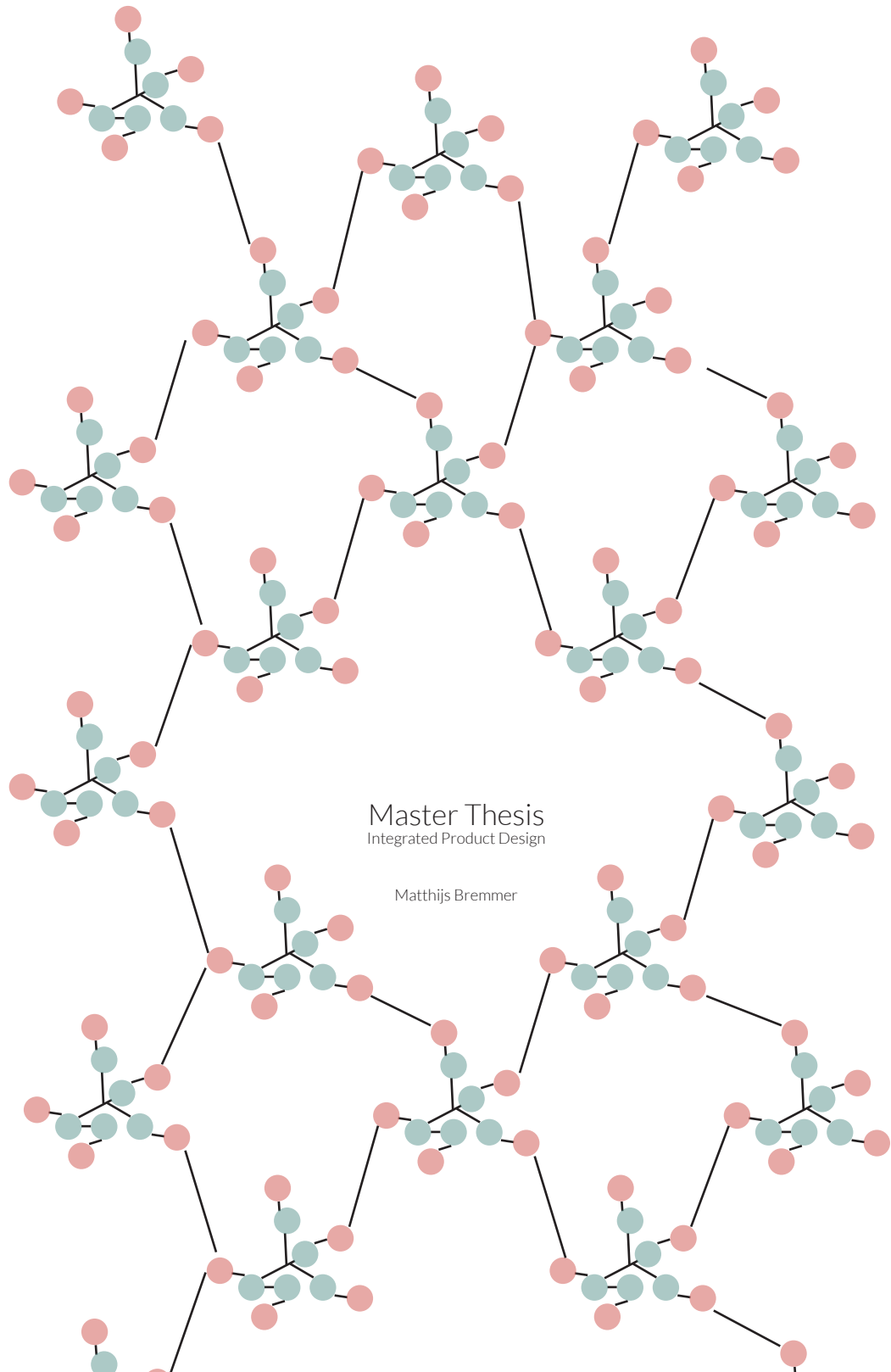


# 3D printing with bio-based materials

Designing a toolkit to guide makers in sustainable material development



**(Albert) Matthijs Bremmer**

4261984

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Master Thesis

MSc Integrated Product Design

Faculty of Industrial Design Engineering

**Supervisory team**

Prof. dr. ir. Bakker, C.A.

Dr. Faludi, J.

MSc. Sauerwein, M.

**Delft University of Technology**

Faculty of Industrial Design Engineering

Landbergstraat 5

2628 CE Delft

The Netherlands

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“Ever Tried.  
Ever failed.  
No matter.  
Try again.  
Fail again.  
Fail better.”

- Samuel Becket



# Preface

---

In front of you lies the result of my graduation project. The quote on the left page summarizes the project on multiple levels. It was an advice that was given to me many times during the process, as my project more often than not was a fight with myself. But it also describes the mindset a material developer who wants to develop a 3D printable bio-based material must have.

Many of the experiments that are carried out in this stage of development that bio-based 3D printable materials are in currently will fail, as there is so little to hold on to. But through perserverance one will reach satisfying results. In the final design I have tried to keep those “failed” experiments in mind, by not seeing them as failed experiments but as useful datapoints.

It was a fight to stay persistant myself and I could not have done it with the ongoing support from my supervisors Mariet, Jeremy & Conny and the support from Gerben and my parents. Thank you for checking in with me so often and dragging me through this process.

Matthijs

# Executive Summary

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Materiom is an online initiative that has been growing an online open source database of bio-based material recipes over the last couple of years. A new category of recipes to be found on this website is the category of bio-based 3D printable materials. The first 3D printable material recipe to emerge in the database was developed at the TU Delft. Since then the recipe was further developed into an alginate mussel shell composite. It was time for the next step: How can more of these kinds of materials be developed? Are there any 'universal truths' to be found in developing these materials and how can makers/designers be guided in this process?

To tackle this problem a multileveled analysis was performed. A literature review of 3D printing with bio-based materials showed that in different fields of expertise, ranging from the medical field to the field of food, the first steps had been made into printing with bio-based materials. Printing experiments were then performed with a selection of binder materials mentioned in these papers. With iota-Carrageenan and kappa-Carrageenan in combination with different kinds of fillers such as rice starch and ground cacao shell, successful prints were made. Furthermore, interesting hand-printing results were obtained with chitosan and alginate. More importantly, however, an indicative range of material ratios was found that could help future material developers save time in their search. Finally, some basic guidelines for the printer set-up regarding nozzle size, print head speed and extrusion speed were formulated based on the findings in the experiments.

These results led, together with the insights gained from analyzing makerspaces, cookbooks and the Materiom website, to a design proposal for an online material development tool. Currently the Materiom website provides ready-made recipes. As noticed during the analysis phase, however, not every experiment leads to a complete recipe. Also, based on the experience of a material developer, complete recipes are not always what is needed. Recipes can be inflexible and hinder experimentation and innovation. The material development tool provides therefore room for data on different levels. This means that the community can share a larger amount of data and it leaves room for more experimentation and innovation.

This idea is brought in practice by dividing data into three different parts: Material information, material compatibility information and complete recipes. Together with a formulaic recipe that gives a starting point for experimentation and printer set-up guidelines, material developers have a range of tools that they can use according to their needs and likings for new material development. And when the experiments have been carried out, the data can be shared with community even when the results were not satisfying and thus saving time of their successors.

## MATERIAL BUILDER

Welcome to the material builder tool. This is a set of webpages where you can find help with to create bio-based materials for 3D printing. There are different sections that can be useful for different levels of experience with material creation. The recipe database is similar to what Materiom already offers. On the material creation page a more generalizable picture is sketched of printing pastes based on gels. In the component matcher, you can find which materials go well together, in case you already have some materials in mind and finally the printing guidance helps with setting up a 3D printer for bio-based material prints.

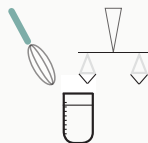
### Materials in focus



### Directories



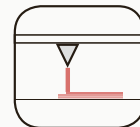
Recipe database



Material Creation



Component Matcher



Printing Guidance

Database of material recipes which are ready to make.

Starting points for ingredient ratios to speed up the beginning of the experiment

Component matching data for the more adventurous material developer

Guidance on the transition from hand printing to machine printing

Based on data from the community that can be collected through a data entry form

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# 1 PROJECT FORMULATION

## 1.1 INTRODUCTION

This graduation thesis is the result of the research and design work done on the subject of bio-based 3D printable pastes. This report describes the results of the analysis of printing with bio-based pastes and the context in which this happens. A design proposal is made to bridge the gap that currently exists between available knowledge in research papers and experimental material design performed by designers and material enthusiasts in the makerspace environment. Furthermore, the design opens more opportunities for community-based contributions to an open-source database of knowledge on this subject.

## 1.2 PROBLEM DESCRIPTION

The interest in 3D printing - also known as additive manufacturing (AM) - is quickly rising. The economic impact is predicted to be over \$500 billion per year by 2025 (Manyika et al., 2013). With the rise of this production technique new opportunities and challenges arise. Both positive and negative aspects regarding the sustainability of AM are being researched at the moment.

On one hand, Sauerwein et al. (2019) identified several aspects of AM that could be beneficial to sustainability. The Government Office for Science in the UK (Walport & Laphorne, 2014) summarizes those opportunities in the following quotes: "AM can have a profound impact on the way manufacturers make almost any product". Furthermore, they state that AM "will become an essential 'tool' allowing designs to be optimised to reduce waste; products to be made as light as possible; inventories of spare parts to be reduced; greater flexibility in the location of manufacturing; products to be personalised to consumers; consumers to make some of their own products; and products to be made with new graded composition and bespoke properties"

On the other hand, a lot of work needs to be done to improve the environmental impact of AM. Fused Deposition Modelling, an extrusion based printing technology, has a specific energy consumption that is up to 100 times higher than traditional bulk production processes like injection moulding (Yoon et al., 2014). Printing energy is currently the driving factor in the sustainability assesment of 3D printing (Faludi et al., 2015), but there are concerns as well regarding toxicity (Merlo et al., 2015). Material choice is in this aspect an important factor, as the material choice determines the process that can be used and thus the printing energy and possible toxicity (Faludi et al., 2019).

Together with the rise of AM, there is also a rise in the diy culture. Furthermore, makerspaces are becoming more popular. These makerspaces are mentioned as drivers for innovation (BRON!). Initiatives like Materiom are catering to these developments by providing open source information on the production of bio-based materials.

## 1.3 ASSIGNMENT

Recently, Vette (2018) developed a 3D printable and recyclable paste made from a mix of mussel shells and alginate that tackled some of the aforementioned challenges. The recipe was an improvement of an earlier recipe developed by Sauerwein, Doubrovski and Vette that was published in the open source database of Materiom (2018). Materiom is an online platform that promotes the distribution of knowledge regarding bio-based materials.

The impression that this recipe was only the start of a far greater set of bio-based 3D printable materials led to the inception of an assignment that:

1. Has the aim to explore other bio-based 3D printable pastes;
2. Translates the gathered knowledge on bio-based 3D printable pastes into a tool/ approach that can guide designers in the exploration & development of bio-based 3D printable materials.
3. Helps Materiom to build an open source database of bio-based 3D printable material recipes

## 1.4 METHODOLOGY

The general method followed through this assignment is the triple diamond method. A phase of divergence, followed by a converging phase.

### *Analysis*

In the analysis phase the design context was investigated in order to get a better understanding of it and design a solution that fits to the problem. The analysis was performed through a mixture of literature research, desk research and lab experiments. First, a broad literature research was performed on 3D printable bio-based materials in different fields. Google scholar and Scopus were searched for papers on bio-based 3D printing with combinations of the terms: "Bio-based", "Bio", "3D-printing", "AM", "Additive manufacturing", "Paste printing", "Paste extrusion", "Food printing". This divergent search into 3D printing with bio-based materials was followed by a convergence towards specific bio-based paste printing and specific investigation into the subjects of cross-linking and material specific research. To achieve this, the broad set of literature that was found was filtered, to exclude filament printing and other non-paste printing methods. Furthermore, some literature on cross-linking was reviewed. The literature was then analyzed on recipe proposals and their performance. Based on the results from the literature review, a set of lab experiments were performed with the resources that were prominent and promising in the reviewed literature. Next to the material focused research, some desk research was performed on the context and user. The current Materiom website was investigated and literature on makerspaces was reviewed. Furthermore, interviews were taken with the people from Materiom. Finally, design engineering students were interviewed during and after a 3D printing workshop with bio-based materials on their experiences and the pitfalls that they encountered during material experiments. The combination of the outcomes of these investigations led to the formation of a design assignment, to which the following phases were tested.

### *Ideation*

The outcomes of the analysis phase are used as inputs for the ideation phase. The ideation phase is at first diverging, resulting in a broad spectrum of ideas that could possibly fit the solution of the assignment. These ideas are then clustered and assessed and the

most fruitful ideas are taken to the next phase. The selection of ideas is based on the knowledge gathered in the analysis phase and whether the ideas fit the design goal that was formulated at the end of the analysis phase.

### *Conceptualization*

The most promising ideas from the ideation phase are the starting point for the conceptualization. The ideas are elaborated in higher detail and developed into concepts. The concepts are assessed on a set of criteria and feasibility. This process is performed in an iterative and continuous manner.

### *Evaluation*

The evaluation part is mentioned as last here, but that is not a representation of the actual process. Evaluations are being performed throughout the project, to check whether the outcomes a certain part of the project still alligns with the goals of this project.





## 2. ANALYSIS

Different types of analysis were performed to explore the subject of 3D printing with bio-based materials. The design context, equipment used, the materials themselves and the user were analyzed in order to get a good overview of the design space of the assignment. The findings are then used to form a design goal.

### 2.1 ADDITIVE MANUFACTURING

#### Introduction

Additive Manufacturing [AM] is a relatively new set of manufacturing processes in which, opposed to more traditional subtractive manufacturing like milling or turning, a part or product is built up layer by layer. In this chapter, the most common groups of AM are discussed, the process that is chosen for this project is explained and machinery to execute these processes are introduced.

#### Types of additive manufacturing

In order to get an overview of the available additive manufacturing methods and the focus of this report, four major AM categories will shortly be discussed. The most widely used additive manufacturing processes can be categorized into four main groups (Dai et al., 2019):

1. Extrusion-based methods such as fused deposition modeling (FDM) and direct ink writing (DIW);
2. Particle fusion based methods like selective laser sintering (SLS);
3. Photopolymerization based methods (e.g. stereolithography);
4. Inkjet printing.

Extrusion-based methods use the principle that a viscous liquid or a molten material, basically the ink, is extruded through a nozzle. The nozzle is following a path that has been defined by the 3D printing software, based on the CAD model of the product. Layer by layer the product is built up. For good printing results, the ink should quickly transform into a solid or have solid-like behaviour after extrusion. This can be achieved by (1) quick melt-to-solid transitions (this is used for e.g. ABS, PLA, etc.). (2) Shear-thinning or thixotropic behaviour, in which the ink flows under the extrusion pressure but becomes more viscous once the pressure is removed (the opposite behaviour of a water/corn starch bath). (3) Cross-linking, which is the interconnecting of different polymer chains. This will be discussed in 2.2. (4) Evaporation of solvents (Guvendiren, 2016).

Particle fusion based methods make use of a laser that heats particles over their melting temperature and fuses them together. For each new layer of which the product is built up, a new layer of particles is added. Then the laser travels along the path that describes that specific layer and fuses the particles where needed. This process is repeated until all layers of the product are built up and fused (Kruth et al., 2015).

Photopolymerization based methods are using light to start the polymerization process. A UV or a laser beam moves over a bath of photopolymerizable liquid. Upon contact with

the beam the liquid polymerizes, forming a solid. Once a layer of the product is finished, the plateau on which the product is placed, lowers itself into the liquid bath and a new layer can be added (Guvendiren, 2016).

Inktjet printing is a 3D printing process in which small droplets of ink (1-100 picoliters) are deposited onto a printing surface/substrate. On this surface the droplets solidify, to form 3D structures. The solidification happens through the same mechanisms as that were described for extrusion-based methods; e.g. evaporation/gelling/temperature controlled transition (Derby, 2010).

For this project, the first category of AM is chosen; extrusion-based 3D-printing. This is done because of the resources at the university and because the research this project is building on was also on extrusion based 3D printing. In order to strive for low print energies, a paste extrusion process with no material heating is chosen, as the heating process is often energy intensive (Faludi et al., 2019). This means that there are three basic directions into which will be looked for good printing results: shear thinning behaviour, cross-linking and evaporation of solvents. In the printing with pastes chapter (2.3) these options will be discussed more in detail.

## Machinery

The variety of equipment that can be used for extrusion-based 3D printing is large. A couple of different options are briefly discussed with their key characteristics.

### *Paste extrusion printers*

Paste extrusion cannot be executed by the more traditional and widely used filament extrusion printers like the Ultimaker range. The last few years, there have been developed a couple of add-ons and standalone 3D paste extrusion printers that can deal with paste extrusion. Furthermore, some tutorials are available for self-built conversions. Below a couple of the different options for paste extrusion are discussed and a summarization of the properties of those printers can be found in [table 1](#). The key properties are (minimum) nozzle size, paste capacity, printbed size and price.

### *Stoneflower 3D*

The company Stoneflower 3D offers both a conversion kit for filament-based 3D printers as well as a standalone paste extrusion 3D printer. The kit can for instance be incorporated in the Ultimaker series of 3D printers, while the 3D printer is a standalone device.

### *Ceramic 3D printing KIT*

The Ceramic 3D printing KIT made by Stoneflower 3D is a clay 3D printing add-on that can be used in combination with a 3D printer or a CNC-router. It consists of a print head that homogenizes the clay and removes air bubbles, a ram extruder/syringe pump and a control unit that controls extrusion speed and print head acceleration. The price of this conversion kit varies between €499 and €800,- depending on the size of the ram extruder (0.5L or 1.5L). Furthermore there is an additional micro printing set for €29 that allows for nozzles of 0.5, 1.0, 1.5 and 2.0 mm. The maximum printing dimensions depend on the size of the 3D printer or CNC-router to which it is attached.

### *3D printer Stoneflower*

The 3D printer from Stoneflower is a paste 3D printer that uses the same principles as the ceramic kit, but integrated in a standalone format. The maximum printing dimensions are 50x50x50 cm<sup>3</sup>, with an option to increase the height to 80 cm. The print speed can be varied between 30-100 mm/s and nozzle size from 0.5-7mm. The capacity of the ram extruder is 1.5l. The price of this printer is €3599.

### Structur3d Discov3ry

Like stoneflower, the Structur3d Discov3ry is a conversion kit for filament 3D printers. It can be added to almost any 3D printer. It works with 60 CC syringes with the luer lock system and the syringe tips range from 0.1-1.4mm. The maximum printing dimensions are based on the 3D printer to which it will be attached. The price of this add-on is \$1299.

### Zmorph

Zmorph offers paste extrusion print heads for their Zmorph VX 3D printer. The nozzle size is either 2mm or 4mm and print speeds range between 10-100mm/s. The price of this print head is €180, but it can only be used in combination with the Zmorph VX that starts at €2299.

### Wasp Clay kit

Wasp offers a clay kit add-on that is suitable for most 3D filament printers. Its nozzle size can be varied between 1.2 and 2 mm using their stainless steel nozzles. The paste tank can hold 3000 or 5000 ml of paste. The extruder has an outward pressure of max 40 bar. The kit comes without an air compressor, so this has to be bought separately. The price of the kit is €690.

Name	Min nozzle size	Capacity	Price	Printbed size
Stoneflower 3D Kit	0.5 mm (with microprint set)	0,5 l / 1,5 l	€499/€899 microprinting set: €29	Dependent on printer
Stoneflower 3D Printer	0.5 mm	1,5 l	€3599	500x500x500 mm <sup>3</sup>
Structur3d Discov3ry	0.1 mm	0.06 l	\$1299	Dependent on printer
Zmorph VX 3D	2 mm	0.1 l	€180 (printhead) €2299 (printer)	up to 250x235x165 mm <sup>3</sup>
Wasp Clay Kit	1.2 mm	3 l / 5 l	€690	Dependent on printer
3D Potter Potterbot - Scara v4	1 mm	1 l - 4 l	\$2850 - 17950	254x254x305 mm <sup>3</sup> - ø1829x1143 mm <sup>3</sup>
Vormvrij LUTUM V4.3	0.6 mm	0.7 l / 1.4 l	€6595	430x450x500 mm <sup>3</sup>

Table 1 - 3D printers and their main characteristics

### 3D Potter

3D potter offers a wide range of 3D clay printers, using direct extrusion. The nozzle size is variable from 1-10mm. The printing dimensions range from 25.4x25.4x30.5 cm<sup>3</sup> to ø182.9\*114.3 cm<sup>3</sup>. The printing speed can be varied between 35-60 mm/s, 55-130 mm/s and 30-100 mm/s depending on the model. The extruder capacity ranges from 1000-

4000ml. Prices range from \$2850-\$17950.

## LUTUM

Lutum offers a 3D clay printer with the interesting option of dual material nozzles. This can be used to work with two component clays, like two component glues. In the experiments section the use of sodium silicate liquids is discussed. This material seems to react with certain binders which makes working with it in a single nozzle system difficult. A dual nozzle system could possibly solve this issue.

## Summary & Conclusions

From the different types of AM, extrusion based 3D printing will be used in this project or more specifically paste extrusion printing. The underlying mechanisms that are important for this printing method are either evaporation, cross-linking or thixotropic behaviour of the paste. For the creation of well-performing pastes, it is important to search for these mechanisms and to use these properties to make well-performing 3D printable pastes. Furthermore, from the analysis of the different types of 3D printing hardware, it can be observed that specifications regarding e.g. minimum nozzle size, but also pressure settings vary wildly. Therefore, universal guidelines must be created in the design process to guide makers in setting up their 3D printer

## 2.2 PRINTING WITH PASTES

### Introduction

In this project, extrusion based 3D printing is used as printing method, more specifically paste-extrusion printing. Pastes come in a wide range of sorts, but can be described as viscous fluids made from mixing one or more solid ingredients with a liquid. In this project the main focus is on composite pastes. Composite paste materials are built up from at least two types of ingredients; binders and fillers.

Binders are viscous liquid substances or solid substances that are mixed with liquids to form viscous liquid substances that can harden after a chemical or physical process (e.g. a chemical reaction or evaporation). Binders bind for example the filler materials (e.g. fibres or powder) that are added to the binder. A well-known example of a binder is a glue.

Fillers are solid materials that are added to a binder mixture to improve the mechanical properties. Fillers can be particulates or fibers, where fibers have high aspect ratios – fibers are long compared to their width – and particulates lower aspect ratios. Fibers are used to improve e.g. tensile strength.

### Knowledge gap

Currently very little literature is available on the subject of bio-based 3D printing materials for industrial design applications – see also the subchapter below. Furthermore, the literature that is currently available is not catered to designers. This chapter aims to give an overview of what is currently known in different fields of practice. The gathered information is used as input both for the following material experiments and for the guiding tool. Such that designers do not have to dig through this often complicated literature.

### 3D paste printing in industrial design applications

For product applications, several studies have been set up to test and develop new bio-based 3D printing materials (e.g. Dai et al., 2019; Faludi et al., 2019; Sauerwein &

Dubrovski, 2018; Zeidler et al., 2018). In the next paragraph some of algae-based gels will be discussed that were mentioned in the aforementioned papers.

Some of the materials that are not mentioned in the medical literature are polyvinyl alcohol and sodium silicate liquid (Faludi et al., 2019; Zeidler et al., 2018) and rice flour, gelatin and wheat dextrin as multipurpose materials (Faludi et al., 2019). Sauerwein & Dubrovski (2018) used a water sugar mixture as binder, while Dai et al. (2019) mentioned the earlier described methylcellulose as binder among other cellulose ethers.

## Hydrogels in the medical field

Most research on paste extrusion printing is currently conducted in the medical field (e.g. Reddy et al, 2015; Wei et al., 2015; Wang et al., 2015; Ng et al., 2016; Negrini et al., 2018 & Li et al., 2018). Although some constraints that are present in the medical field do not necessarily apply in the production of products via 3D printing and a 1-to-1 translation of results is therefore not always possible, the field holds a large body of knowledge on 3D printable materials and especially hydrogels. An aspect that the medical field has to deal with for example is the avoidance of high pressures – this can lead to cell death. This constraint does not hold for (industrial) product production applications. The knowledge from the medical field can therefore be used as inspiration, but the possibilities in product applications are possibly wider. Some of the biobased hydrogels in the medical field are discussed below.

### (Macro)Algae-derived

Alginate is a polysaccharide that is made from different types of brown seaweeds. Alginate can form hydrogels, where the brittleness/elasticness is based on the ratio between the G and M blocks, the two types of building blocks from which the polysaccharide is built (Li et al., 2018). Lowering the pH of an alginate solution can increase the viscosity the solution, as more hydrogen bonds are formed. The alginate can be ionically cross-linked through addition of positively charged cations ( $\text{Ca}^{2+}$ ,  $\text{Ba}^{2+}$ ,  $\text{Mg}^{2+}$ ). Furthermore, covalent cross-linking is possible upon poly (ethylene glycol) diamine (Lee & Mooney, 2012).

Carrageenan is, like alginate, a polysaccharide, but derived from red seaweeds. There are three types of carrageenans; kappa, jota and lambda. Kappa- and lambda-carrageenan can cross-link in the presence of cations, e.g. potassium. Kappa-carrageenan has the favourable mechanical properties of the two cross-linkable carrageenans (Li et al., 2018).

Agar (or agarose) is a natural occurring polysaccharide obtained from red algae. It is mainly used together with other binder ingredients, as it is hard to use on its own as agar has a wide temperature range where it starts to liquify (20-70°C) (Li et al., 2018). It has been used with succes, however, in combination with for instance alginate (Wei et al., 2015).

### Animal-derived

Gelatin is a protein derived from collagen and is soluble in water. Hydrogels can be formed below temperatures of 29°C. Therefore, the interest from the biomedical field into unmodified gelatin is rather small, as this is below the human body temperature (Li et al., 2018).

Collagen, a protein, is one of the main components of connective tissue in mammals. Collagen is not very suitable for 3D printing due to the slow gelation mechanism. The deposited collagen hydrogel stays in a liquid state for up to 10 minutes (Smith et al., 2004)

Chitosan is a linear polysaccharide that can be obtained by deacetylation of Chitin. Chitin is obtained from the exoskeleton of crabs and shrimps and is the second most important biopolymer after cellulose (Rinaudo, 2006). Chitosan is often used in hydrogels in the medical field, due to the good biocompatibility. To improve its mechanical properties,

chitosan can be cross-linked with a couple of cross-linkers, like epichlorohydrin and diisocyanate. Furthermore, in literature, different chitosan mixes are proposed, e.g. with cellulose and polyvinyl alcohol (Rinaudo, 2006). Due to the fact that chitosan is the only positively charged bio polysaccharide, it can form hydrogels with negatively charged polysaccharides like alginate (Li et al., 2018).

### Cellulose-derived

Methylcellulose is derived from the biopolysaccharide cellulose. In contrast to cellulose, methylcellulose is soluble in water (Suntornnond et al., 2017). Methylcellulose is very viscous and is good stackable. This makes it suitable as printing material. For instance, Li et al. (2017) prepared a highly thixotropic hydrogel made of a methylcellulose-alginate mix.

### Cross-linking

An important factor for the toughness of polymeric materials is cross-linking. Cross-linking is basically the forming of a connection between – or bonding of - different polymer molecules. Due to cross-linking the different polymer chains of which a material is built become better connected. Not only – weak - intermolecular forces “hold” the molecules together, but there is ionic and/or covalent bonding as well. Covalent bonding happens when two atoms share an electron. Ionic bonding is the connection between positive and negatively charged atoms (ions), this can occur when an electron from one atom moves to a nearby other atom, effectively creating a positively (where the electron left) and negatively (to where the electron went) charged atom that are attracted to each other. This interconnectedness results in a less brittle material, as the material consists of a network of polymer chains. Instead of relatively easily breaking alongside a linear polymer chain (brittle behaviour), the polymer chains are better interconnected, leading to tougher material characteristics (Nielsen, 1969).

### 3D printing with food pastes

In the food world there is an interest in paste extrusion printing as well (e.g. Godoi et al., 2016, Lille et al., 2018 & Liu et al., 2018a/b, Sun et al., 2015). To a certain extent, similar materials are used in food printing to obtain hydrogels. Alginate, gelatin and starches are for example proposed by Godoi et al. (2016). Apart from those earlier mentioned hydrogels, Liu et al. (2018b) produced a mashed potatoe paste with xanthan gum and kappa-carrageenan that showed favourable shear-thinning behaviour and well matched geometries when compared to the CAD model. This paste was also compared to pure xanthan gum/ guar gum/ kappa-carrageenan samples and performed better on dimensional accuracy and surface quality (Liu et al., 2018a). Lille et al. (2018) printed different mixtures of milk powder, starch, bean and oat protein concentrate and cellulose nano fibers. Pastes made of 10% starch + 15% skimmed milk powder; 0.8% cellulose nano fibrils + 50% semi skimmed milk powder; 60% semi skimmed milk powder & 35% oat protein concentrate showed best results.

### Summary & Conclusions

In different areas of expertise there has been a rising interest in printing with pastes. The knowledge on printing with paste-like mixtures is most advanced in the medical field, but there is a growing interest from the design field and the food (design) field. Gelling agents have mostly been investigated. Some gels are temperature-controlled others are controlled by pH. In the medical field many crosslinkers have been identified, but the usefulness in industrial design applications seems limited due to the high prices. Materials that are recurring in all three different fields are: carrageenan (both iota and kappa), alginate, gelatin and starches. Because the requirements for materials in industrial applications are less stringent regarding bio-compatibility/digestion/health than for the food or medical sector, the use of Sodium Silicate liquids is an interesting option as well.

### Introduction

This chapter will provide deeper insights in the design context for this assignment. This not only contains the user group, but as well an overview of the company the design is for as well as the broader theory of Circular Economy.

### Makerspaces

The context of this assignment is the makerspace, also known as fab labs. Fab labs or makerspaces are places that give individuals the opportunity to design and/or fabricate their own products by offering an array of digital manufacturing equipment (like 3D printers). Opposed to other prototyping firms/services, the maker in a makerspace often operates without the intervention of a technical specialist (Kohtala, 2013). Fablabs lean on a peer-to-peer educational community in which makers can learn from each other. These aspects make the makerspace an interesting context for this project, as makers cannot always rely on an expert that can advise them what exactly to do when searching for/designing with bio-based 3D printable materials.

Furthermore, makerspaces have been mentioned as places that support sustainable innovation (Kohtala, 2016) and enable decentralized production, which in itself has the potential to support CE, because this can influence the user-product interaction and 'prosumer' behaviour (Lofthouse & Prendeville, 2017). Stimulating bio-based material development in makerspaces by giving makers the tools to design their own materials can thus hopefully help sustainable innovation.

### Materiom

The company for whom this design process is executed is Materiom. Materiom is an open-source database for bio-based material recipes that wants to accelerate the development in this area. They want to achieve this by lowering barriers in the new materials market and give communities the means to nourish themselves. To achieve this goal they cooperate with makerspaces, companies and local communities and support them in the development of new bio-based materials based on the characteristics of the natural resources that are available in the respective regions where those communities operate in. Their open-source database makes it very easy to produce the materials by following recipes (see figure 1.). By enabling makerspaces to develop bio-based materials based on the local natural resources, local communities can be nourished and local production can be stimulated ("Imagine if", 2019). Local production is seen as a sustainable alternative for centralized production (Ford & Despeisse, 2016) and provides the opportunity to close the loop in a circular economy by linking local resource streams to local production and recycling (Sauerwein et al., 2018).

### Circular Economy

The Circular Economy is a term that is widely used in different definitions as is described by Kirchherr et al. (2017). A partial description of the Circular Economy, apart from the concepts of refusing rethinking and reducing products, is that the Circular Economy is an economic system in which products that are at the end of their service life "turn into resources" for new product/service lives (Stahel, 2016). This "turning into resources" for a new life manifests itself in different forms. The most preferable and simple manifestation of this is reuse where the product stays intact. The least favorable manifestations are recycling where the product is broken down into different resource streams that are used to build new products or even incineration, in which the internal energy of the product is used for energy production (Kirchherr, 2017).



Earlier on, a wide range of binders was discussed that have been presented in a wide range of articles, from the medical field to food and architecture. Little of these articles had any industrial design related implementations. Material tests were therefore performed in order to relate this literature to the practice of designers and how they can create their own materials. This chapter will give an overview of the selection of the materials to be tested, the experiment design and the experiment results.

### Material selection

Based on the findings in the literature analysis, a set of binders is selected for further investigation. Most of the known literature is aimed at the medical world; a one-to-one translation of the findings in these papers to product-oriented applications is therefore unlikely. The findings of the literature review are therefore only indicative and provide a starting point for the material exploration experiments.

As binder materials the following materials were selected

- i-Carrageenan
- k-Carrageenan
- Agar
- Alginate
- Methylcellulose
- Sodium silicate liquid
- Chitosan

As additive, Microfibrillated cellulose (MFC) was selected, as some of the literature showed promising results of MFC as a strength enhancer.

As a filler, the following materials were used:

- Rice flour
- Ground mussel shells
- Walnut shell powder
- Cacao shell powder
- Pistachio shell powder

Most of the tests have been performed with rice flour, as this was readily available and most of the other powders were very time intensive to produce. Furthermore, the focus of the research was mostly on binder materials, as these often work with a wide range of filler materials.

### Experiment design

The experiments were set up in loose fashion. In the first experiments an idea of ingredient ratios was established. Furthermore the influence of the order of ingredient addition was investigated. The paste quality seemed to be independent of the order of material addition. This enabled finetuning of pastes to a desired consistency even when the initial trial recipe was slightly off. Furthermore the differences in paste behaviour for different fillers were tested. And finally, machine printing was performed with a selected set of materials. In these experiments it was tried to obtain general values for printer settings that could be translated to different printer set-ups.

Recipe	HP	MP	Viscosity	Print Quality
RF : W : i-Car 6 : 8 : 0.14	✓		More viscous than cake dough, but flows nicely	Considerable shrink
sRF : W : i-Car 3 : 5 : 0.14	✓		Same as 1	More shrink than 1
TS : W : i-Car 2 : 3.5 : 0.1	✓		Same as 1	Similar to 2
RF : W : Agar 3 : 4.2 : 0.45			Seems shear-thinning	Overextrusion of water, thus flowing after print
MS : W : i-Car 3 : 4.25 : 0.14	✓		Similar to 1-3	Quite hard and good
RF : W : 80A : Ch 3.5 : 3 : 0.3 : 0.3			Very fluid-like	Flowing after print
RF : W : 80A : Ch 5 : 4 : 0.2 : 0.2			Too thick, mud-like	N.A.
RF : W : 80A : Ch 5 : 6.75 : 0.2 : 0.2			Similar to 1	Too liquidy -> flowing
RF : W : k-Car 3 : 9 : 0.4	✓		Gummy-like	Very good, good flow, little sagging. Brittle result
RF : W : MC 2.5 : 6 : 0.4			Very sticky	Flowing after extrusion
RF : W : MC 4.5 : 6 : 0.4	✓		N.A.	Better than 10
RF : MFC 3 : 4			Easily flowing under pressure	Uneven extrusion speed, flowing after extrusion
"Former" + W : k-Car + 2.5 : 0.2	✓		Same as 12	More consistent speed than 12, holds up nicely
RF : SSL 2.05 : 3.5			Too viscous for printing with tip	Very strong material after extrusion without tip
MS : SSL 13 : 7	✓		Better (lower) viscosity than 15	Hard, good, slightly saggy

Table 2a - Experiment results

MP = Machine Print  
 HP = Hand print  
 RF = Rice Flour  
 W = Water  
 i/k-Car = iota/kappa-Carrageenan  
 TS = Tapioca Starch  
 MS = mussel shell

80A = 80% vinegar  
 Ch = Chitosan  
 MC = Methylcellulose  
 SSL = Sodium Silicate Liquid  
 CS = Cacao Shell  
 WS = Walnut Shell  
 PS = Pistachio Shell

Recipe	HP	MP	Viscosity	Print Quality
RF : W : k-Car : SSL 5.5 : 5 : 0.2 : 2	✓		<i>Improving over the course of printing</i>	<i>Improving over the course of printing</i>
MS : W : k-Car 3 : 9 : 0.4			<i>Similar to 1</i>	<i>Stronger than 1, good print</i>
CS : SSL 2.3 : 15			<i>Lumpy &amp; too viscous</i>	N.A.
CS : W : k-Car + SSL 3 : 10 : 0.35 + 10	✓		<i>No lumps, very viscous but printable</i>	<i>Nice extrusion, no sagging</i>
WS(large) : SSL 2 : 5			<i>Overextrusion of liquids</i>	<i>Overextrusion SSL, no good print</i>
WS(<125micron) : SSL 1 : 4			<i>too thick</i>	<i>Not printing due to chemical reaction</i>
PS : SSL : k-Car 1.5 : 10 : 0.5			<i>too thick</i>	<i>Same as 25</i>
W : RF : k-Car 12 : 9 : 0.6	✓	✓		<i>Good print.</i>
W : CS : k-Car 12.5 : 5 : 0.55	✓	✓	<i>Some thickening over time of the paste</i>	

Table 2b - Experiment results

## Results

### Recipes with starches

#### Iota-carrageenan-based recipes

Test 1 used a paste made of rice flour, water – normal, cold, tap water & iota-carrageenan. Kappa-carrageenan was selected for its cross-linking possibilities, but due to availability, the first tests were performed with iota-carrageenan.

The first paste was produced by first mixing the water [8g] and the rice flour [6g]. The water/rice flour ratio was achieved after some time and based on gut feeling. When the solution was more viscous but still fluid, the binder was added. Roughly 1 wt.% of the total paste weight of carrageenan was added [0.14g], this is on the lower side of the recommended percentage in literature [1-3%]. The consistency of the paste was slightly more viscous than cake dough.

Printing: The first paste flowed nicely. There was an even disposition of material but quite some extrusion force was needed. After drying, there was a considerable shrink in the prints, probably due to the high water content (>50%). This shrinking also led to cracks in the material. See figure XY for the printing results

Test 2 and Test 3 also used iota-carrageenan, but with different fillers. Test 2 used sticky rice flour [asian food store] and Test 3 used tapioca starch. The ratios were found in the same way as in Test 1, e.g. water+filler and then binder. For Test 2 ratios were found of sticky rice flour/water/carrageenan = 3g/5g/0.14g. For Test 3 ratios were found of tapioca starch/water/carrageenan = 2g/3.5g/0.1g. Thus both tests had higher water contents than the first tests. Printing results seemed, however, to be quite comparable, see figure XY & XY.

#### Agar-based recipes

Test 4 explored a different binder: Agar. 3g of rice flour was added to [3.5g] of water and mixed. Then [0.45g] agar was added in tranches of [0.15g]. After the third addition of [0.15g] agar the paste became too thick, thus an extra [0.7g] of water was added. Under stirring, it appeared as if there was some shear thickening behaviour, but this could not be tested.

Printing: The paste did not print. Under pressure, the water contents of the paste were extruded, but the solid components stayed in the syringe.

Test 5 explored the same binder as 1,2&3 but now with a different filler type. Instead of the starch or flour like fillers from 1,2 & 3 a calciumcarbonate-based filler made of ground mussel shells was used. The recipe consisted of 3g mussels on [4.25g] water and [0.14g] carrageenan. Flow of the paste was similar to 1,2 & 3 but the resulting print seemed to be harder – and more brittle - than the first three tests.

#### Chitosan-based recipes

After the first five tests it was found that it was more convenient to first make the gel. It is also easier to control the consistency and viscosity of the final paste, as a mix of water and filler is very responsive to a little addition of binder, while a mix of binder and water is less responsive to the addition of a little more filler.

Test 6 explored the binder chitosan. As chitosan is only soluble in acidic fluids, [0.3] g of 80% vinegar was added to [3g] of water. To this, [3.5 g] of rice flour was added. The resulting paste was printable and flowing nicely through the nozzle, but quite quickly after disposition the paste flowed. More tests are needed to see how this paste can be made more suitable for 3D-printing.

In test 7 a mix of [4g] water, [0.2g] vinegar and [0.2g] chitosan was tested with [5g] of rice flour, but this paste was too thick for printing. Water was added to the paste ([2.75g] in total) but the resulting paste did not hold up well, similar to test 6.

#### Kappa-carrageenan-based recipes

From experiment 8 onwards, the iota-carrageenan is replaced by kappa-carrageenan. In literature it was found that kappa-carrageenan was cross-linkable in a sodium or kalium bath in contrast to the iota-carrageenan for which no easy cross-linking reaction is available.

Test 8 explored k-carrageenan in a higher amount compared to experiment 1. ~3 wt.% [0.4g] of k-carrageenan was dissolved in [9g] of water. To this mix [3g] rice flour was added until a gummy-like texture was achieved. The paste was very viscous, but seemed to be thinning under stirring, which suggests that this might be a shear-thinning fluid. Under extrusion there was a continuous flow and no sagging. Even with hand printing a single walled structure of more than 10 layers was easily achieved, see figure 1. The structure seemed slightly grainy.



Figure 1 - Single walled pyramid

### Methylcellulose-based recipes

Test 9.1 explored the binder Methylcellulose. [0.4g] of methylcellulose was dissolved in [6g] of water. Dissolution was not perfect, as air bubbles and small lumps formed. After addition of the rice flour [2.5g], however, the paste seemed to have become homogenous without air bubbles trapped inside it. The resulting paste was very sticky and the output under extrusion was nice and continuous. After disposition, though, the paste flowed.

In test 9.2 an alternative paste for test 9 was created with the same amounts of water and methylcellulose, but with [4.5g] of rice flour. This paste seemed to sag slightly less, but the sagging was not eliminated entirely, see figure 2.



Figure 2 - Sagged prints, no printing lines visible anymore

### Microfibrilated cellulose-based recipes

Test 10.1 explored a microfibrilated cellulose solution [3wt.% MFC] as a water replacement. [4g] of the microfibrilated cellulose solution was mixed with 3 g of rice flour. This paste was flowing really easily and showed minimal sagging after printing. At the end of the hand printing experiment, the water content of the remaining paste seemed to be lower, however. It looked like more of the water contents were extruded compared to the rice flour. Furthermore, the extrusion speed was not constant.

To tackle the problems found in test 10.1, another paste was made in test 10.2. The same paste as in 10.1 was used, but a gel of [0.2g] k-carrageenan and [2.5g] water was added. The results were promising. The paste showed the same positive characteristics, but the

extrusion speed was more consistent and the paste did not seem to be thickening over the course of the extrusion.

### Sodium silicate liquid-based recipes

In test 11 another type of binder was investigated. Thus far gelling agents have been tested, that transform water into a gel after dissolution. Here, a sodium silicate liquid was used instead. [3.5g] of sodium silicate liquid was mixed with [2.05g] of water. The paste was too viscous for hand extrusion through a nozzle, but printing without a nozzle tip was promising. The paste flowed well under extrusion and was solidifying quickly after disposition. The resulting print seemed very cohesive.

Test 12 explored the same binder as 11, sodium silicate liquid, but with another filler, mussell shells. [7g] of sodium silicate liquid was mixed with [13g] of mussell shells. Compared to test 11, much more solid content was needed (~65% solid contents in test 12 compared to ~37% solid contents in test 11). This is promising, as less liquid can evaporate and shrink after drying will be less. Printing results were nice, although there was some sligh sagging after disposition. Once dried up, shrinkage was small and the material was quite hard.

### Mixing it up

In test 13.1-13.3 different pastes of all binders that were used before were produced.

In test 13.1, sodium silicate liquid [2g] was mixed with a water [5g] k-carrageenan [0.2g] gel and rice flour [5.5g]. At first the paste was too liquid, but over the course of the extrusion, the paste started performing better. Was the excessive water extruded first? Or was there overextrusion and would it eventually end up in clogging? No decisive answer was found yet to this question.

Test 13.2 added to 7g of mix 13.1 microfibrilated cellulose [3.5g] and k-carrageenan [0.2g]. The paste seemed smoother than 13.1, but did not flow at all after disposition.

Test 13.3 added even more ingredients to the existing mix. 4.5g of 13.2 was mixed with an additional 1.5g of mussel shells. The printing went nicely, but at the end a dried rest of paste was left in the syringe. Some overextrusion was thus happening.

Test 14 explored the difference between rice flour and mussel shell as filler. The same recipe was followed as in 8, but the rice was replaced by mussel shell powder.

After the first hand printing tests, a print with the stoneflower paste printer was executed to see whether a paste behaved differently.

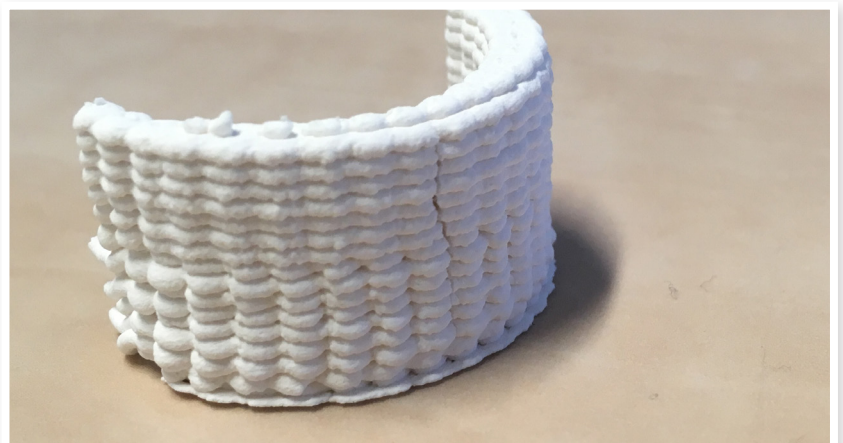


Figure 3 - 3D printed rice flour carrageenan cylinder part



First, the paste of test 1 was replicated with k-carrageenan instead of i-carrageenan.

A mix was made of [12g] water, [0.21g] k-carrageenan and [9g] rice flour. This paste was printed into a cylindrical form. This first print was flawed due to some issues that occurred in Cura - the slicing did not reproduce all parts of the cylinder, but the overall print quality was quite ok. The material showed some small cracks due to shrinkage and it was brittle, see figure 3. In this case, there was however, again overextrusion of the fluids at play. The print stopped because of clogging before the end of the file.

### Cellulose-based recipes

Based on the findings of Faludi et al. (2019), that showed relatively good printing results with pecan shell flour and sodium silicate liquid, pastes were produced consisting of at least sodium silicate liquid as binder and cellulosic fillers in the form of agricultural waste (walnut shells, cacao shells and pistachio shells).

In experiment 16 & 17, the combination of cacao shell powder with a particle size smaller than 0.075 mm and sodium silicate liquid was investigated. The ingredients of both recipes were the same – 2.3 g of cacao shell powder and 15 g of sodium silicate liquid - but the order of addition was changed, as the first try (addition of sodium silicate liquid (SSL) to cacao shell powder) resulted in lumps. The results of switching the addition order did not change. The lumps seemed to appear because the SSL reacted with parts of the cacao shells, which made those reacted fractions undissolvable. This reaction was also noticeable by the smell of the paste. While the cacao shells initially smelled very much like chocolate, after addition of the SSL, the smell turned into something like ammonia. The resulting pastes were barely extrudable through large tips. A part of the contents of the syringe in the form of a cylinder was dried to see what the resulting material would look like.



The dried material seems very strong and also tougher than earlier prints. Therefore it would be interesting to see whether there are ways to get a good extrusion.

Figure 4 - Cacao shell flour with K-Carrageenan and sodium silicate liquid

In experiment 18, it was tried to avoid the formation of lumps. The idea was to first form a gel with kappa-carrageenan (0.35g) and water (10g) and mix the cacao shells (3g) in this gel resulting in a very viscous gel with evenly distributed cacao shell powder. This was tried as in earlier experiments no lump-forming was observed after using this method with starches. Next, the SSL (10g) was added to the mix. In the first try, a very watery solution was the result. As this did not seem logical the experiment was repeated. Now the resulting paste had a nice texture – it was still a fluid but very viscous - and was easily extrudable and not sagging after extrusion, see figure 4. Probably, a human error was made in the first experiment and water was added in the second stage instead of SSL.

Next, in experiment 19, walnut shell powder was used as a filler ingredient. First, the walnut shell powder (2g) as purchased (Berivita) was used in combination with SSL (5g).

The particle size was rather large. Upon extrusion only the SSL was being extruded while the solid contents remained in the syringe. Probably the particle size was too large for extrusion.

To test this hypothesis, walnut shell powder (1g) with a particle size not exceeding 0.125mm was used in experiment 20. SSL (4g) was added to this, but this paste behaved similarly to the cacao shell powder of experiment 18. Again an ammonia-like smell was observed upon mixing and the walnut shell powder turned very dark.

As both cellulose-based materials seemed to react with the SSL, at least when the particle size was small enough, A test was done to see whether SSL gells like water upon addition of kappa-carrageenan. 10g of SSL was mixed with 0.5g of kappa-carrageenan. This resulted in only a marginally gelled mix. To this mix 1.5g of pistachio shell powder ( $\leq 0.125$  mm) was added. The resulting paste was very gluey and barely extrudable through a tip, possibly due to the large friction. Furthermore the paste flowed after extrusion, resulting in blobs of material.

Printer set-up

During the 3D printing experiments data was gathered on the flow rate of prints that executed well. In table 3 a summary of these settings is presented.

As the way 3D printers can be set up varies wildly, it is not very useful to communicate the settings for e.g. material flow in the StoneFlower paste extruder - almost the lowest setting. Rather, one wants to communicate printer setting data in a way that can be easily translated to other settings.

Parameter	nozzle I.D. = 1.55 mm
Layer width	~ 1,5 mm
Layer height	~ 1.1-1.2 mm
Printing head speed	5.8 mm/s

Table 3 - Initial printer setup

The settings presented in table 2 can be easily carried over to other printing systems or adapted to that system. The rules for setting these parameters are:

- (1) layer width is roughly equal to the nozzle size
- (2) layer height is roughly 80% of the nozzle size
- (3) a good starting point for the printing speed = 5.8 mm/s - this setting can be adjusted in the printer's software

Based on these settings, it is now possible to construct recommendations for parameter settings for a wide range of paste extruders.



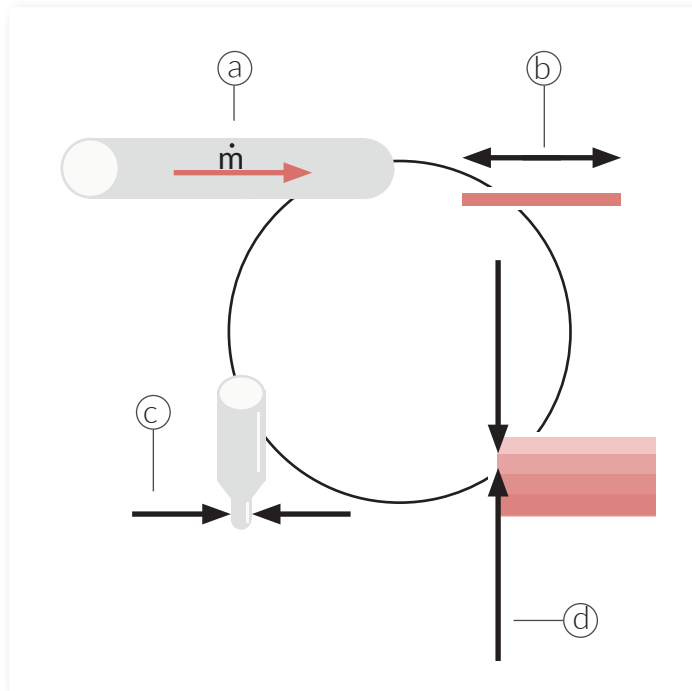


Figure 5 - Schematics of printing parameter relationships

In figure 5 a schematic overview is given of the printing parameters that are interconnected. The material flow (a) [mm<sup>3</sup>/s] is distributed over length (b) [mm/s] in nozzle width (c) [mm] and layer height (d) [mm].

When one now has determined the nozzle that will be used, the initial layer width and layer height can be set. The material flow is then calculated via the following formula:

$$a \text{ [mm}^3\text{/s]} = b \text{ [mm/s]} \cdot c \text{ [mm]} \cdot d \text{ [mm]}$$

The resulting material flow can be set in the 3D printer. When no numerical value can be entered, a test print can be performed for e.g. 10 minutes to obtain the numerical value of the print speed.

## Summary & Conclusions

Overall, for most material combinations it was possible to produce a paste that was at least hand-printable. Chitosan, methylcellulose and agar are still challenging, however, to get printable. More tests, also with cellulose-based filler materials, instead of starch-based materials are needed. Furthermore experiments with slightly acidic solutions should also be tried as some fillers work better in a slightly acidic environment.

Kappa-carrageenan seemed to be performing best so far from the algae-derived filler materials. The cross-linking step by addition of potassium ions has to be performed, though. Without cross-linking, the material is still very brittle. Furthermore, shrinkage so far has also been considerable, so more effort should be put into keeping the paste printable while minimizing the amount of solvents used. More tests will be done, also with other solvents like isopropanol alcohol and glycerine.

Sodium silicate liquid is a very interesting binder, as the resulting prints seem to be the hardest produced so far. In many instances, however, it can be hard to produce a paste that is still extrudable through smaller tips. For the cellulosic materials sodium silicate liquid also seems to react with the very small particle sizes. This did not seem to happen for the larger walnut particles (size unknown), but the smaller (0.125/0.075mm) walnut and cacao particles started to smell strange after addition of sodium silicate liquid. This reaction also seemed to cause problems for complete dissolution of the filler in the binder, as lumps were formed after mixing the two. This can be prevented by first making a gel with the filler and then add the sodium silicate liquid. This process has to be better understood however, as the addition of k-carrageenan to sodium silicate liquid did not result in a gel.

Underextrusion of the solid contents of a paste seems to happen when too little binder is available in the paste. This underextrusion eventually ends in clogging as the resulting paste in the syringe gets thicker and thicker.

### Introduction

In order to design tools for makers/designers to help them develop their own 3D-printable bio-based materials, one has to know how makers operate. Six groups (or twelve subgroups) of 3rd year industrial design engineering students were asked what their way of working was while working with new bio-based materials and to describe the results they obtained at the end of the workshop. This was done after a workshop that was given by Mariet Sauerwein on 3D printing with bio-based materials. In the workshop each subgroup was given one filler material (saw dust in various forms/sand/cacao shells/coffee grounds/dried leaves/egg shells/olive pomace) and alginate as gelling agent. Furthermore the recipe of Mariet's mussel shell-alginate paste was shared.

In order to see how makers (3rd year industrial design engineering students in this case) proceed from here, their process was evaluated afterwards. Furthermore, they were asked to describe the qualities of the paste and the dried printing results to build a vocabulary that works well for describing paste and print samples. Finally, some general notes were taken on their final recipes. This all was done via a semi-structured interview, which can be found in appendix XY.

### interviews

#### Methods used by makers

Generally speaking, the methods used by the students to develop the recipes could be divided into two categories:

1. One of three variables (wt.% of a certain ingredient) was kept constant while one of the other two was gradually increased or decreased. The starting point for the first recipe was the recipe of the mussel shells and alginate. This action was continued until a maximum – of print quality – was reached.
2. One of the three variables was again kept constant while with the ratio of the other variables the extremes around the initial mussel recipe were tested. Where the one extreme was an almost watery paste and the other and almost solid paste. After the first two test, the the average of the two extremes was produced. Then for the next step the favourable side of this middle value was considered, repeating the approach from above.

It was interesting to notice that in general, most groups kept the alginate fraction constant and started playing with the water-filler ratio first. Furthermore, there was no experimentation with anything other than the given (cold) water or with the order of adding ingredients.

#### Vocabulary

For favourable paste consistencies, different wordings were used. Some of the used wordings are: peanut butter texture, wet cookie dough texture. But many others found it hard to describe. Often descriptions were used like: "The spoon would keep standing in it, but it was not completely solid"

For the dried product, most would describe the material as brittle. It was once described with the texture of an old "spice nut" to give an idea of how the material would break into many small crumbs when breaking. Another, slightly tougher material that could yield slightly, was described as a one day old spiced biscuit. Furthermore, breaking the material was described as clean or crumbly.

## Insights recipes

From the overview of the different filler materials a couple of nice insights were gathered. Materials that had cellulose contents seemed to be tougher than for instance sand or eggshell. For eggshell, the larger particle size (0.125 mm) seemed to perform better than the smaller particle size (0.075mm). The resulting print was better stackable and stronger after drying. Also, the coffee grounds appeared to be too large to extrude through a tip, but the resulting dried material appeared very strong. For very voluminous fillers, weight seems to be no good indication for the amount of filler needed in a mix. In these cases a volume measurement is a better indication of the needed amount of filler. Finally, the mixing of a cellulose based filler material with a non-cellulose based material was tried in a couple of groups. It seemed as if these mixes resulted in something that was better than the separate parts. The paste flowed easier through the tip and the shrinkage was smaller than the respective ingredients (pine wood saw dust and sand in this case).

## 3. IDEATION

The input gathered in the analysis phase is used as the starting point for ideation. Firstly, this chapter will summarize the findings and insights from the analysis phase and state the design goal. From there onwards, the idea generation process will be described and the outcomes of this process will be summarized. Finally, the ideas are clustered, assessed and the most fruitful ideas are selected for further development.

### 3.1 IDEATION INPUT

#### Results analysis

In the analysis phase of this project a better understanding of bio-based 3D printable materials, the makerspace community and the platform of Materiom. The main insights and findings can be categorized in x different categories

#### Recipe creation related insights

- Binders that work: Methylcellulose, kappa/iota Carrageenan, alginate
- Fillers are mostly interchangeable:
- Filler volume is more important than filler weight in the recipe description
- Overextrusion of liquids can often be fixed with the addition of binder material

#### Printing setup related insights

- There is a fixed relation between nozzle size/layer height/material flow
- Syringe – nozzle distance has to be minimized
- Translation from hand to machine prints is not straightforward

#### Context related insights

- Use community to accelerate innovation
- Peer to peer communication is key for fablabs

#### Goal/intention related insights

- Balance between guidance and experimentation is needed
- Method should be usable by a wide range of people (some experience with 3D printing is required)
- Makers should be enabled to share more than only finished recipes.

#### Design goal

The insights gathered in the analysis phase have resulted in a design goal for this assignment:

**“Design a digital material development tool that uses the inputs from the maker community to its full extent and encourages makers to experiment, while giving enough guiding to make the barrier to experimentation as low as possible ”**

The tool should be digital, as this is fitting to what Materiom is currently doing and this ensures that the tool can be dynamic and that it can grow and develop together with the insights from the community.

The tool should make maximal use of the insights from the community it is serving. This means that possibly a wider viewpoint must be taken on which kind of data is presented, compared to what Materiom is offering right now.

The tool should encourage experimentation, while being easy to use as well. By making the tool more supportive than prescriptive the outcomes of the experiments of the community will become more varied. This means that a wider range of experiments will be carried out and possibilities for new findings will be greater.

The barrier for using the tool should be as low as possible. Currently Materiom offers very detailed and easy to follow recipes. We do not want to alienate the current Materiom users, so the understandability of the tool should be comparable to the recipes that they currently offer.

## 3.2 INSPIRATION

### Inspiration

During the material experiments, the similarities between cooking and the preparation of material recipes were very clear. They both share a set of ingredients, tools and step-by-step instructions for preparatio. A comparative overview was therefore made of a set of differently designed cooking related books to search for design inspiration, see table 4.



Figure 6 - Jamie's Italy

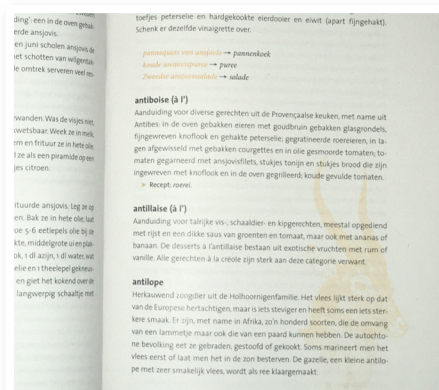


Figure 7 - Larousse Gastronomique

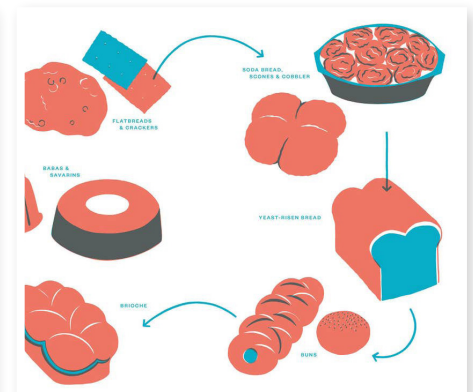


Figure 8 - Lateral Cooking

### Larousse Gastronomique

Larousse gastronomique, see figure 7, is also referred to as the cooking bible. It is a set of books that is formatted like an encyclopaedia. Alphabetically all terminology related to cooking is described. For terms related to recipes, recipes are also included or related recipes are stated. At the end of the alphabetical section a set of menus from high cuisine chefs is presented.

### Lateral Cooking

Lateral cooking, figure 8, is set up differently than the regular cooking book. The book by Nicky Segnit is divided into twelve parts in which the recipes are in some way

interconnected. Within each category a set of fundamental recipes is presented. Below each fundamental recipe variations from around the world are described, for example different flour types, the substitution of flour by other ingredients or additional spices. Then finally, other directions with the recipe are visualized, e.g. from flatbread one can go to quesedillas or roti.

The next fundamental recipe builds further on the latter recipe. The idea is to generate a greater understanding of the interconnection of recipes and to have a basis for an endless amount of recipes described in six fundamentals

Jamie’s Italy

Jamie’s Italy, shown in figure 6, is the most conventional cooking book of the three books described. The element that stands out here is the connection of the recipe to the resources needed to produce the recipe. Furthermore the local community where the recipe stems from is described.

Larousse Gastronomique	Lateral Cooking	Jamie’s Italy
Complete	Exciting	Low key
‘Knowledge creator’	‘Insight creator’	‘Empathy creator’
Precise	Experimental	Impressionist

Table 4 - Guided experimentation through tables and explanatory texts

3.3 IDEA GENERATION

Process

Based on the three types of cooking books described in table XY, three sets of ideas were developed. The cooking books were used as analogies. The three sets of ideas differed on the level of the closedness/openness of the presented solutions.

Idea sets

On the next pages, three directions are shown in increasing amounts of guidance. Guidance has advantages, such as a low entry barrier and a clearer communication/translation of ones test results towards a different user. On the other hand, too much guidance inhibits experimentation and innovation as the users will be repeating what has been done.

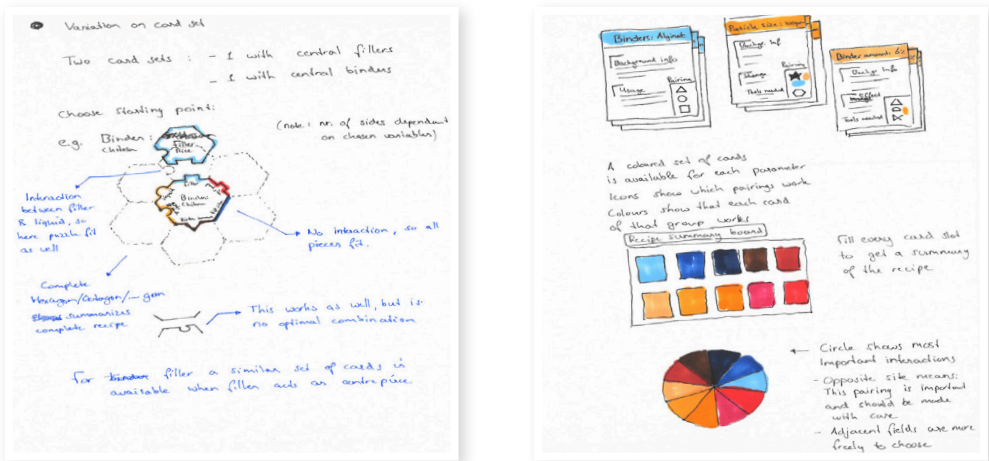


Figure 9 & 10 - Free experimentation - Material combo cards & puzzle pieces

The first set of ideas revolves around the free experimentation, see figure 9 & 10. This is exciting and can be very playful - playing cards or puzzle pieces are giving minimal guidance in material combinations. This set of ideas results in the wildest set of experiments, but can be overwhelming for new users.

### Guided experimentation

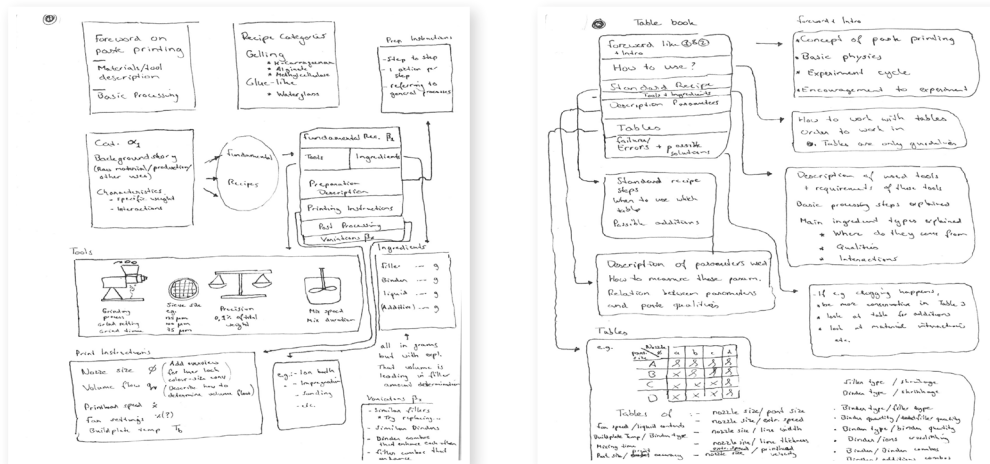


Figure 11 & 12 - Guided experimentation through tables and explanatory texts

The second set of ideas is more oriented towards guidance in building blocks, see figure 11 & 12. The recipe is broken down into different segments and for each segment an explanatory element is created. This can be tables that inform the user on ingredient ratios, explanatory texts on e.g. cross-linking or detailed information on a specific ingredient. It is up to the user to combine the given information and form recipes with it

### Complete guidance

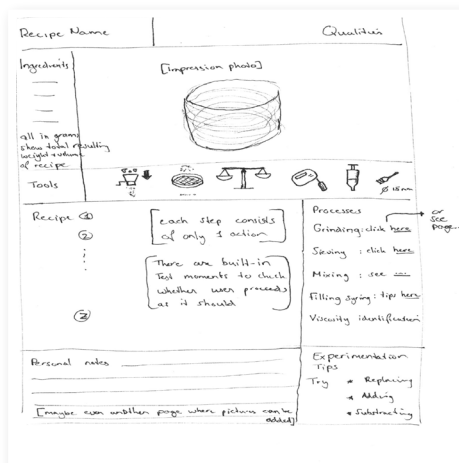


Figure 13 - Complete guidance through a recipe

The final idea direction is the use of complete recipes, see figure 13. This makes the usage of user input more straightforward; recipes collected from the community members can directly be communicated towards the rest of the community. This enables, furthermore, the widest range of people to make use of the material tool as the entry barrier is lowest in this situation.

Apart from the three levels of guidance some thought is also put into the search process. How does the user end up at a certain recipe or set of instructions. Two main options were proposed:

### “Material type” driven searches

The user searches on resources that are for instance locally available or a material group that he/she wants to work with. This makes the search usable when the resource materials are given or limited, but it can be limiting when a certain type of material is desired. For that it might be better to do:

### “Final material qualities” driven searches

The user searches on material qualities. This is the more design-like search. Based on e.g. the desired hand feel of the designer, the search engine presents him/her with a set of materials that comply to that criterium.

Although the selection of final material qualities would be more suitable to many design applications, currently the set of available materials and their application is very limited. Therefore this search option could be an option in the future, but it is not convenient for now.

## Further development

Within the three categories described above, ideas were formed to communicate on how to create bio-based 3D printable pastes. On the next pages, some of these ideas are highlighted. The origin of the idea is described, the design proposal is described and then the idea is informally evaluated whether it is fit to be part of further developments.



## Origin Idea

Tables can be hard to read and often only one of the entries is of interest, especially for binary cases where it is stated whether a combination of a certain filler and binder work together.

## Design Proposal

The user is presented with puzzle pieces that have different forms that communicate whether that specific parameter combination works, see figure 14. Each puzzle piece represents a specific parameter. The fitting of two puzzle pieces communicates the chance of success. A “recipe” is constructed from a set of parameter combos

## Evaluation

The data needed to make such precise predictions is not available yet. At this moment the best we can do is say whether something works or not.

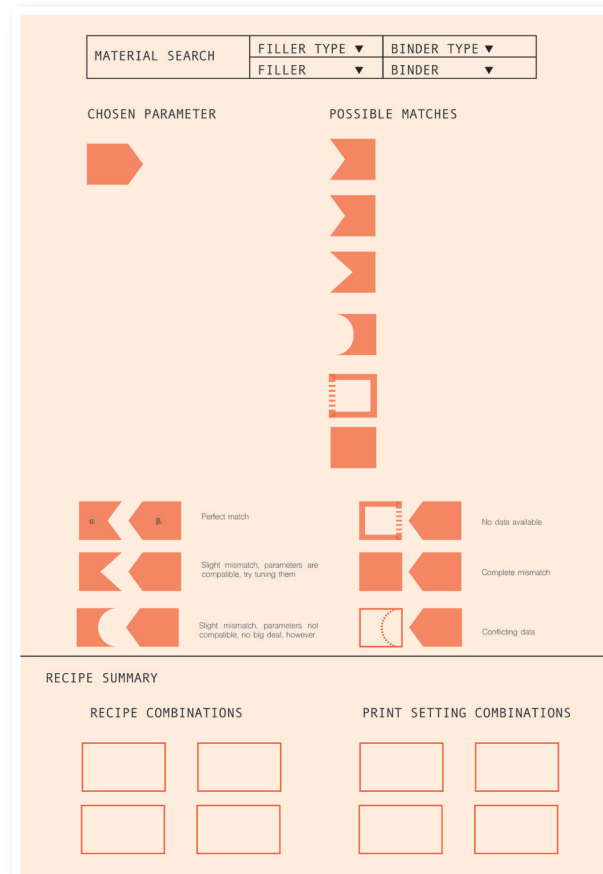


Figure 14 - Material matching through form fitting

## Origin Idea

Modular synthesizers work in such a way that the user gathers the elements that he/she needs (oscillators/effects/etc.) and then connects these elements such that a working instrument is created. What happens when this way of working is translated to the process of creating bio-based 3D printable pastes?

## Design Proposal

Patching modules for parameter combinations. Patching cables can connect different options. Well working options result in a green light (or a positive sound). A recipe can be constructed from connecting different modules, see figure 15.

## Evaluation

The process of trying to patch combinations and then see whether something works can be tedious. A better solution is to show which combinations work when one variable is selected.

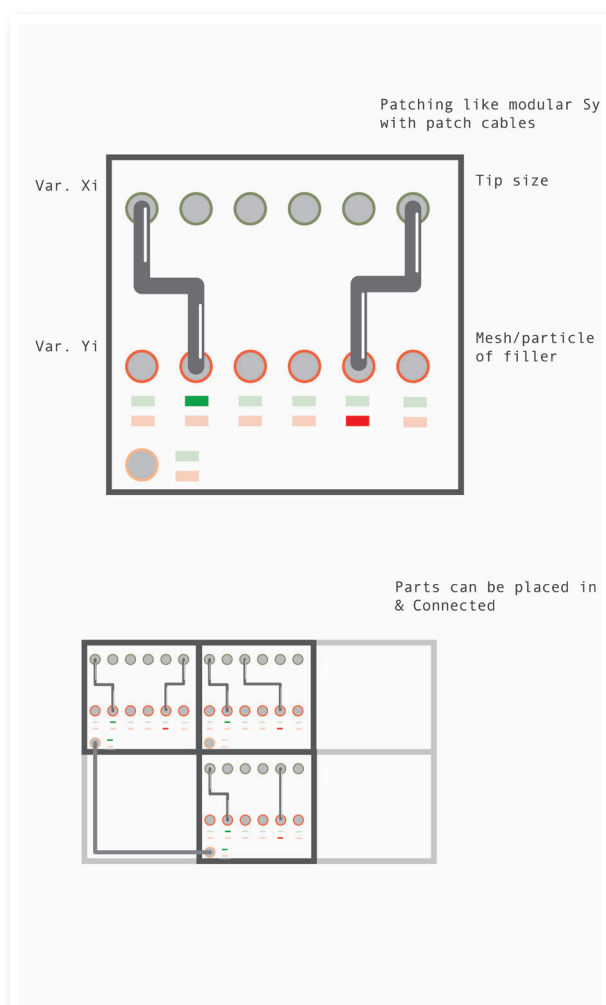


Figure 15 - Material matching inspired by patching in synthesizers

MATERIAL SEARCH	FILLER TYPE ▼	BINDER TYPE ▼
FILLER ▼	BINDER ▼	
<b>RECIPE VARIABLES</b> <div> <div>No Cellulose-based fillers</div> <div></div> <div></div> <div></div> <div></div> </div> <div>Decreasing importance of match</div>		
<b>PRINT QUALITY VARIABLES</b> <div> <div>Tip size: 0.8, 1.0, 1.2, 1.4</div> <div></div> <div></div> <div></div> <div></div> </div>		
<b>RECIPE SUMMARY</b> <div> <div>Binder</div> <div>Filler</div> <div>Carrier</div> <div>Additive 1</div> <div>pH</div> <div>Wt. ratios</div> <div>Tip Size</div> <div>Part. size</div> <div></div> <div></div> </div>		

During the material experiments, the observation was made that some parameter changes (e.g. material quantity/nozzle size/etc.) were much more critical than others. In the big amount of parameters that can be changed during the creation of 1 bio-based 3D print this can be overwhelming. This idea was aimed at the category guided experimentation.

## Design proposal

In order to make the relative importance of the different variables clear to the user, a visual communication style was created that lets the user focus on the parameters that matter. In this way someone dedicates his/her time to the area where it matters most, see figure 16.

## Evaluation

For some parameters it is very clear which is more important than another. This is not the case for all of them, however. This could lead to inconsistencies in the tool. The idea itself can be helpful in guiding users through the process, but must be supported by other elements

Figure 16 - Heat chart mapping importance parameters

## Origin Idea

When one wants to experiment with bio-based materials, some basic understanding of the materials is useful. What has been tried with this material? What are directions that one can take? What problems are to be expected? And how can they be solved?

## Design Proposal

Material sheets of the building blocks of bio-based 3D printing, see figure 17. On the material sheet the key characteristics of a specific material are summarized. Furthermore failures and successes with the material are highlighted. Experimentation tips are presented and in case of problems, a brief trouble shooting sheet is presented. The sheet can also link to existing recipes in the database

## Evaluation

This proposal is close to the way Materiom is communicating right now. There are categories of materials with recipes within that category. The additional information is new and could spark experimentation. The translation from data available to these tips is quite hard, however.

"SODIUM-ALGINATE"																	
<b>Key Characteristics</b> Chemical Name: NaC <sub>4</sub> H <sub>3</sub> O <sub>6</sub> Type: <b>Binder</b> Source: <b>Algae</b> Similar to: X, Y pH: 7-8 Specific weight: a.b g/cm <sup>3</sup> Distribution area: S.-America, E.-Asia, S.-Africa, S.E.-Australia																	
<b>Overview</b> <ul style="list-style-type: none"> <li>Gelling agent: thickens watery solutions</li> <li>Quantities used are mostly 2 - 6 wt.% of the total paste</li> <li>Can be cross-linked in the presence of sodium ions</li> <li>Gel is stable up until 150°C</li> </ul>	<b>Associated Tools</b> 																
<b>Works well with</b> <ul style="list-style-type: none"> <li>Water</li> <li>Cellulose- and Calcium-based fillers</li> <li>Additions like methylcellulose, chitosan, MFC</li> </ul> <a href="#">add your personal findings here</a>	<b>Does not work with</b> <ul style="list-style-type: none"> <li>Alcohol-containing solutions</li> <li>Acidic (pH&lt;5) or basic solutions (pH&gt;11)</li> <li>Additions like waterglass</li> </ul> <a href="#">add your personal findings here</a>																
<b>Experimentation tips</b> <ul style="list-style-type: none"> <li>Adding a second filler from a different material type can improve printing/mechanical performance</li> <li>Agar seems to be an interesting binder to add to the mix for tougher gels</li> <li>...</li> </ul>	<b>Trouble shooting</b> <table border="1"> <thead> <tr> <th>No extrusion?</th> <th>Too much shrinkage?</th> </tr> </thead> <tbody> <tr> <td>Increase amount of gel</td> <td>Decrease water contents</td> </tr> <tr> <td>Check filler particle size</td> <td>...</td> </tr> <tr> <td>Add methylcellulose</td> <td>...</td> </tr> <tr> <td colspan="2">Sagging?</td> </tr> <tr> <td>...</td> <td>...</td> </tr> <tr> <td>...</td> <td>...</td> </tr> <tr> <td>...</td> <td>...</td> </tr> </tbody> </table>	No extrusion?	Too much shrinkage?	Increase amount of gel	Decrease water contents	Check filler particle size	...	Add methylcellulose	...	Sagging?		...	...	...	...	...	...
No extrusion?	Too much shrinkage?																
Increase amount of gel	Decrease water contents																
Check filler particle size	...																
Add methylcellulose	...																
Sagging?																	
...	...																
...	...																
...	...																
<b>Example recipes</b>																	

Figure 17 - Material info sheet

### 3.4 Evaluations & Conclusions Ideation

---

During the ideation many different turns were taken and a variety of ideas were tested that not always seem logical or connected to each other. In this concluding chapter the connection to the next phase is explained.

The ideas that were presented, represented possible solutions on how to communicate data on material development. Currently this is done on Materiom through recipes. Those recipes can be very valuable, as they present a clear and complete overview. The recipes should therefore stay as a part of the material development tool.

Recipes are, however limiting in that they are prescriptive and do not encourage experimentation. Also, in the case that a user of the Materiom wants to share his/her knowledge, recipes might be limiting. Not every experiment leads to a final recipe, but useful information can still be gathered even without that recipe as result. It would therefore be a waste to let these insights be forgotten.

Apart from the recipes, guidance on material combinations will be presented to tackle the disadvantages of recipes. Furthermore, basic material information is useful as well to give the makers a better understanding of the material.

## 4. SCHEMATIC CONCEPT

In this chapter the elements of the tool will be schematically discussed. In the ideation phase it was noticed that recipes by themselves do not fulfill all the goals set in the design goal. Therefore a more differentiated set of data is needed.

### 4.1 SCHEMATIC SITE STRUCTURE

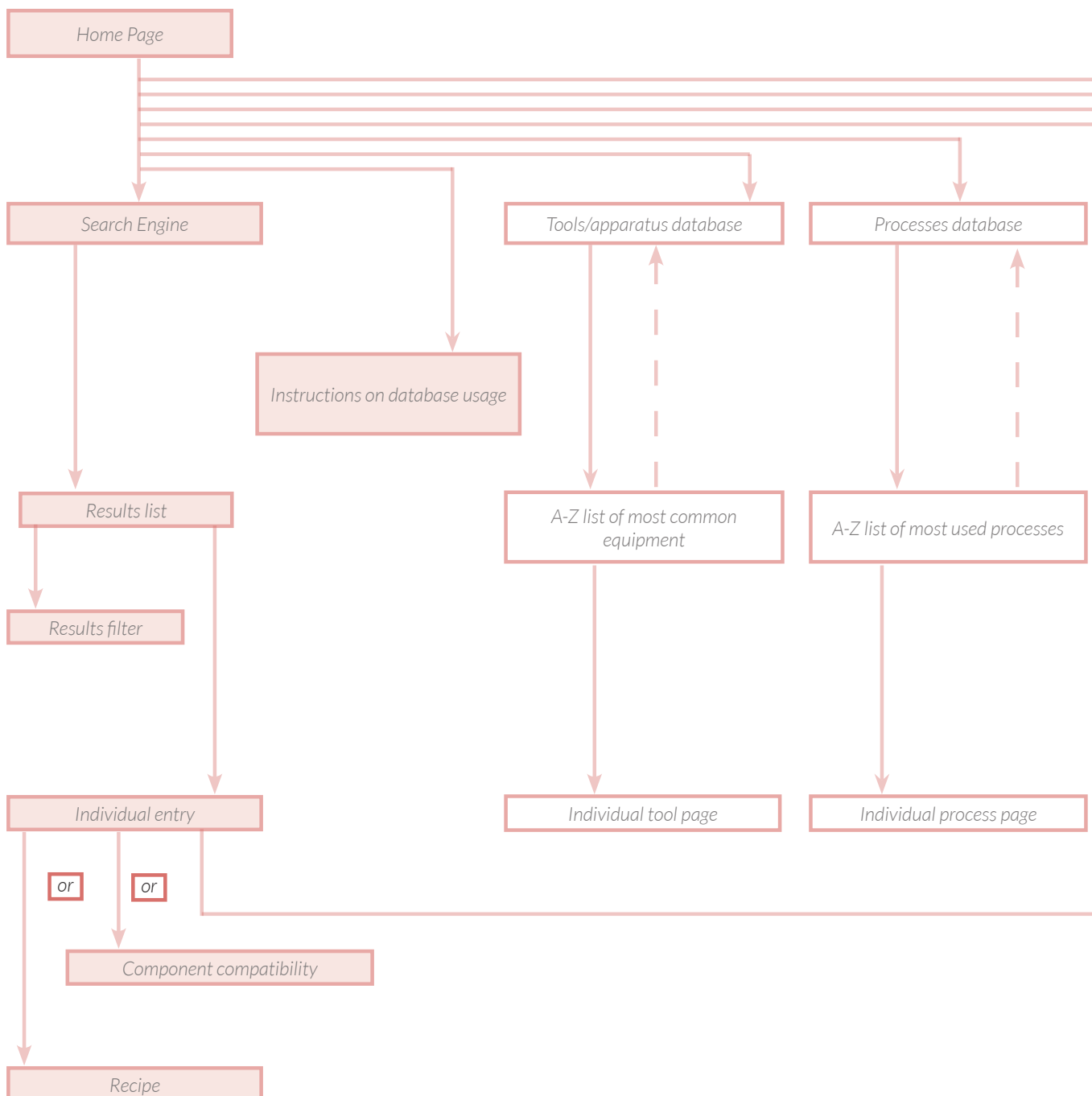
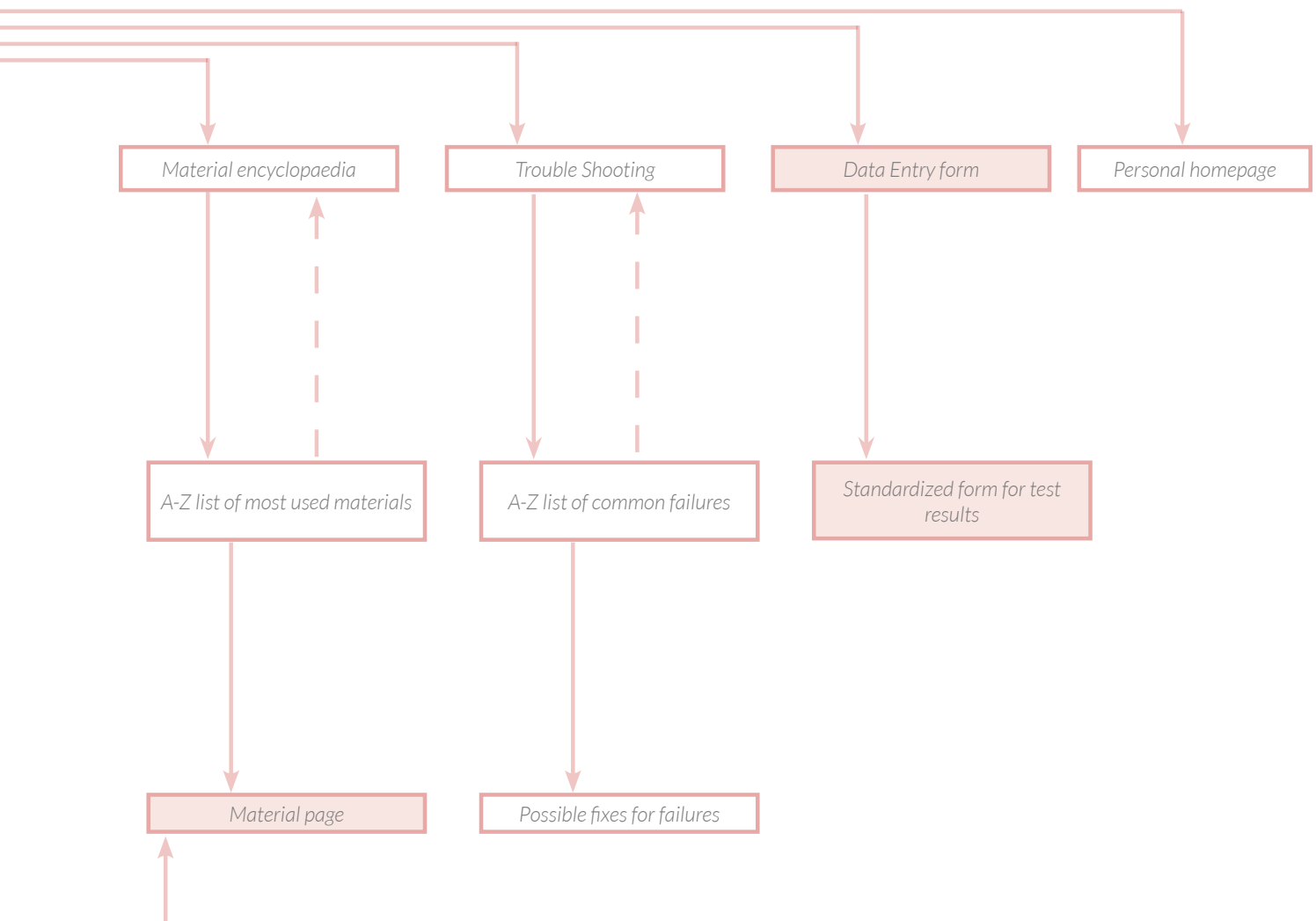


Figure 18 - Schematics of the site architecture

## Schematics material development tool

In figure 18 on these two pages an extended (ideal) schematics is given of the structure of the material development tool. Due to time constraints only the essential elements will be developed in detail. Those elements are shown in the coloured blocks.



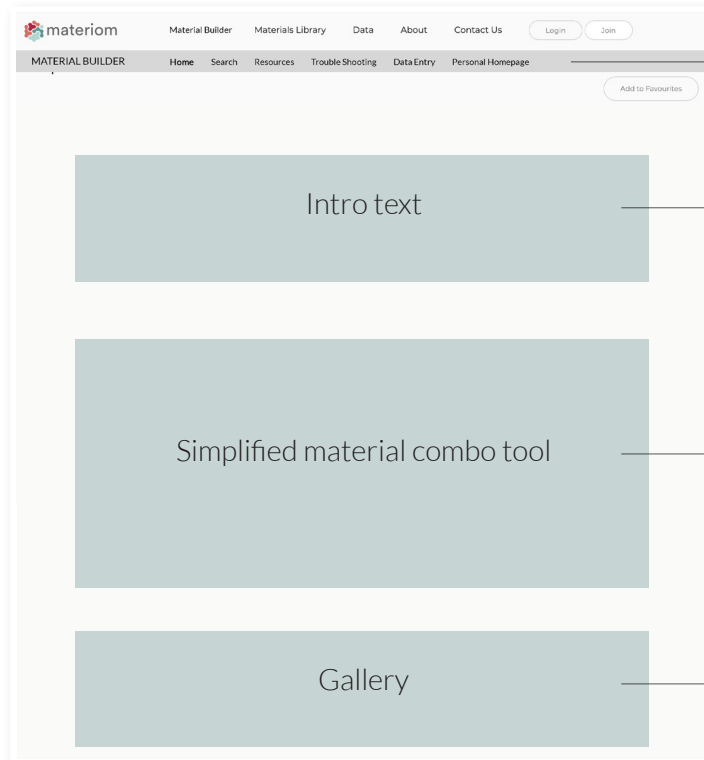
## 4.2 TOOL ELEMENTS

### Introduction

In the website structurization a couple of page types were introduced. In this chapter, the most important pages will be drawn out in a schematic fashion and the page elements are briefly described.

#### Splash page

Shows the content of the online tool. Gives a super short summary of printing with bio-based pastes. It is the inviting starting page, that gives new users the inspiration and most basic explanation to start with developing their own materials & is an easy navigation tool for more experienced users.



Menu to navigate through the different elements of the tool (advanced user).

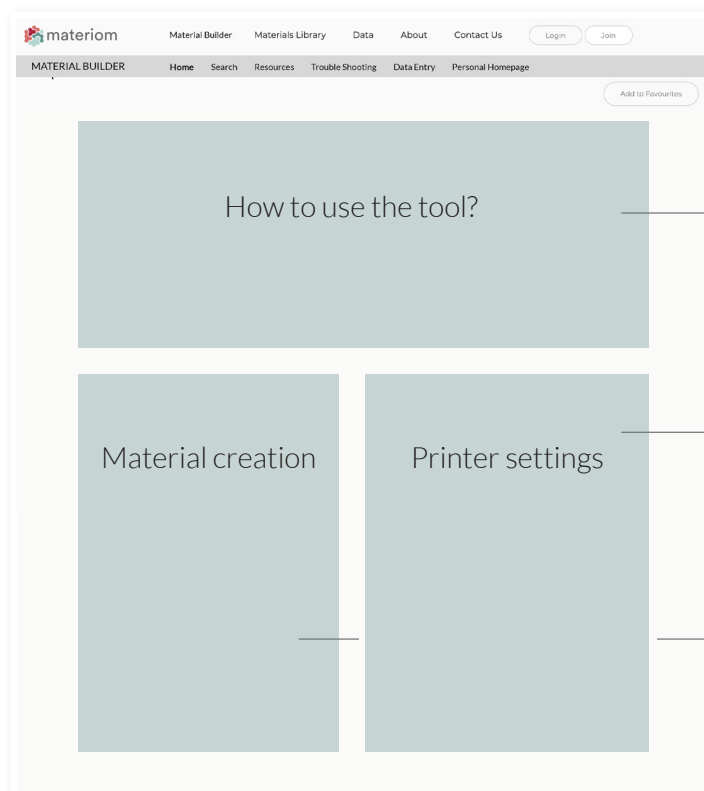
Short explanation of working principles of the tool

Basic version of material combination tool that is understandable for everyone. This is the entry for new users, experienced users can use the more specific tabs in the menu.

Gallery that showcases some of the entries in the database.

#### Instruction page

Gives instruction on how to use the material builder tool. Furthermore it describes in a formulaic way how to create materials and how to set up a 3D printer for printing with the bio-based pastes.



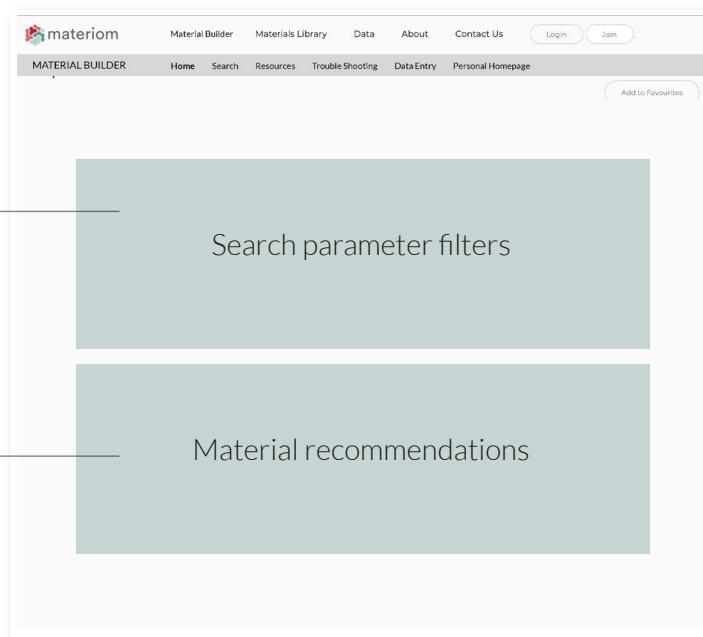
Short explanation of working principles of the tool. How does a beginner start? What is available for more advanced users?

Instructions on how to set up your own 3D printer. Which settings are important? With which values should you start?

Formulaic explanation of the creation of bio-based pastes. This recipe shows the commonalities between different material recipes. It improves the knowledge of the user on how a generic recipe is built up. This makes experimenting an easier task.

The user specifies what he/she is looking for: Material group (e.g. cellulose materials), material type (binder/filler), successfulness of prints etc.

Based on the user activity, new materials / material groups are proposed



## Search Engine

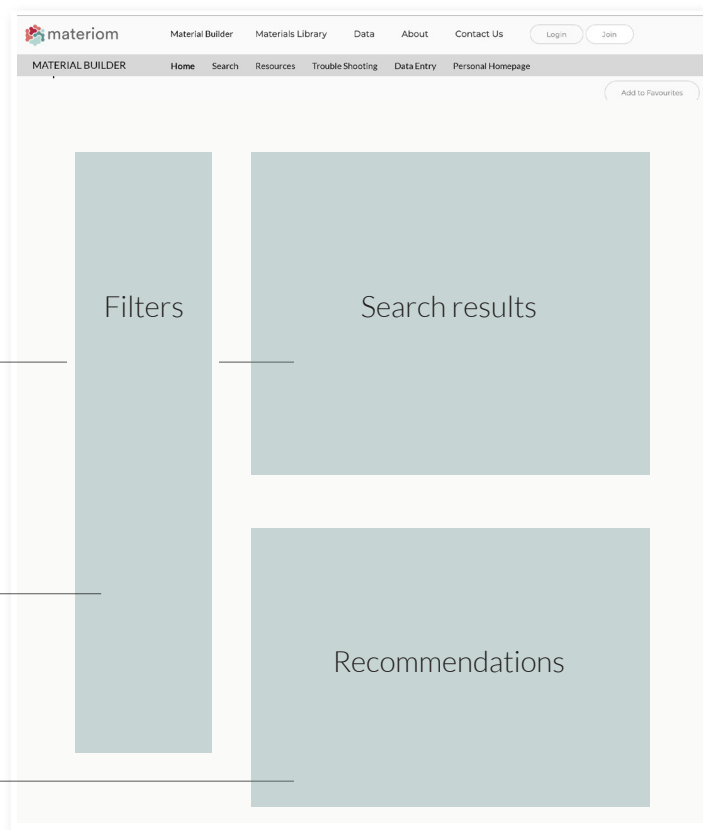
The search engine enables the user to find the recipe (set)/ table/ material group that he or she is looking for.

List of results. Mentioned in the title line:

- o Small picture of result
- o Binder material
- o Filler material(s)
- o Additives (if any)
- o Print succesful or not (+whether hand/machine print was tested)

Search results filter. The list of results can be further decreased by additional filtering options in the side bar. Think of: min-max particle size/ heating needed/rough or soft texture/etc.

Recommendations for additional research

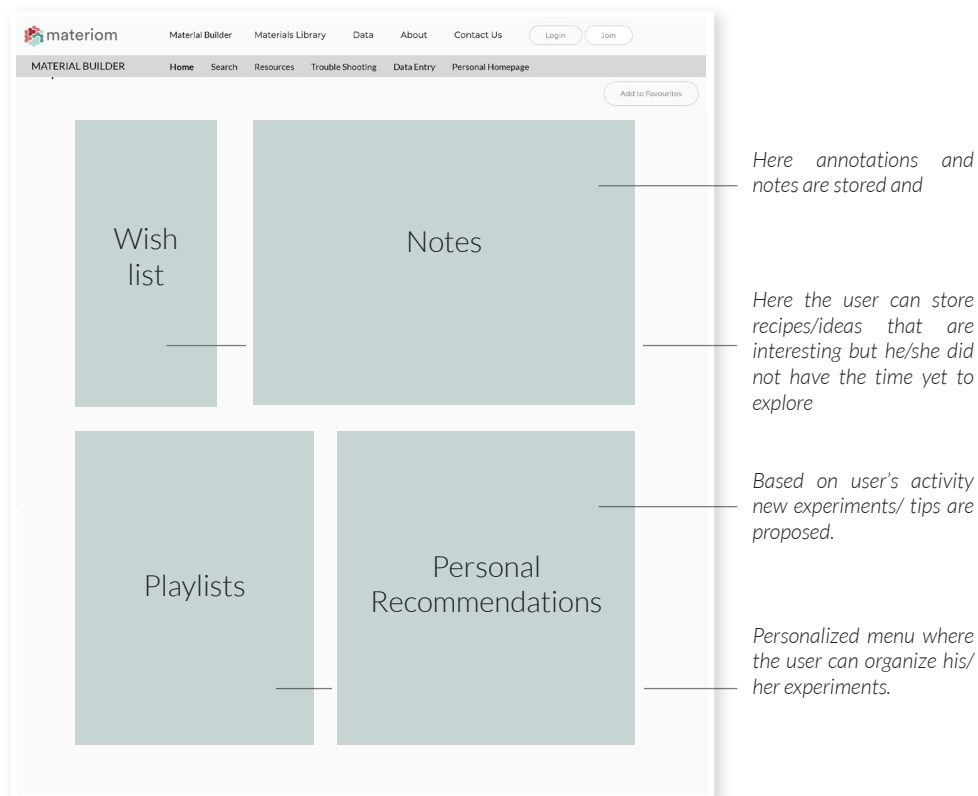


## Search results

Shows all available data that was collected related to the entered search terms. Furthermore, it includes recommendations from earlier researches when those recommendations connect (e.g. material-wise) with the entered search terms.

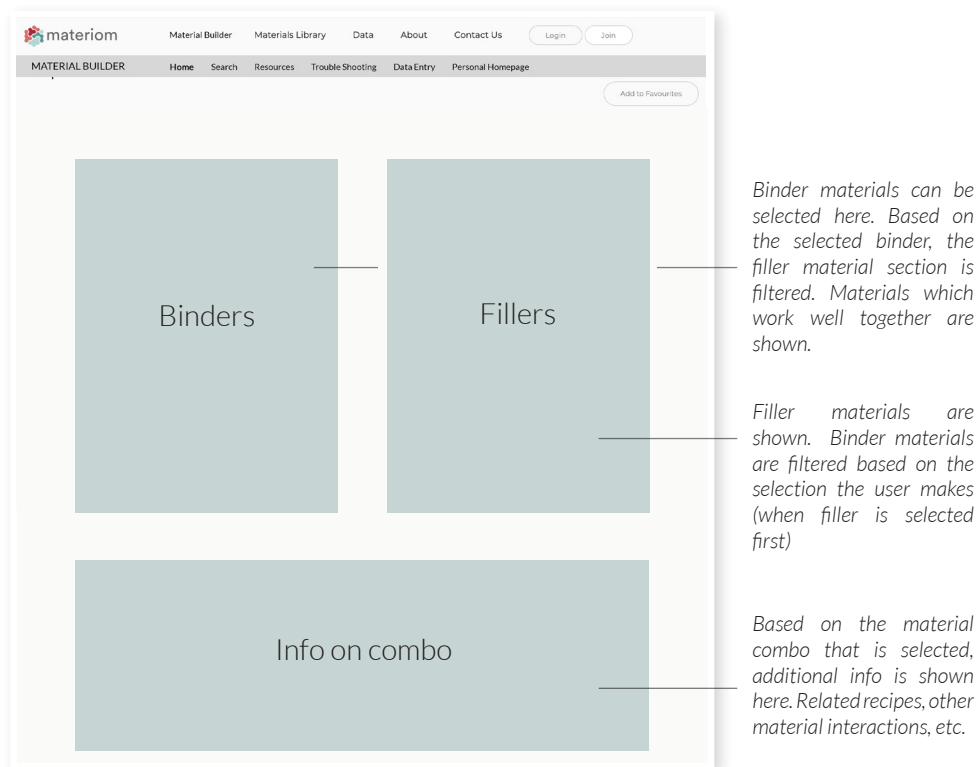
## Personal homepage

This is the user specific environment, where the user can store his/her recipes, to do's etc. Recipes can be stored in "playlists" and new experiments are proposed based on the user's activity. This provides the user with a handy overview of their activities so far and can help to link outcomes of different experiments.



## Component matcher

This gives a visual representation of materials that will work together and which do not. Some material combinations lead to a chemical reaction. This can be favourable, but also unfavourable. Known interactions and their effects are described.





List of ingredients needed for this recipe. All ingredients are listed in grams or in milliliters. Furthermore the material yield of the experiment is listed.

Needed tools and processes to prepare the recipe are listed here.

Step by step description of the steps taken in the recipe. 1 action per step.

Step by step description of the steps taken in the recipe. 1 action per step.

Empty space to collect/ summarize notes on the recipe/experiment

## Recipe

Shows summary of the tools/processes/ ingredients that were used in that specific experiment. Furthermore there is space to add personal notes to the experiment and tips from the former researchers/ designers on what might be interesting to look at.

Empty fields to fill in the binder and filler used in the experiment + other materials (when applicable).

Empty format to fill in the process steps that were executed during the experiment.

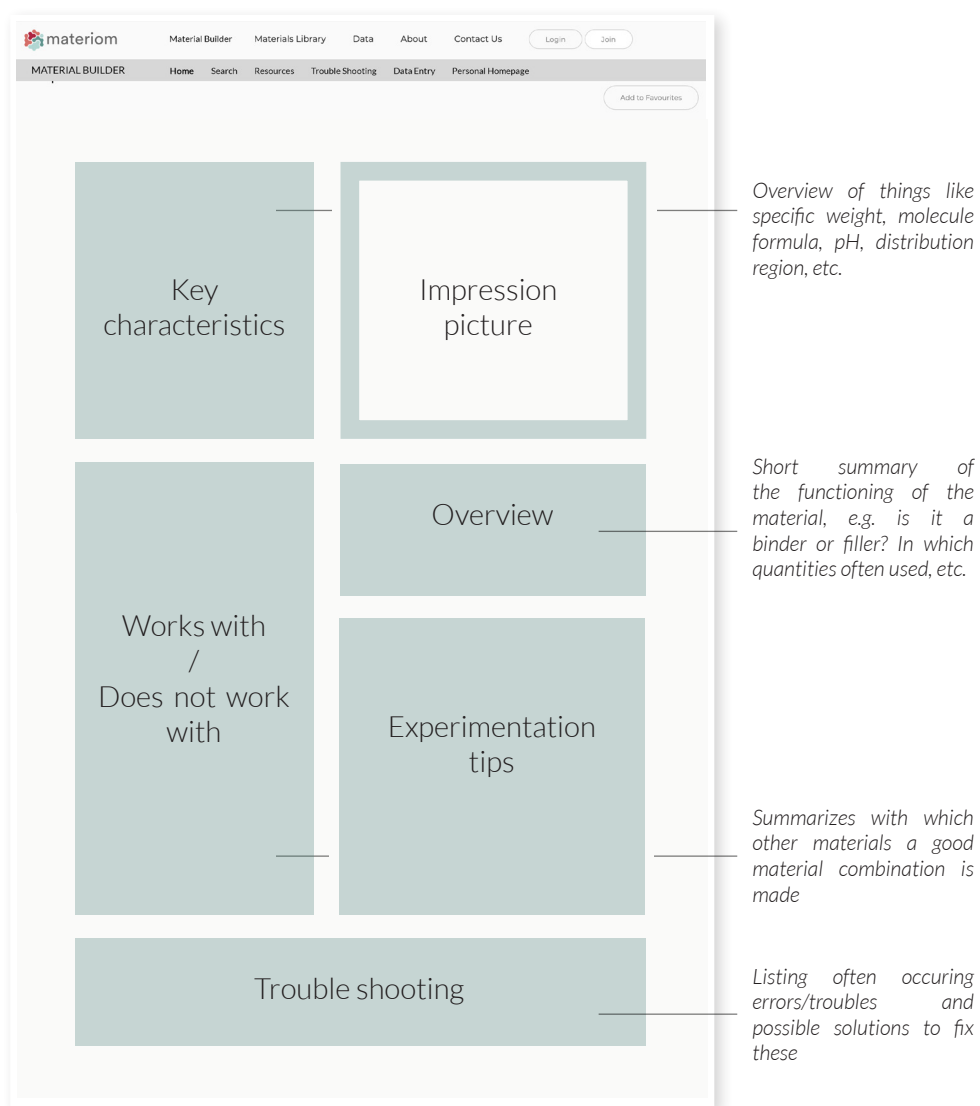
Notes can be placed here to inspire for further experimentation. E.g. in case that the experiments triggered questions or led to presumptions that could not be tested

## Data entry form

The data entry form is the basis for all the input for the digital tool. It briefly asks the researcher/ designer to summarize the process that they have gone through in clear defined questions.

## Material info sheet

Shows the story behind the materials: In which parts of the world can this material be found/ how is it processed/ what are common usages/ which other materials behave similarly





## 5. Final design

On the following pages, the final design is presented. In the accompanying text, the different elements will be explained and the reasoning behind design elements will be clarified.

The final design is the result of iterations of the ideas presented in the last two chapters. The final design revolves around a couple of concepts. The first one is that currently during material development much more data is collected than shared through Materiom. Some experiments are not finalized into complete recipes and some experiments do not yield positive results. By enabling the users of the Materiom platform to also share and search for these data, much more information will become available. Secondly, the transition from hand printing to machine printing is often difficult. A printing guidance page is designed to make this barrier slightly lower. Thirdly, many of the gel-containing recipes have ingredient ratios that follow a similar kind of pattern. On the material creation page, the user is made aware of this pattern to give him an idea for the initial ingredient ratios of his experiment.

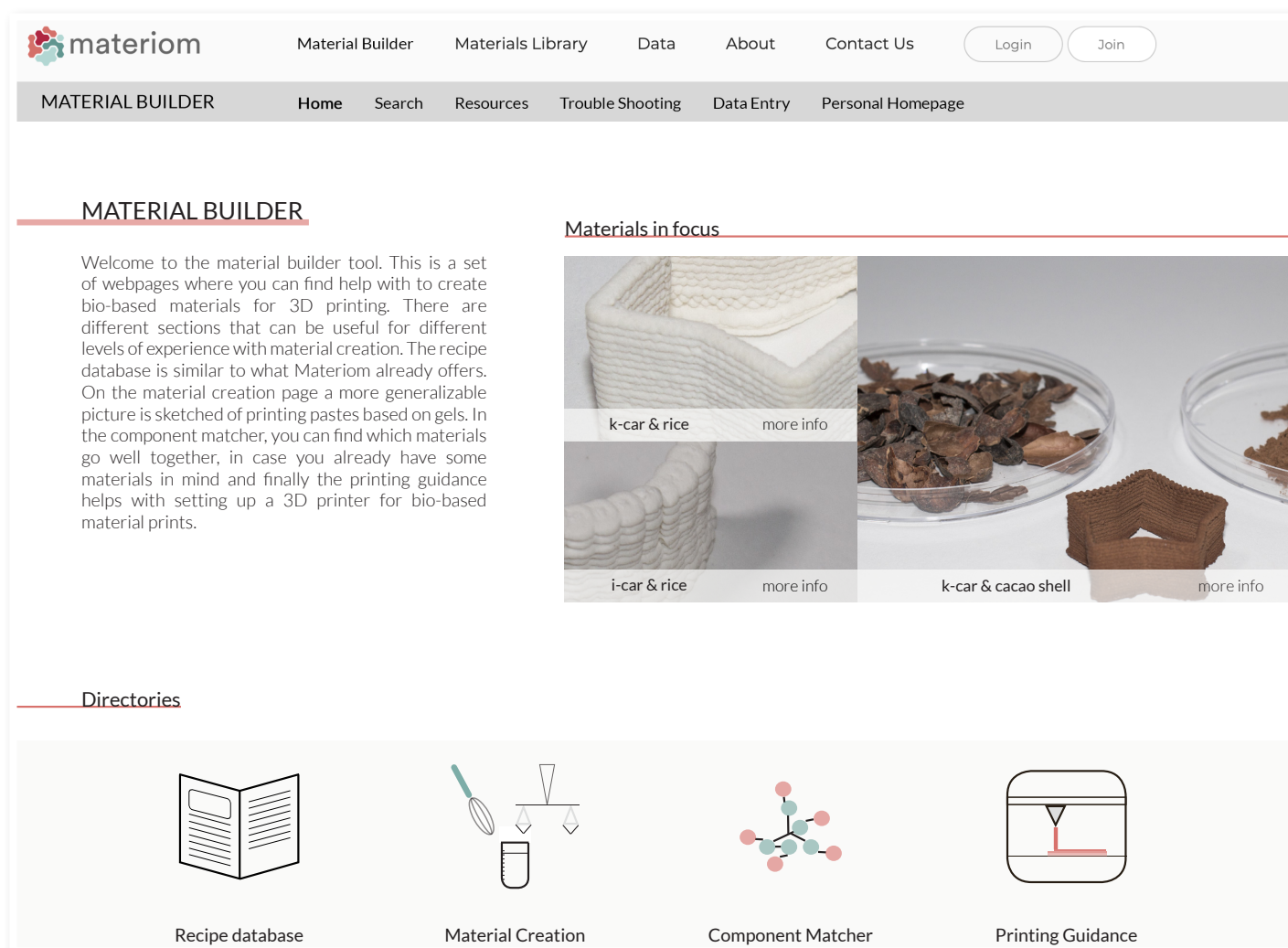
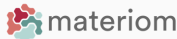


Figure 20- Material Builder Homepage

Intro page

In figure 20 the welcome page is shown. This is the first page one sees when the Material Builder is opened. A short introduction is given on what is to be expected of the tool, three material recipes are highlighted and in the bottom four hyperlinks lead the user to the different elements of the tool.

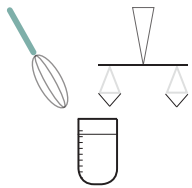
On the webpage shown in figure 21 the user is introduced into the basics of paste creation. Furthermore an formulaic recipe is presented which can function as a good starting point for material ratios. Finally, the component matcher is introduced and a link to the component matcher is shown.


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Login

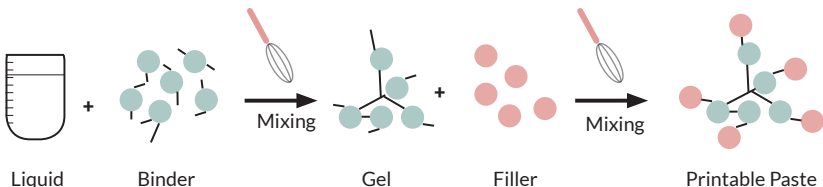
Join

MATERIAL BUILDER
Home
Search
Resources
Trouble Shooting
Data Entry
Personal Homepage



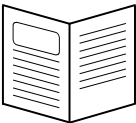
Starting with creating your own bio-based 3D printable materials can be hard. Documentation is hard to come by and few and far between. This page gives an overview of the basic steps that one has to go through to create a printable material. The process is subdivided into three separate sections. The first section discusses in a general manner what a composite paste often consists of in terms of ingredients. The second part discusses some of the starting component ratios and finally, the component matcher is introduced. The interactive component matcher can also be found on a separate page, see the menu at the top of this page.




Material Creation



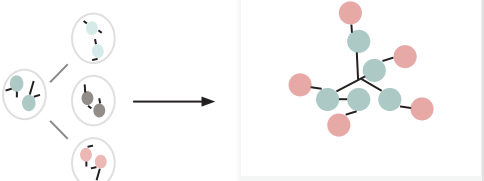
Liquid      Binder      Gel      Filler      Printable Paste

There are many different options to create a printable material, this section focuses on composite pastes that primarily use a gelling agent for the cohesion of the mix. In this case first a gel is prepared from a binder material in combination with a liquid - mostly water. These ingredients are mixed until a homogenous gel is formed. Next, a filler material that is responsible for the structural rigidity after drying is added and again mixed until a homogenous composite paste. Suitable pastes are structured enough to carry their own weight and liquid enough to be extruded through a small nozzle. Ideally their behaviour is opposite of that of a cornstarch bath, such that they flow more easily if a pressure is applied to them.



 = [50-70]wt.%
 = [50-70]wt.%
 = [50-70]wt.%

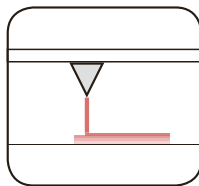
Now the basic process of mixing is understood, it is handy to know about what kind of ingredient ratios we are talking. Generally speaking, around 4-8 wt.% of binder material is used, 40-60 wt.% of filler material is used and the remaining part is water which is something between 30-55 wt.%. These rules however must not be taken too strictly. For very light voluminous powders, like e.g. wood saw dust, the volume is often a better predictor than the weight. In the case of those light powders, a good start is somewhere around the 50 vol.% range.



Ideally components of a composite paste enhance each other's behaviour. But at least components should be compatible. In the component matcher you can search for either binder materials or filler materials and see whether they have been paired up before by other users of Materiom. The component matcher also shows whether the match-up was successful to give you an indication whether your plan might work or to inspire you for material combinations.

Figure 21- Material Creation page

## Printing Guidance



The step from hand printing to printing with a 3D printer is often troublesome. The paste can be changing due to the longer processing time compared to hand printing. Furthermore, the

layer height, print speed, nozzle size and flow settings must be spot on to prevent the layers to become disconnected due to too little material flow or bloated due to material overflow. This page

helps with getting initial values for these settings and shows how the different values are correlated. Finally a

### mass flow rate

The volume flow rate is a setting in the printer setup. Sometimes it can be controlled directly with a numerical input, but it can also be a function of pressure or an unknown value entered through a pot meter. In those last two cases it is possible to measure the volume flow by letting the machine extrude for a minute and calculate the corresponding average flow per second.

### print head speed

The print head speed is a setting that can be found/set in the slicing software that is used to prepare a file that can be read by the 3D printer. It can be the case that the software automatically generates a value, overwrite this function and start somewhere in the range of 6 mm/s for a tip size of 1.5 mm

### nozzle size

Depending on the printer setup the nozzle size can be selected from the nozzles that come with the printer or, in case of using syringes, luer lock nozzles can be used. For a luer lock size conversion, see below.

### layer height

Like the print head speed the, the layer height can be chosen in the slicing software. As a general rule of thumb one can say: layer height = 0.8 • nozzle size



Figure 22- Printing guidance

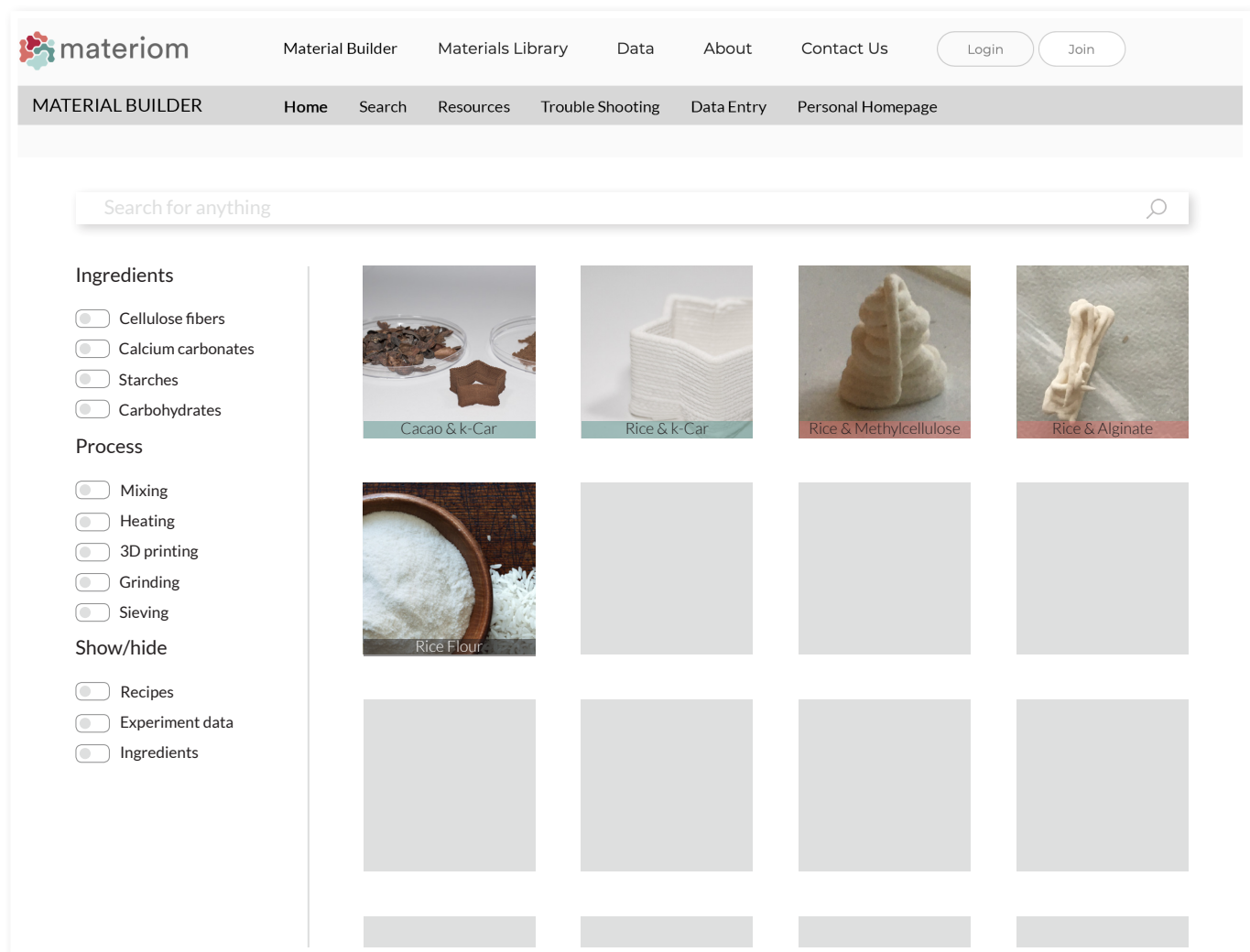


Figure 23- Search page

### Intro page

In figure 22 the printing guidance page is shown. The different variables that are tunable in each printer setup are described on this page. The relationship between these different variables is explained and initial values are presented. At the bottom, a small calculator is placed which calculates the missing variable when the other three variables are chosen.

### Search page

On the webpage shown in figure 23 the search page is shown for the database. The user can search for key words and/or filter the results on material type, processes used and the type of data that he/she is looking for. The recipes, experiment data and material data are differentiated with the coloured name labels.



### Ingredients

Binder	:	<b>k-Carrageenan</b>	0.5 g
Filler	:	<b>Rice flour</b>	8.0 g
Liquid	:	<b>Water</b>	12.0 g

### Yield

25 mL

20 g

### Collection

**Seaweed recipes**

### Processing

**Mixing, 3D printing**

### Difficulty



### Equipment



### Experimentation tips

Try a potassium solution instead of water: k-Carrageenan cross-links with potassium.

Rice flour can be exchanged for many other types of starches. Try for instance potato starch

If using another filler: Match the filler quantities on volume

### Preparation


1. Weigh the rice flour, water and k-Carrageenan and keep them separate
2. Add the k-Carrageenan to the water under continuous stirring continue stirring until no lobes of k-Carrageenan are visible any more
3. Add the rice flour and mix until you reach a smooth consistency
4. Prepare your printer set up, click for tips & tricks here

### Personal notes

Add personal experimentation notes here, e.g.:

Paste can be thickening sometimes. Experiment in this case with higher quantities of binder material (k-Carrageenan). Machine extrusion makes this problem more apparent.





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MATERIAL BUILDER

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Material type

☒ All
☐ Binders
☐ Fillers
☐ Multi-purpose materials

Data type

☒ All
☐ Full recipes
☐ Experimental results
☐ Material basics





Selected material	Compatible materials	Material type	Data completeness	Links to data
kappa-Carrageenan 		Filler	● ● ●	Recipes <a href="#">①</a> Experiment results <a href="#">①</a> <a href="#">②</a> <a href="#">③</a> Material basics <a href="#">①</a>
		Filler	● ●	Recipes Experiment results <a href="#">①</a> Material basics <a href="#">①</a>
		Filler	● ● ●	Recipes <a href="#">①</a> Experiment results <a href="#">①</a> <a href="#">②</a> Material basics <a href="#">①</a>


Figure 25- Material Matcher, pictures obtained from the internet, see reference 1), 2), 3)

## Recipe page

In figure 24 an example of a material recipe is shown. Ingredients, the material yield and difficulty of the recipe are shown. Furthermore an overview of the needed tools is given, together with preparation instructions. Except for the yield of the recipe and the needed tools, nothing is new here. The addition of experimentation tips and the possibility to save the recipe with personal notes are new additions.

## Material matcher

On the webpage shown in figure 25 the material matcher is shown. Materials can be found through the same kind of search function as the database search bar. Once a material is selected, compatible materials are shown. These compatible materials are divided into three different categories, based on the data that is available: Full recipe data, experiment results and material basics. Links to the data entries are shown to view the data into more detail.



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## SODIUM ALGINATE

### Key Characteristics


Chemical Name	pH
$\text{NaC}_6\text{H}_7\text{O}_6$	7-8
Type	Specific weight
Binder	a.b g/cm <sup>3</sup>
Source	Distribution area
Algae	South-America, East-Asia, South-Africa, South-East-Australia



### Material behaviour

Gelling agent	thickens watery solutions
2 - 6 wt. %	is mostly used of the total paste weight
Cross-linking	in the presence of sodium ions
150°C	maximum temperature upon which gel is stable

### Is found in these recipes



### Works well with

- Water
- Cellulose- and Calcium-based fillers
- Additions like metylcellulose, chitosan, MFC

### ~~Does not work with~~

- Alcohol-containing solutions
- Acidic (pH<5) or basic solutions (pH>11)
- Additions like waterglass

### Experimentation tips

Adding a second filler from a different material type can improve printing/mechanical performance

Agar seems to be an interesting binder to add to the mix for tougher gels

As chitosan is the only negatively charged polysaccharide. It possibly can form good bonds with alginate

Figure 26- Material Info Page

## Material page

In figure 26 the welcome page is shown. This is the first page one sees when the Material Builder is opened. A short introduction is given on what is to be expected of the tool, three material recipes are highlighted and in the bottom four hyperlinks lead the user to the different elements of the tool.

## Data entry form

On the webpage shown in figure 27 the user is introduced into the basics of paste creation. Furthermore an formulaic recipe is presented which can function as a good starting point for material ratios. Finally, the component matcher is introduced and a link to the component matcher is shown.

To help your fellow material developers, we need your data. This sheet is specifically meant to be filled in for 3D printable composite pastes. You can help us by filling in some data that you gathered during your experiments.

Add as many different ratio test as you like, but use one form only for one specific material combination. Furthermore, tell us a bit about the processes and equipment used. Finally, if during your experiments you felt like there might be some

nice pairings with other materials or you did not have the time/means to perform an experiment ideas, please share. This enables other material developers to have a headstart with experimenting. We look forward to your input!

### Materials used

Material 1

Material 3

Material 2

+ add more materials

### Material ratios

Test 1

Success: Y ☐ N ☐

Material 1

 g

Material 2

 g

Material 3

 g  
mL  
%

Test 2

Success: Y ☐ N ☐

Material 1

 g  
mL  
%

Material 2

 mL

+ add more tests

### Equipment used

Tool 1

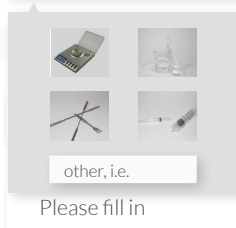
Tool 3

Tool 2

+ add more

### Equipment used

Process 1



other, i.e.

Process 2

Process 3

Ground  
Mixed  
Heated

### Ideas on experimentation

---

---

---

---

---

Figure 27- Data Entry Form

## 6. Evaluation

In this chapter a brief evaluation is made of the results that were obtained during this project. It will be checked whether the design fit the design goal theoretically and limitations will be discussed.

### 6.1 Fit with design goal

In Chapter 3.1 the design goal was described as: “Design a digital material development tool that uses the inputs from the maker community to its full extent and encourages makers to experiment, while giving enough guiding to make the barrier to experimentation as low as possible.” The final design fit to this goal in the following ways:

Data gathered by the maker community is used as much as possible. Not only full recipes are collected, but also experiment results that might not have a completely positive outcome

Experimentation is encouraged & the barrier for experimentation is kept as low as possible through the material creation page, the printing guidelines, and the fact that not only full recipes are presented but also material combinations. The material creation page in combination with the printing guidelines give the user initial values to start with for printing and material parameters, while experimentation tips on recipe pages encourage the users.

Due to the corona virus a user tests was sadly not performed to check whether these assumptions also held in the real world practice of material experimentation

### 6.2 Recommendations

As mentioned, no user test was conducted with the final design. This should first be done before any of the elements can be implemented.

Furthermore, part of the design is to collect and communicate much more data with the community than that is currently done. Smart data collection and labeling algorithms should be selected to make this data useful for the maker. The data entry forms might have to be revised to fit to these algorithms.

The success of a system such as proposed depends on the amount of data that is available. Currently, very little is known about printing with bio-based pastes. This means that the material development tool currently asks more from the user than that it can give back. Therefore, it should be high priority to collect a collection of experiment results on 3D printing with pastes.



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2.) Picture retrieved from <https://www.google.com/url?sa=i&url=https%3A%2F%2Fibake.com.au%2Frice-flour-5kg%2F&psig=AOvVaw0vYf4He4JuTcBz-ilvfDCJ&ust=1590106441049000&source=images&cd=vfe&ved=0CAIQjRxqFwoTCLC2j6DWw-kCFQAAAAAdAAAAABAG>

3.) Picture retrieved from <https://www.google.com/url?sa=i&url=https%3A%2F%2Fnz.candlesupplies.nz%2Fproducts%2Fwalnut-shell-powder&psig=AOvVaw2oqtfn17PSqEI3ivjwRqFN&ust=1590106484521000&source=images&cd=vfe&ved=0CAIQjRxqFwoTCKCNI7XWw-kCFQAAAAAdAAAAABAD>



### **Open source and biobased 3D printable materials for the circular economy**

Are you an IPD student and excited about biobased materials? Do you find the makerspace community fascinating? And do you have an interest in 3D printing? Join us in the exciting exploration of open source and biobased materials!

This graduation assignment is in collaboration with Materiom. Materiom is an online platform that provides open data on how to make materials sourced from locally abundant biomass for the circular economy. The platform provides the opportunity to upload recipes to share with makerspaces and the general community.

In this assignment, we ask you to explore and develop an approach that will help people (makers) to develop biobased materials into 3D printable materials. What kind of understanding is needed? What steps should be taken?

The starting point will be the current recipes on Materiom. Most of them are not designed for 3D printing, but can you develop them into a 3D printable material? If so, how? If not, why not? The approach has to operate in a makerspace and thus be understandable for the makerspace community, relying on common resources.

This assignment provides you the opportunity to explore biobased materials and generate an understanding into 3D printable material, as well as to collaborate within the network of the Materiom platform, for example with Fab @ CIC Boston and the Universidad de Santiago de Chile .

If you are interested, please contact:  
Mariet Sauerwein: [m.sauerwein@tudelft.nl](mailto:m.sauerwein@tudelft.nl)



# IDE Master Graduation

## Project team, Procedural checks and personal Project brief

This document contains the agreements made between student and supervisory team about the student's IDE Master Graduation Project. This document can also include the involvement of an external organisation, however, it does not cover any legal employment relationship that the student and the client (might) agree upon. Next to that, this document facilitates the required procedural checks. In this document:

- The student defines the team, what he/she is going to do/deliver and how that will come about.
- SSC E&SA (Shared Service Center, Education & Student Affairs) reports on the student's registration and study progress.
- IDE's Board of Examiners confirms if the student is allowed to start the Graduation Project.

### ! USE ADOBE ACROBAT READER TO OPEN, EDIT AND SAVE THIS DOCUMENT

Download again and reopen in case you tried other software, such as Preview (Mac) or a webbrowser.

### STUDENT DATA & MASTER PROGRAMME

Save this form according to the format "IDE Master Graduation Project Brief\_familyname\_firstname\_studentnumber\_dd-mm-yyyy". Complete all blue parts of the form and include the approved Project Brief in your Graduation Report as Appendix 1 !



family name Bremmer  
initials A.M. given name Matthijs  
student number 4261984  
street & no. Goudsesingel 242 B  
zipcode & city 3011 KE, Rotterdam  
country The Netherlands  
phone +31640854105  
email matthijsbremmer@live.nl

Your master programme (only select the options that apply to you):

IDE master(s): ☒ IPD ☐ Dfl ☐ SPD

2<sup>nd</sup> non-IDE master: \_\_\_\_\_

individual programme: \_\_\_\_\_ (give date of approval)

honours programme: ☐ Honours Programme Master

specialisation / annotation: ☐ Medesign

☐ Tech. in Sustainable Design

☐ Entrepreneurship

### SUPERVISORY TEAM \*\*

Fill in the required data for the supervisory team members. Please check the instructions on the right !

\*\* chair Prof. dr. ir. C.A. Bakker dept. / section: Design f. Sustainability  
\*\* mentor PhD J. Faludi dept. / section: Design Engineering  
2<sup>nd</sup> mentor \_\_\_\_\_  
organisation: \_\_\_\_\_  
city: \_\_\_\_\_ country: \_\_\_\_\_

comments  
(optional)

⋮

Chair should request the IDE Board of Examiners for approval of a non-IDE mentor, including a motivation letter and c.v..



Second mentor only applies in case the assignment is hosted by an external organisation.



Ensure a heterogeneous team. In case you wish to include two team members from the same section, please explain why.

**APPROVAL PROJECT BRIEF**

To be filled in by the chair of the supervisory team.

chair Prof. dr. ir. C.A. Bakker date 24 - 09 - 2019

signature

**Conny  
Bakker  
r - IO**

 Digitally  
signed by  
Conny  
Bakker - IO  
Date:  
2019.09.24  
21:23:56  
+0200
**CHECK STUDY PROGRESS**
 To be filled in by the SSC E&SA (Shared Service Center, Education & Student Affairs), after approval of the project brief by the Chair.  
 The study progress will be checked for a 2nd time just before the green light meeting.

Master electives no. of EC accumulated in total: \_\_\_\_\_ EC

 Of which, taking the conditional requirements  
 into account, can be part of the exam programme \_\_\_\_\_ EC

 List of electives obtained before the third  
 semester without approval of the BoE

☒ **YES** all 1<sup>st</sup> year master courses passed

☐ **NO** missing 1<sup>st</sup> year master courses are:

name \_\_\_\_\_ date \_\_\_\_ - \_\_\_\_ - \_\_\_\_ signature \_\_\_\_\_

**FORMAL APPROVAL GRADUATION PROJECT**
 To be filled in by the Board of Examiners of IDE TU Delft. Please check the supervisory team and study the parts of the brief marked \*\*.  
 Next, please assess, (dis)approve and sign this Project Brief, by using the criteria below.

- Does the project fit within the (MSc)-programme of the student (taking into account, if described, the activities done next to the obligatory MSc specific courses)?
- Is the level of the project challenging enough for a MSc IDE graduating student?
- Is the project expected to be doable within 100 working days/20 weeks?
- Does the composition of the supervisory team comply with the regulations and fit the assignment?

 Content: ☐ **APPROVED** ☐ **NOT APPROVED**

 Procedure: ☐ **APPROVED** ☐ **NOT APPROVED**


comments

name \_\_\_\_\_ date \_\_\_\_ - \_\_\_\_ - \_\_\_\_ signature \_\_\_\_\_

## Open source & bio-based 3D printable materials for the circular economy project title

Please state the title of your graduation project (above) and the start date and end date (below). Keep the title compact and simple. Do not use abbreviations. The remainder of this document allows you to define and clarify your graduation project.

start date 16 - 09 - 2019

20 - 02 - 2020

end date

### INTRODUCTION \*\*

Please describe, the context of your project, and address the main stakeholders (interests) within this context in a concise yet complete manner. Who are involved, what do they value and how do they currently operate within the given context? What are the main opportunities and limitations you are currently aware of (cultural- and social norms, resources (time, money,...), technology, ...).

In our current society oil reserves are declining, sustainability becomes more important and the size and impact of plastic pollution is more and more understood (Eriksen et al., 2014). Therefore, the use and development of alternatives for oil-based plastics is very relevant (Wool & Sun, 2011). New and more sustainable materials for state of the art production are needed. Bio-based materials are a promising contender to (partially) fill the spot of oil-based plastics (Álvarez-Chávez et al., 2012), but at this moment still a lot of innovation is needed. This holds especially true for the use of bio-based materials in additive manufacturing (AM) practices. In AM there are still big innovations needed regarding print quality and (structural) strength for bio-based materials to be able to compete with e.g. ABS (Faludi et al., 2019).

Materiom is an open-source database for bio-based material recipes that wants to accelerate the development in this area. They want to achieve this by lowering barriers in the new materials market and give communities the means to nourish themselves. To achieve this goal they cooperate with makerspaces, companies and local communities and support them in the development of new bio-based materials based on the characteristics of the natural resources that are available in the respective regions where those communities operate in. Their open-source database makes it very easy to make the materials yourself by following recipes (see figure 1.). By enabling makerspaces to develop bio-based materials based on the local natural resources, local communities can be nourished and local production can be stimulated ("Imagine if", 2019). Local production is seen as a sustainable alternative for centralized production (Ford & Despeisse, 2016) and provides the opportunity to close the loop in a circular economy by linking local resource streams to local production and recycling (Sauerwein et al., 2018).

By providing makerspaces and designer communities with the tools to make their own recipes for 3D-printable bio-based materials, it will become easier for those makerspaces and other communities to experiment with the resources that are abundant locally. The likeliness of finding good alternatives for the materials currently in use will rise with the amount of experimentation and the accessibility to do so even with relatively small means/knowledge. Furthermore, this way each community can have a local solution for material streams that are characteristic for their specific environment. But most importantly for sustainability is the development of 3D-printable materials that can bond at room temperature. Multiple researchers (Böckin et al., 2019; Cerdas et al., 2017; Faludi et al., 2015) have shown that print energy consumption is the crucial factor for the sustainability of 3D printing. Print energy can be drastically reduced by using printing methods that do not use the principles of thermoplastics or any heat treatment after printing. Due to vast diversity of makerspaces and communities around the world, both in culture and in local resources, thought has to be put into how all those different communities can be effectively reached.

space available for images / figures on next page

## Personal Project Brief - IDE Master Graduation

introduction (continued): space for images

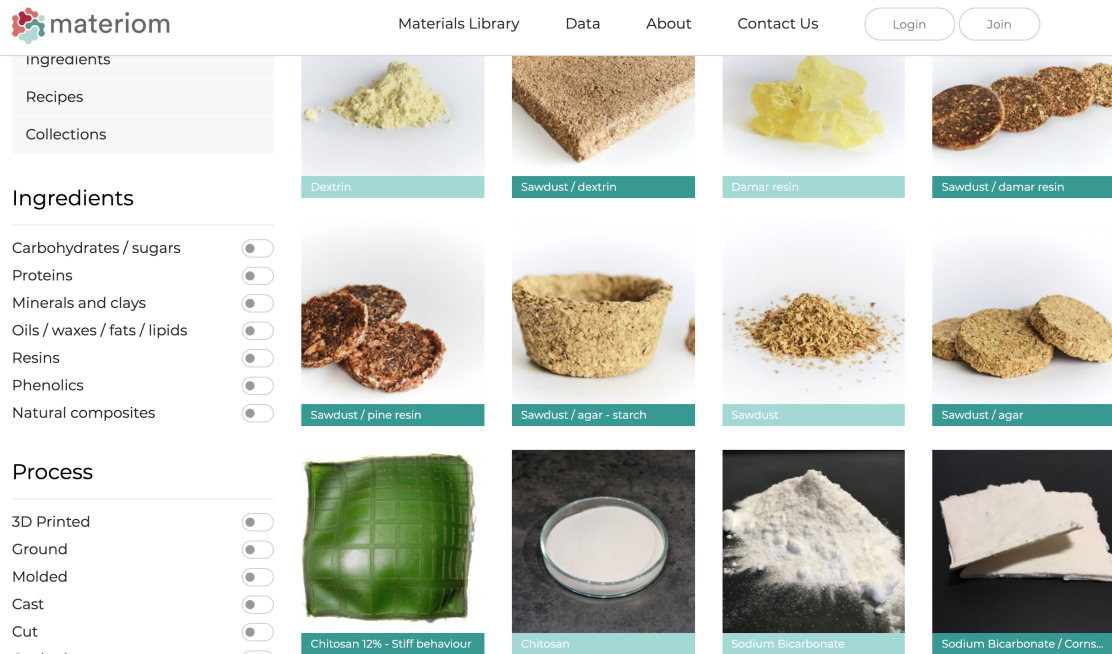


image / figure 1: Open source database of material recipes

image / figure 2: \_\_\_\_\_

**PROBLEM DEFINITION \*\***

Limit and define the scope and solution space of your project to one that is manageable within one Master Graduation Project of 30 EC (= 20 full time weeks or 100 working days) and clearly indicate what issue(s) should be addressed in this project.

Currently bio-based material recipes for 3D printing are relatively scarce. The database of Materiom for example consists only of two variations of a single recipe for the use of ground mussel shells as the basis for an extrusion paste ("Materials Library", 2019). In literature some more recipes are to be found (Rael & San Fratello, 2018; Faludi et al., 2019), but they are far from widely accessible to makers. At the moment, structural strength, printability and print quality of bio-based 3D printable materials are often inferior compared to ABS as Faludi et al. (2019) have shown. But maybe more importantly, the range of materials for 3D printing is rather small. A wider range of materials for 3D printing is therefore desirable.

In this project, it will be investigated what is needed for relatively layman makers in makerspaces to develop new materials for printing by non-toxic paste extrusion at room temperature. What are the problems that have to be tackled when developing a new 3D printable material? In order to achieve the best quality prints as possible, predictors for print quality will be researched. The influence of particle size, binder medium and general rheological characteristics of the extrusion paste on the print quality are investigated. Finally, it is researched what these characteristics mean in a makerspace environment. What are methods and tools that are easily applicable in relatively low-tech environments that help to establish high quality printing pastes?

In order to ensure that the new materials are more sustainable than the materials currently in use, paste extrusion printing at ambient temperature as AM process is chosen because of the lower print energy (Böckin et al., 2019; Faludi et al., 2015). Existing 3D-printing materials contain health hazards due to emissions of nanoparticles and gasses (Kim et al., 2015; Mendes et al., 2017), so this has to be tackled as well in the new material development. The focus will therefore lie on printable composite materials that exist of a filler and a binder. Fillers are generally particles that give the 3D printed material its strength, e.g. cellulose fibers. Binders act as the medium that keep the fillers together and in one piece. This can be seen as the glue that keeps the 3D printable paste together.

**ASSIGNMENT \*\***

State in 2 or 3 sentences what you are going to research, design, create and / or generate, that will solve (part of) the issue(s) pointed out in "problem definition". Then illustrate this assignment by indicating what kind of solution you expect and / or aim to deliver, for instance: a product, a product-service combination, a strategy illustrated through product or product-service combination ideas, ... . In case of a Specialisation and/or Annotation, make sure the assignment reflects this/these.

In this thesis an approach will be developed that helps people in makerspaces to develop their own bio-based 3D printable materials. A tool will be created that guides makers in creating their own 3D-printable materials.

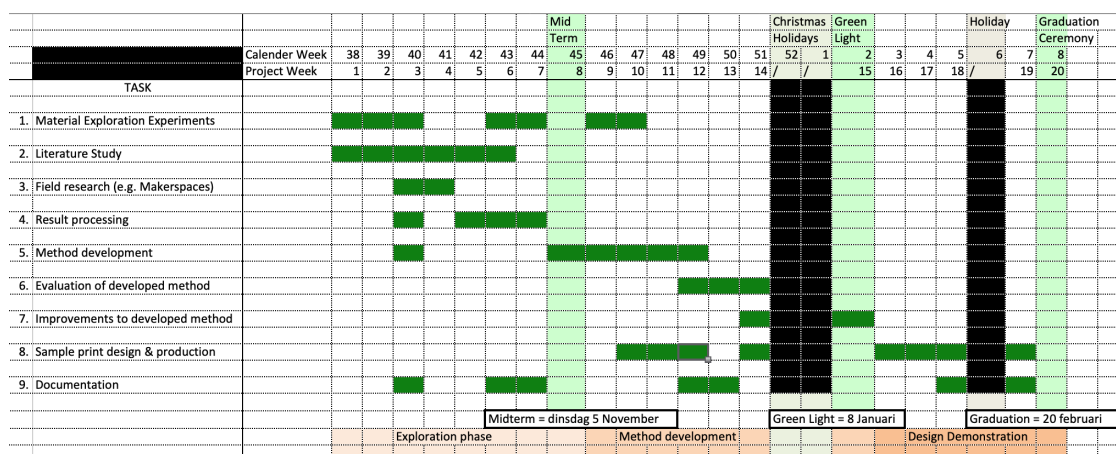
The primary result of this thesis will be an easily executable method for makers to develop a printable composite material. The maker will be guided in the processing and testing of the filler & binder that are needed for this composite paste. Preferably the filler and binder are from a local and renewable resource stream.

The resulting method will have the form of a decision tree. Step by step, the user/maker is guided through a set of questions that help to define the material properties of the used material(s). By following the right answers, ultimately a narrow solution space is reached for the material development. In this solution space probable ranges for the different material parameters (e.g. particle size, weight ratio of binder to filler, possible successful binders, nozzle size, extrusion speed and extrusion pressure) are shown. Within these ranges that are likely to show good results the user/maker is encouraged to experiment with different values.

Simple material test procedures are then provided to give the user/maker guidelines how to come to an evaluation of the printing material quality; does the material show shear thinning properties? is the material viscous enough? is there a likelihood of clogging? A separate troubleshoot sheet is provided for the case that the material is not performing well. Different common types of failures are described with possible solutions (e.g. increasing particle size).

To illustrate the results to which the method can lead, a couple of probe prints are included. These probes are printed in materials that were developed by following the method that was designed. Those probe prints will not only show the print quality but also possible experiential qualities that are hard to quantify. The probe prints will be designed using parts of the material driven design method.

20 - 2 - 2020      end date



In the second phase, the aim is to combine the knowledge gained in the explorative phase into a general model. What are the general principles at play & how can material propositions be formed & tested based on these principles? Which material characteristics does a maker have to test and how does he/she do this in order to get an idea about the quality of the print paste and how to improve this? A general method will be designed and tested that uses the insights from the first phase. This method will then be evaluated: Does it help in the creation of new 3D printable materials? Parallel to this method development, a start will be made with the third phase; the design of the sample prints. What can the new method achieve in terms of prints and what is the best way to showcase the results?

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## MOTIVATION AND PERSONAL AMBITIONS

Explain why you set up this project, what competences you want to prove and learn. For example: acquired competences from your MSc programme, the elective semester, extra-curricular activities (etc.) and point out the competences you have yet developed. Optionally, describe which personal learning ambitions you explicitly want to address in this project, on top of the learning objectives of the Graduation Project, such as: in depth knowledge a on specific subject, broadening your competences or experimenting with a specific tool and/or methodology, ... . Stick to no more than five ambitions.

This project was mainly set up for three reasons:

Firstly, this project lies in the area of new, bio-based, material development. Apart from the fact that this field has drawn me from a young age, materials are the building blocks with which a designer has to work. A thorough understanding of materials is therefore, in my opinion, crucial to be(come) a good designer.

Secondly, the development of new materials is an important topic in the current society as there will be a shift over the coming decades towards a circular and sustainable economy in which there is little place for oil based plastics. This makes this field exciting and relevant to work in. Furthermore, as mentioned above, 3D printing can never be green with the current energy intensive printing methods, therefore new materials are needed to accomplish this.

Finally, it is a research-focused project in which a rigorous working method and deep understanding of the knowledge required for the project is needed. Personally, I like the concise way of working in a research project.

In this project, I want to show my qualities in the rigorous analysis of a problem and the data that come with it. The aim is to develop a method that spans an as wide as possible range of materials. In order to achieve that, a broad and reliable set of data/knowledge has to be gathered and meaningful and justified connections between different observations/datapoints/insights have to be made. My background as a BSc in Mechanical Engineering will contribute due to the slightly more analytic mindset that is trained in that study.

### Personal ambitions

Linked to the reasons for setting up this project, is my personal ambition to acquire in depth knowledge about bio-based materials. What are the principles at work that make a success or a failure of a new material? What are the strengths and weaknesses of new materials at the moment? What properties need to be improved and which can be exploited? What are the barriers that prevent mass-market adaptation?

Secondly, but linked to the former ambition is the ambition to apply the material driven design method. I have never worked with the method, but a paper on the MDD-method sparked my interest. It was for me a new and extended view on materials, in which not only ultimate tensile strength and surface smoothness was valued.

### References

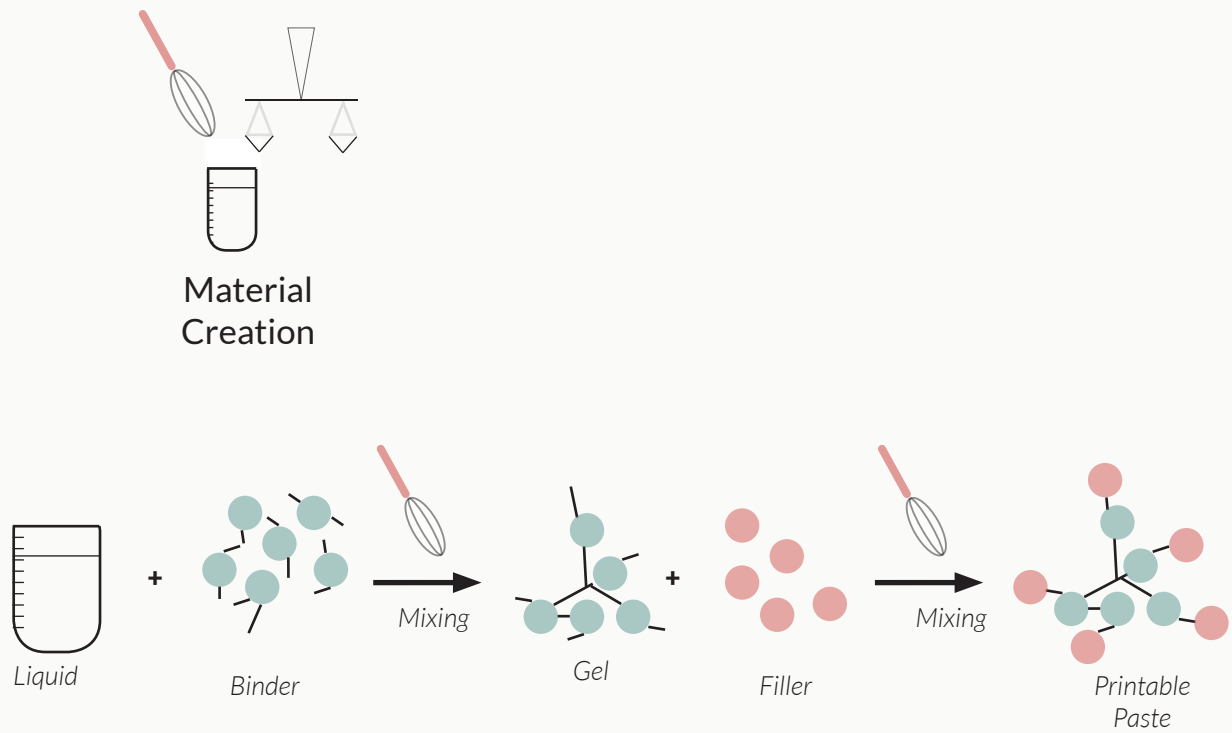
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## FINAL COMMENTS

In case your project brief needs final comments, please add any information you think is relevant.

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	Filler	Binder		Summary	
	 Walnut shell	 Algininate		Best match so far Slightly flexible High printing resolution	Try with methylcellulose hard fillers
	 Pecan shell	 Chitosan		#data entries = 12	<a href="#">Recipes</a>
	 Cacao shell	 K-Carrageenan		xxx xx x	Try with xxx xxx
	 Rice			#data entries = n	<a href="#">Recipes</a>
	 Mussel shell				

