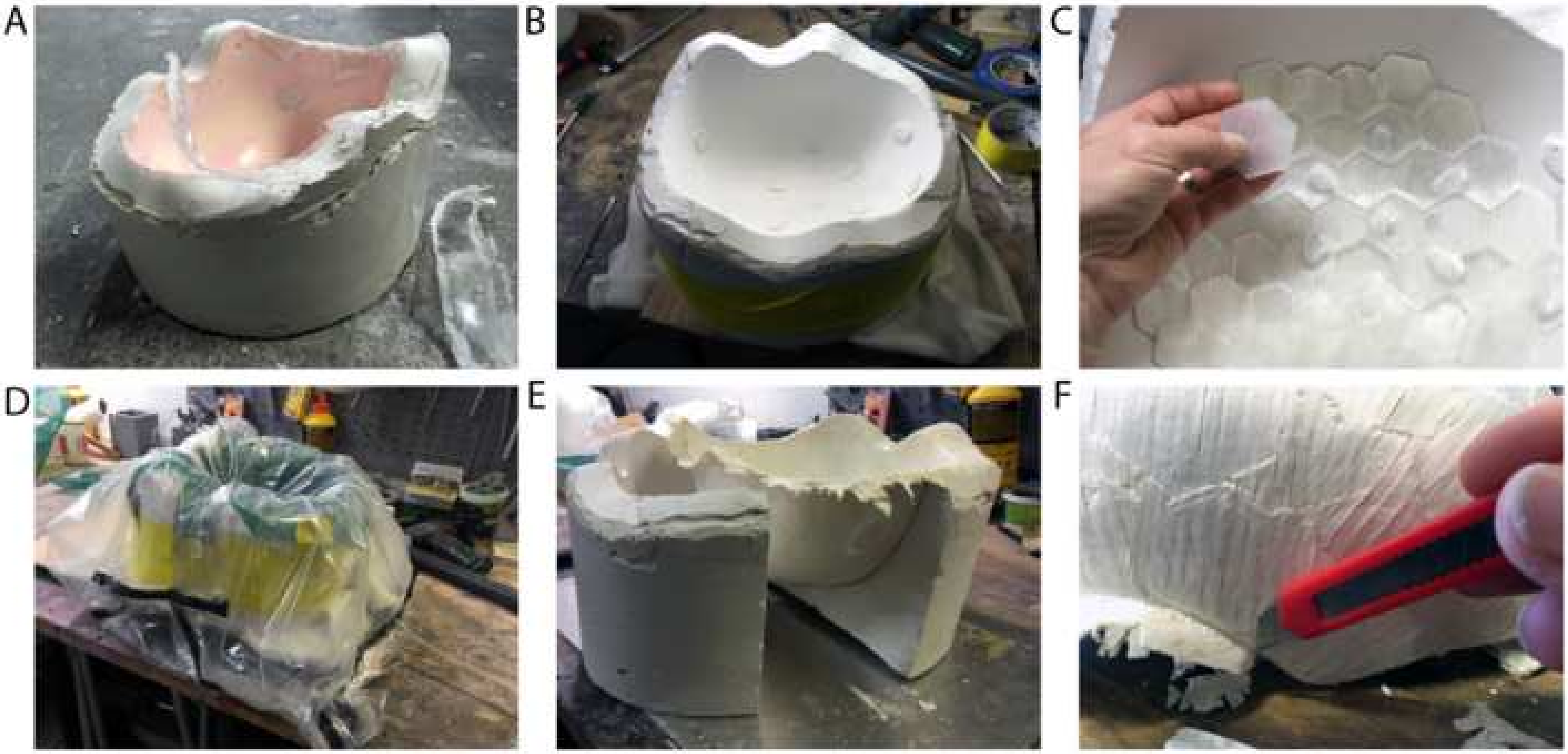
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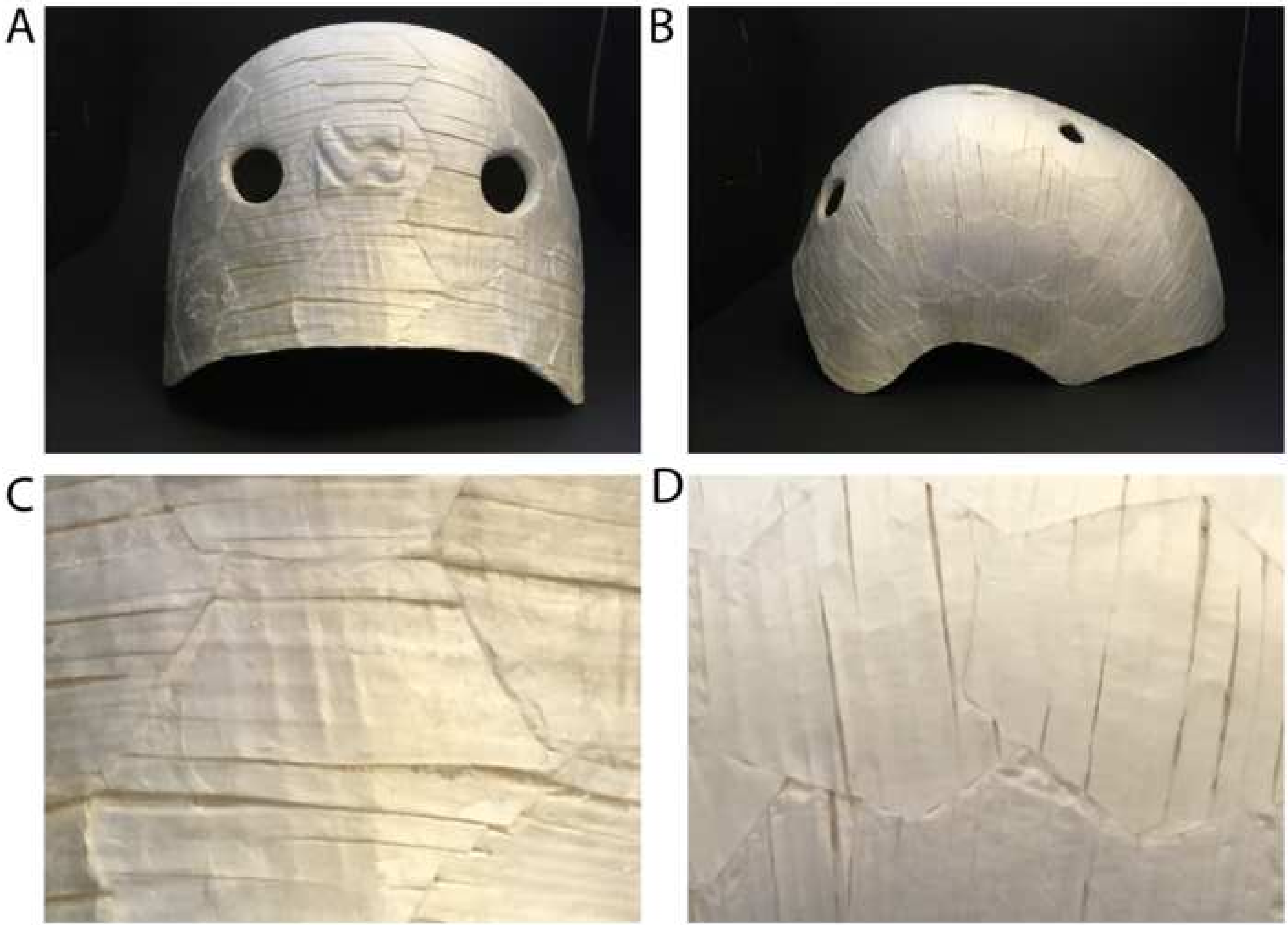

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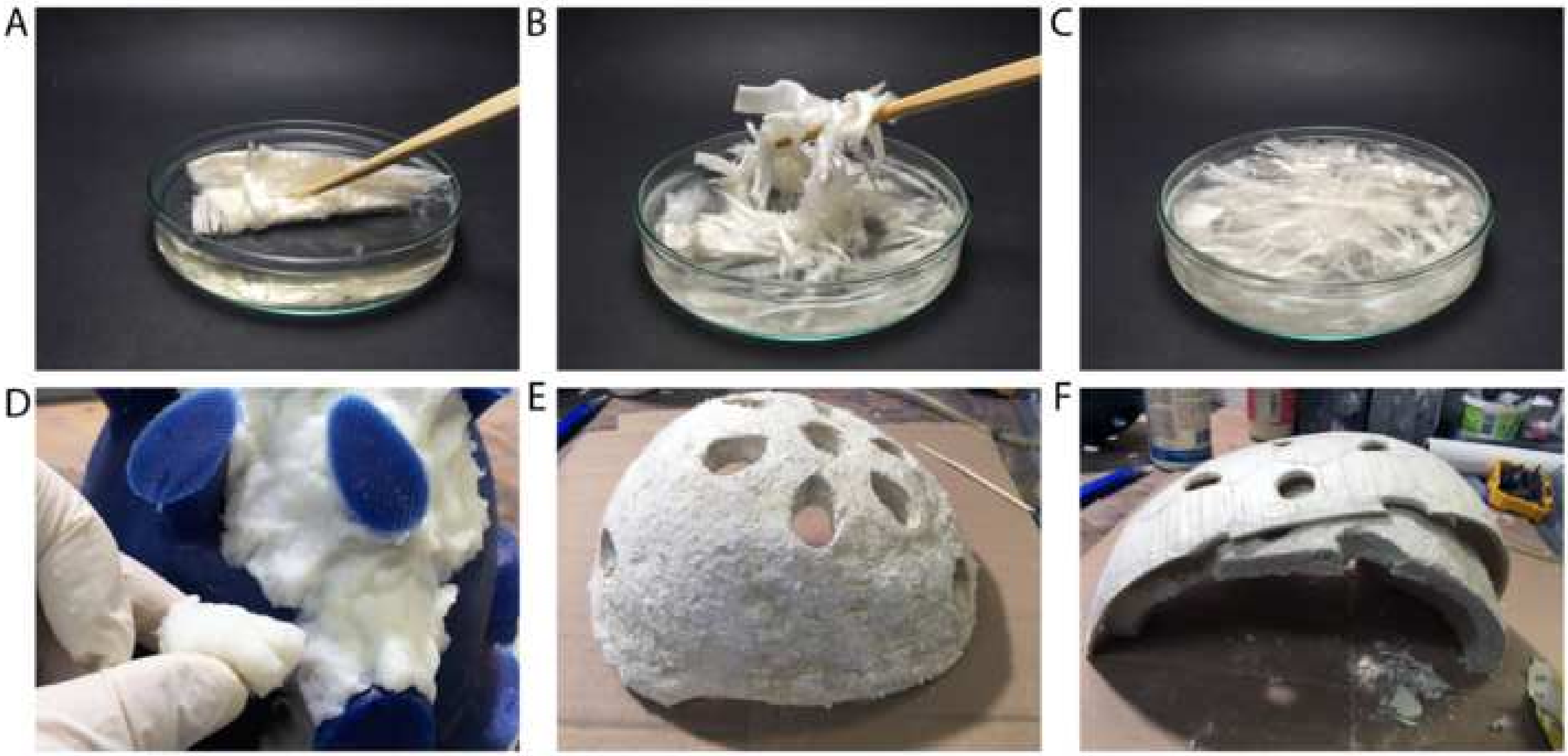

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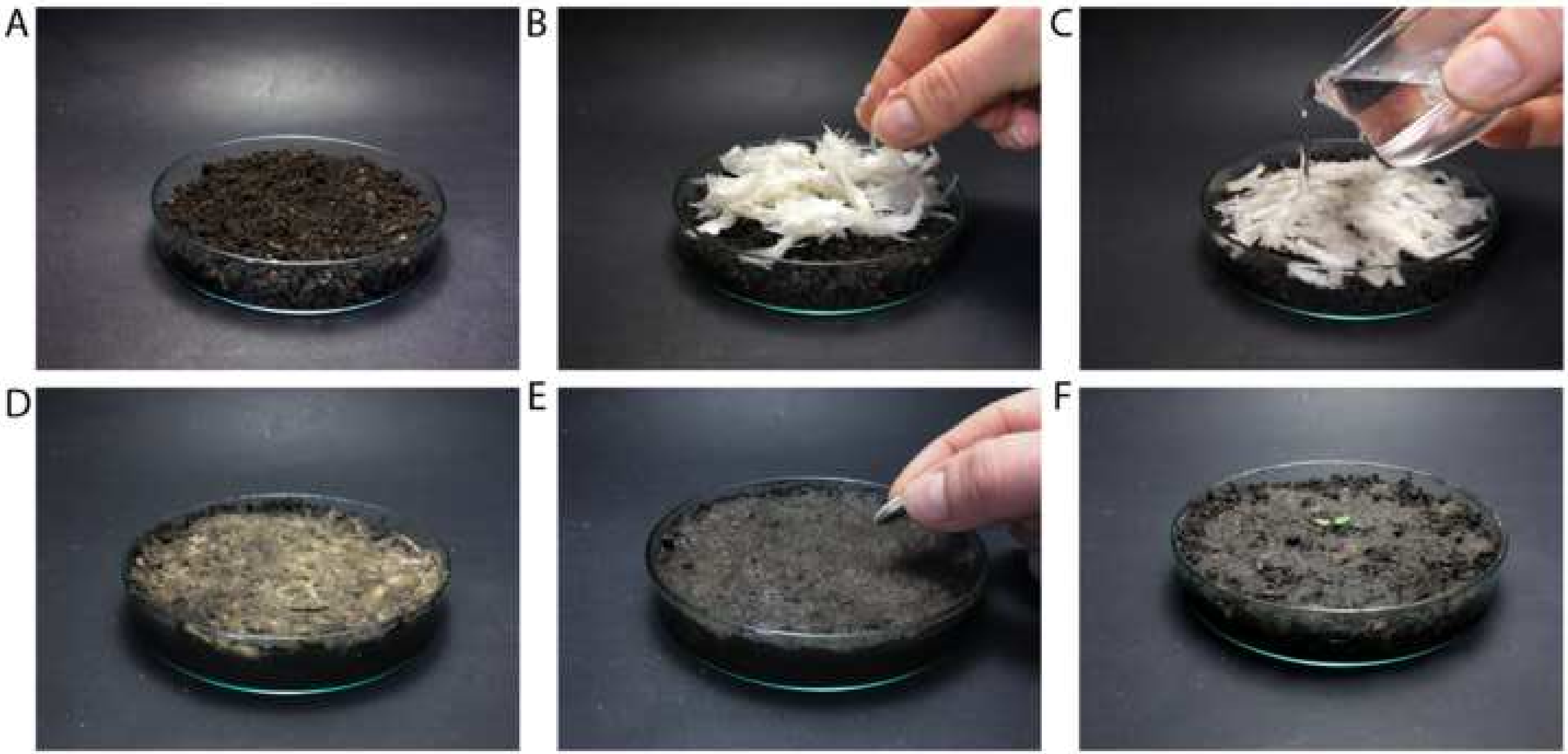

Table 1

FVC	20%	33%	50%	66%	85%
Density (g/cm^3)	0.3	0.5	0.72	1	1.3
Tensile elastic modulus (GPa)[7]	5	10	15	25	35
Tensile strength (MPa)[7]	60	90	120	180	250

Name of Material/Equipment	Company	Catalog Number	Comments/Description
Acetic acid	VWR Chemicals	20104.312	
Breather	Suter Kunststoffe AG	923.015	
Flow mesh/bleeder	Suter Kunststoffe AG	180.007	
Gypsum	Suter Kunststoffe AG	115.3002	
Hydrogen peroxide, 30%	VWR Chemicals	23622.298	
Oven	Binder GmbH		
Press	Imex Technik AG		
Seal tape	Suter Kunststoffe AG	31344	
Stainless steel mesh	Drawag AG		
Starch	Agrana Beteilungs AG		
Textile, peel ply	Suter Kunststoffe AG	222.001	
Vacuum bag	Suter Kunststoffe AG	215.15	
Vacuum bag, elastic	Suter Kunststoffe AG	390.1761	elastic vacuum bag for complex shapes
Vacuum pump	Vacuumbrand		
Vacuum tubing	Suter Kunststoffe AG	77008.001	
Wood veneers	Bollinger AG		

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Title of Article: Fabrication and design of wood based recyclable high-performance composites

Author(s): Marion Frey, Meri Zirkelbach, Clemens Dransfeld, Eric Faude, Etienne Trachsel, Mikael Hannus, Ingo Burgert, Tobias Keplinger

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Signature: [signature] Date: 27.5.2019

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Dear Dr. Steindel

We would like to thank the reviewers for their feedback on our manuscript JoVE60327 “Fabrication and design of wood based high-performance composites”. Please find below our replies to the reviewers’ comments. Changes in the text of the manuscript are highlighted.

Best regards,

Tobias Keplinger & Marion Frey
On behalf of all authors

Response to Reviewer:

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For example: VWR Chemicals

Protocol:
1. For each protocol step, please ensure you answer the “how” question, i.e., how is the step performed? Alternatively, add references to published material specifying how to perform the protocol action. If revisions cause a step to have more than 2-3 actions and 4 sentences per step, please split into separate steps or substeps.

Specific Protocol steps:
1. 1.4: Figure 2c does not exist.
changed accordingly
Figures:
1. Please cite Figure 1 within the manuscript text.

changed accordingly
2. Please include a title and a description of each figure and/or table. All figures and/or tables showing data must include measurement definitions, scale bars, and error bars (if applicable). Please include all the Figure Legends together at the end of the Representative Results in the manuscript text.

References:
1. Please ensure references have a consistent format.

Table of Materials:
1. Please ensure the Table of Materials has information on all materials and equipment used, especially those mentioned in the Protocol.

Reviewers' comments:

Reviewer #1:

Major Concerns:

What species of wood was used? Hardwood, softwood? Tropical? Important as some woods have a high content of inorganics and extractives.

The used wood species is mentioned in line 100-101: "*Here we use radial cut spruce veneers with a thickness of 1.5 mm*"

The authors state, "Processing methods for pulp and slurry based products cannot be used for fabrication of delignified densified wood composites, as they do not preserve the beneficial wood fiber alignment that is crucial to obtain materials with excellent mechanical properties." What does this mean? If whole wood fibers are densified, fiber alignment is lost. If delignified wood fibers are densified, fiber alignment is lost. This statement makes no scientific sense.

The retained fiber alignment in the densified cellulose composites is key for the excellent mechanical properties. This alignment is in contrast to conventional pulp and slurry based products. In our process we do not utilize separated wood fibers – we densify structurally intact cellulose scaffolds. Therefore the fiber alignment can be kept.

We adapted this paragraph to provide more clarity.

"Direct densification of wet delignified wood samples leads to limited densification degree, cracks and fibre alignment distortions due to the wet sample inherent free water that creates a counter pressure during densification." What does this mean? This statement makes no scientific sense.

Delignification of wood results in the removal of lignin between wood fibers and in the cell wall. After delignification and in wet state, delignified wood fibers are therefore not in contact with neighboring fibers anymore, as "free water" fills the empty space between fibers. This free water is beneficial because it enables deformation of the cellulose scaffold in water-saturated state, but when it comes to densification, the free water creates a counter pressure and fibers start sliding past along each other which results in fiber flow deviations.

The authors need to read research done in Japan on wood densification as well as many other references not listed. In this regard, the literature is poorly reviewed and missing a lot of research done in wood densification over the years. The earliest reference in the manuscript is 1998 and densification of wood has been done for over 80 years.

We are aware of the numerous works dealing with wood densification. However, this publication focuses on the densification and processing of cellulose scaffolds.
To account for the reviewers suggestions, however, we have added further relevant literature to the manuscript.

Delignification of wood veneers: The authors claim that they have remove "all" of the lignin but they did not present any analytical evidence that is was accomplished. They go on to state, "Complete delignification leads to a mass reduction of around 40 % and a volume reduction of around 20 % after drying at 65% RH.6. Besides lignin, part of hemicelluloses get removed too." What was the lignin, hemicellulose and cellulose content before and after the delignification process?

In the protocol section, we cite Frey et al 2018 and Segmehl et al 2018 – in this publications, the respective data can be found.

Storage and "cellulose prepreg" production: What about the hemicellulose content. Mold and other microorganism attack would be a big issue or the fragile veneers would have to be used immediately.

Based on our experience it is possible to store delignified wood samples in wet state for up to three weeks. For long-term storage, immersion in ethanol is recommended. These points are mentioned in the manuscript in lines 128-129.

"Densification of wet delignified wood (Figure 5A-C) is demanding, as free water in the scaffold creates a counter pressure upon densification and allows the material to flow during processing. This causes fiber deviations and cracks in the final material (Figure 5B,C)." But, this seems to be the main reason to do this process and now the authors are saying, it has problems?
"One possibility to bypass these limitations is to use pre-conditioned (95 % RH and 20 ˚C), delignified wood. In this condition, delignified wood is still reasonably shapeable and its densification does not lead to fiber alignment distortions and defects. Pre-conditioned material, however, is more rigid compared to the wet state, which makes it difficult to obtain small curvature radii without material damage. For small curvature radii, wet draping followed by conditioning in an already shaped state prior densification can be used. But, conditioning is rather time consuming and therefore not recommended for large scale applications." Now I am not sure which process is recommended for what applications? Very confusing!

We realize that the used terminology may have led to this confusion. We have two wet states of the samples (water-saturated and pre-conditioned at 95% RH at 20 ˚C), which we make more clear in the revised manuscript. The two different main processes relate to these wet states.

1) densification in closed molds by simple pressing: when using a closed mold process the delignified samples must be pe-conditioned at 95 % RH and 20 ˚C to retain the sample integrity. A pre-conditioning of the delignified wood however results in a more rigid material compared to the water-saturated, which makes it difficult to obtain small curvature radii without material damage.

2) vacuum shaping: in this setup water-saturated samples can be used, which makes it possible to obtain very small radii of curvature. Wet processing is possible by the vacuum approach because drying and densification is conducted simultaneously.

The big issue with this process is the moisture sensitivity of the product. The authors did not look at moisture sorption at any relative humidity, liquid water sorption or swelling of the product. This composite could only be used where there was no moisture present in the application.

Yes, this is true. Therefore we address this issue in lines 296-303: "*On the one hand, the hydrophilicity of the material is a disadvantage, as it leads to reduced mechanical* *performance when in contact with water. A simple method to protect the composite from liquid water comprises hydrophobic coatings as we have shown in Frey et al. 2019*[7]*. On the other hand, a hydrophilic behavior of the material can also be beneficial when it comes to end-of life use and recycling aspects*."

We've added a sentence in the conclusion, which emphasizes, that proper coatings/modifications need to be applied before using the composite under moist conditions.

"Densification down to 1/4th of the initial thickness resulted in a final thickness of 2.5 mm of the 8-layer thick composite part." No properties of this composite are given….MOE, MOR, indentation, tensile, stiffness, elongation, etc.

We have now included Table 1 for an overview on density, tensile elastic modulus and tensile strength for varying FVCs. The results are based on our previous work on densification of delignified wood veneers by densification in a press. A FVC of approximately 66% is obtained in the vacuum processing and therefore marked in the table in blue.

"For both processing methods, we recommend the use of an adhesive system in between delignified wood layers in order to decrease the risk of delamination." What type of adhesive? This is critical for recycling and environmental issues. Adhesives are expensive.
"For the given example, we choose starch, as it is a well-known bio-based glue for pulp and paper products such as paper bags and is water based." The addition of starch makes this composite even more moisture and mold sensitive.

We agree, that starch as hydrophilic adhesive does not protect the composite from moisture ingression. Coatings or hydrophilization are therefore needed as mentioned above. However, starch is very appealing as it is a bio-based glue well-known in pulp and paper products.

"In general, vacuum processing of delignified wood has the potential for an eased and fast production of large-scale densified cellulose fiber composites. Possible industrial applications include automotive components such as door panels, floor, and dashboards." Moisture sorption would result in a loss of properties i.e. strength, stiffness, hardness, mold, etc.

We have now included the importance to apply a proper coating/modification in the conclusion.

"Our material could replace metals or fiber reinforced composites in order to reduce weight for better fuel efficiency and to improve recyclability." It is easy to make this type of statement without any data to support it. This process works on a small laboratory scale but it is difficult to see how this would be used in an industrial process.

Table 1 reveals the low density (1 g/cm^3) of our densified delignified wood and corresponding mechanical properties. A specific elastic modulus of 25 $10^6 m^2 s^{-2}$ is comparable to steel (25 $10^6 m^2 s^{-2}$) or glass fiber reinforced polymers (18 $10^6 m^2 s^{-2}$) and therefore clearly justifies our statement. In addition, our vacuum processing is inspired by open-mold technologies already widely used in the fiber reinforced composites production on an industrial scale.

Reviewer #2:
Manuscript Summary:
It is an interesting work by using the vacuum processing for the delignified wood to make

artwork. The writing skills and logic is very good.

Minor Concerns:
1、The title of "Fabrication and design of wood based recyclable high-performance composites" is too big， please simplified it by specifical technology used in the paper

We removed “recyclable” from the title to shorten it.

2、Please add some mechanical properties of wood composite by Table or curves， such as density， MOE,MOR,shearing strength， et al.

We have now included Table 1 for an overview on density, tensile elastic modulus and tensile strength for varying FVCs. The results are based on our previous work on densification of delignified wood veneers simple densification in a press. A FVC of approximately 66% is obtained in the vacuum processing and therefore marked in the table in blue.

Reviewer #3:
Manuscript Summary:
The manuscript presented two fabrication processes, closed mold densification and vacuum densification, to produce high-performance cellulose composites based on delignified wood. Meanwhile, the authors suggested strategies how the composites can be re-used or decomposed at the end-of-life cycle. It is an interesting research for scientific community. However, the manuscript should make minor revision before acceptance.

Minor Concerns:
1. Line 148-151. "Densify the material in the closed mold either by using screw clamps …" I recommended authors provide the parameter of pressure.

A rather low pressure in the range of a few MPa is already sufficient to densify the wet material. Therefore we control the final thickness of the composite part by placing spacers between the mold surfaces. We’ve included this in a note in 3.3.

2. Line 176-177. "Apply vacuum for drying and simultaneous densification of the composite…" The authors should provide the time of vacuum.

The time strongly depends on the thickness and size of the sample. We added a note in 5.1. to discuss the influence of thickness on drying/densification time.

3. In section Protocol 5. Manufacturing of laminated composite parts. I recommended authors provide the number of layers and the amount of adhesive.

We’ve now included the amount of adhesive in a note 5.2. We are on purpose not defining a specific number of layers because this can be adjusted depending on the targeted thickness of the composite part.

4. Regarding delignification process of cellulose composites based on delignified wood please refer to 10.1021/acsnano.8b06409.

The mentioned reference of the reviewer reports the fabrication of a composite based on the densification of lignocellulosic pulp and brushite. As this report is conceptionally very different to our approach, we think it does not fit here.

Reviewer #4:
Manuscript Summary:
This manuscript describes a method for using partially delignified wood veneers as oriented, bio-based, semi-finished materials (e.g., reinforcement/preforms) using techniques common to fiber reinforced thermoset fabrication but little used in this context. The authors appropriately state that, while such techniques run counter to the currently popular bottom-up fabrication techniques, they efficiently use the inherent favorable structural attributes of natural materials (e.g., high degree of orientation). Given the movement towards bio-based materials, the protocol is a noteworthy contribution. The manuscript is generally well written. However, there is still some editing remaining, which I have refrained from doing as it lies outside of this portion of the review process.

Major Concerns:
None

Minor Concerns:
Several issues that should be remedied are listed below.
*Line 117 refers to Figure 2C, which does not exist.

Changed accordingly.

*Line 128: The first time that the abbreviation for ethanol (EtOH) is used, it needs to be defined. Similarly "UD" in line 243.

Done

*Do the authors wish to discuss Figures 6e,f - presumably about demolding and trimming? Nothing is mentioned in the text on them.

This is true, thank you for this comment. We now added a description of Figures 6e,f in the text.

*Lines 309-313: The possible applications listed (e.g., door panels, floors, dashboards, metal/fiber reinforced composite replacements) cannot be justified without additional information - presumably further processing with other materials (e.g., thermosets), which is not mentioned. As fabricated, these materials are not strong enough or durable enough, for example, to be used in these applications. I don't believe that the authors intend that they are but, as written, it appears to suggest that this is the case. The authors should at least suggest generally what further steps might be necessary to use materials made from their protocols in the applications mentioned.

We now added the need for addressing durability with proper coatings, gluing systems or chemical modification before use in the mentioned applications.

*Reference 6 should be updated.

Thank you for spotting this error. We have changed it accordingly.