Waste-based biopolymer slurry for 3D printing targeting construction elements

Rech, Arianna; Chiujdea, Ruxandra; Colmo, Claudia; Rossi, Gabriella; Nicholas, Paul; Tamke, Martin; Thomsen, Mette Ramsgaard; Daugaard, Anders E.

Published in: Materials Today Communications

Link to article, DOI: 10.1016/j.mtcomm.2022.104963

Publication date: 2022

Document Version Publisher's PDF, also known as Version of record

Link back to DTU Orbit


General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.
Waste-based biopolymer slurry for 3D printing targeting construction elements

Arianna Rech a, Ruxandra Chiujdea b, Claudia Colmo b, Gabriella Rossi b, Paul Nicholas b, Martin Tanke b, Mette Ramsgaard Thomsen b, Anders E. Daugaard a, * 

Abstract

The construction industry consumes significant quantities of mineral-based materials, thereby causing pollution and raw material depletion. To meet projected demands for new buildings, while simultaneously reducing the use of non-renewable materials and fossil-based carbon, there is therefore a strong drive to develop alternative, renewable materials to replace those currently used in this sector. In the present study, we investigate the formulation of waste-based biopolymer slurries for 3D printing as a path towards more sustainable construction elements. The printing formulation was based on xanthan gum as a binder with lignocellulosic waste fibres and particles for reinforcement. At the same time, flow properties were controlled using a combination of bio-based plasticiser (glycerol) and water. First, the amounts of water, plasticiser and waste fillers were optimised to minimise shrinkage and deformation while obtaining the best possible mechanical properties after drying. Different reinforcing fillers were investigated, and it was proven that a minor addition of inorganic material (vermiculite) could significantly improve the material’s mechanical properties during printing and in its final form after drying. Furthermore, it was shown how using calcium ions could enhance the binding effect of the xanthan gum and how combining it with an increase in binder concentration could improve the early-age strength of the printed material. The optimised formulations were proven suitable for 3D printing. The addition of vermiculite improved weathering resistance and stability during printing (shrinkage and height), which was illustrated in a bench-scale printing test.

1. Introduction

The construction industry consumes 50% of the world’s raw materials, contributes to 39% of all global CO2 emissions, and produces 35% of total waste [1–5]. Due to its impact, the development of new building materials from renewable sources or waste materials, which are also biodegradable or recyclable, has enormous potential to contribute to a reduction in CO2 emissions and raw material consumption globally [3, 6–8].

Most research on sustainable architecture has focused on reducing the energy a building uses once it is built rather than on employing sustainable construction materials [6]. When they are indeed mentioned, wall and insulation materials are the main components [6]. In terms of sustainable wall materials, the literature has focused on cement reinforcement, substituting mineral admixtures with natural fibres (cellulose, hemp, etc.) or biopolymers, and the use of recycled waste from construction. Hence, there is a lack of studies on entirely or mainly bio-based alternatives [6, 9–13]. Alternatively, for sustainable insulation materials, many studies focus on entirely or mainly bio-based materials, where the use of natural and recycled fibres, agricultural waste composites, fungi composites, waste materials, etc. is investigated [6, 14–17]. In addition, emerging research highlights the need to address embedded carbon to shift from fossil to plant-based carbon, and to store carbon within buildings by expanding the use of bio-based materials in the construction industry [18–20].

Since 3D printing introduces efficiencies into the construction process, such as time and material savings, reductions in on-site complexity, easier customisation and personalisation, etc., this manufacturing...
approach is considered a state of the art method to address sustainability challenges [3,21].

3D printing in the construction sector currently focuses on concrete [22–27]; however, other materials are also evaluated, such as metals [22,23,28] and polymers [22,23,29,30]. Among the studies on construction 3D printing that can be found in literature, very few examined 3D printing with sustainable materials. Some studies can be found where sustainable fillers are added to a mineral-based matrix (cement-based composites with bio-based fillers) [3,31], others where mineral fillers are added to a sustainable binder (biopolymer-based composites with mineral aggregates) [32] and, lastly, others where only biopolymers are employed for both the matrix and fillers (fully bio-based composites) [29,33,34]. In 3D printing, in general, a few studies exist on biopolymer composites for large-scale 3D printing. These can be divided into wood powder-based composites, where the wood powder is bonded through either petroleum-based glues/polymers or biopolymers [35–38], or fungal-based composites, where fungi are used as bio-based binders and biomass is used as filler [4], or fungus-like materials using biopolymers as matrix and cellulose fibres as filler [39]. In relation to small-scale 3D printing, numerous studies have looked at the development of biopolymer composites targeting the development of more sustainable plastic materials or biomaterials, albeit these have not addressed printing large-scale elements.

There are currently very few studies on renewable materials for construction 3D printing. Most identified studies focus on hybrids, where only a fraction of the final material is renewable. Therefore, this study’s objective was to formulate a slurry-based 3D printable formulation made of bio-based or waste-based material for architectural-scale 3D printing. Specifically, the target was to develop a renewable formulation from readily available low-cost waste materials sourced from local industrial processes, biopolymers, and bio-based plasticizers, which would allow for fast printing at scale, in uncontrolled environmental conditions, and possess mechanical properties comparable to mid-strength natural materials.

2. Materials and methods

2.1. Materials

The biopolymer, xanthan gum, was purchased from buXtrade, Germany. The bio-based plasticizer, glycerol (C3H8O3, ≥ 99.5%), was obtained from Emmeleve A/S, Denmark. The first waste filler, cellulose insulation (Papiruld Iso-Let) derived from recycled paper, was purchased from Papiruld Danmark in Denmark and consisted of an homogeneous mixture of paper pieces and fibres from 1 mm to 2 cm, as shown in Fig. S1. The second waste filler, sawdust (wood flour fine, 0.5 mm) was purchased from Hedensted Gruppen in Denmark, and it was formed from elongated wood fragments varying from 2 mm to 0.5 mm in length, as displayed in Fig. S2. The reinforcing filler, vermiculite (granules medium), was purchased from Manutan, Denmark. The crosslinker, calcium chloride (CaCl2, anhydrous, granular, ≤ 7 mm, ≥ 93%), was obtained from Sigma Aldrich, Denmark. Deionized water was used as a solvent in all formulations.

2.2. Slurry preparation

First, xanthan gum, glycerol, and water were mixed using a high-speed mixer operated at 20,500 rpm (UltraTurrax, IKA, Germany) until a uniform pale yellow viscous liquid was produced. Subsequently, cellulose insulation and sawdust were added sequentially to the mixture and homogenised for 5 min by shifting around the high-speed mixer operated at 20,500 rpm. The result was a uniform grey slurry. If other fillers, such as vermiculite, were added to the slurry, the addition was done at the same time as the cellulose insulation and sawdust. Lastly, if anionic crosslinker was used in the formulation, an aqueous solution containing the crosslinker was added instead of pure deionized water during the initial phase of slurry preparation. The proportions of each ingredient and all the different experimented formulations are represented in Tables S1–S6 in the Supporting Information. When scale-up experiments were conducted, deionised water was substituted with tap water, and the mixing technique was adapted to the scale, using tools such as a hand blender, a dough mixer, or a cement mixer, allowing similar results to be obtained with longer mixing times.

2.3. Composite preparation

The composite samples were prepared by spreading the slurry in a rectangular mould (width 35.88 mm, height 109.83 mm, and thickness 2.82 mm) fixed on a PE sheet. After removing the mould, the rectangular-shaped samples were dried at room temperature in a ventilated fume hood for three days. The PE sheet was removed only once most of the water had evaporated. Samples were considered fully dry when a change in weight lower than 1 wt% was observed over two consecutive days.

2.4. Characterisation

2.4.1. Shrinkage

Shrinkage of the different formulations was measured based on the volumetric size variation from the moulded slurry to the dry composite.

The measurement was conducted in triplicates on the samples prepared, as described in Section 2.3.

2.4.2. Density

Density was calculated based on the volume and weight of a portion of the dry composite samples. The test was conducted in three replicates on circular samples that were cut from the moulded composite (Section 2.3) with a diameter of 12 mm and thickness of approximately 1.5 mm (depending on the shrinkage of the tested material).

2.4.3. Mechanical properties

Mechanical tests were performed on a tensile test instrument, namely 3300 Series Universal Testing Systems 3345 from Instron. The tests were conducted using a load cell of 500 N, pneumatic clamps with serrated jaw faces with a load capacity of up to 1 kN and by applying 5 bar of pressure. The tests were conducted at a strain rate of 5 mm/min at room temperature. Young’s modulus was calculated from the slope of the stress-strain curves between 0.1% and 0.5% elongation. The test was conducted in five replicates on rectangular samples that were cut from the moulded composites (Section 2.3) at 30 mm in width, 10 mm in length, and approximately 1.5 mm thickness (depending on the shrinkage of the tested material).

2.4.4. Viscosity

The shear-dependent viscosity of the slurry was measured by rheology on a hybrid rheometer Discovery HR-1 from TA instruments, using a parallel plate geometry with a top plate diameter of 12 mm and a gap of 1 mm. Viscosity was measured on the freshly prepared slurry (Section 2.2) with a flow sweep measurement conducted at 25 °C, with a shear rate range between 0.01 and 100 s⁻¹. The viscosity value represented in the plots was measured at a shear rate of 10 s⁻¹.

2.4.5. 3D printing

3D printing was conducted with an in-house-built extrusion 3D printer prototype based on a robotic arm (ABB, IRB 1600 M2004) and an extrusion 3D printing head (3D printed in-house), displayed in Fig. S3. The system utilised air pressure and a rotating screw to guide the material from the reservoir to the nozzle. During 3D printing, a moving speed of 40 mm/s, a nozzle diameter of 9 mm and a layer height of 4 mm were used. Air pressure controlling the extrusion velocity of the material was adjusted manually (through a pressure regulator) based on the used formulation, and it was decreased by 0.1 bar once the reservoir was half
empty. The 3D printer was fed with freshly prepared slurry (Section 2.2). Two different types of samples were printed: cylinders 90 mm in diameter and with 25 layers to test printability and shrinkage and cylinders 100 mm in diameter and with 40 layers, to test the stability of higher structures.

2.4.6. Accelerated weathering

An accelerated weathering test was executed in a QUV chamber ATLAS UV Test® Fluorescent / UV Instrument. Operations of ASTM G154 cycle 1 were followed, whereby a UV cycle of 8 h at 60 °C and 0.89 W/m² using UVA 340 nm lamps alternated with a condensation cycle of 4 h at 50 °C. The test was conducted on samples within one, three, and six weeks (1000 h) of exposure. Rectangular samples were produced using a mold with a width of 75 mm, a height of 110 mm, and a thickness of 1 mm, using the same procedure described in Section 2.3. The samples were monitored during the test with pictures. After extraction from the chamber, the maximum deformations of all samples, and the mechanical properties of the samples that could withstand the six weeks of weathering, were tested. Lastly, microscopy pictures of the samples were taken over time, using a Dino-Lite Universal microscope, to evaluate visible changes to the surface of the degraded samples.

![Diagram of the ingredients and procedure used to formulate the bio-based slurry and to obtain the final composite material after water evaporation.](image-url)
3. Results and discussion

A schematic illustration of the ingredients and procedures used to prepare the slurry and then the composite is shown in Fig. 1. The slurry was based on deionised water (W), since water ensures the complete dissolution of the binder and provides optimal viscosity and flow properties during processing [29]. Bio-based glycerol (G) was used as a permanent plasticiser with the role of lubricating the slurry and increasing elasticity in the final material. Xanthan gum (XG) was selected as the binder because it is a water-soluble biopolymer with excellent shear thinning properties. Waste materials were used as fillers, targeting high loading in the final composite material. Specifically, two cellulose-based materials were selected: cellulose insulation (CI), which is produced from recycled paper, and sawdust (SD), which is a by-product of woodworking. Cellulose fibres are attractive because they are readily available, cheap, renewable, recyclable, have good insulation properties, and offer good stiffness [13]. Starting from a base recipe, the content of the different ingredients was varied, and their role was investigated to understand how to tune the properties of the bio-based slurry and find its optimal composition. In particular, shrinkage, mechanical, physical, and rheological properties, weathering resistance, and printability were evaluated.

3.1. Role of solvent and plasticizer

Using water as a base for the slurry had several advantages and disadvantages. Water has all the beneficial properties listed previously and can evaporate at room temperature. In addition, a high water content results in a lightweight and porous material after post-process drying. However, this process is slow and causes significant shrinkage and deformation. A system with very high shrinkage and deformation is problematic since it makes it difficult to predict the dimensions and shape of the final 3D printed structures. Another consequence of slow water evaporation is the softness of the material immediately after printing. This low early-age strength limits ultimate height, the number of printable layers, as well as the applicable pressure during printing.

In order to limit shrinkage and deformation while trying to retain the flowability of the slurry, the ratio of liquid ingredients in the slurry (solvent and plasticiser) was evaluated by decreasing water content (from 79 to 0 wt%) and conversely increasing glycerol content (from 1 to 81 wt%). Visual changes in the materials, and mechanical properties, are shown in Fig. 2.

From the dried samples (Fig. 2a), it was clear that a relatively small glycerol content of \( \leq 24 \) wt% resulted in a cohesive material, while higher concentrations led to a greasy, soft material that was difficult to handle. Furthermore, for contents of glycerol above 8 wt%, no distortion was observed during the drying process. When dried samples are
mentioned, it is implied that a certain amount of water is still present in the samples after the drying process due to the high hygroscopicity of the materials used. As seen in Table S7, it was observed that higher water content in the formulation resulted in higher residual water in the composite, with a maximum of 14.8 wt% and a minimum of 7.7 wt% for the highest glycerol content. Fig. 2b shows that with lower water content and higher glycerol content, lower volumetric shrinkage was achieved, starting from a shrinkage of 57 vol% for the higher water content samples and reaching a shrinkage of approximately 7–8 vol% with low or no water in the slurry. As the glycerol remains in the dry material’s matrix, the glycerol increases the density in the final composite material (Fig. 2b). This rise in density was insignificant for an increase in glycerol from 1 to 8 wt%, which changed from 431 to 529 kg/m$^3$ when compared to the three-fold increase when water was entirely substituted by glycerol, reaching 1261 kg/m$^3$. It is well known that the introduction of a permanent plasticiser can reduce the mechanical properties of the final material since its presence increases the distance between polymer molecules, effectively reducing their interactions and allowing higher chain mobility [39]. In particular, glycerol plasticisation also imparts sensitivity to moisture and temperature. As displayed in Fig. 2c, Young’s modulus dropped by a factor of four when increasing the plasticiser to 8 wt% of glycerol and ten with 24 wt% of glycerol. Regarding the slurry’s flow properties, replacing water with higher-viscosity glycerol led to an increase in viscosity, though the impact on processing was insignificant. Fig. 2c shows that variations in glycerol content caused minimal variations in the shear viscosity of the slurry for values between 1 and 24 wt% (128 – 160 Pa s), while higher amounts caused a more drastic rise in the shear viscosity of the slurry until reaching a value of 679 Pa s when water was entirely substituted by glycerol.

While analysing the relationships between these liquid components, it was observed that xanthan gum does not hydrate and swell in glycerol as it does in water (Fig. S5). In water, xanthan gum hydrated quickly (1 min), resulting in a pale yellow gel, while in glycerol, it showed slow and poor solubility. After one day of immersion in glycerol, it formed a white opaque hard gel due to glycerol/xanthan cross-linking, which was also observed by Bilanovic et al. [40]. It was observed that water is required, at least in minor amounts, to enable the quick dissolution of xanthan gum.

It was decided to proceed with the formulation containing 8 wt% glycerol and 73 wt% water (G8W73), which helped to avoid distortion during drying and reduce shrinkage without significantly affecting the binder hydration and the density and viscosity of the slurry.

3.2. Role of waste fillers

Recycled fibres were selected as fillers to promote the reuse of waste material and to have a cheap bulk material as the main component in the final composite since construction materials are expected to have a...
limited cost. Cellulose insulation consists of recycled newspaper with additives such as borax and boric acid as a flame retardant and to prevent fungal growth; additionally, it has some interesting features for application in the construction industry as thermal and sound insulation [41]. Sawdust is a by-product of sawmill industry processes and has already been used in concrete to obtain lightweight and low-density construction materials [42,43]. In order to evaluate the proper balance between these two fillers in the formulation, their content was varied; cellulose insulation content was increased from 0 to 18 wt% of the overall formulation, while sawdust content was simultaneously decreased from 18 to 0 wt%. The resultant samples and their effects on material properties are shown in Fig. 3. From the dry samples (Fig. 3a), it was possible to deduce that without cellulose insulation, the material was not cohesive and that cellulose also contributed to binding the ingredients together. Furthermore, it was noted that higher cellulose insulation content resulted in higher distortion during drying.

The ability of cellulose fibres to align in an ordered way and form hydrogen bonds allows the formation of a compact material, while the presence of sawdust, which is composed of thicker, shorter and stiffer fibres, prevents this alignment and compactness. These characteristics of the two different fillers influenced material properties, as illustrated in Fig. 3b and c. Volumetric shrinkage (Fig. 3b) increased with the cellulose insulation, reaching a maximum of 68 vol%, and decreased with sawdust, reaching a minimum of 29 vol%. Density (Fig. 3b) varied from 214 kg/m³, when only sawdust was used as a filler, due to the prevention of fibre alignment, to 552 kg/m³, when only cellulose insulation was used due to the compactness given by fibre alignment and bonding. The Young’s modulus (Fig. 3c) rose from 31 MPa when sawdust was used as a filler to 816 MPa when only cellulose insulation was used. Lastly, shear viscosity (Fig. 3c) was almost constant in the presence of cellulose insulation (100–148 Pa s) but drastically increased when only sawdust was used as a filler (4127 Pa s), which demonstrated the hypothesis of poorer mobility and the alignment of sawdust fibres.

Based on this analysis, it was observed that cellulose insulation acted not only as a filler but also gave cohesion to the final material because of the strong interaction between cellulose fibres. Instead, sawdust served as a low-weight bulky filler, which did not improve mechanical properties but resulted in higher porosity and higher free volume in the final material, thereby allowing for decreased deformation and shrinkage. Combined with the considerations for shrinkage and targeting the highest possible strength, the formulation with 11 wt% cellulose insulation and 7 wt% sawdust (CI11SD7) was selected for further work.

3.3. Reinforcing effect of vermiculite

Additional fillers were investigated to evaluate how to tune the properties of this material and obtain better mechanical performance. Among the different fillers, vermiculite, a medium-swelling clay with fireproofing and sound insulation properties, was found to be a good candidate. [44] The effect of vermiculite in different concentrations on the mechanical properties of the final composite material is shown in Fig. 4. Vermiculite at 5 pph in the presence of 8 wt% glycerol in the formulation effectively doubled Young’s modulus of the material (Fig. 4a), increased tensile strength, and reduced elongation. The addition of vermiculite also engendered a change of consistency in the slurry, which showed higher cohesiveness and a paste-like texture. Considering this point, adding 5 pph of vermiculite ((2)V5) was retained as an option to be considered when higher mechanical strength and higher cohesiveness are required in the composite.

3.4. Use of crosslinkers for binder support

Enhancement of the binding effect of binder and fillers was investigated by using calcium ions, from calcium chloride, as an ionic crosslinker. Calcium ions were selected because of their demonstrated effectiveness in cross-linking xanthan gum hydrogels and gels, showing a solid contribution in enhancing their mechanical properties [45–47], and because of improved gelation following the interaction between polysaccharides and divalent cations, known as the “egg-box” model [48]. The effect of this ionic crosslinker and the increased xanthan gum concentration was verified and compared through a tensile test (Fig. 5). Higher xanthan gum concentration resulted in a slightly increased Young’s modulus and tensile strength and significantly increased elongation. Calcium ions, overall, demonstrated a more linear behaviour when in the presence of 2.6 wt% of xanthan gum, wherein both Young’s modulus and tensile strength increased together with ion concentration, while elongation was not significantly affected. Higher early-age strength in the material with increased xanthan gum and the presence of the ionic crosslinker was proven with a gap test, with a significant reduction in deformation observed over time (Fig. S7).

Based on these considerations, 2.6 wt% of xanthan gum and a 0.2 M concentration of calcium chloride ((2)XG2.6Ca²⁺/0.2) were chosen as optimal to improve early-age strength during 3D printing.

3.5. Weathering resistance of the developed composites

The developed formulations were evaluated for their applicability in construction 3D printing, mainly focusing on the impact of the different
compositions and how they affect the material’s durability. Having water as the primary solvent of the slurry can have positive effects; for example, water can be used to reclaim the ingredients of the composite material (Fig. S10), but it can also have adverse effects in terms of poor weathering resistance. Fig. 6 illustrates the changes in the selected formulations during the accelerated weathering test to compare the impact of glycerol content, the use of an ionic crosslinker, xanthan gum concentration, and the use of vermiculite. Further information on the weathered samples is provided in Figs. S11, S12 and Tables S6, S8. The weathering resistance. The samples with lower glycerol content (G1W79) were washed away over concentration, and the use of vermiculite. Further information on the weathered samples is provided in Figs. S11, S12 and Tables S6, S8. The samples with lower glycerol content (G1W79) were washed away over the 6 weeks, thereby revealing poor weathering resistance and significantly lower performance than the samples with increased glycerol. The use of a crosslinker or an increase in xanthan gum concentration did not improve weathering resistance. Among the tested formulations, the vermiculite ((2)XG1.8Ca<sup>2+</sup>0.2V5) sample displayed higher resistance to accelerated weathering exposure since it did not develop cracks. The degradation of the least (G1W79) and the most ((2)XG1.8Ca<sup>2+</sup>0.2V5) resistant samples were compared through microscopy pictures (Fig. S11), indicating how cellulose insulation and sawdust leaked over time in G1W79, leaving a thin layer of cellulose. For ((2)XG1.8Ca<sup>2+</sup>0.2V5) in this regard, the fillers seemed to become more compact over time, resulting in higher cohesion in the material. Lastly, the mechanical properties of (2)XG1.8Ca<sup>2+</sup>0.2V5 were observed during the degradation test (Fig. S12). After 1 week of exposure, an increase in stiffness was observed, which could be explained by the leaching of the plasticiser (glycerol) due to water spray in the weathering chamber. However, after three weeks of weathering exposure, stiffness decreased due to degradation. Overall, even though water greatly influences the material, as expected, a small addition of vermiculite (5 pph) significantly improves its weathering resistance.

### 3.6. 3D printability of the developed slurries

In order to verify the printability of the investigated formulations, the optimised recipes (G8W73, (2)V5, and (2)XG2.6Ca<sup>2+</sup>0.2) were 3D printed with the use of a robotic arm extrusion 3D printer. The results of this test can be observed in Fig. 7, which shows images of the 3D printed samples freshly printed and dry, and Fig. S13, where shrinkage of the printed structures is displayed. Samples with 25 layers, which had a height-to-width ratio close to 1, revealed that all of the developed recipes are printable, stable, and reproducible. However, when observing shrinkage behaviour during drying, it was notable that G8W73 and (2)V5 performed very similarly, even though (2)V5 exhibited the least amount of shrinkage, while (2)XG2.6Ca<sup>2+</sup>0.2 displayed the highest shrinkage and variation among the produced samples. Samples with 40 layers, whose height was almost two times the width, showed more significant differences in performance among the formulations. G8W73 started to tilt during the first 5 min after printing, revealing that the low early-age strength of the lower layers could not withstand the height and weight of the structure. (2)XG2.6Ca<sup>2+</sup>0.2 showed improved early-age strength compared to G8W73, as expected from the increase of xanthan gum and crosslinker; however, during the drying period, it deformed in the lower part of the structure. (2)V5 displayed high stability and shape retention, even for the higher structure, as no deformation was observed during drying, and the lowest shrinkage observed among the samples, probably due to the improved mechanical properties and material cohesion following the addition of vermiculite. Among the formulations, G8W73 was printed at the lowest pressure (1.2 bar), while (2)V5 needed the highest driving pressure (2 bar), albeit both of these recipes dispensed uniformly from the printing nozzle. (2)XG2.6Ca<sup>2+</sup>0.2 required low driving pressure (1.4 bar), but the high stickiness of the formulation caused adhesion to the walls of the reservoir, thus making the extrusion more complicated. Based on the results obtained, all of the recipes proved to be printable; however, (2)V5 was the best-performing formulation, as the small addition of vermiculite helped overcome or minimise some of the typical problems associated with slurry 3D printing: deformation due to shrinkage, low early-age strength and a reduced capacity to support self-weight.

These formulations were also used in different scaled-up architectural prototypes to demonstrate their printability in various geometries and scales, an example of which is presented in Fig. 8. This demonstrates the potential of this material for use in architectural-scale 3D printing.

### 4. Conclusions

This study confirms the large untapped potential in the exploitation of waste- and bio-based materials for 3D printing of new construction materials. Optimised formulations of bio-based slurries, in which plasticiser, water, waste fillers, and binder contents were balanced to achieve optimal flow in the slurry and higher stiffness in the final composite, were identified. Particularly, the formulation G8W73 (C11SD7) was identified as a good compromise between shrinkage and deformation and exhibited good mechanical properties, low density, and good flowability. Similarly, the addition (5pph) of vermiculite imparted highly beneficial properties in terms of increased stiffness and cohesiveness ((2)V5). By combining a broad range of waste materials, we demonstrate how the proper selection of components enables using locally sourced waste materials for 3D printing new construction materials. In addition, the demonstration prints also show that this can take place on a relevant scale. Implementation of this kind of system has the
potential to increase the use of renewable raw materials for construction materials, thereby offering the potential to reduce CO2 emissions and waste generation in the construction industry.

**Fig. 6.** QUV weathered samples after different exposure times (0, 1, 3, 6 weeks) for the different formulations. (Note: when XG concentration is not mentioned, it is 1.8 wt%).

CRediT authorship contribution statement

Arianna Rech: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review &
Fig. 7. 3D printing tests were conducted on the most promising formulations. Printed cylinders before and after drying for G8W73, (2)V5, and (2)XG2.6Ca2+0.2, printed in 40 and 25 layers. (Note: the G8W73 40-layer dry sample is missing, as it could not support the weight of the higher structure).

Fig. 8. Upscaled 3D printing projects part of the Predicting Response project [49]: (a) 3D printing of a geometry 1.9 m long, 0.87 m wide and 0.1 m thick; (b) demonstrator 3D printed and assembled, 3 m wide, 1.8 m tall and 1 m deep (Photo credits to Anders Ingvarsten, 2022, Centre for Information Technology and Architecture).

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

The work was conducted as part of the Predicting Response project which was financially supported by Independent Research Fund Denmark through grant no. DFF – 9131 – 00034B.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.mtcomm.2022.104963.

References
