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3D printing of biodegradable parts using renewable biobased materials

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Abstract

The paper presents ways of utilising the additive manufacturing process 3D-printing using renewable biobased materials, with a focus on adapted packaging for sensitive components. This application provides an attractive scenario for sustainable production and the re- or upcycling of waste material such as wood flour, rice husk or miscanthus fibre. Packaging of prototypes with sensitive or filigree structures or made of fragile materials is currently difficult, because standardized packages are often not suitable and cost for adapted packages are high. Within this study, CAD-data based adapted packaging made of renewable raw materials is addressed. The 3D-printing process was modified for using conditioned biobased fibre and a special binder. Furthermore, a software tool was developed to create adapted packaging designs by using the individual part geometry.
In this way and in combination with AM processes, sustainable and biodegradable packaging for complex components can be generated within a short time. The manufactured packaging is conform to the demands of packaging for fragile goods and can be produced just-in-time.

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Keywords: Additive Manufacturing; 3D Printing; Binder Jetting; Renewable Materials; Biodegradable Materials;

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1. Introduction

Additive manufacturing (AM), also called 3D printing by the wider public, is a group of manufacturing technologies that can, and already has, change the way parts and products are being made [1]. Contrary to the established subtractive technologies such as milling or grinding, AM builds up material to form a part, stepwise and layer by layer. It allows for new designs to be realised which were unthinkable of using only subtractive processes, such as structures inspired by biological growth or internal cavities. While much research and development has been conducted using metallic and polymer materials for AM, some material sectors are so far not being addressed. This study works towards closing this gap and focuses on bio-based materials and their application in 3D printing.

2. 3D Printing

3D printing (3DP) in its original terminology is a process within the group of Additive Manufacturing. In 3DP, also named Binder Jetting, an inkjet print head is used to inject a liquid (mostly a binder substance) into the top layer of a powder bed, causing fusion or agglomeration of the particles. By lowering the platform and raking a new defined layer of fresh, unfused powder on top, the process can be repeated (Fig. 1, a). This enables the generation of three dimensional parts in a powder bed. Because of the bed's stabilisation mechanism, there is no need for support structures as they are required for example in resin-based processes such as SLA. Furthermore, the process is fast compared to the polymer-melt based FDM/FFF processes which are colloquially coined '3D Printing'. With state-of-the-art printers, around 20 mm of build height are possible per hour on a full platform of the size of 300 x 200 mm.

Fig. 1: (a) 3D printing process principle [2], (b) multi-coloured 3D printing example part [3]

After the part has been printed, it is taken out of the powder bed and blown clean of the unused powder. Mostly, the part is then infiltrated or at least (spray) coated to enhance its mechanical properties and add water proof features. Commonly used powder materials include gypsum, starch and ceramic materials (which need to be sintered after 3DP). Because the process is based on desktop inkjet print head technology, multi-coloured parts are possible. Its application was first to be found in the prototyping/design sectors where the visual properties were more important than industrial-grade mechanical strength (Fig. 1, b).

3. Biodegradable materials

3.1. Motivation

While many parts aim for maximum lifetime, for some applications a very defined lifetime is more beneficial. Packaging, for example, needs to protect the goods until they have reached their final destination; afterwards its use has ended and it needs to be disposed of. Similar boundary conditions exist for visual aids or decorative parts such as theater stage or film set props, architectural models or event equipment. For most of these applications, the volume of material used is high, as is the amount of waste generated. Commonly used materials are polymers in solid or foamed state, which degrade very slowly or need to be incinerated in a controlled environment. From a sustainability point of view, an environmentally friendly disposal route is highly anticipated. The application of materials that are quickly biodegradable could significantly contribute to this. Additionally, the use of biobased and existing natural waste material could further increase the environmental benefit [2,4,5].

3.2. Properties

As in any production scenario, the material needs to be suitable for or tailored to the requirements of the application. The properties relevant for the applications mentioned above are low density to reduce part weight, defined or even adjustable degradation timespan, optional water resistivity, good temperature damping, for some cases biocompatibility and sufficient mechanical properties such as compressive or bending strength. Natural resources however have a large fluctuation of properties, which needs to be considered in designing the process and part.

4. 3DP of biodegradable materials

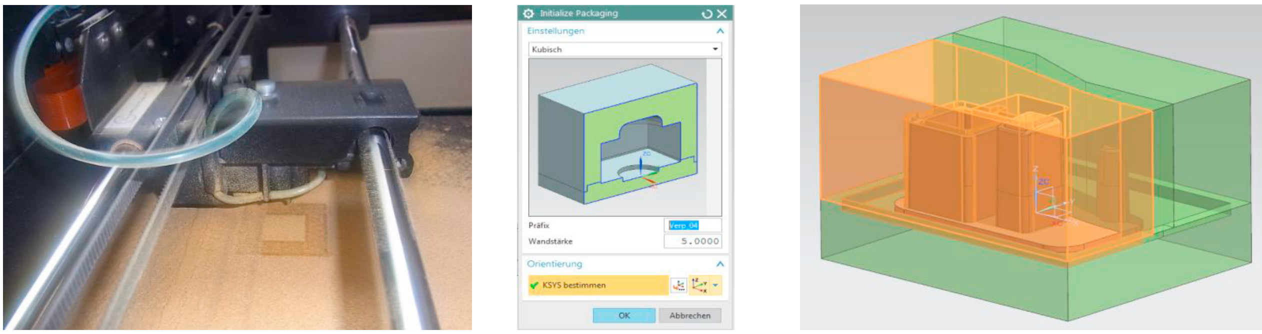
Additive manufacturing as a key enabling technology can shift the design boundaries towards more complex parts without incurring additional cost or process requirements. This makes it especially suitable for customized parts and parts of low batch size. Furthermore, given the small dimensions and low external requirements of some AM machine tools, it can co-locate production and use case site, reducing logistics and transportation needs.

While many users and makers apply FFF (fused filament fabrication) machines because of the low cost and ease of use, other AM technologies should not be neglected. This study focuses on 3D Printing, or Binder Jetting, process which has significant benefits in terms of speed and flexibility when using biobased materials.

4.1. Printing technology and equipment

For this study, different AM machine tools are applied: ZCorp Z510, Z402 and Z310 models incorporating HP inkjet print heads (Fig. 2 a)), capable of printing in single- and multi-coloured. These aged, but commonly used printers suffer from technological limitations such as general stability issues (clogging of nozzles, operating system instabilities) and are limited in their capability of adapting to fluctuating material properties. While designed for operation in defined, proprietary materials, the large spectrum of natural material properties poses a challenge. Also, they are limited in their build size envelope to roughly 250 x 350 x 200 mm³. However, the binder jetting principle allows for an easy incorporation of novel materials without the need to produce filaments.

To easily create adapted packaging, a plugin for Siemens NX CAD software was programmed (Fig. 2 b)). By using the CAD data of the to-be packaged part, it enables the user to design a multi-part package, tailor-made to the part and its boundary condition (such as no-touch-areas, specific holders etc.). After only a few clicks, the plugin generates the package part (Fig. 2c)) and the stl-file, which can be directly transferred to the 3D printer.



a) b) c)

Fig. 2: a) 3DP print head in operation b) plugin to create packaging data c) CAD data for multi-part package (part visible), created by plugin

4.2. Selection and modification of printed materials

To work towards circular economy and low environmental footprint targets, the re-use of de-facto waste materials and their upcycling is a stepping stone. Here, we aim at using waste material that has no other use apart from energetic/ burning or to use locally available materials with little or no transportation need and low cost.

4.2.1. Miscanthus particles

Miscanthus is a fast growing reed (C4-plant) that has little requirements regarding soil quality. Because it is very lightweight and has good damping properties. It is, apart from energetic use, mostly used for fibre casting e.g. to create egg cartons. For this application it has to be shredded, milled and sieved to a usable particle distribution (Fig. 3 a)). For 3DP a small particle size that is considered a waste product of the fibre casting process is used.

4.2.2. Wood flour

Wood flour are residues of mills and wood-working plants. While the longer fibres are used for wood pellet production, the short fibres suitable for 3DP. Depending on the type of wood, properties vary significantly. Wood types assessed included e.g. willow, beech, maple (Fig. 3 b)), larch (Fig. 3 c)), oak, spruce, lime.

4.2.3. Seashell powder

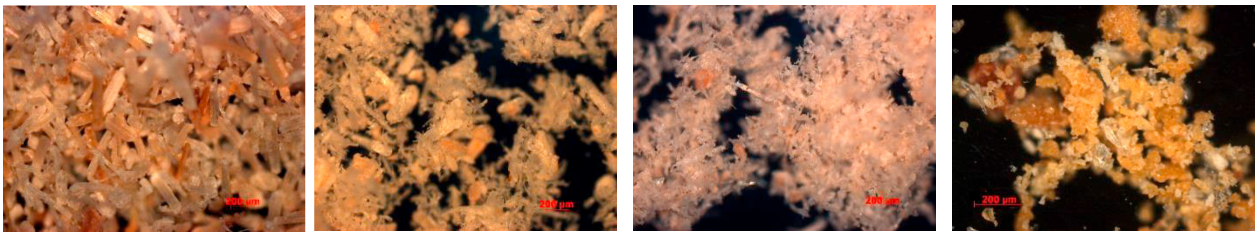
Seashells are abundant in some regions and lack a significant use case. Their availability however could promote applications that can make use of their properties. For use in 3DP, they are milled and ground to relevant size.

4.2.4. Fruit stone flour

Drupes, or stone fruits, contain a solid stone that is not edible. Some of these are used by the pharmaceutical industry to produce oil from the soft inner parts, the hard shell is usually disposed of. For 3DP apricot stone is investigated. Its hard parts are milled and sieved to a relevant particle spectrum. Compared to wood flour and Miscanthus they have a relatively high density (Fig. 3 d)), which can lead to good part strength and accuracy due to the small possible particle size.

4.2.5. Perspective: rice husk

Rice husk is abundantly available in many regions, making it an interesting candidate to investigate. So far, there have been few trials without significant results. Certainly, rice husk will be investigated in the future development.



a) b) c) d)

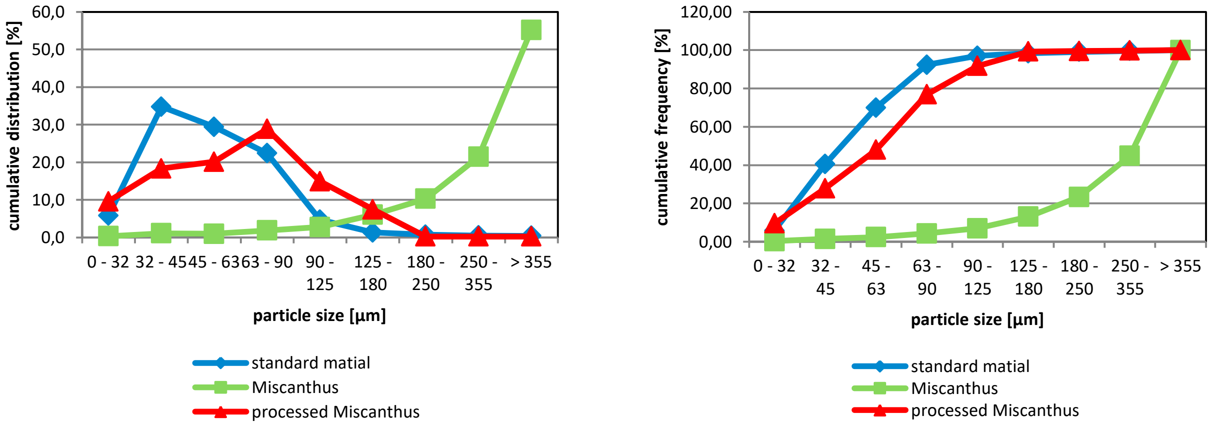
Fig. 3: microscopy images of material particles a) Miscanthus b) Maple c) Larch d) Apricot stone

4.2.6. Powder preparation

Powder for 3D printing needs to fulfill certain requirements to be successfully printed. The required accuracy (layer thickness for *z*, print resolution for *x* and *y*) defines the upper boundary for its particle size. Handling and processing properties such as tendency to agglomerate, electrostatic charging, flowability etc. which diminish below a certain particle size need to be considered, too. Hence, the particle distribution needs to be carefully set.

To adjust the material accordingly, sieving and sizing was conducted until the distribution curves aligned well with those of the standard 3D printing (manufacturer supplied zp151) material. Fig. 4 shows the original distribution of both standard (blue/diamond curve) and natural (green/square curve) material – in this graph Miscanthus powder

– and the distribution curve after sizing (red/triangle curve). The good alignment is prerequisite for successful 3D printing.



a) b)

Fig. 4: a) cumulative distribution and b) cumulative frequency of standard material, Miscanthus and processed Miscanthus powder

Fig. 5 shows images of Miscanthus powder after harvesting, milling and processing.

a) b) c)

Fig. 5: Miscanthus powder after a) harvesting and shredding b) milling c) sieving (ready for 3DP)

4.3. Binder selection and evaluation

In 3DP, a binder is necessary to stabilise the powder particles. Binder selection is based on the targeted application and capabilities of the printer and print head. For biodegradable parts, the binder must be biodegradable, too; it should also be non-toxic, easy to handle and readily available. In the context of renewable materials, the binder should also be based on natural or renewable resources. Because of these requests, common binders for 3DP were not acceptable.

A further challenge posed by the powder material is its inertia to react with known liquid binders. To overcome this challenge, solid binders were investigated. To use them they are mixed with the powder before printing; activation takes place by printing an aqueous solution into the powder bed.

Binders were tested and evaluated with regard to printing accuracy, part and process stability as well as easiness of handling. Amongst the binders successfully evaluated were lignin sulfonate, sodium silicate (waterglass) and polyvinyl alcohol, whereas the latter provided the best results (Fig. 6).

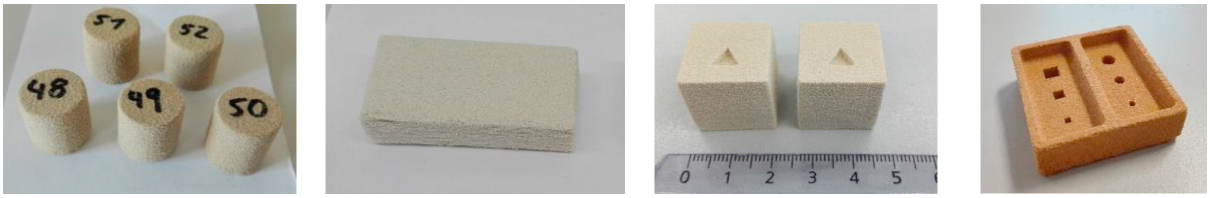
To increase mechanical strength and add functionality such as water resistance, post processing by spray coating or infiltration is possible. Again, the application defines the material choice – for increased stability a common epoxy resin is promising, but not suitable for fast biodegradation.

a) b) c)

Fig. 6: 3DP test parts using different binder materials a) lignin sulfonate (40 wt %) b) sodium silicate (6 wt %) c) PVA (9 wt %)

4.4. Geometries

To identify and measure properties of the printed parts, standard geometries were printed and evaluated. Test geometries included cylinders and bars for mechanical strength analysis, cubic parts for shrinkage tests and parts incorporating geometrical features of different sizes for accuracy assessment (Fig. 7).



a) b) c) d)

Fig. 7: test sample geometries a) (Miscanthus) cylinders b) (Maple) bar c) (Larch) shrinkage test part d) (Apricot stone) geometry test part

Furthermore, to evaluate capabilities of the different materials, demonstrator parts were printed, amongst them a 3D puzzle, a model chair, a complex bridge structure, architectural models and adapted packages created with the plugin. Figure 8 shows examples of parts in different materials. The Miscanthus bridge structure in Fig. 8 c) was spray coated with a sodium silicate solution, which enhanced mechanical strength significantly.

a) b) c) d)

Fig. 8: test sample geometries a) (Miscanthus) ball/hollow hemisphere b) (Apricot stone) 3D symbol c) (Miscanthus) thin walled bridge structure d) (Sea shell) thin walled shell model

Due to the limitations of the printer, parts larger than 250 x 250 x 200 mm³ could so far not be produced in one piece. To create larger parts, they as of now must be joined from smaller parts. Clearly, the setup of a larger printer (larger than 1 m³) is targeted, which would open up new applications in e.g. stage prop and furniture production.

5. Results

5.1. General printability / Binder/material combinations

The applicability of renewable resources for 3D printing was proven. By selecting appropriate powder/binder combinations such as Miscanthus/PVA, solid, lightweight parts were produced. Process parameters and machine setup has to be set in a way to minimize clogging of print head nozzles and to consider the lifetime of the powder mixture.

5.2. Mechanical properties

By selecting/combining powder material and binder, mechanical properties can be widely adjusted. However, without post-treatment by infiltration or coating, the mechanical strength of most parts is small compared to polymer based (FFF/SLA/SLS) parts. Their low density and soft texture shift the application more towards non-mechanically loaded designs.

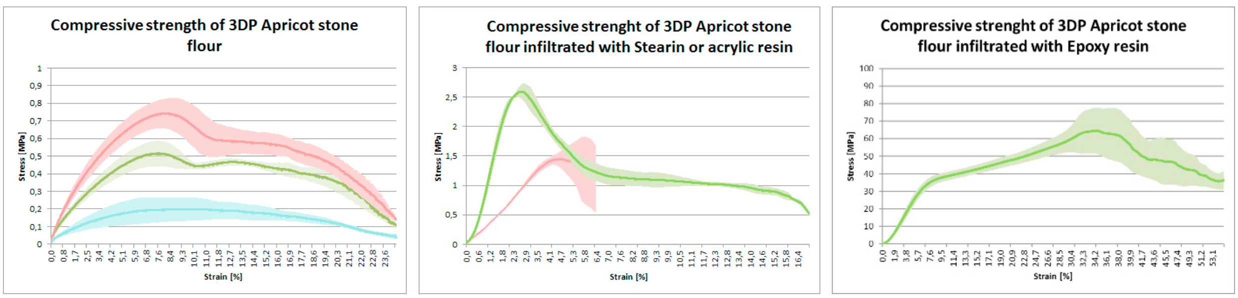
To evaluate the influence of infiltration and different coatings to mechanical strength, compressive strength tests have been conducted. Tests were carried out on cylindrical test samples of dia. $d = 20$ mm and height $h = 20$ mm using a Zwick 100 kN test rig at $v = 0.02$ mm/s feed. The samples were infiltrated with either acrylic resin, epoxy resin or stearin. As a comparison, as-built samples without infiltration were tested, too.

a) b) c) d) e)

Fig. 9: a) compressive strength test setup; samples after test: b) Miscanthus c) Apricot stone, uncoated d) Apricot stone, coated e) Sea shell

While a spray coating with Acrodur enhanced the compressive strength slightly compared to the as-built samples, Lupamin coating had a negative effect (Fig. 10 a)). The best result in terms of compression strength could be achieved applying epoxy resin infiltration (Fig. 10 c)), followed by stearin and acrylic resin (Fig. 10 b)).

Notably, changing the build direction by 90° led to a significant decrease in strength to below 20 %. An explanation is given by the fact that the inter-layer connection is far weaker than in-layer.

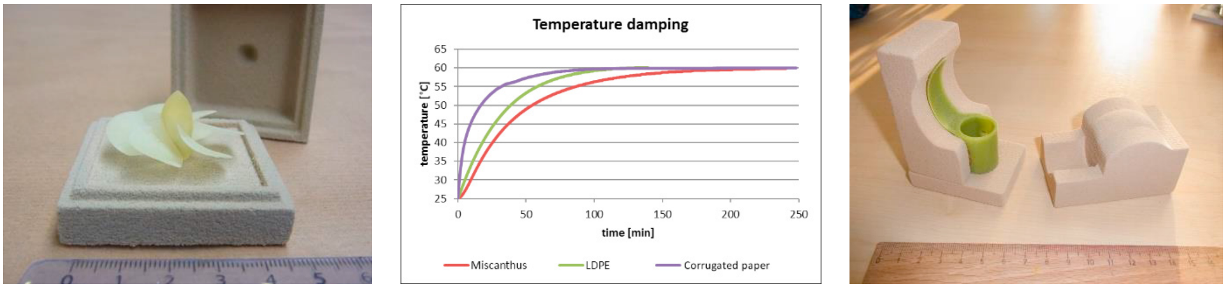


a) b) c)

Fig. 10: compressive strength a) Acrodur coated (red/top); uncoated (green/center); Lupamin coated (blue/bottom) b) stearin (green/top); acrylic (red/bottom) infiltrated c) epoxy infiltrated

5.3. Adapted packaging application

Both the low density and the good mechanical damping as well as the low thermal conductivity render the materials suitable for adapted, i.e. product-specific packages. Shipping tests undertaken on fragile, thin-walled wax components for investment casting and incorporating shock and temperature sensors showed that the packages are suitable and can compare to state of the art materials like polystyrene (Fig. 11).



a) b) c)

Fig. 11: adapted packaging a) SLA part b) temperature curve during shipment c) wax part

6. Summary

Renewable materials have been successfully used in 3D printing; it could be shown that a large variety of substances can be adapted. Their specific properties have been analysed and suitable applications were found. By utilising them for adapted packaging, a biodegradable solution based on the re-use or upcycling of waste material was found.

7. Outlook

The next step in applying renewable materials in Additive Manufacturing must be local sourcing to further reduce the transportation cost. Trials have already been started, also to increase the build size and speed of the printer. Further applications such as temporary furniture will be addressed.

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