CELLULOSIC BIOCOMPOSITES FOR SUSTAINABLE MANUFACTURING

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Overview

The aim of this research work is to create a general-purpose manufacturing technology considering foremost its sustainability characteristics. Our approach is bio-inspired in that we attempt to replicate some of the principles of natural synthesis, characterised by exclusive use of locally available resources, utilising material ingredients which are often insignificant by themselves, performing assembly of complex hierarchical structures in low-energy environments and producing artefacts integrally embedded within their ecological cycles.

We implement those principles with the development of a new family of biocomposites combining cellulose and chitin, the first and second most abundant biopolymers on earth, and assembling spatial artefacts by selective deposition of materials through additive manufacturing at room temperature. The technology developed can produce large-scale objects at low-cost that are fully bio-sourced and biodegradable. Applications investigated within the building industry range from: interiors such as furniture and fittings; to construction such as insulated panels, pre-finished building members and recyclable moulds; even to restoration of timber elements and ornamentations in heritage buildings.

Objectives

Biological polymers produced by plants and animals are in the realm of billions of tonnes annually. A key feature of biological materials is in their innate embedding within ecological cycles. This offers an untapped opportunity for a fundamentally sustainable approach to manufacturing, if we manage to control their synthesis and assembly.

The constraints guiding the design of our bio-material process include: (a) use of ingredients ubiquitous in every natural ecosystem aiming to promote regional as opposed to transcontinental production and transport; (b) ensuring available material component sourcing at low-cost from renewable resources and even industrial by-products or waste recovery, aiming to enable scalability for general manufacturing that competes with commodity plastics and potentially supports circular models of production and consumption; (c) low-energy fabrication process at room temperature without thermoforming or high-pressure processes to achieve a small environmental footprint in production; and (d) ecologically embedded material synthesis avoiding material transformation by chemical modification, as for each intervention an additional reversing step is required and one away from natural recovery.

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Materials

Cellulose is the most abundant biological material on earth (Reiterer, 1999). It is the main component of timber and plant matter in general. Chitin is also a highly abundant bio-material encountered mainly in the animal kingdom, in the exoskeleton of arthropods such as shrimps, crabs and lobsters as well as in insects including bees, grasshoppers and maggots. Despite their molecular similarity and being some of the most common bio-polymers on earth, cellulose and chitin rarely appear in the same organism.

Composition of these two ingredients gives rise to a family of lightweight biocomposites named fungal-like adhesive materials (FLAM), after oomycotes or egg-shaped fungi, a species of eukaryotic organisms, historically misclassified as fungi, whose cell walls contain cellulose and chitin. Experiments with different ratios of the two components were performed to determine the best mechanical but also rheologic parameters suitable for manufacture. One may consider cellulose as the fibrous reinforcement within a chitinous matrix in a conventional composite materials sense. Therefore, too high cellulose to chitin ratio produces viscuous materials which are difficult to extrude and dry to become brittle, while too high chitin produces materials that slump too much and shrink uncontrollably. Surprisingly, the same ratio as in the cell-walls of oomycota (1:8) produced both strong, extrudable and controllable composites. A unique aspect of FLAM is their ability to adhere to themselves even after drying which allows for infinitely restorable objects. Before drying, they can also be rehydrated and reused. To protect them from water after curing, they require coating like natural timbers.

FLAMs can be produced from either industrial grade, pure cellulose, used as fillers or wood fibre waste from timber manufacturing. The properties of FLAM (370 kg/m3 density | 0.26GPa Young's modulus) are in the range of high-density synthetic foams, such as machinable polyurethanes, and low-density natural timbers such as balsa and cedar. Using pure cellulose, the material appears as compressed paper while using wood fibre, its appearance is similar to particle boards. However, unlike common cellulosic materials and composites such as cellophane, plywood, particle boards and wood-plastics, FLAMs involve no petrochemical adhesives or toxic chemical substances such as strong solvents. In addition, unlike conventional 3D printing materials such as wood particulate plastics and bio-plastics such as PLA, it is 100% biodegradable without requiring specialised composting or recovery processes. In the eyes of nature, FLAMs are no different from mushrooms or timbers. Additional information pertaining the materials science of FLAMs, including extensive study of material characterisations, was first presented by Sanandiya et al. (2018), and precursor research on chitinous biocomposites – on which this project builds – by Fernandez (2009) and Fernandez and Ingber (2014).

Manufacturing

Fabricating with organic composites is vastly different from working with inorganic or inert materials such as plastics or concrete. Foremost it requires understanding and addressing issues which spring from the innate variability of its constituent components. Just as no two pieces of wood are ever the same, inevitable variation of its cellulosic content produces material property changes, such as viscosity for instance, which affect both assembly as well as curing. Moreover, the composite in its wet state is an adhesive which makes it very difficult to handle as it is highly tacky and shear thinning. Therefore, co-evolution of the material formulation alongside its manufacturing method was integral.

The digital fabrication system is comprised of an industrial robot equipped with a volumetric auger dispenser for sealants and adhesives, a bulk material supply (15 litre batches) and a programmable control logic controller for flow rate modulation. Details on the setup are presented by Dritsas et al. (2018). Early implementations follow the canonical material extrusion AM principles, where objects are built by filament layers arranged vertically, not unlike Fused Deposition Modelling of rapid prototyping or Direct Ink Writing of tissue engineering. The material is dispensed as a viscous paste and fuses with consecutive layers.

3D printing FLAM is a low-energy process compared with FDM and SLS as it does not require thermal input. Filaments are directly printed at room temperature and by evaporation at ambient conditions water is removed from the colloid as objects transform to rigid solids. Working with a broad range of nozzle diameters (1 to 7mm) motivated departing from conventional three-axis printing to leverage production time and resolution. We investigated fusion of fabrication paradigms, namely five-axis dispensing to coat over scaffolds, subtractive techniques to introduce features below the nozzle diameter, and forming operations displacing material while in a wet-state with net-zero material change.

Space-filling algorithms were developed to maximise surface to volume ratios for accelerating evaporative hardening. Production of large objects in a single printing session gave its place to understanding how to segment for improved throughput: as the process is not bottlenecked by thermo-dispensing such as in large diameter FDM, printing can be much faster, for example 50mm/sec at 7mm diameter, while curing takes place separately. It became evident that it was thus more meaningful to consider scaling in terms of time, within a heterogenous sequence of production steps, instead of targeting physical scale using additive manufacturing exclusively.

1. Natural composite pillar overall photography and details. Photo: Frank Pinckers.

2. Fungus-like adhesive material with cellulosic waste from timber manufacturing at wet-state.

3. Detail of large-diameter nozzle extrusion in canonical 3-axis mode.

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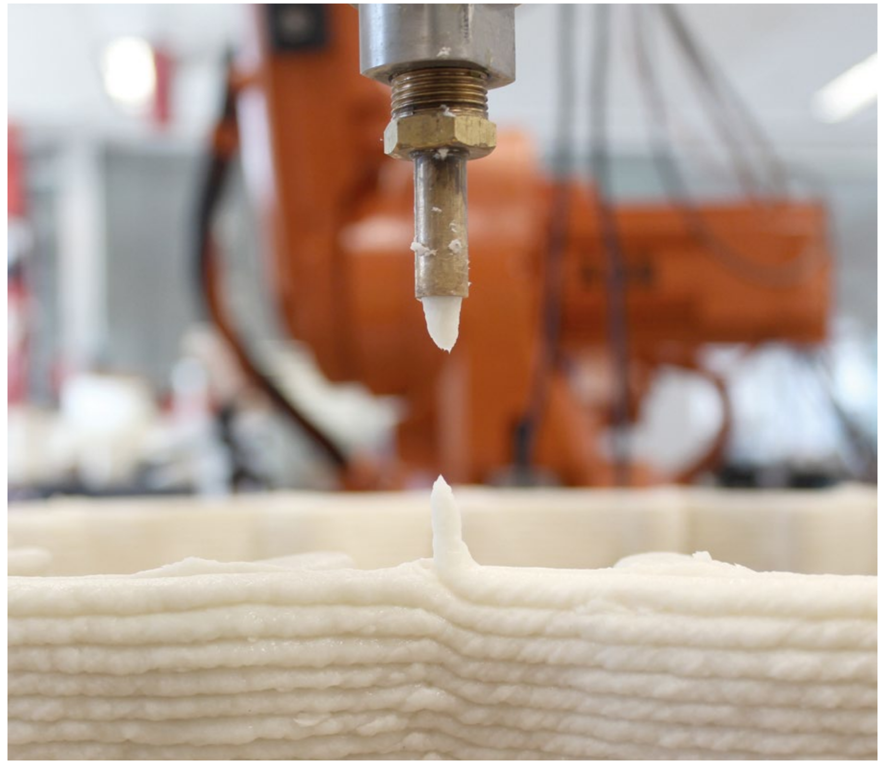

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Challenges

FLAMs are suspensions of fibres (chitin to cellulose 1:8) in an organic matrix comprised of chitosan and water (3%). The uncured composite's properties are in the class of non-Newtonian viscoelastic materials. The material behaviour, both during extrusion and while curing, is highly non-linear as properties including density, viscosity and elasticity depend on both the shear force applied during extrusion as well as time. Challenges pertaining to fabrication with shrinkage anisotropy being the highest (5% along the extrusion direction, 12% in the transverse and 32% vertically), are presented in detail by Dritsas et al. (2019).

To overcome these issues, instead of a mechanistic, such as transient multi-physics computational fluid dynamics, we employed experimental modelling methods leveraging the ease of 3D printing to collect data via optical metrology and 3D scanning, for statistical analysis. This approach allowed us to absorb the variability of controllable but also the uncontrollable design parameters related to material and environmental conditions and to bypass the highly involved development of an analytical CFD model which would incorporate material state transition, moisture convection-diffusion, and transient forces affecting the 3D printed geometry.

Control of the extrusion process parameters such as the robot's feed rate, material flow rate and layer height, presented by Vijay et al. (2019), were modelled through a face-centred Central Composite Design of Experiment (Montgomery, 2009) to associate the resulting filament dimensions and their mechanical properties. The model is expressed as a set of multiple quadratic equations that capture not only the main parameters but also all the possible combinations of their interactions. To derive desirable operating points from the response surface models, we employed multi-response optimisation methodology (Derringer and Suich, 1980).

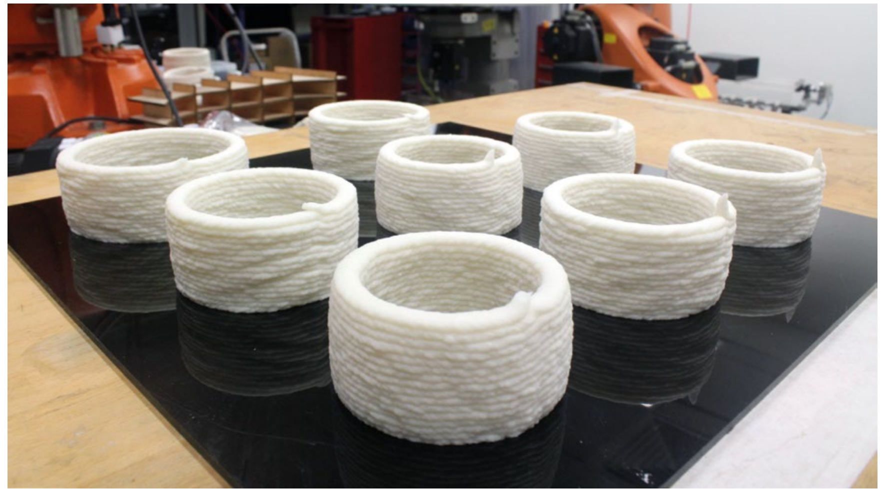
Interestingly, the study shows that we can retain constant filament dimensions whilst controlling their tensile strength dynamically via motion-flow rate modulation. This implies that the material process has innate functionally-graded characteristic potential, beyond conventional geometric density modulation by 3D printing, such as variably-sized spatial lattices. Non-linear regression models using machine-learning techniques were employed to relate notional-to-resultant geometries and enable prediction-correction of object deformation during curing. We call the process pre-set modelling which is equivalent to elongating structural members in building design in anticipation of progressive compression deformation during construction.

Evaluation

The necessity for statistical data to develop predictive models resulted in numerous early prototypes of simple measurable geometries. Single filaments were printed to assess filament profile uniformity against feed and flow rates; pairs of adjacent filaments determined the requisite overlapping fraction sufficient for adhesion; straight walls measured vertical compaction rate and intralayer fusion; square profiles determined shrinkage anisotropy; and surfaces were printed to collect curvature and deflection features.

The first large-scale prototype was an airfoil (NACA 0015) printed in two halves (50% infill, 1 hour printing time each), fused and coated with the same material and hand polished (1.2m length, 5kg weight). Its objective was to demonstrate the versatility of the material used for 3D printing, its self-adhesive properties past curing, and compatibility with conventional casting and wood-working techniques. While the material's surface characteristics are not suitable for airfoil applications, it may be an alternative to current core designs using natural materials such as balsa.

To demonstrate the free-form fabrication capability, material strength, assess the reliability of the extrusion system and understand the workflow, we developed an architectural-scale prototype pillar of 0.6-1.0m diameter and 5m height. The form was split into 50 vertical segments of 250mm height, taking 30-120min each to print. Segments are comprised of two adjacent filaments with wall thickness of 25mm. Alternative wall structures were tested such as incorporating buttress fins or an internal web pattern for increased stiffness but the double filament wall design was the most time efficient. The amount of time required for printing was 60 hours with total wet material weight c. 480kg and cured weight 105kg. The cost of materials was c. £220 | £2.1/kg.



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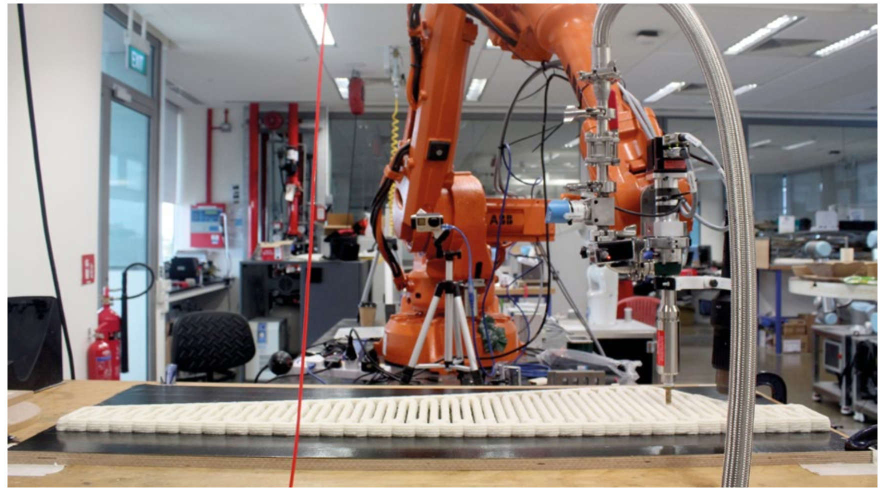

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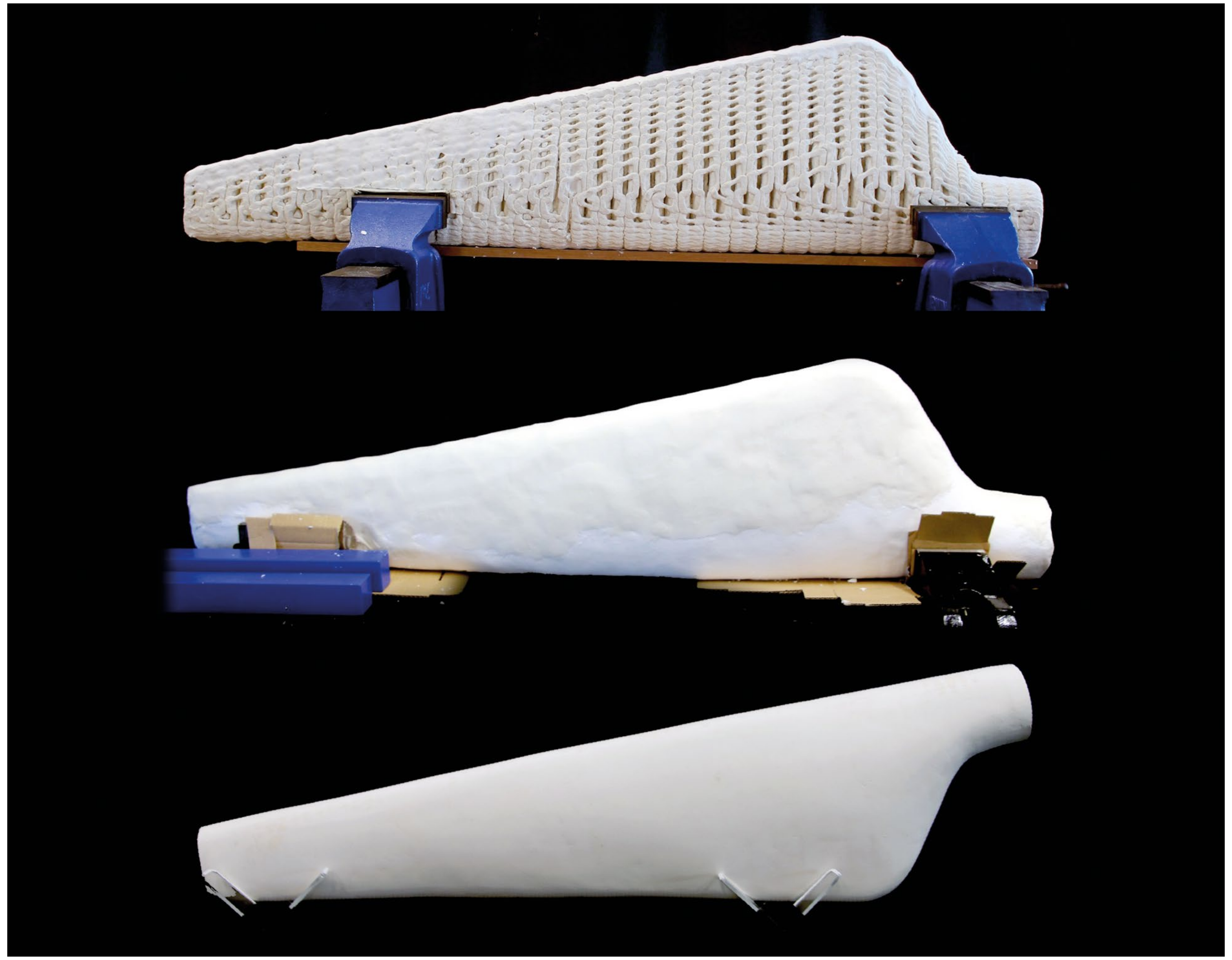

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4. Prediction correction non-linear regression modeling for shrinkage compensation.

5. Prediction correction non-linear regression modelling for shrinkage compensation.

6. Printing half of the airfoil prototype required approximately one hour.

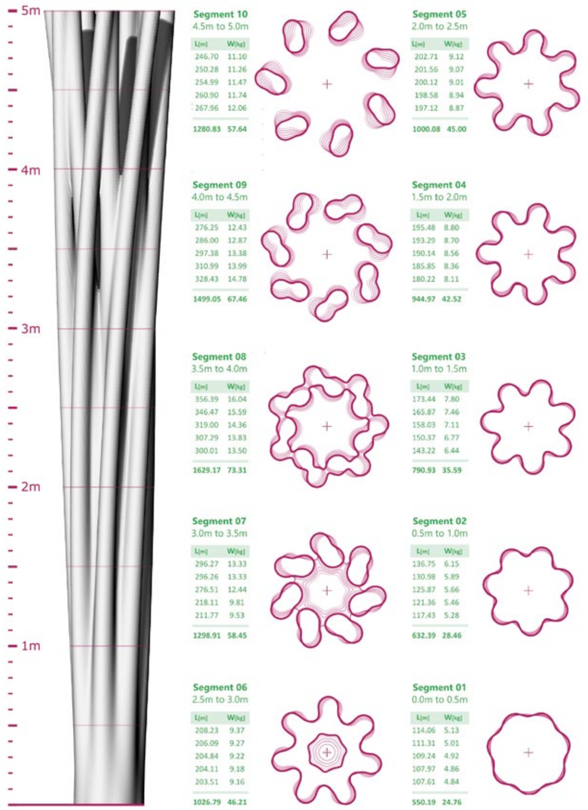
7. Two halves bonded with FLAM, coated and sanded using woodworking methods.

Apart from human-related operating errors that required recycling the material and reprinting two segments, the process was highly robust. The predictive-corrective models used to adjust notional geometry to account for shrinkage gave overall good results, but additional work is required to reduce error over the diameter of segments to be under 1% or 5mm. Nevertheless, if segments are assembled within the first 48 hours after 3D printing while the material is still moist, parts can be fused seamlessly. The artefact suggests direct applications for non-load bearing interior fittings as well as an approach for free-form structural element mould-making, perhaps even pre-finished as it can be easily sanded and/or coated.

Conclusions

Additive manufacturing became the dominant paradigm during the past few decades representing the future of industrial production (Thompson et al., 2016) and even building construction. Despite unmatched benefits such as rapid design-to-production, capability for free-form geometry, design customisation and efficient material use (Tofail et al., 2018), challenges in materiality, scalability, sustainability, affordability and reliability still persist (Royal Academy of Engineers, 2013). Sustainability in additive manufacturing (Baumers et al., 2011) is a domain that only recently came to the foreground (Gebler et al., 2014).

We presented a new technology addressing several of those challenges. Its significance is in an approach which departs from optimising resource uptake within current production workflows or developing eco-friendly materials that suit existing modes of production, such as injection moulding and 3D printing PLA, currently the most popular bioplastic sourced from agricultural food sources which nevertheless requires specialised composting recovery.



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Instead, the work is informed by biology and the life-sciences where materials are understood as being embedded within their production and consumption cycles. FLAMs are produced by widely available natural bio-materials: both cellulose and chitin are sourced from waste of the timber and fishing industries and most importantly they remain unmodified, meaning they are natural. Nevertheless, precisely because they were not designed with priority on ease of manufacture, they require development of a specialised approach for their fabrication and control.

Digital methods of modelling and fabrication offer a level of precision that was impossible to achieve in the past. Additional research work is currently underway in both material characterisation including thermal, acoustical and fire properties, as well as process-control simulation and optimisation to enable adoption and use in construction and general-manufacturing. We believe the use of ubiquitous natural materials and digital fabrication may offer an environmentally-benign manufacturing and design paradigm towards a circular and sustainable society.

8. The design of the natural composite pillar is comprised of seven arcs fused using implicit surface distance field methodology producing a geometrically- and topologically-complex surface model.

9. Prototype of natural composite pillar reinforced with convetional rebar cage and cast with concrete.

10. Natural composite pillar. Photo: Frank Pinckers.

Acknowledgements

The research was supported by the SUTD-MIT International Design Centre (IDG21600101), the National Additive Manufacturing Innovation Cluster (NAMIC2016026), Digital Manufacturing and Design Centre (RGDM1620303).

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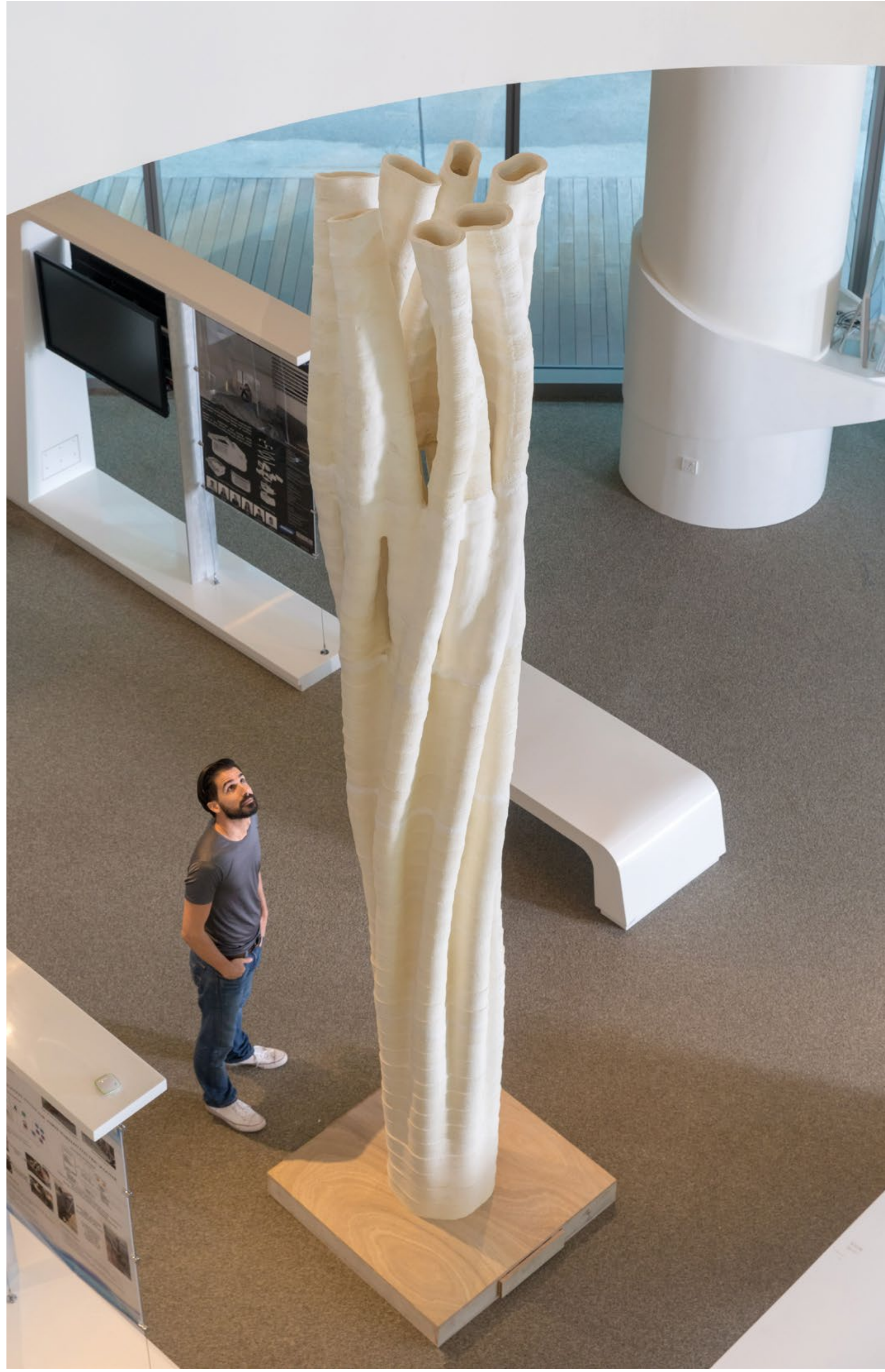

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