3D printed fiber reinforced lignin
Exploring the options to use wood in an additive manufacturing process

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ABSTRACT

Problem definition and objective
Additive manufacturing has already been developed to use various sorts of plastics, metals, composites and concrete. Until now, the most used composite in building materials, natural wood, has not been used in an additive manufacturing process. The potential of using wood in an additive manufacturing process lies in the complex shapes which could be made, the savings of materials, the wood properties which could be used in a more optimal way and the ease to reproduce an object with high accuracy without the costs of labor. This research will investigate if there are options to use wood in an additive manufacturing process such as 3d printing. The question to answer is; Can wood be used in an additive manufacturing process?

Study design
This research is based on a broad literature survey on the topics of wood, additive manufacturing and state of the art of 3d printing wood. This is followed by an exploration phase where various material mixes are made and tested in a qualitative way in order to get an understanding of the behaviour of the material. The materials are extruded with medical syringes and later on with a 3d printer to see how they behave in this process. Finally a design is made what shows the potential of this material in 3d printing.

Focus
This study focusses on the use of the materials cellulose and lignin, which are combined in a 3d printable material. The process is based on the soluble character of lignin, and therefore does not work with a temperature gradient. The materials which are used are Skogcell 90Z bleached kraft WFBR and Indulin AT lignin, both retrieved from softwood in the kraft process and are fully biobased. This research describes a limited amount of material mixes, based on the input materials.

Results
Theory about wood and its main components is translated into a production process which leads to a wood based 3d printable material. Material mixes with a content up to 20% of cellulose are tested and printed with a syringe. Materials containing up to 11% of cellulose are printed with a 3d printer. The result, a wood based 3d printing material.
Can wood be used in an additive manufacturing process?

[Main research question]
General introduction

Additive manufacturing has become a more used production technology for small production batches of various products. Where complex shaped products were expensive and time-consuming to build, nowadays even shapes which can not be built with conventional methods can be build with an additive manufacturing process. Thanks to the ability to split up objects in layers and compose them layer by layer, even the most complex shapes have become possible to build.

The additive manufacturing process, which can be described as a production process based on adding layers of one material and binding these without the use of an additional type of material such as a glue, has been developed to use various sorts of plastics, metals and concrete. Surprisingly, natural wood, one of world’s most used building materials, is not developed yet to be used as a stand alone material in an additive manufacturing process.

Current production processes which are used for natural wood are subtractive manufacturing processes. These processes can be described as processes where material is subtracted in order to shape the material to the need of its final design. Examples of these techniques are cutting, milling, planing or even simple things such as drilling. The result, lots of waste material what can’t be used as natural wood and is used as bio-fuel or is processed into wood based building materials.

With the technical development of mankind in the past two centuries, nowadays resources are depleting more rapidly than ever before. Although timber is a natural material that can be harvested from trees, the timecycle to grow a tree is much longer compared to the amount of timber that is needed within this timespan. As result, rainforests are harvested with major effects. Animal species are threatened with extinction and climate change is encouraged.

The use of wood in an additive manufacturing process would not only release new production options, it could also have technical and environmental benefits which in the end will benefit human mankind. Wastewood is nowadays often burned in order to use it for energy, but what if these piles of wastewood could be reused into a 3d printable material, and products can be made from it? This research will investigate if there are options to use wood in a 3d printing process.
Problem statement

Additive manufacturing is a production technology which has developed into a widely used technology. Especially for small production batches additive manufacturing is a profitable production technology. Also it is used for the creation of mock-ups or prototypes. Another benefit of additive manufacturing is the fact that the labor can be outsourced to the machines which are used for this production technology. The 3d cad models which are needed for this production technology can easily be shared and reproduced at another printing device.

Another advantage of additive manufacturing lies in the fact that complex shapes can be made which are not feasible or not remunerative with conventional production technologies such as milling, cutting drilling or turning. These stated technologies are also known as subtracting manufacturing technologies which means they remove material in order to shape the material to the final needed shape. The waste material is often used for wood based building materials or even used as fuel. Additive manufacturing adds material which means it only uses the amount of material which is needed. This means there is no waste material. This could potentially save a lot of wood, which could be beneficial for the climate and ecosystem.

Additive manufacturing is developed to use various sorts of plastics, metals, composites and concrete. Until now, the most used composite in building materials, natural wood, has not been used in an additive manufacturing process. The potential of using wood in an additive manufacturing process lies in the complex shapes which could be made, the savings of materials, the wood properties which could be used in a more optimal way and the ease to reproduce an object with high accuracy without the costs of labor.

This research will investigate if there are options to use wood in an additive manufacturing process such as 3d printing.
GOAL
Exploring the opportunities of the use of wood in an additive manufacturing process.

SUB GOALS

- Getting a clear understanding of wood

Understanding wood as a material will be important in order to judge if wood can be used in an additive manufacturing process. Understanding the properties of wood but also the elementary building blocks of wood will give more insight in the materials and therefore provide important information for this research.

- Getting a clear understanding of additive manufacturing

This subgoal is important to understand the definition of additive manufacturing. Furthermore, this will lead to a clear understanding of the benefits of these processes and an understanding of why these methodologies are applied. This sub goal can only be achieved if the additive manufacturing processes are being studied which might be needed in case a new process or existing process needs to be developed or adjusted.

- Getting knowledge of what has been done with additive manufacturing and wood.

This subgoal is important because it will take care of the already existing methodologies on the theme of using wood in additive manufacturing. Getting knowledge of these will also potentially lead to processes which might form a basis for further research in the field of using wood in additive manufacturing.

RESEARCH QUESTION

Based on the goal of this research we can state the following research question:

Can wood be used in an additive manufacturing process?

SUB QUESTIONS

The research question of this report can be split up in several sub questions which are also linked to the sub goals of this research. The following sub questions can be stated:

- What is wood, and what are the properties of wood and its components?
- How can the components of wood be combined in a printable material?
- What is additive manufacturing and which advantages does this production methodology have?
- What is the state of the art technology in the usage of wood in an additive manufacturing process?

The subquestions regarding wood are needed to reach the subgoal on wood. Being able to answer these questions will lead to an answer on an important part of the main question. The subquestions regarding additive manufacturing will help get to the subgoal about additive manufacturing and thereby also help in solving the main research question. Questions about the state of the art technology of using wood in additive manufacturing processes will help to find existing processes which might be a good starting point for this research.
PROCESS

METHOD DESCRIPTION

Phase 1: Literature & exploratory reading
In order to get a clear mindset on the goal of this research, exploring the opportunities of the use of wood in an additive manufacturing process, literature was needed. First of all, literature on the state of the art of using wood in an additive manufacturing process was retrieved from sources. The sources which were used are Google scholar in order to guarantee that the information was validated. This resulted in various sorts of research papers which have described progress on the topic of the use of wood in AM. Sources which were used are research papers, internet videos for processes which were not described in research papers, and contacts with researchers of Wageningen University. Terms which were used in Google scholar to find research papers are:

- 3d printed wood
- Wood filament
- Wood in additive manufacturing
- Wood based material in 3d printing
- 3d printed cellulose
- 3d printing of wood fiber

This step lead to several papers which stated what already was done in the topic of this research. However, the amount of papers which were found were scarce. The next step which was taken is doing research on the material wood. In order to do so, various books about wood were read, research papers were read and a researcher from Wageningen University was contacted. Search terms which were used are:

- Wood
- Wood properties
- Structural properties of wood
- Aesthetic properties of wood
- Composition of wood
- Cellulose
- Lignin
- Timber

The next step was to do research on additive manufacturing, in order to get a clear view on this technology and its advantages. Also the definition of additive manufacturing is needed in order to understand which technologies are applicable. Sources such as validated research reports and scientific books were used to gain information.

In order to get information the following search terms were used:

- 3D printing
- Additive manufacturing
- Rapid prototyping
- Additive fabrication
- Stratified additive manufacturing

Phase 2: Literature survey
The literature survey of the report will elaborate on the sub questions which are needed to be answered in order to give a proper answer on the main questions.

- What is wood, and what are the general properties of wood?
- Which properties do the components of wood have?
- What is additive manufacturing and which advantages does this production methodology have?
- What are the advantages of additive manufacturing?
- What is the state of the art technology in the usage of wood in an additive manufacturing process?

This is to give a clear understanding on the material wood, the technology of additive manufacturing and the state of the art in using wood in additive manufacturing processes.
Starting with the literature on the sub questions regarding wood. Several research papers were studied which all could partly provide an answer to the sub questions. In order to get specific information on the substances of wood and their properties, a literature study was done. The library of the university in Wageningen has an extensive collection of papers and books on the topic wood. This university has a specialized department on wood, biomaterials and forestry. General information about the aesthetics and structural properties of wood were collected based on the types hardwood and softwood. This was done in order to be able to assess the print samples later on in the process.

In order to answer the questions regarding additive manufacturing new research papers will be studied. The goal is to get a clear understanding of the definition of Additive manufacturing, understand the benefits and disadvantages of the processes and investigate which processes are part of additive manufacturing. All research papers which were used were retrieved from Google scholar and validated. Next to that a scientific book on additive manufacturing was used to get information about additive manufacturing, and the methodologies which are commonly used. All information as result of this process will be used for the literature survey in this report in order to contribute to the theoretical framework for the research.

After the previous topics some investigation on the state of the art of the use of wood in an additive manufacturing process is needed. The information which was found in phase 1 could be used but also new information was needed in order to keep the research as broad as possible. The results of this part will be elaborated in the report, in order to see if there are some interesting starting points for further research and development.

**Phase 3: Experiment phase**

Based on the results of the literature survey some basic components of wood were obtained from the University of Wageningen and used in tests. For these tests, Kraft lignin, Indulin AT was obtained from Wageningen university and is used in powder form. The cellulose which was used in the project is Skogcell 90Z bleached kraft WFBR. Based on leads from talks with a researcher of the university of Wageningen and the literature survey, a process was designed in order to create a material suitable for 3d printing. The material would be composed of an amount of lignin, cellulose, water and acetone. Based on the idea of creating multiple different mixes, qualitative tests were done to get an idea about how the material behaves.

During the experiment phase material samples were made. Every sample was put in a petri dish and later on a part of it was used to try to extrude it. For this a medical syringe with a nozzle size of 1,6 mm was used. The syringe itself had a cross section of 18 mm(inner radius) and could hold 20 ml of fluid. Every sample was tried to be extruded, and the material resulting from that was also put in a petri dish to dry. After this step was finished, every sample was checked in a qualitative way for homogeneity, viscosity and adhesiveness. The tests which were done are based on qualitative observation. The test setup was as following:

For homogeneity tests, a small amount of material was put in a petridish. The next step is to tilt the petridish and observe if a separation of fluid and dry material can be seen. In case this does not happen the material mix is marked as homogeneous.

The second test on viscosity is also done with a petridish and a small plate. A small amount material is put in a petridish. The next step is to tilt the petridish and observe if a separation of fluid and dry material can be seen. In case this does not happen the material mix is marked as homogeneous.

The force needed to move the top plate with a constant speed is proportional to the shear strength within the material. The relation between the force and speed can be explained with the formula and figure 1 on the next page:
The shear stress in the material is noted with $\tau$ and is assumed to be proportional to the force which is applied on the top plate. The velocity of the top plate is noted with $u$, and this parameter is kept similar in all tests. The amount of space inbetween the petridish and the plate is determined to be about 5 mm. The viscosity of a material is noted with $\mu$, and will be different for every material. Since $\frac{du}{dy}$ is kept similar for all tests, and the force applied on the top plate might be different in order to do so, the viscosity has to change. If a high force is needed to move the material at speed $u$, then $\mu$ will automatically have a high value as well. If a lower force is sufficient to move the top plate at speed $u$, then $\mu$ will be lower.

Because these tests will be done based on qualitative input and output, the results are kept simple. For material with a low pressure applied, the score will be low. For materials which needed a high force to move the plate at speed $u$, the output score of the test was high. If materials needed an intermediate amount of force, the score was medium. The results of this test were important in order to get a understanding of the flow properties of a material mix. These properties are important because they tell something about the easiness to extrude but also about the abilility to stack the material on top of another layer of the same material.

The third test is done to get an understanding of the stickiness of the material. It is important to get a understanding of this property, because this will be key in the bonding of the material inbetween layers, but also inbetween the fibers in the material mix. The test is setup as following: A small amount of material is taken between your thumb and middle finger. The next step is to compress the sample a bit, so it sticks on your fingers. The next step is to move the thumb and middle finger away from each other, so the material forms a “bridge” inbetween the fingers. If the material is not sticky it will lose grip and fall of your fingers immediately. Therefore these mixes get a score “-”, which can be seen as low. The material which is sticky and has good adhesive score and will form a long “bridge” will get the score “++”, and are interpret as high stickyness material. All these test were done to get a better understanding of the material. However, in order to get accurate data on these specifications, more research is needed.

The goal of this phase is to create a material that can be extruded with a medical syringe. During this experimental phase, data was tracked, telling how much of each component is in the material mix. These results, together with the experience of extruding the material, and the extruded samples resulting from that, were used to decide which material samples would be suitable to try extruding with a machine.

The printed material samples were tested with a microscope to get a better understanding of the extruded material. The material samples were checked for fiber alignment, the distribution of lignin within the material and waterproofness was checked. For fiber alignment, multiple material samples were checked underneath a microscope. With this test, the placement of fibers after extruding was studied. The test on lignin dispersion was done with some material samples. In this test, it was checked if the lignin was nicely positioned around fibers. The test on waterproofness was done with a petridish and a material sample which was put in the water for 168 hours. These simple tests were done to get a better idea of the output material from the printing process.
Phase 4: prototype & production
In phase 4 a homemade printer was used to do tests on extruding the material samples from phase 3. The printer is controlled via a minitronics board obtained via Reprapworld.nl. The machine has stepper motors of the type NEMA 17 with a holding torque value of 4.08 kg.cm. The stepper motors were also acquired from Reprapworld.nl. The resolution is 1.8 deg per step with 1/16th microstepping. The bed is a standard bed without heating. The maximum print size is limited to 180 x 120 x 60 mm. During this phase, simple shapes like a cylinder and triangle were printed to see how the material behaves in a 3d printing process. The circle had a diameter of 38 mm and the triangle had sides of 40 mm. The triangle was used to check if the material had a constant flow. Based on the findings in phase 4, a design is made for an object that was printed. The idea is to design an object that shows the potential of the 3d print technology based on components of wood. As a design a node was made with a complex geometry. The node is assumed to be impossible to make, with conventional production methods, from one piece of wood without the need to separate it, or spill a lot of material in the production phase.

Research framework planning
On the next page, the planning for the research framework is shown. The planning shows when all actions take place.

Figure 2: Scheme of process applied in research.
Figure 3: Time schedule used for research.
Wood

Trees are complex living organisms which are like humans. When young, they are small and humble, and grow fast in case they receive the right nutrition. Full grown trees grow slowly and produce different tissue compared to the young ones. Young trees originate from fertilized eggs and become seed encased embryos or they originate through vegetative propagation. Growth can only happen because the trees respire, and they need a balanced mix of minerals in order to stay healthy. And like humans, they metabolize this food, but trees can also synthesize their own food. In case a tree gets wounded, they take care of processes so that healing will happen. Full grown trees are strong and they will maintain this for a long period. But after this long period, the tree loses strength and might not be able to heal the wounds anymore. At this stadium the tree becomes vulnerable for diseases and finally, the tree dies. This is a short explanation of the growth cycle of trees. This literature survey is not about the growth cycle of trees, but about its result of growth; wood.

Classification
Trees can be defined in two sorts; the ones which deliver softwood, and the ones who deliver hardwood. As the name might suspect, hardwood is wood with a higher hardness compared to softwood. But this is not entirely true. Some species of softwood deliver wood with a higher hardness compared to hardwood. For example balsa wood, which is a hardwood species, which has a low hardness compared to many softwood species. The precise difference between the two lies in the cell biology of the two sorts. What also results in clear distinctive types of trees. Both hardwood and softwood species are categorized in the division of Spermatophyte’s (seed plants). This means both produce seeds in order to reproduce. However, The way they do this is completely different. Softwood is categorized in the subdivision of Gymnosperms, which basically means naked seeds. Hardware is categorized in the subdivision of Angiosperms, what means they produce seeds within fruits (Bowyers, Shmulsky & Haygreen, 2007).

These differences are visible, for example needle like leaves characterize softwood species, whereas hardwood species have leaves. The needles from softwood species remain during winter, so the tree stays green, whereas leaves from hardwood species change color and eventually drop off in autumn. The softwood species grow often scaly cone shaped fruits which bear the seeds. Hardwood species produce seeds within a fruiting body, for example an acorn for an oak and or a chestnut for a chestnut tree. The differences of the divisions are visible in table 1.1 which is shown on the next page.

Hardwood species are categorized in the subdivision of angiosperms. This subdivision has two classes named monocotyledons and dicotyledons. Hardware producing species fall within the class of dicotyledons. Monocotyledons are mostly known as palm trees or yucca’s (agave family).

Wood structure
Now that the appearance and divisions have been discussed, we can also discuss the differences in wood structure. Not only the appearance of softwood and hardwood are different but also its cell types, the arrangement of cells and its relative numbers. The fundamental difference between hardwood and softwood is the fact that hardwood contains a type of cell which is called the vessel element. This type of cell occurs in most hardwoods, but is almost never seen in softwoods (Bowyers, Shmulsky & Haygreen, 2007). Hardwoods are classified as ring or diffuse-porous, depending upon the size and distribution of vessels in a cross section. Woods that form very large-diameter vessels part of a year and smaller ones thereafter are called ring-porous. An example of this can be seen in figure 5, where end grain oak is photographed. Woods that form vessels of the same size throughout the year are classified as diffuse-porous. An example of this can be seen in figure 4. In this picture, end grain padouk can be seen. The differences are clear. Padouk, has vessels spread all over the cross section where oak has vessels which are positioned in rings (in this picture lines as part of the rings).
Table 1.1: Trees in the plant kingdom

<table>
<thead>
<tr>
<th>Divisions:</th>
<th>Thallophytes</th>
<th>Bryophytes</th>
<th>Pteridophytes</th>
<th>Spermatophytes (seed plants)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Algae</td>
<td>Mosses</td>
<td>Ferns</td>
<td>(seed in fruit)</td>
</tr>
<tr>
<td></td>
<td>Fungi</td>
<td>Liverworts</td>
<td>Horsetails</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Rushes</td>
<td></td>
</tr>
</tbody>
</table>

Subdivisions:

<table>
<thead>
<tr>
<th>Orders:</th>
<th>Gymnosperms</th>
<th>Coniferales</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(naked seed)</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Orders:</th>
<th>Cycadales</th>
<th>Ginkgoales</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(palmlike)</td>
<td>(rare)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Families:</th>
<th>Taxaceae</th>
<th>Pinaceae</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Yew</td>
<td>Fir</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Hemlock</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Pine</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Spruce</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Larch</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Families:</th>
<th>Taxodiaceae</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Redwood</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Baldcypress</td>
<td></td>
</tr>
</tbody>
</table>

25 families in the United States

source: Bowyers, Shmulsky & Haygreen (2007)

Figure 4: Picture of endgrain Padouk. This type of wood is known to be diffuse-porous. The vessels are spread all over the cross section.

Figure 5: Picture of endgrain Oak. This type of wood is known to be ring-porous. The vessel openings can be seen in lines which form a ring in the cross section of the trunk.
A close look at wood shows it is made of tiny cells or fibers. The main difference between hardwood and softwood was already explained on the previous page. But if we look at cell size of softwood and hardwood we can notify some more differences. Where hardwoods have relative short cells and vessel elements, softwoods have relative long cells which are shown in figure 6. The cells are pin shaped and have a hollow core which is called Lumen. In the cell wall are some small openings situated which are called Pits. The cross sectional shape of softwood fibers is almost square. In figure 7 the arrangement of the wood cells is shown. Figure 7 shows also the 3 ways to cut wood, transversal, also known as cross section, tangential and radial surfaces. The combination is helpful to identify pieces of wood.

Figure 6: typical cell in softwoods
source: Bowyers, Shmulsky & Haygreen (2007)

Figure 7: softwood cells arranged in piece of wood
source: Bowyers, Shmulsky & Haygreen (2007)
Hardwood has different sizes of cells and not only fibers. Figure 8 shows the relative size of hard wood fibers compared to the ones in softwood. Also the vessel elements are shown. The differences between softwood and hardwood are not only the vessel elements, but also the size of the fibers. Fibers from hardwood are smaller, and have different cross sectional shapes. The ones from hardwood are almost round. Fibers from hardwood are also more thick walled and have bordered pits with less-developed borders than softwood tracheids (Esau 1965, p239). The function of hardwood fibers is more specialized compared to the ones from softwood. In softwood, the longitudinal fibers take care of transport of fluids and are responsible for strength. In hardwood, this happens in a different way. The vessel elements take care of the fluid transport and the fibers are responsible for strength. The strength and thus density of hardwoods is therefore mainly related to the portion of fibers per volume of wood ((Esau 1965, p239. (Bowyers, Shmulsky & Haygreen, 2007)).

Composition
As building material, wood is one of the simplest and most used products. It can easily be shaped and fastened. At the same time wood is also one of the most complex materials which are used. It is made of tiny cells, varying in type depending on hardwoods or softwoods. Both have a precise structure of tiny openings, membranes, and layered walls. The ease with which wood is converted to a product depends on the knowledge of its structure. Now the cellular nature of wood is introduced, the focus will be a bit more on the molecular level of wood.

Wood consists of 3 basic elements; carbon, hydrogen and oxygen. Out of these elements the following organic polymers are made; Cellulose, hemicellulose and lignin. Depending on the sort of wood, hardwoods or softwoods, these polymers have a certain included percentage. Hardwood and softwoods have in general 40 till 44 percent of cellulose included. Hardwoods have about 15 till 35 percent of hemicellulose included whereas softwoods have 20 till 32 percent of hemicellulose.

Figure 8: Differences between wood cells.
source: Bowyers, Shmulsky & Haygreen (2007)
Hardwoods have 18 to 25 percent of lignin included whereas softwoods have 25 to 35 percent of lignin included. On this page the polymers will be explained a bit more.

**Cellulose**
Trees and plants can produce their own food, which is done by photosynthesis. This process delivers the goods which are needed for the forming of cellulose, what is done in the wood cells.

Photosynthesis is the process by which water and carbon dioxide are combined in the leaves of green plants, using the energy from the sun to form glucose and other simple sugars, with oxygen as a byproduct. Figure 9 shows the process of photosynthesis. According to Kozlowski & Pallardy (1997) this glucose and simple sugars are converted into starch, glucose 6-phosphate or fructose 6-phosphate and then to sucrose($C_{12}H_{22}O_{11}$). These sugars are then transported in the form of sap to the branch tips where they will be further transported to the main bole, branches and roots. When the sugars arrive at the individual wood cells, the sucrose is combined with water to form glucose and fructose. Trees use glucose and fructose to make leaves, wood and bark.

According to Peng, Kawagoe, Hogan & Delmer (2002) Cellulose is synthesized within living cells from a glucose-based sugar nucleotide. A nucleotide is a compound derived from combining a sugar with a phosphate group and a base that is a constituent of RNA or DNA. Complex and separate mechanisms are thought to control initiation of cellulose-chain formation, chain elongation and termination of the synthesis process. The next effect is that the glucose molecules are joined up end to end, with the elimination of a water molecule in order to form a chemical linkage between neighboring units. These strings of cellulose can be as long as up to 10000 units. Cellulose is a well known material for humans. Paper is made out of the cellulose from wood and cotton is also 99% pure cellulose. Cellulose is not a source of food, humans can’t digest it because the enzymes in our body can’t break down the bonds between the molecules. To do so, the bonds between the cells need to be hydrolyzed, which we are not able to. In part 3 more information can be found about cellulose.

**Hemicellulose**
As explained in the previous paragraph, glucose is one of the sugars which is produced with photosynthesis. However, next to glucose some other sugars are produced as well, such as galactose, mannose, xylose and arabinose. Within the younger and developing cells, different products are produced as described before. In these cells, hemicellulose is produced with these sugars. The hemicellulose is known as an lower weight polysaccharide, with smaller chain lengths (up to hundreds of times).

**Lignin**
Lignin is a complex and high molecular weight polymer built upon phenyl propane units. Lignin is composed of carbon, hydrogen and oxygen and not related to carbohydrates but is phenolic in nature. Lignin is known to be stable and difficult to isolate from wood. The exact configuration of lignin is not known because lignin occurs in a large variety of forms. Lignin acts as a natural glue between cells and within cell walls. The function of lignin is to give wood cells rigidity which is done by bonding cellulose and hemicellulose. Also lignin adds to the wood's toxicity which protects wood from decay and insects. The effect of lignin can be seen when we compare wood with seaweed or cotton. Both cotton and seaweed exist out of cellulose, but don’t have ligning. Both are nonrigid, which wood also would have been in case the lignin was not inside it. In part 4 of this report, more information will be given about Lignin.
Figure 9: Process of photosynthesis explained

- Sunlight
- Photosynthesis
- Sugars
- Water
- Carbon dioxide
- Oxygen
- Sucrose
- Cellulose + lignin
Appearance
Wood is a widely chosen material simply enough because people like its appearance. Now we know what wood is made of, and the differences between hardwoods and softwoods are explained, its appearance will be explained.

Annual rings
Ask a child how old a tree is, and he or she will probably tell you to count the annual rings in the cross section. And they are right, the rings do tell you how old a tree is. But the rings itself can tell a lot more. A growth ring can be defined in two parts. Figure 10 shows an transverse cut of a tree. In the cross section the annual rings are visible. An annual ring is often split in two parts, the wide part is the early wood, the narrow part is known as latewood. But what are the reasons for doing this?

Annual rings are the result of a tree which grows for a year. During the year, the weather changes as result of different seasons. During spring, the trees start growing as result of more daylight, higher temperatures, followed by summer. Because of the longer days, more photosynthesis will take place, with as result more food for the tree. This amount of food will be synthesized into sugars which will result in the gain of cellulose. The trees start growing in a rapid pace. As result the tree grows fast which means that new fibers will grow with a low density and cells will have a relative large cross section. When autumn starts, days become shorter, temperatures are slightly becoming lower and in case of hardwoods, trees are dropping their leaves as result of less food. The tree is preparing for the cold winter where minimal amount of food is available. As result the trees are growing in a lower pace. The cells will have a smaller cross section and cells will grow with a higher density, resulting in the darker part of the annual ring. Figure 11 shows a cross section of earlywood and latewood cells. In this figure the early and latewood are clearly distinctive. The growth ring is like a mirror of the past. It clearly shows the weather forecast of the past.

However, what happens with a tree in the tropics, where no winter is known. Do these trees grow annual rings? And how can the early and late wood be distinguished?

In the tropics, some trees only grow one growth ring annually. But more oftenly seen is that trees in the tropics grow more than one growth ring each year. Often there is no difference visible between earlywood and latewood. This means, it is not possible to determine the age of such trees which have lived in the tropics.
Heartwood and sapwood
When a cross section of a stem is studied, some things might stand out. Things you might see are annual rings, with earlywood and latewood. But something else what may stand out is the difference in color of the inner-wood, which is called heartwood and the outer wood what is called sapwood. What are the differences and why do they often have a different color? Heartwood might be known as the part of the stem which is heavier, stronger, more highly figured and more resistant to decay. Some of these notions are true but others are not.

When a young tree starts to grow it has often no heartwood. After a number of years heartwood typically starts to form in the center of the tree. The exact number of years is still unclear. Hillis (1987) reported that heartwood is starting to form after 14 to 18 years. Another research of Dadswell & Hillis (1962) on eucalyptus showed that heartwood may begin to grow after 5 years. Gominho & Pereina (2000) did also research on eucalyptus and found that all examined trees, which had an age of 9 years, were found to have heartwood. Hoadley (1990) reported that species such as black locust and catalpa maintain no more that 3 years of sapwood, while Dadswell & Hillis (1962) reported that some species of wood like beech or European ash may not begin to form hardwood before they reached an age of 60 to 100 years, and that some species apparently never form heartwood.

Bowyers, Shmulsky & Haygreen (2007) describe the differences between heartwood and sapwood as almost totally chemical. The chemicals which are in the heartwood, give it its specific properties. The difference in color between heartwood and sapwood is caused mainly because of the chemicals which are in the heartwood. The chemicals, mostly polyphenols which are found in temperate zone trees are colorless in regular light. However, they are unstable and degrade with time. The color is often darker as a result of an oxidation process. Figure 12 shows a piece of oak where the color difference between heartwood and sapwood is clearly visible.

Another difference is the fact that heartwood might be difficult to penetrate with liquids. This is also partly caused by the chemicals which are in the wood. For example the presence of extractable oils, waxes or gums. These might close tiny passages in the cell walls. Another reason is the fact that cells may rearrange over time with the result that cell to cell passages are not connected anymore.

Another difference is the volumetric weight. Heartwood might have a higher weight compared to sapwood. This is mainly caused by the extra chemicals which are present in the heartwood. These chemicals might also affect the ease to dry the wood and the odor of the heartwood.

Figure 12: clear differences visible between heartwood and sapwood in this piece of oak.
Structural behaviour

Now that we explained what wood is, how its made, what the components are and what the visual characteristics are, something will be explained about the structural behavior. On the previous pages the cell typologies of wood were explained. Also, the alignment of the cells has been shown. This specific alignment of cells has an effect on the structural behavior of wood.

A material which has the same mechanical properties in each direction is termed isotropic. Many materials such as metals, plastics and cement are known to show this behavior. However, wood behaves differently because it has fibers aligned in one direction, causing different properties in the transverse direction compared with the direction parallel to the grain. This makes wood an anisotropic material with different behavior in at least two directions. The properties of wood are different in longitudinal, tangential and radial directions. The largest differences can be found between longitudinal direction and radial or tangential directions. Radial and tangential directions do not differ greatly. The differences, explained in ratio’s are as follow. Strength properties of wood parallel to the grain are 10 to 20 times higher compared to strength properties in the direction perpendicular to the grain. Knowing this, it is quite important how to apply wood in structural usage.
Timber is one of the world’s most used building materials and produced by nature. Trees are the world’s oldest living organisms, starting as a small humble plant growing to the largest structures delivered by nature. Trees are like humans, they breathe, in order to live. And by doing so they deliver oxygen to the environment, but also generate sugars which are used to produce wood cells. So they have a metabolism like humans do. But trees are also capable of producing their own food, something humans can’t do on their own.

If we look into the material wood, it can be noticed that the material is rather complicated up to the nano scale level of detail. Trees are composed of cellulose, hemi cellulose produced by the younger cells, and lignin which functions as a natural binder. Within the world of trees there are however two main sorts of trees. One sort is softwood, also called needle trees. This sort will remain green during the year which means they don’t drop their needles. They also produce seeds which are in general grown in cones. These cones have a snake-skin like surface and in between the scales the seeds are visible, so not protected. The other sort is hardwoods which are different in the fact that they have leaves, and drop these if necessary. They also grow fruits in which they put their seeds, well protected.

If we look on a cell detail level, the two sorts hardwoods and softwoods are quite different. Softwoods have rather large cells which have two functions; taking care of sap streams and taking care of strength. In case of hardwoods these tasks are separated and the cells are way smaller compared to the cells of softwoods. Also hardwoods have vessel elements which take care of sap streams within the tree.

The aesthetics of wood are the result of rings in the cross section. These originate from annual growth with in case of hardwoods, earlywood rings and latewood rings. This has to do with the seasons during the year which makes this as an historic mirror which shows a good image of the surrounding where the tree has lived, and how the weather was over the years.

The mechanical behavior of wood is quite special because wood as material has an anisotropic character. This means wood behaves in the direction of the grain in a different way compared to the direction orthogonal to the grain. This is one of the main reasons why knowledge about wood is important in order to build with it. The anisotropic character results in a strength ratio of parallel to grain/orthogonal to grain of 20/1.

Wood itself is almost colorless and derives its colors as result of extractables which are hidden in the wood. These extractables are rather important because they serve as protector in the wood for for example decay or insect resistance and it gives the wood its odor.
Additive manufacturing

In this part of the report the focus will be on additive manufacturing. What is additive manufacturing and what are the advantages of additive manufacturing?

Additive manufacturing is the name which is used for the technology 3D printing. The technology is based on the principal of manufacturing a part by depositing material layer-by-layer. Current technologies are different because these are based on subtraction (milling or drilling), formative processes (casting or forging), and joining processes such as welding or fastening (Conner et al., 2007; Berman, 2012). Gibson. I, Rosen. D & Stucker. B, (2015) add to that that the name of additive manufacturing is also called rapid prototyping, what is popularly called 3D printing. The term rapid prototyping is used for processes where the emphasis is on creating something quickly. The output of such an process is a prototype or basis model from which further models and eventually the final product will be derived. Gershenfeld (2012), Berman (2012) and Reeves (2008) validate this by saying; "the additive manufacturing process has proven to be compatible with industrial manufacturing beyond prototyping". Nowadays, rapid prototyping is in many cases not the correct word since in a wide variety of industries, products developed with rapid prototyping, are used as a final product instead of a prototype. The reason for that is the development in technologies which are used and the improved output quality. Another comment adding to this is that the term rapid prototyping also overlooks the basic principle of these technologies in that they all fabricate parts using an additive approach. According to Gibson. I, Rosen. D & Stucker. B, (2015) a technical committee within ASTM International agrees that new terminology should be adopted. Instead of rapid prototyping, Additive manufacturing should be used. Concluding, Additive manufacturing is a collective name for additive production processes, such as 3d printing, which are typical processes which are used to produce products layer-by-layer.

Process of additive manufacturing

The basic principle of this technology is that a three-dimensional Computer-Aided Design (3D CAD) can directly be produced without the need of a detailed process planning. In this chapter the process will be described more into detail. The steps which are in the additive manufacturing process are as following:

1: Cad model
2: Conversion to STL
3: File transfer to machine
4: Machine setup
5: Build
6: Remove
7: Post process
8: Application

These steps will now be elaborated more into detail.

Step 1: CAD Model
All additive manufacturing processes start with a computer model. This model, a 3D CAD model, can be made with almost every professional solid modeling software, as long the output is a 3d solid model or surface representation. This surface representation or solid model can also be generated by scanning a physical object.

Step 2: Conversion to STL
Almost every additive manufacturing machine does accept STL files. STL is a file format which can be generated with CAD software. The file describes the geometry of the CAD model and forms the basis for the printing process.

Step 3: file transfer to machine
Once the file is transferred to the machine, it needs often to be manipulated. This to correct the size, position on the bed and orientation for building. This to make the print possible and time effective.
Step 4: Machine Setup
Once the STL file is positioned in the right position, printing parameters can be chosen. These parameters are like material constraints, energy source, layer thickness, timings, bed temperature, nozzle temperature, cooling of the material after printing, etc. etc. Before starting the print process, it is wise to check if all needed maintenance is done on the machine. The machines use often delicate technologies such as lasers or leveling devices, all require different levels of maintenance.

Step 5: Build
Now all configurations have been done, the device can start printing. The printing process itself is an automated process which can happen nearly without supervision. The only check which is needed is superficial monitoring of the machine, to make sure no errors have taken place, running out of materials, software issues or power related issues. Figure 13 shows an image of the FDM process, which is a commonly used printing methodology based on plastics.

Step 6: Remove
Once the print is finished the 3D model needs to be removed from the printer bed. In some cases, some safety measures need to be taken in order to make sure temperatures are low enough or moving parts will be shut off.

Step 7: Post-processing
3D printers are capable of printing highly complex shapes. In order to do this, printers need sometimes to print extra supporting material. This stage is about removing support material and cleaning the model.

Step 8: Application
After cleaning of supporting material of excess material, the printed part might be ready for use. In some occasions the printed parts need additional treatment in order to upgrade the surface quality. This can be done with priming or painting in order to give an acceptable surface quality. The printed part might also be assembled together with other products.

Why additive manufacturing?
Now the process of additive manufacturing is described, we can look for an answer as to why apply this technology. What makes additive manufacturing interesting to use?

If additive manufacturing is compared with technologies such as injection molding or cutting based technologies we can see some interesting differences based on production costs and production speed. Injection molding is a technology which is used a lot for mass production. The technology requires costly molds in order to produce products. In contrast to that, additive manufacturing is a technology which doesn’t need molds in order to start production. The starting costs of the additive manufacturing are reasonably low. This makes additive manufacturing costs effective for small production runs (Berman, 2012). Other sources stating that additive manufacturing is an industrial manufacturing process with the potential to significantly reduce resource and energy demands as well as process-related CO₂ emissions per unit (Baumers, Tuck, Wildman, Ashcroft & Hague, 2011; Baumers, 2012; Campbell, Williams, Ivanova & Garrett, 2011).
However if the production scale increases, injection molding will become cheaper and more time effective. The benefit of this is that complex shaped products, which are expensive to produce molds for, can cheaply be produced with additive manufacturing.

If additive manufacturing is compared with subtractive technologies such as CNC milling, we can conclude that cnc milling has also low startup costs. The difference however, can be found in the use of material. Since subtracting technologies take material away in order to shape the product. In contrast with that, additive manufacturing adds material to shape a product. The result, additive manufacturing uses less material and can reduce up to 40% in comparison with subtracting technologies (Berman, 2012). Additional to that, most of the excess material which is used in additive manufacturing can be reused. Petrovic et al., (2011) stated that up to 95% - 98% of waste material can be recycled in 3-D printing. In comparison with subtractive technologies, where up to 96% of the raw material can be wasted when creating a product (Wagner, 2010).

Production costs in additive manufacturing do not change in case production amounts scale up. In case of injection molding or cutting based technologies, production costs per unit do decrease in case the production scale increases. However, in small production runs, the production process in additive manufacturing can start directly, without the need to wait for the set-up time for for example the production of molds.

The break-even point of cost effectiveness in production scale is according to Sedacca (2011) in-between 50 to 5000 parts, based on plastic injection molding. Another source states that additive manufacturing is cost effective up to a production run of 1000 parts (“Print me a Stradivarius: How a new manufacturing technology will change the world,” 2011).

To summarize, the advantages of 3d printing in comparison to other technologies are as following:

**Additive manufacturing pro’s**

+ Can economically build custom products in small quantities as if mass production were used.

**Sources of cost effectiveness include:**

- No need for costly tools, molds, or punches
- No scrap, milling or sanding requirements
- Automated manufacturing
- Use of readily available supplies
- Ability to recycle waste material

+ Ability to easily share designs and outsource manufacturing.

+ Speed and ease of designing and modifying products.

+ Faster compared to hand built models, and more accurate

+ Lower energy demands

+ Less CO₂ emissions

**Additive manufacturing con’s**

- Only cost effective on small production scale ( < ~1000 parts)

- No reduction in production costs if production series increases

- Relative long production time for parts, compared with injection molding
Additive manufacturing methods

Now the main advantages of Additive manufacturing have been elaborated it is time to go elaborate on the different typologies of additive manufacturing. Hopkinson, Hague & Dickens (2006) have explained 18 different methodologies, of which 15 will be elaborated, which can be accommodated in 3 different categories. These categories are:

- Liquid based systems
- Powder-based systems
- Solid-based systems

Within the category of Liquid-based systems the following processes are known:

- Stereolithography
- Jetting systems
- Direct light processing technologies
- High-Viscosity Jetting
- The MAPLE process

Within the category of Powder-based systems the following processes are known:

- Selective laser Sintering (Polymers)
- Selective laser Sintering (Ceramics and metals)
- Direct metal laser sintering
- Three-Dimensional printing
- Fused metal Deposition systems
- Electron beam melting
- Selective laser Melting
- Selective masking Sintering

Within the category of Solid-based systems the following processes are known:

- Fused depositioning modelling
- Sheet stacking Technologies

In this chapter the stated technologies will be explained in order to understand these processes.

1. Stereolithography
This process is known as the founding process withing the field of additive manufacturing. Figure 14 shows a schematic image of the process. An ultraviolet laser is used to initiate the curing reaction in a photocurable resin. A CAD file is used to drive the laser in order to cure a selected portion of the surface of a vat resin, in order to cure and solidify the resin on a platform. The platform is then lowered and a new layer of liquid resin is deposited over the previous layer. The laser then scans a new layer which will be bond to the previous layer. In some cases support material is needed, which will be generated by the printer in such a way it can be removed easily after the print is finished.

Figure 14: Schematic of the stereolithography process

2. Jetting systems
Two different technologies that use inkjet technology to create parts from photocurable resins have been commercialized. These are the polyjet process and the invision process. This report will explain the Polyjet process.
The process uses an array of printing heads to simultaneously selectively deposit an acrylate-based photo-polymer. Each layer is then cured by trailing UV lamps that pass over the deposited material. Supporting material will simultaneously be deformed through a second series of jets and cured to a gel state with the UV lamp, so it can be removed with water jets or similar after building.

3. Direct Light Processing Technologies
This process is special because the process builds parts that grow downwards rather than upwards. The process builds parts from an acrylate-based photocurable resin, but it does so by using a two-dimensional matrix of mirrors rather than a 1D array of print heads to selectively cure the material. The process makes use of Digital mirror devices to switch on and off mirrors that reflect UV light from a source on the build area. Figure 15 shows a schematic of the process.

4. High viscosity jetting
The principle involves continuous change in a layered pattern according to a very thin slice of the object to be printed. This uses a mechanism based on displacing a small drop of a printable material to a desired location on the substrate. The fundamental unit consists of a single jet, which is controlled by pressured air, the distance from the substrate and the length of the jetting pulse. The accuracy of the process is dependent on the size of the hole which is being used for the jet. Figure 16 shows a schematic of the process.

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Figure 15: Schematic of Direct light processing technologies

Figure 16: Schematic of High-viscosity Jetting process
5. The MAPLE process
Maple stands for Matrix Assisted Pulsed Laser Evaporation. The MAPLE process uses a high-repetition-rate, 355nm UV laser beam which is focused on a transparent material or ribbon that has a thin layer of build material on the underside. As the laser energy is directed to the ribbon the build material transfers to the receiving substrate. Figure 17 shows a schematic of the process.

![Figure 17: Schematic of the MAPLE process](source: Hopkinson, Hague & Dickens (2006))

**Powder based processes**

6. Selective laser sintering (polymers)
Selective laser sintering process is in many ways similar to stereolithography, but the powdered raw material is sintered or melted by a laser that selectively scans the surface of a powder bed to create two-dimensional solid shapes. A fresh layer of material is then added to the top of the bed so that the laser can scan a new two-dimensional shape which bonds the new layer to the material underneath it. The uncured material serves as supporting material which can easily be removed because it remains as powder. These steps are repeated until the final shape is achieved. The bed is typically heated to a temperature that is typically a few degrees below the sintering temperature. This is mostly done by infrared heaters and helps the process by reducing thermal gradients between sintered and non-sintered material. The goal of this is reducing the amount of energy which is needed by the laser to sinter the powder. Figure 18 shows a schematic image of the Selective Laser Process.

![Figure 18: Schematic of Selective Laser Sintering (polymers) process](source: Hopkinson, Hague & Dickens (2006))

7. Selective Laser Sintering (ceramics and metals)
This process uses similar technology compared to the polymer variant. However, the material is different. The technology uses metal powder which is coated with a polymer resin which acts as binder. The function of the coating is acting as binder material which can be fused by the laser. After the shape is made in the printer, the final part is put in an oven in order to burn away the polymers which are in the material. This process causes that cavities occur in the material. The heat treatment also helps the metal parts to sinter and bind. After this stage the final shape is treated with bronze, in order to infiltrate the porous parts.

8. Direct metal laser sintering
This process is different compared to the previous two processes in the fact that the material which is used is different. The material can be used without the need of a polymer coating. The metals for the direct metal laser sintering process involve either melting or liquid phase sintering of the metal powder, which is typically a mixture of various components having different melting points.
9. Three-Dimensional Printing

Using 1D jetting technology the process has a relatively high throughput in terms of creating green parts similar to those by metal selective laser sintering. The post processing is similar to that for selective laser sintered parts, but surface finish usually requires some form of machining to create a surface suitable for tooling. The process is therefore suitable for applications where fine surface quality is not required. Figure 19 shows an schematic image of the process.

10. Fused metal deposition systems

This process uses the principle of blowing metal powders into a melt pool created by a laser. In general these processes have relatively slow deposition rates and produce parts with poor surface finish, but they do offer the potential to process functionally graded materials in high melt temperature metals such as titanium. Image 20 shows an schematic image of the Fused metal deposition system process.

11. Electron beam melting

This process uses a similar approach to selective laser sintering but replaces the laser with an electron beam, with the following reasons. First of all the electron beam may be directed by changing the electromagnetic field through which it passes. This eliminates the need for scanning mirrors and can significantly increase scanning speed. Secondly, the power developed by the electron beam is very high, allowing the process to fully melt a wide range of metals including titanium alloy using a very fast scanning rate. However, the process is limited to conductive materials and surfaces. As with many other layer-based processes, it often requires extensive finishing, especially for tooling applications.

12. Selective laser melting

This technology uses a laser to fully melt stainless steel parts in a similar manner to laser sintering. The process is particularly adept at producing very small components, including ones with complex lattice structures.
13. Selective masking Sintering

The Selective masking sintering process relies on printing a mask of infrared radiation reflecting material on to a glass sheet and placing the sheet over a powder bed. The next step is applying infra red radiation to the glass sheet and selectively allowing it to pass through the mask in order to sinter the powder directly underneath it. Figure 21 shows an image of the Selective masking Sintering process.

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Solid based processes

14. Fused Deposition Modelling (FDM)

The Fused Deposition Modelling process creates parts by extruding materials through a nozzle that moves in X and Y direction to create a two- dimensional layer. The machine might have 2 different nozzles which deposit material that forms the parts and material that forms the supporting material where required. Nozzle sizes can vary and is depending on the accuracy which is needed. A small nozzle size gives a high resolution approach of the desired shape, but will take a high time-frame to build parts. In contrast to that, a large nozzle size will result in a quick printing process but the resolution of the printed part will be lower. Support material can be manually removed, or in case a soluble material is used, removed with water or an acid in order to dissolve the material. This process is one of the most used processes in additive manufacturing and can be done with rather small desktop machines. Figure 22 shows an image of the FDM process.

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Figure 21: Selective masking sintering process source: Hopkinson, Hague & Dickens (2006)

Figure 22: Fused Deposition Modelling process source: Hopkinson, Hague & Dickens (2006)
15. Sheet Stacking Technologies
This methodology is based on stacking two dimensional sheets in order to form a three dimensional object. The materials which could be used are various, for example paper or cardboard. Before the sheets are bond together, the upper sheet will be cut into the desired shape. This cutting can be done by lasers or a milling machine. Because each layer is cut in the desired shape, the waste material can be removed manually after the print is finished. In order to bind the layers, an adhesive is used. One of the disadvantages of this method is that it is hard to remove excess material in case a model has thin walls. The removal of material may cause damage to the desired model. Figure 23 shows an image of the Sheet Stacking Technologies process.

Figure 23: Sheet Stacking Technologies process source: Hopkinson, Hague & Dickens (2006)
Summary

Additive manufacturing is a technology which is used to create objects based on a CAD model. The method does this by splitting the cad model into thin layers which are then built layer-by-layer. The processes which could be applied are various, according to the literature survey on different typologies. However, regarding the aim of this research, not every method will be suitable. Depending on the outcome of the research a new technology needs to be designed, or a current technology can be used, maybe even a combination of both.

In general additive manufacturing has a lot of potential, regarding its benefits in comparison with existing technologies. Also additive manufacturing has some disadvantages. The main advantages are stated below:

+ Can economically build custom products in small quantities as if mass production were used.
+ Ability to easily share designs and outsource manufacturing.
+ Speed and ease of designing and modifying products.
+ Faster compared to hand build models, and more accurate
+ Lower energy demands
+ Less CO₂ emissions

The technology has also its disadvantages or ‘play rules’ which should be kept in mind because another production method may be more economically feasible. The main disadvantages are stated below:

- Cost effective on small production scale (< ~1000 parts)
- No reduction in production costs if production series increases
- Relative long production time for parts, compared with injection molding.
Cellulose
Part 3
As mentioned in part one, cellulose is the material of which cells are made in wood. This material can be seen as world’s most available material and is a sustainable resource for various materials. This chapter will explain a little more about the processes needed to extract cellulose from wood. Secondly it will explain a bit more about the material properties of cellulose.

**From wood to paper, the pulping process**

Paper production is basically a two step process in which a tree, the primary source of cellulose, is converted into a pulp, and then the pulp is processed into paper. The first step of the process is used to treat the wood in such a way that the unusable parts within the wood are removed from the usable elements in wood. In order to make paper, cellulose is used and lignin is the part that is considered to be waste material. Pulping can be done in various ways, for example mechanically or chemically as is done in the kraft pulping process. After the pulping, the pulp needs a next treatment like bleaching in order to give the paper its nice white appearance, also depending on the type of paper and the end goal of the paper. After this step, the pulp is dried and pressed into sheets.

The paper and pulping industry is one of the most important industries in the world. Nowadays the industry serves about 5 billion people worldwide (Bajpai, 2015). Nowadays, the process is dominated by machines whereas in the early history of paper it was a time consuming process. The machines of today can make paper at a rate of 2000m/minute width a with of about 8 meter at once. The paper sheet is mainly made of cellulose fibers with some additional components pressed into a sheet. But what does the process look like? The paper-making process has been known since 105 AD and was then invented and reported to the Chinese emperor Ts’ai Lun. The process as we know today is modernized and developed but in essence the process still consists of the same steps;

- Pulping the wood, and separate the fibers.
- Beating and refining the fibers
- Diluting to form a thin fiber slurry
- Suspending in solution

- Forming a sheet of fibers on a thin screen
- Pressing the material in order to increase the density
- Drying the pressed sheet in order to remove moisture
- Finishing the sheet in order to provide the requested Surface quality

In figure 24 the pulping process of a modern production plant is schematized. Now we will explain the steps a little more in order to get a better understanding of the chemical production process of cellulose.

**0 preparing wood**

Wood is the material which is used the most in the paper pulping industry. First step in the process is cutting the trees and preparing them for the paper-pulping process. One of the first steps after cutting the tree is debarking the tree. After the debarking process a tree will be chipped into smaller parts in order to make the material suitable for the pulping process. Chips will get screened for size and too big chunks of wood will be chipped again. The chips will then get stored, until they enter the pulping process. These steps are needed for almost every pulping process and can be seen as an independent pre process for chipping.

**1 pulping**

The chips will now be transported to the pulping process which is shown in figure 24. During this process the cellulose is separated from the lignin using a chemical process (Kraft, Soda or Sulphite). The first step is cooking the wooden chips in a solution of aqueous chemicals under elevated temperature conditions and a higher pressure level. The kraft process uses alkaline cooking liquor of sodium hydroxide and sodium sulphide as the chemicals. These chemicals are used to digest the wood and the mix is called white liquor and is mixed with the chips in the digester system (1). The kraft process is by far the most common pulping process used in the industry (about 75% of all pulp produced). The reason for the position of the kraft process is the resulting fiber strength of the end product and the advantages of chemical recovery in the process.
2 separating
After the cooking process the chips are softened and will enter the knotter system (2). This happens under pressurized circumstances. As the mass of softened, cooked chips impacts on the tangential entry of the knotter system, the chips disintegrate into fibers, named pulp, and the residual black liquor.

3 pulp washing
The next step is to put the pulp and black liquor in a series of washing machines which will separate the pulp fibers from the black liquor. The aim for this step is to separate the pulp from unwanted solubles with as little washing water as possible. After this step the black liquor and cellulose fibers are separated, and the black liquor gets stored to be used as cheap fuel with the aim to recover the chemical compounds (10) & (11).

4 grading the pulp
The pulp resulting from step 3 gets graded in the screening system (4). Here the pulp gets separated in different qualities depending on the grade of product. The aim of this process is to separate the pulp from unwanted particles like uncooked chips and small fiber bundles that have not been separated by the chemical process, but also to separate the different fiber qualities.

5 bleaching
Unbleached pulp fibers are known to have a dark brown color and are generally used for packaging products like cardboard. These fibers have in general a higher amount of lignin which is still in the pulp. Bleached pulps are commonly used for the production of white papers (Bajpai, 2015).
The bleaching process is carried out in multiple steps which vary in delignification and dissolved material extracting steps. In the oxygen delignification system (6), oxygen or hydrogen peroxide-based solutions are used in order to reinforce the extraction operation of residual lignin in the pulp. The result of bleaching is that the properties of the fibers change. Bleached fibers, those where residual lignin had been removed, have an increased flexibility and strength, and have a lower hemicellulose content. The bleaching step is a delicate process because it could also damage the fibers when it has been done too severe. In the next step the fibers enter the bleaching system (7) where chemicals such as chlorine are used to bleach the pulp.

6 Cleaning
In the next step, refiners and cleaners (8) clean the bleached pulp from residual chemicals and deliver the cellulose fibers in such a way it can be used for the production of paper. This step also modifies the morphology and surface characteristics of the fibers. The fibers are now ready to be used for paper production.

Material properties of cellulose

The molecular formula of cellulose is \((C_6H_{10}O_5)_n\) and is a homopolymer of glucose. The chemical composition of cellulose is shown in figure 25. As the molecular formula already explains, cellulose is made of Carbon, Oxygen and Hydrogen. Cellulose is non-toxic, biodegradable and a homo-biopolymer. In its pure state cellulose has a white color and a density of 1.52 - 1.54 g/cm\(^3\). Cellulose is known to have a High tensile and compressive strength, exact numbers are depending on the original source (wood, hemp, flax, cotton).

Cellulose is known to be a non-thermoplastic polymer. This means it won’t melt, but will burn. Cellulose is very much stable in thermal conduction and is known to be insoluble in water.

Figure 25: Overview of Cellulose molecule
Source: Bowyers, Shmulsky & Haygreen (2007)
As mentioned earlier in part one, lignin is one of the natural components in wood and can be found in-between and in cellulose cells. In this chapter more information will be given about lignin. First of all, available quantities of lignin will be described, secondly the process of isolation of lignin will be described, and finally some material properties of lignin will be described.

**Lignin in abundance**

Paper is a product that is made from trees, and in this production process, the pulp process, cellulose and lignin are separated. The cellulose is used for paper, and the lignin is left over as part of a low quality residue. Luckily, more and more research is done in order to see what can be done with the leftover lignin in order to increase profitability (Doherty, Mousavioun & Fellows, 2011). Nowadays, most of the lignin captured in residue of the kraft process is burned as fuel. The material has a huge energy and carbon reserve but due to its complex and recalcitrant nature it is often regarded as waste. Hu, Zhang & Lee, (2018) state that an annual quantity of 150 billion tons of lignin is produced that has an energy potential of 3000 EJ/ yr, equivalent to 0.082 % of all the solar energy received by earth’s surface and about 5.4 times the amount of energy consumed on earth.

The total amount of lignin produced in paper mills, ending as waste, is estimated to be around 50 million tons per year. Lignin from softwood pulping is available in commercial quantities and it can be assumed that this amount will rapidly increase in the future. Therefore development of material systems based on softwood kraft lignin should be beneficial for the future sustainable society and add value to a renewable resource (Gellerstedt, 2015).

**Material properties**

As mentioned in the chapter above, lignin is often considered to be recalcitrant and complexly organized. Figure 28 shows how a lignin molecule could be found in wood. Because lignin occurs in a variety of forms the exact configuration of a molecule is still uncertain. Forss & Fremer (2003) describe lignin as a molecule made of mostly glycolignin, aromatic polymers, and an ordenen polymer made of a repeating unit consisting of 18 phenyl-propane units. What might be noticed, looking at figure 28, is the large amount of repetitive elements in the lignin molecule. Lignin is made of small building blocks which could have a setup as is shown in figure 26 and 27. The building block which is shown in figure 26 is a building block which could be found in hardwoods and in softwoods. Figure 27 shows a building block as can be found in hardwoods (Bowyers, Shmulsky & Haygreen, 2007). The difference between the molecules can be found in the extra CH₃O element which is attached to the building block.

![Figure 26: Lignin building block, part of a lignin molecule](source: Bowyers, Shmulsky & Haygreen, (2007))

![Figure 27: Lignin building block, part of a lignin molecule](source: Bowyers, Shmulsky & Haygreen, (2007))
Figure 28: Lignin molecule as can been found in wood.
Source: Bowyers, Shmulsky & Haygreen, (2007)
However, for this thesis, material properties of the material lignin as a residual product of the paper industry are important. Fitigau, Peter & Boeriu (2013) did research on lignin based on the structural analysis and checked the solubility of various types of lignin. The types of lignin which were used are Soda lignin recovered from wheat straws, P1000 Lignin recovered from Sarkanda grass, Alcell TM organo-solv lignin recovered from mixed hardwoods, Kraft lignin, Indulin AT, recovered from softwoods, and Alkali-pretreated wheat straw lignin. In their research, they checked the solubility of lignin in a fluid made of acetone and water. They tested the solubility of the different types of lignin in various mixtures of the fluid made of acetone and water. The results showed clear results which are represented by figure 29.

**Recovery process of lignin from black liquor**

Black liquor is the residual product of the kraft pulp process, which is nowadays mostly used as cheap fuel as explained on the previous pages. In order to refine the lignin which is captured in it, a process like the ANDRITZ lignin recovery process which is described in figure 30 is needed. The system is designed to remove about 35-45% of dry solids with a typical pH of 12 - 13. After the filtration and washing steps, the dry content of lignin is typically raised to a level of about 60-62% before it enters the drying stage. After the drying stage, where the moisture will be removed, about 95% of the solids will be composed of lignin. When the process for lignin recovery is applied in its most complete form, it consists of the following sub processes:

1. **Precipitation**

   The black liquor, as residual product from the evaporation plant in the kraft process, is treated in the acidification reactor with CO$_2$ with the aim to lower the pH value. This results in precipitation of the lignin from the black liquor. The next step is to separate the lignin from the Lignin-lean black liquor with a membrane filter press (Pehu-Lehtonen (2017)).

2. **Acid wash**

   The separated lignin contains impurities like sodium which are leached from the compressed precipitated lignin with the help of a dilution wash of sulfur acid. A technique which is called displacement washing is then used to remove the sodium sulfate formed during the acid wash. After this step, the lignin is put through a second membrane filter press in order to dewater the slurry (Pehu-Lehtonen (2017)).

3. **Drying**

   After phase 2, the lignin which is still a bit moist is put through a disintegration in order to minimize the size of chunks of lignin so the surface area is increased before the lignin enters the dryer. Rated from the black liquor and ready to use for multiple purposes. The heat source which is used for this drying process is mainly the flue gas from a recovery boiler or a lime kiln which also acts as a passive drying media. After these processes, the lignin is separated from the black liquor and ready to use for multiple purposes (Pehu-Lehtonen 2017).

![Figure 29: Graph showing effect of acetone concentration on lignin solubility at 23 celcius degrees](image)

Source: Fitigau, Peter & Boeriu (2013)
Figure 30: The separation process of lignin from black liquor based on an ANDRITZ lignin recovery process which is suitable for kraft pulp processes. Source: Pehu-Lehtonen (2017)
Reference projects

Part 5
Reference projects

In this chapter some reference projects will be elaborated. The aim of this research is to explore the possibilities of using wood in an additive manufacturing process. With this aim in mind, some reference projects were found which potentially can give a good starting point for this research to do further research into.

1. Wood based bulk material in 3D printing processes for applications in construction.

The first article is written by Henke & Treml, dated from 2012. The title of the article is Wood based bulk material in 3D printing processes for applications in construction.

Henke & Treml 2012 used wood based bulk material, such as sawdust, wooden chips etc. In an additive manufacturing process in order to form large scale solids. The way they did this was applying thin layers of bulk material on a bed, with a bonding material in between. This way they formed solids of almost any desired shape.

The goal of their research was to investigate whether it was possible to combine mineral materials with plant based materials in an additive manufacturing process in order to derive materials which are relatively lightweight and reasonable good insulators compared to mineral materials such as cement. The research was done with special regard to applications within the field of construction.

Materials and methods

The research was conducted with wooden chips, made of spruce, as they are used for the production of particle boards. The particle size was tested and stated between 0,8 mm and 2 mm. The density of the chips was 192 kg/m³. Gypsum, methyl cellulose, sodium silicate and different types of cement were used as bonding materials.

In case of the binding materials gypsum, methyl cellulose and cement the wooden chips were first mixed with dry binding material. The mixing ratio was varied in order to fill voids. The next step was to apply a thin layer of material on the bed and use a mask in order to spray aer-

olised water onto the material which was used as activator. The mass of activator which was needed was controlled by a scale. In case of the binding material sodium silicate, the chips were applied on the bed, and the binding material was applied locally into the chips. After the first layer was bonded, the platform moved down by the thickness of one layer and the process repeated with a smaller mask. Once the final layer was finished, the bed was moved back in its starting position and the solid body could be dismantled. Figure 31 shows the process and figure 32 and 33 show the results.

Results

Henke & Treml (2012) described that tests with methyl cellulose led to poor mechanical behavior. Also the first trials with sodium silicate turned out to be difficult, because the grain boundary voids couldn’t be filled adequately. Good results were achieved with gypsum with a chips/gypsum ratio of 0,33 and water/gypsum ratio of 0,75 and the process led to a good contour accuracy and good consolidation of the chips.

The best results were achieved with cement used as a binder material. In this process a broader range of mixing ratios were tested. On basis of these tests a chips/cement ratio of 0,15 and a water/cement ratio of 0,8 was used to deliver the best results.

However, Henke & Treml (2012) stated that further optimization was needed in order to make this technology suitable for a broader range of applications. For example more optimization of the raw material and optimization of the process itself was needed.

Conclusion

This research what was done by Henke & Treml (2012) proved that there is a way to use wood in an additive manufacturing process. However, they used wood as a fill material instead of producing wood itself. Also the material behavior of the final material is different compared to the material behavior of wood. For example the anisotropic character of wood can’t be found in this product as well as the aesthetic character of wood.
Figure 31: Image of process used by Henke & Treml (2012)  
source: Henke & Treml (2012)

B = mixture of bulk and binder, P = platform, M1-Mx = masks, A = activator, D = drive and control

Figure 32: Image of material created by Henke & Treml (2012). Cone is produced with gypsum as binder material  
source: Henke & Treml (2012)

Figure 33: Inner structure of cone with bonded with cement (top) and gypsum (down)  
source: Henke & Treml (2012)
2. Hydrodynamic alignment and assembly of nanofibril resulting in strong cellulose filament

This research was done by Håkansson et al., (2014) and they used nanofibrils in order to create cellulose filaments. Håkansson et al., (2014) stated the following:

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"Cellulose nanofibrils can be obtained from trees and have considerable potential as building block for bio-based materials. In order to achieve good properties of these materials, the nanostructure must be controlled. Here we present a process combining hydrodynamic alignment with a dispersion-gel transition that produces homogeneous and smooth filaments from a low-concentration dispersion of cellulose nanofibrils in water. The preferential fibril orientation along the filament direction can be controlled by the process parameters. The specific ultimate strength is considerably higher than previous reported filaments made of cellulose nanofibrils. The strength is even in line with the strongest cellulose pulp fibers extracted from wood with the same degree of fibril alignment. Successful nanoscale alignment before gelation demands a proper separation of the timescale involved. Somewhat surprisingly, the device must not be too small if this is to be achieved."
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Håkansson et al., (2014) claim to have developed a continuous and potentially industrially scalable and parallelizable method that prepares strong and stiff cellulose nanofibril-based filaments. The process is realized using a millimeter sized flow focusing system, designed in a nozzle. The alignment of the nano fibrils is a result of Brownian diffusion, a process which was first seen by the botanist Robert Brown and later on described by Albert Einstein and published in a paper about this phenomenon in 1905. The theory explains why particles in water are moving and the definition of Brownian diffusion is as following:

Brownian motion is the random motion of particles suspended in a fluid (a liquid or a gas) resulting from their collision with the fast-moving atoms or molecules in the gas or liquid. Because shape of the nozzle, the fluid in the channel is forced to increase speed, given a continuous pressure at the beginning of the chanel and a decrease in radius along the nozzle. This increase of speed, causes the nanofibrils to orientate in the flow channel. This process can be seen in figure 34. According to tests done by Håkansson et al., (2014) the speed is important, and lower speed in the channel causes less alignment of fibers. At the end of the chanel, when all fibers are aligned, the fibers need to be fixed before Brownian diffusion can mess up the alignment of the fibers. This is done by creating a gel dispersion in the fluid of Na+ and NaCL. The gel is ejected into a water bath in order for the electrolyte to diffuse out of the gel. After 24 hours the gel filament is transferred to an acetone bath where it is fixed, after which the gel filament is taken out of the bath and fastened in both ends to dry. After drying, the filaments are assumed to be homogeneous cellulose materials.

The process parameters, such as the working mechanisms and the timescale in the process is generic and also applicable on other anisotropic materials which have a fixed shape, such as other types of fibrils, fiброins or even organic polymers. Using this technology in the correct way, anisotropic materials could be made with similar properties to the natural material.

Conclusion

This technology could be the key to forming wood in an additive manufacturing process. Wood has an anisotropic character, which can only be printed if wood fibers, (i.e. cellulose) are aligned in an printing process. Using this technology, fibers can be aligned in a similar way like they also appear in natural wood. However, this research achieved the alignment of fibers and managed to produce homogeneous cellulose with the help of a gel formed out of Na+ and NaCL. Wood exists out of cellulose, hemi cellulose and lignin and an interesting question would be if this process is also possible with lignin as fluid. Further research on this question is needed in order to give a correct answer to this question.
The nanofibrils in the focused flow are illustrated as rods (the fibril length in relation to the channel width is exaggerated by approximately a factor of 300). The diffusion of Na\(^+\), from addition of NaCl in the focusing liquid, is illustrated with a grey tint. The rows of small images above and below the central image illustrates the hydrodynamical, molecular and electrochemical processes involved. (a) Brownian diffusion (illustrated with the dashed arrows) affects the orientation of a single fibril, (b) hydrodynamically induced alignment (illustrated by solid, grey, arrows) occurs during the acceleration/stretching, (c) Brownian diffusion continues to act after the stretching has ceased, (d) Brownian diffusion is prevented by the transition to a gel. The lower row of small images illustrate how the electrostatic repulsion (illustrated by the red area representing the Debye length) decreases from e to h as the Debye length is decreased with increasing Na\(^+\) concentration or by protonation of the carboxyl groups on the fibril surface.

source: Håkansson et al., (2014)
3. Stratified Additive Manufacturing using Wood Veneer

This process is based on the technology explained in the previous chapter. The technology which is used is the methodology of sheet Stacking Technologies. This methodology is based on stacking two dimensional sheets in order to form a three dimensional object. The materials which could be used are various, for example paper or cardboard. In this case veneer could be used, which is paper thin wood. Before the sheets are bond together, the upper sheet will be cut into the desired shape. This cutting can be done by lasers or a milling machine. Because each layer is cut in the desired shape, the waste material can be removed manually after the print is finished. In order to bind the layers, an adhesive is used. In this case it would be nice to use a lignin based glue, but further research on that topic needs to be done.

MCor builds paper based process machines (http://www.mcortechnologies.com).

On the next page the process is elaborated step by step. The images are retrieved from a video which was posted by Mcor.

This process is nice but it has its disadvantages. For example, the process doesn’t deal with excess material. In case wood is used, how do we deal with the excess material?

Figure 35: step 1: a CAD model is needed of the design which has to be built. In this case a hammer.

Figure 36: step 2: hammer is divided into slices with the same thickness as the material, In this case paper. The colour is printed onto the paper in order to give it some esthetics.

Figure 37: step 3: all sheets are put in the machine, which is now ready to start producing.

Figure 38: step 4: the machine takes a sheet and puts adhesives on it. At the location of the hammer a lot, next to it, on the excess material, less.
Figure 39: step 5: the machine takes a new slide and puts it on top of the previous layer.

Figure 40: step 6: the machine presses the layers together.

Figure 41: step 7: the machine starts cutting so the object can be removed easily from the excess material. Now the process can repeat from step 4 till 7, until the process is finished.

Figure 42: step 8: Now the machine is finished, 'a pack of paper' is ready to be unpacked.

Figure 43: step 9: Now the printed object can be separated from the excess material.

Figure 44: step 10: Once finished, the product is ready to use.
Sumary

This report is about the question if wood can be used in an additive manufacturing process. The use of wood in an additive manufacturing process would open new options in the production of parts. Also additive manufacturing can outsource labor and help in a sustainable way by reducing waste material. Especially the sustainable part in combination with opening new possibilities in production are relevant topics.

Based on the literature study on wood we can conclude that wood is a building material delivered by nature. The world’s largest and oldest living organisms are able, with the help of sunlight, to produce sugars which are converted to building material for trees. With these building materials they produce cellulose and hemicellulose. With these components and lignin, which comes as a third ingredient, trees are able to reach to the sky. Trees exist in two main divisions, hardwoods and softwoods. Both have their own characteristics. Softwoods for example, also named needle trees, have relatively large cells which have a double function. These functions are transportation of fluids in the stem and maintaining strength. In hardwoods this is organized differently. First of all, hardwoods have vessel elements which take care of the fluid transportation in the stem, secondly the wood fibers take care of strength and third, wood cells which occur in hardwoods are relatively small compared to the ones in softwoods. Besides these differences, there are some other distinguishing facts. Softwoods have small needle like leaves, which will remain during winter. The reproduction of these species happens with seeds which are on the surface of typical cones. The seeds are not protected as they are in case of hardwoods. They produce small fruits which act as a small chamber where inside the seeds are hidden. Furthermore, hardwoods have big leaves which in most of the occasions will drop during winter.

The aesthetics of wood are determined by a set of factors. First of all trees have annual rings, although sometimes they are not visible. These annual rings can be split up in earlywood rings and latewood rings. The occurrence of these two types of annual rings depends on the seasons during the year. Secondly trees have heartwood and sapwood. These are mostly clear distinguished by their colour. Heartwood is mostly colored dark as result of extractives in the wood which protect the tree from decay and insects. Sapwood is rather light colored and can’t withstand decay or insect attacks simply enough because it doesn’t have extractives in it. Furthermore, wood porosity causes aesthetic effects on the wood because you can mostly see these tiny holes in the transversal surface but also in the radial surface of the stem. Wood is known as an anisotropic material which means it behaves differently in the direction along the grain and the direction perpendicular to the grain. This is something that can be clearly noticed when structural tests are done on wood. The ratio between the two can vary up to a ratio of 20 where the loading capacity along the grain is the highest.

Additive manufacturing has developed over the past decades into a widely used technology not only for prototyping but also for delivering market ready products. The technology is widely used with materials such as plastics, metals and mineral materials such as cement. Strangely enough, the combination of wood and additive manufacturing has not been developed yet. Additive manufacturing is a production technology which is based on a CAD model. This computer model is a graphical model of the desired product which is used as input for additive manufacturing technologies. The model is converted into a STL file which can be used by additive manufacturing machines. The machines split the model up in small layers, which are then built layer-by-layer. This process is called additive manufacturing. The advantages of this production methodology are stated below:

+ Can economically build custom products in small quantities as if mass production were used.
+ Ability to easily share designs and outsource manufacturing.
+ Speed and ease of designing and modifying products.
+ Faster compared to hand build models, and more accurate
+ Lower energy demands
+ Less CO₂ emissions
The technology has also its disadvantages or ‘play rules’ which should be kept in mind because another production method may be more economical feasible. The main disadvantages are stated below:

- Cost effective on small production scale (<~1000 parts)
- No reduction in production costs if production series increases
- Relative long production time for parts, compared with injection molding

Over the years many different technologies have been developed for example FDM printing, SLS printing and layer stacking technologies. However, wood has hardly been used in these processes.

Based on some reference projects we can conclude that wood has been used as a filling material in additive manufacturing processes. However this is not exactly in line with the goal of this research. From the second reference project, it seems that researchers have invented a way to make high quality cellulose filaments, which have an anisotropic character. This is an interesting development which might result in the fact that people will be able to print wood in the future. However this filament was formed in a gel of Na⁺ and NaCL. Further research is needed to check if it is possible to use this process in combination with lignin.

The third reference project is based on the methodology of paper stacking methods. Although this process is done with paper, it could also be done with paper like wood; veneer. However, this process is not that sustainable since it still produces a lot of waste material. But in this case wood can still be used in an additive manufacturing process.
Exploration phase

Part 6
Exploration phase

Based on the information which was found in the literature review, this part of the thesis will elaborate on the design of experiments which was done. The aim of this research is to explore the opportunities of the use of wood in an additive manufacturing process and therefore this thesis focuses on answering the following question; “Can wood be used in an additive manufacturing process?”

In order to answer this question, experiments were done with the basic components of wood with the aim to produce a material sample which can be printed with a 3D printer. This chapter describes the materials which were used, the design of the process, the production phase and explains a little more about the small tests which were done to validate the outcome of the production phase.

Materials

As basis for the experiments Kraft lignin, Indulin AT was obtained from Wageningen university and is used in powder form. The cellulose which was used in the project is Skogcell 90Z bleached kraft WFBR and is a product from the kraft pulping process. The cellulose is delivered as sheets of paper and is bleached. The cellulose was also obtained from Wageningen university. For this experiment acetone was used (C₃H₆O) and was obtained from Sigma Aldrich. Demineralised water was used as well and was obtained from the demineralization machine in the laboratory of stevinlab 2. The quality of the demineralised water is; < 8 µS/cm. The experiments were conducted in a climatized room in the stevinlab 2 building. The room had a temperature of 21 degrees and an air humidity of about 45 RH. For the experiment itself various lab equipment was used which can be seen in figure 45. Petri dishes (1) with a diameter of 80 mm were used as plates for samples. These petri dishes were obtained from sichma Aaldrich. Next to petri dishes, 2 different types of laboratory beakers (2) were used for preparation of material samples and as storage bowl for wood components. These laboratory beakers had a volume of 600 ml and 2000 ml and can be obtained from sigma aaldrich. In order to test if material samples can be printed, they were first tested with a medical syringe (3) in order to see how the materials behave. 2 types of medical syringes were used, one small syringe with a inner diameter of 18 mm and a maximum content volume of 20 ml and a nozzle of 1,6 mm. The second type of syringe was slightly bigger with a inner diameter of 28 mm and a maximum content of 60 ml and a nozzle size of 1,6 mm. These were obtained from a local pharmacy in Delft. In order to determine exact quantities of materials a lab scale (4) was used of the brand KPZ waagen, type kpz 2-05-3. For mixing the materials, a simple kitchen mixer (5) was used and some spoons(6) as well. Furthermore scissors were used to cut the sheets of cellulose into smaller pieces to make them fit the planned process. Next to syringes, refillable plastic tubes (7) with a nozzle of 2 mm were used to do some tests with printing as well. These plastic tubes can be obtained from BLM, a building related materials and tools shop. To extrude the material out of these plastic tubes, an electric driven extruder (8) was used. Also a material shredder (9) was used to treat the cellulose sheets with. This machine was produced by FRITSCH and comes with different sizes of sieves. The sieves which were used in this project had rectangular gab sizes of 2 mm. For the test some plastic strips are used to do the viscosity tests(10). Also, plastic pots were used in order to store prepared samples, or store printed samples(11).

Although wood is composed of cellulose, hemicellulose and lignin, for this thesis was chosen to work only with cellulose and lignin. Hemicelulose, which is also a component of wood, was kept out, for the reason it is almost similar compared to cellulose, the main difference is the shorter link length as explained in chapter 1. The idea behind it is, that when the printing with cellulose is working, some of the fibers could be replaced with smaller hemicellulose fibers without experiencing problems with printing.
Figure 45: Used tools and equipment for research.
Experiment setup:
The experiments are conducted in a climatized room within the laboratory of the civil engineering department (stevins lab 2) of the technical university of delft. The room is set at a temperature of 20 degrees with an humidity of 45 RH. Figure 49 shows the flowchart of the steps which needed to be taken. These steps will now be elaborated. The solution which is used in the experiment is made of 20/80 wt. % of demi water and acetone. This ratio was advised by the researcher from Wageningen university. The aim was to do a test with the following ratio’s

Experiment 1
- Kraft lignin 10 % wt. 20 gram
- Cellulose 15 % wt. 30 gram
- Water/acetone mix ((20/80)) 50 wt % 150 gram

In order to do so, one liter of water-acetone mix was created and was put in a laboratory beaker and was temporarily capped with a foil so the acetone couldn’t evaporate. Next step was to take the a sheet of cellulose as can be seen in figure 46.

Figure 46: sheet of Skogcell 90Z bleached kraft WFBR

The cellulose, which is delivered as paper sheets, needs to be pulped with water. In order to do this, the paper will be chipped to small parts of approximately 25 mm length and a width of about 15 mm. This can be seen in figure 47. Then the scale is used to determine the needed amount of chips.

Figure 47: chips of Skogcell 90Z bleached kraft WFBR with approximate size of 25 mm x 15 mm

In the next step, the needed amount of water-acetone solution was taken from the laboratory beaker and put in a separate laboratory beaker. Once finished, the right amount of cellulose chips was added to the laboratory beaker with 150 grams of water-acetone solution in it. This can be seen in figure 48.

Figure 48: Laboratory beaker filled with cellulose chips and water acetone solution according to prescribed amounts. beaker is capped to prevent evaporation of the acetone.

After this step, the first problem emerged. The cellulose chips were not pulped by the water-acetone solution. Even after 48 hours and mixing the chips and water-acetone solution with the kitchen mixer the chips didn’t transform to fibers. Apparently, cellulose is hard to pulp in acetone, and the amount of water in the mix was not enough to pulp the chips. Another reason might be the molecule size of acetone. The molecule size of acetone is bigger compared to water, a result of this fact might be that acetone is not able to penetrate into the chemical structure of paper.
Creating water-acetone solution 20/80 WT% and put it in a closed container so it won’t evaporate

Cutting cellulose chips

Take needed amount of chips

Take scale and determine needed amount of water-acetone solution

Mix the chips in with the needed amount of water-acetone solution and create pulp

Figure 49: Flowchart of experiment steps, the red ellipse shows the step where the first problems emerged.
As a result of this first setback in the process, the process had to be redesigned. The new process is shown in figure 53. The process has a new step added which takes care of the “pulping”. In order to do so, various options have been investigated. The easiest option was the shredder which was mentioned before. The machine can transform the chips to fine fibers of a size of approximately 2 to 3 mm. This can be seen in figure 50 and 51.

With the shredded fibers, the next step of the process was entered. The needed amount of water-acetone mix was added which created a pulp. The next step was adding the needed amount of lignin in order to create a paste which could potentially be extruded. However, the material needs to be consistent for extruding, what means that clumps of material should be avoided. With this having said, new problems showed up in the process. In the mixes with a little water-acetone solution added, the material was difficult to mix without resulting in small clumps of cellulose which were not covered in lignin, see figure 52. These inclusions caused problems in extruding the material from the syringes since the syringes got clogged. In the mixes with relatively a lot of fluid added, this didn’t seem to be a problem, but then the problem was the homogeneity of the mix since fibers and dissolved lignin didn’t form a paste.

In order to solve this problem, the process had to be redesigned again. The solution was to mix the cellulose and lignin before the acetone was added to the mix. Since the material is dry and exists of small “dust” like parts, a little water was added before mixing. This was done to prevent the dry matter from nebulizing whilst mixing. In order to control the amount of fluid added to the mix, the step of adding the water/acetone mix had to be split up as well. In the new designed process this step was split up in two phases where in the first step, water was added before mixing the dry content, and in step 2, after mixing the dry matter, the acetone was added in order to dissolve the lignin so it creates a paste. In figure 54 the process is explained in a new flowchart, which shows the process which has been applied in order to get the first material samples which were suitable for extruding with a syringe.
Creating water-acetone solution 20/80 WT% and put it in a closed container so it won't evaporate

Cutting cellulose chips

Take needed amount of chips

Use the shredder to create cellulose fibers

Take scale and determine needed amount of cellulose fibers

Take scale and determine needed amount of water-acetone solution

Mix the shredded fibers with the needed amount of water-acetone solution and create pulp

Take scale and determine needed amount of lignin and put it in a petri-dish

Adding Lignin to pulp-water-acetone mix

Mixing pulp-water-acetone mix

Drape mix in petri dish

Put little amount of mix in a syringe of 20 ml

Try to extrude the material from the syringe

Do tests with the draped and extruded material

Use scale to take needed amount of demi water in laboratory beaker

Use scale to take needed amount of acetone in laboratory beaker

Use the shredder to create cellulose fibers

Take scale and determine needed amount of water-acetone solution

Failure

Figure 53: Adjusted flowchart of experiment steps, New step for shredding the cellulose chips was implemented. Next problem showed up at the stage where the red ellipse is positioned.
Figure 54: Adjusted flowchart of experiment steps. This process was used to create the material for the test prints.
Now the process has been developed, the following sequence will shortly elaborate on the products which are created with this process.

As the process describes, the first steps take care of the right amount of the dry materials, shown in figure 55. These are put in a laboratory beaker and mixed together with a little demi water.

The results of this step can be seen in figure 56. After this step the acetone needs to be added to the mix of cellulose, water and lignin. The dry material transforms in a dark brown clay like material which is shown in figure 57. This material will be draped in a petri dish (figure 58).

From the petri dish, some material will be put in a syringe. This was done with a spoon, and in such a way that as few air bubbles as possible were included in the syringe. This was done to ensure a good material flow from the syringe. Figure 59 shows a medical syringe which is filled with test material.
Samples

Based on this process various mixes of the basic components were produced and tested. The exact mixes are described in figure 60. For every test mix, a number was written down in order to keep track of it during the process. Furthermore, the quantities of water, acetone, lignin and cellulose was noted for each test sample. Based on these parameters some ratio’s were produced which tell a little more about the material.

The percentage of cellulose was determined with the following equation:

\[
\text{Percentage of cellulose} = \frac{\text{Mass of cellulose}}{\text{Total mass ingredients}}
\]

This ratio gives an idea about how much cellulose is in the material mix before drying.

The second ratio determines how much lignin is in the mix compared to the amount of acetone, using the following equation:

\[
\text{Percentage of lignin/acetone} = \frac{\text{Amount of Lignin}}{\text{Amount of Acetone}}
\]

The third ratio compares the amount of cellulose with the amount of lignin in the mix. This shows the amount of cellulose in the mix after it has completely dried.

\[
\text{Percentage of cellulose in mix} = \frac{\text{Amount of cellulose}}{\text{Amount of cellulose} + \text{Lignin}}
\]

Furthermore the amount of water and acetone in the mix was expressed as a ratio of the total combination of acetone and water. Also the total amount of fluid in the mix was expressed as a ratio in order to see how this ratio influences the printability of a material. This ratio was determined using the following ratio:

\[
\text{Percentage of cellulose/lignin} = \frac{\text{Total of fluids in mix}}{\text{Total of materials in mix}}
\]

Figure 60: table with test data

<table>
<thead>
<tr>
<th>type of lignin</th>
<th>amount of water (gr)</th>
<th>amount of acetone (gr)</th>
<th>amount of lignin (gr)</th>
<th>amount of cellulose (gr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>test 1</td>
<td>kraft</td>
<td>16</td>
<td>64</td>
<td>20</td>
</tr>
<tr>
<td>test 2</td>
<td>kraft</td>
<td>15</td>
<td>60</td>
<td>5</td>
</tr>
<tr>
<td>test 3</td>
<td>kraft</td>
<td>15</td>
<td>60</td>
<td>10</td>
</tr>
<tr>
<td>test 4</td>
<td>kraft</td>
<td>15</td>
<td>60</td>
<td>7,5</td>
</tr>
<tr>
<td>test 5</td>
<td>kraft</td>
<td>15</td>
<td>60</td>
<td>11</td>
</tr>
<tr>
<td>test 6</td>
<td>kraft</td>
<td>0</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>test 7</td>
<td>kraft</td>
<td>40</td>
<td>20</td>
<td>40</td>
</tr>
<tr>
<td>test 8</td>
<td>kraft</td>
<td>40</td>
<td>20</td>
<td>40</td>
</tr>
<tr>
<td>test 9</td>
<td>kraft</td>
<td>26</td>
<td>16</td>
<td>40</td>
</tr>
<tr>
<td>test 10</td>
<td>kraft</td>
<td>20</td>
<td>26</td>
<td>40</td>
</tr>
<tr>
<td>test 11</td>
<td>kraft</td>
<td>45</td>
<td>30</td>
<td>40</td>
</tr>
<tr>
<td>test 12</td>
<td>kraft</td>
<td>5</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>test 13</td>
<td>kraft</td>
<td>1,5</td>
<td>5</td>
<td>10</td>
</tr>
<tr>
<td>test 14</td>
<td>kraft</td>
<td>6</td>
<td>13</td>
<td>8</td>
</tr>
<tr>
<td>test 15</td>
<td>kraft</td>
<td>6</td>
<td>15</td>
<td>10</td>
</tr>
<tr>
<td>test 16</td>
<td>kraft</td>
<td>40</td>
<td>46</td>
<td>160</td>
</tr>
<tr>
<td>test 17</td>
<td>kraft</td>
<td>5</td>
<td>22</td>
<td>40</td>
</tr>
<tr>
<td>test 18</td>
<td>kraft</td>
<td>10</td>
<td>26</td>
<td>40</td>
</tr>
<tr>
<td>test 19</td>
<td>kraft</td>
<td>10</td>
<td>22</td>
<td>40</td>
</tr>
<tr>
<td>test 20</td>
<td>kraft</td>
<td>10</td>
<td>28</td>
<td>60</td>
</tr>
</tbody>
</table>

Figure 61: picture sample 1

Figure 61 shows sample number one. The first sample was made of 5 gram cellulose, 20 grams of lignin and a total of 80 grams liquid. The mix of the liquid was 16 grams of water and 64 grams of acetone. Mixing this resulted in a non homogeneous mix where fibers and fluid clearly could be distinguished. Because the relative high amount of fluid, the viscosity was low and the bonding of the material as well. After a relative long drying period, where fluids had to evaporate, a dark brown material was left over. The surface is relative hard and has a good scratch resistant.
<table>
<thead>
<tr>
<th>percentage of</th>
<th>lignin / acetone %</th>
<th>cellulose / lignin %</th>
<th>acetone %</th>
<th>water %</th>
<th>% fluid</th>
<th>homogeneity</th>
<th>viscosity</th>
<th>bonding</th>
<th>notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>4,76%</td>
<td>31,25%</td>
<td>20,00%</td>
<td>80,00%</td>
<td>20,00%</td>
<td>76,19%</td>
<td>--</td>
<td>low</td>
<td>-</td>
<td>too much liquid</td>
</tr>
<tr>
<td>8,57%</td>
<td>8,33%</td>
<td>60,00%</td>
<td>80,00%</td>
<td>20,00%</td>
<td>85,71%</td>
<td>--</td>
<td>high</td>
<td>--</td>
<td>too much cellulose for printing</td>
</tr>
<tr>
<td>2,86%</td>
<td>16,67%</td>
<td>20,00%</td>
<td>80,00%</td>
<td>20,00%</td>
<td>85,71%</td>
<td>--</td>
<td>low</td>
<td>--</td>
<td>too much liquid resulting in shrinkage</td>
</tr>
<tr>
<td>5,71%</td>
<td>12,50%</td>
<td>40,00%</td>
<td>80,00%</td>
<td>20,00%</td>
<td>85,71%</td>
<td>--</td>
<td>high</td>
<td>--</td>
<td>too much cellulose for printing</td>
</tr>
<tr>
<td>1,15%</td>
<td>18,33%</td>
<td>8,33%</td>
<td>80,00%</td>
<td>20,00%</td>
<td>86,21%</td>
<td>--</td>
<td>low</td>
<td>-</td>
<td>too much shrinkage</td>
</tr>
<tr>
<td>0,00%</td>
<td>100,00%</td>
<td>0,00%</td>
<td>100,00%</td>
<td>0,00%</td>
<td>50,00%</td>
<td>+</td>
<td>low</td>
<td>+</td>
<td>too much shrinkage</td>
</tr>
<tr>
<td>0,99%</td>
<td>200,00%</td>
<td>2,44%</td>
<td>33,33%</td>
<td>66,67%</td>
<td>59,41%</td>
<td>-</td>
<td>medium</td>
<td>-</td>
<td>too much liquid resulting in shrinkage</td>
</tr>
<tr>
<td>2,91%</td>
<td>200,00%</td>
<td>6,98%</td>
<td>33,33%</td>
<td>66,67%</td>
<td>58,25%</td>
<td>-</td>
<td>medium</td>
<td>+</td>
<td>too much shrinkage</td>
</tr>
<tr>
<td>4,65%</td>
<td>250,00%</td>
<td>9,09%</td>
<td>38,10%</td>
<td>61,90%</td>
<td>48,84%</td>
<td>+</td>
<td>medium</td>
<td>+</td>
<td>printable material</td>
</tr>
<tr>
<td>5,49%</td>
<td>153,85%</td>
<td>11,11%</td>
<td>56,52%</td>
<td>43,48%</td>
<td>50,55%</td>
<td>++</td>
<td>medium</td>
<td>++</td>
<td>printable material</td>
</tr>
<tr>
<td>8,00%</td>
<td>133,33%</td>
<td>20,00%</td>
<td>40,00%</td>
<td>60,00%</td>
<td>60,00%</td>
<td>+</td>
<td>medium</td>
<td>++</td>
<td>printable material</td>
</tr>
<tr>
<td>9,09%</td>
<td>100,00%</td>
<td>20,00%</td>
<td>66,67%</td>
<td>33,33%</td>
<td>54,55%</td>
<td>++</td>
<td>high</td>
<td>++</td>
<td>printable material</td>
</tr>
<tr>
<td>10,81%</td>
<td>200,00%</td>
<td>16,67%</td>
<td>76,92%</td>
<td>23,08%</td>
<td>35,14%</td>
<td>++</td>
<td>high</td>
<td>+</td>
<td>printable material</td>
</tr>
<tr>
<td>12,90%</td>
<td>61,54%</td>
<td>33,33%</td>
<td>68,42%</td>
<td>31,58%</td>
<td>61,29%</td>
<td>++</td>
<td>high</td>
<td>+</td>
<td>cellulose seems to be too large to print</td>
</tr>
<tr>
<td>11,43%</td>
<td>66,67%</td>
<td>28,57%</td>
<td>71,43%</td>
<td>28,57%</td>
<td>60,00%</td>
<td>++</td>
<td>high</td>
<td>+</td>
<td>cellulose seems to be too large to print</td>
</tr>
<tr>
<td>4,84%</td>
<td>347,83%</td>
<td>7,25%</td>
<td>53,49%</td>
<td>46,51%</td>
<td>33,27%</td>
<td>++</td>
<td>medium</td>
<td>++</td>
<td>printable material</td>
</tr>
<tr>
<td>6,94%</td>
<td>181,82%</td>
<td>11,11%</td>
<td>81,48%</td>
<td>18,52%</td>
<td>37,50%</td>
<td>++</td>
<td>high</td>
<td>++</td>
<td>printable material</td>
</tr>
<tr>
<td>6,17%</td>
<td>153,85%</td>
<td>11,11%</td>
<td>72,22%</td>
<td>27,78%</td>
<td>44,44%</td>
<td>++</td>
<td>high</td>
<td>++</td>
<td>not perfect, printing might be able</td>
</tr>
<tr>
<td>6,49%</td>
<td>181,82%</td>
<td>11,11%</td>
<td>68,75%</td>
<td>31,25%</td>
<td>41,56%</td>
<td>++</td>
<td>medium</td>
<td>++</td>
<td>not perfect, printing might be able</td>
</tr>
<tr>
<td>4,85%</td>
<td>214,29%</td>
<td>7,69%</td>
<td>73,68%</td>
<td>26,32%</td>
<td>36,89%</td>
<td>++</td>
<td>medium</td>
<td>++</td>
<td>printable material</td>
</tr>
</tbody>
</table>

The bottom side of the material sample has an mdf like look and is less scratch resistant. The surface of the bottom side is smooth and the material feels at least as strong as mdf. The material couldn’t be extruded because the homogeneity of the material was too low. Extruding would cause a separation between the fluid and the fibers. The liquid contains 15 grams of water, 60 grams of acetone. The mix of these ingredients resulted in a non homogeneous mix. This can be declared by the amount of fibers which are still visible after drying. The viscosity was rather high and the stickyness of the material was low. These effects were caused by the high percentage of cellulose in the mix. Extrusion of this mix was not possible simply enough because the cellulose fibers clogged the syringe.

Figure 62 shows sample number two. Sample number two is made of 7,5 gram cellulose, 5 grams of lignin and 75 grams of liquid. The liquid contains 15 grams of water, 60 grams of acetone. The mix of these ingredients resulted in a non homogeneous mix. This can be declared by the amount of fibers which are still visible after drying. The viscosity was rather high and the stickyness of the material was low. These effects were caused by the high percentage of cellulose in the mix. Extrusion of this mix was not possible simply enough because the cellulose fibers clogged the syringe.

Figure 63 shows an image of sample three. Sample number three is made of 2,5 gram cellulose, 10 grams of lignin
and 75 grams of liquid. The liquid contains 15 grams of water, 60 grams of acetone. The result of this mix is a sample with a low homogeneity, a low viscosity and a low bonding value. The sample needed a long drying period just like sample one. The only difference is the amount of fluid in the mix. Also when the strength of this sample is compared with the strength of sample one, it can be noticed that the strength of sample number 3 is lower. This can be declared by the amount of fluid in the mix. After drying, when the fluid is gone from the mix, cavities are left in the material. This means that sample number 3 is probably more porous compared to sample number 1, resulting in a lower strength.

Figure 64: picture sample 4

Figure 64 shows sample number four. Sample number four is made of 5 gram cellulose, 7.5 grams of lignin and 75 grams of liquid. The liquid contains 15 grams of water, 60 grams of acetone. The result of this mix is a non homogenous mix with an high viscosity and a low bonding value. The viscosity is relatively high because a relatively high amount of cellulose can be found in the mix. This results in a material which is not suitable for extrusion. The strength of this sample is rather low, which can be expected by the low amount of lignin compared with the amount of cellulose in the mix. As described in the chapters before, lignin is the component which works as a glue and provides strength in wood, something that can be seen in this material as well. Also porosity might be one of the causes of a lower strength, since this mix contained a lot of fluid.

Figure 65: picture sample 5

Figure 65 shows sample number five. Sample number five is made of 1 gram cellulose, 11 grams of lignin and 75 grams of liquid. The liquid contains 15 grams of water, 60 grams of acetone. The result of this mix is a low homogeneity, a low viscosity and a low bonding value. The lignin in this mix was all dissolved in the fluid mix, which resulted in a non homogenous mix. The amount of fluid in the mix was too high in the past samples and therefore a test with lignin was done in sample number six.

Figure 66: Picture sample 6

Figure 66 shows a picture of sample number six. This sample was made of 10 grams of lignin and 10 grams of acetone. No cellulose and water were used for this mix. The result is a mix with a reasonable homogeneity a low viscosity and a reasonable bonding value. The material dried to a solid material with a brown and smooth surface.
Figure 67 shows sample number seven. Sample number seven is made of 1 gram cellulose, 40 grams of lignin and 60 grams of liquid. The liquid contains 40 grams of water, 20 grams of acetone. The results of this mix are an average homogeneity, medium viscosity and a low bonding value. The amount of fluid in this mix resulted in a non homogenous material and after curing, and cavities were visible in the material. This resulted in a lower strength of the material. Also the color of the material was darker compared to the samples before and traces of undissolved lignin can be found on the surface. This can be expected from the amount of water which was combined with acetone. The fluid mix was probably not able to dissolve all the lignin which was used because it had a lower capacity. This can also be expected by the information in chapter 4.

Figure 68 shows sample number eight. Sample number eight is made of 3 gram cellulose, 40 grams of lignin and 60 grams of liquid. The liquid contains 40 grams of water, 20 grams of acetone. This mix resulted in a material sample with average homogeneity, medium viscosity and a reasonable bonding value. Compared to sample 7, this mix contained a little more cellulose, which resulted in a higher bonding value. Normally cellulose does not act as a glue, but the cellulose might have absorbed a part of the water in the mix, which left more acetone for the lignin. This sample was extruded with a syringe, but the material collapsed under its own weight. This can be expected by the amount of fluid in the mix. Resulting in a porous material after drying.

Figure 69 shows sample number nine. Sample number nine is made of 4 gram cellulose, 40 grams of lignin and 42 grams of liquid. The liquid contains 26 grams of water, 16 grams of acetone. The result of this mix was a good homogeneity, a medium viscosity and a good bonding value. This resulted in a material what could be extruded with a medical syringe resulting in layers which were strong enough to support a next material layer. The amount of fluid in this mix was lower compared to the previous what resulted in better material properties.
Figure 70: Picture sample 10

Figure 70 shows sample number ten. Sample number ten is made of 5 gram cellulose, 40 grams of lignin and 46 grams of liquid. The liquid contains 20 grams of water, 26 grams of acetone. This mix resulted in a material with good homogeneity properties, a medium viscosity and a high bonding value. The material was extruded with a medical syringe resulting in a multi layered print, which was able to withstand its own loading. Also the surface quality looked smooth and this sample was the best one so far.

Figure 71: Picture sample 11

Figure 71 shows sample number eleven. Sample number eleven is made of 10 gram cellulose, 40 grams of lignin and 75 grams of liquid. The liquid contains 45 grams of water, 30 grams of acetone. This mix resulted in a sample with reasonable homogeneity, medium viscosity and a good bonding value. During extrusion, fluid and dry material where separated. This resulted in the reasonable homogeneity. Also the amount of cellulose in the mix was a bit high, which resulted in problems with extruding.

Figure 72: Picture sample 12

Figure 72 shows sample number twelve. Sample number twelve is made of 2,5 gram cellulose, 10 grams of lignin and 15 grams of liquid. The liquid contains 5 grams of water, 10 grams of acetone. This mix resulted in a material with good homogeneity, high viscousity and good bonding values. The material couldn’t be extruded with a medical syringe, since too much pressure was needed to extrude it. Therefore a electrical extruder was taken in combination with refillable tubes and a nozzle size of 2 mm. With these tools, the material could be extruded. The reason for the problems with extruding were the low amount of fluid in the mix.

Figure 73: Picture sample 13
Figure 73 shows sample number thirteen. Sample number thirteen is made of 2 gram cellulose, 10 grams of lignin and 6.5 grams of liquid. The liquid contains 1.5 grams of water, 5 grams of acetone. This mix resulted in a material with the following properties; the homogeneity had a high score, as well as the viscosity and the bonding had a reasonable score. This material was hard to extrude by hand with a medical syringe. Even with an electric extruder, it was hard to extrude the material. This would mean that a 3D printer probably is not able to extrude it. This has to do with the minimal amount if liquid in the mix.

Figure 74 shows sample number fourteen. Sample number fourteen is made of 4 gram cellulose, 8 grams of lignin and 19 grams of liquid. The liquid contains 6 grams of water, 13 grams of acetone. The characteristics of this sample are similar to the sample described before. The cellulose levels in this sample are relative high which causes the problems in extrusion.

Figure 75 shows sample number fifteen. Sample number fifteen is made of 4 gram cellulose, 10 grams of lignin and 21 grams of liquid. The liquid contains 6 grams of water, 15 grams of acetone. The result of this mix is a material sample with a high homogeneity, viscosity and reasonable bonding value. This sample was not suitable for printing, the amount of cellulose was too much and the lignin was not enough to create a paste of it.

Figure 76 shows sample number sixteen. Sample number sixteen is made of 12.5 gram cellulose, 160 grams of lignin and 86 grams of liquid. The liquid contains 40 grams of water, 46 grams of acetone. This mix resulted in a printable material. The material was extruded with a small medical syringe. The material didn’t seem to be strong enough to take its own weight. That can be seen on the tilted crosssection of the print. The material collapsed after some layers were stacked on the first layers.
Figure 77 shows sample number seventeen. Sample number seventeen is made of 5 gram cellulose, 40 grams of lignin and 27 grams of liquid. The liquid contains 5 grams of water, 22 grams of acetone. This mix resulted in a material with a high homogeneity, viscosity and bonding value. The high viscosity caused a problem with extruding, and caused the fact that the material was not extrudable with a medical syringe. Also the amount of cellulose was a problem in extruding. the amount was reasonably high compared to mixes which proved to be successful in extruding.

Figure 78 shows sample number eighteen. Sample number eighteen is made of 5 gram cellulose, 40 grams of lignin and 36 grams of liquid. The liquid contains 10 grams of water, 26 grams of acetone. This material mix resulted in a sample with good homogeneity, a medium viscosity and a high bonding value. This resulted in a material with good properties although it was hard to extrude the material. This had to do with the combination of lignin and low amount of fluid, which resulted in a dry paste which is hard to extrude.

Figure 79 shows sample number nineteen. Sample number nineteen is made of 5 gram cellulose, 40 grams of lignin and 32 grams of liquid. The liquid contains 10 grams of water, 22 grams of acetone. The material was extrudable like material sample 18. However printing this material somehow seemed to be more positive. Looking at the material numbers, this material should slightly be more difficult to extrude.

Figure 80 shows sample number twenty. Sample number twenty is made of 5 gram cellulose, 60 grams of lignin and 38 grams of liquid. The liquid contains 10 grams of water, 28 grams of acetone. This sample had a good homogeneity, a medium viscosity and a high bonding value. The material was relative easy to extrude, and strong enough to hold its own weight without collapsing. This means the viscosity of the material was good enough to extrude, but also strong enough to build on. The reason why this material was easier to extrude compared to the previous ones is the amount of lignin in the mix. The extra lignin created a more smooth paste which was easier to extrude.
Tests
The next step is testing the material for three different criteria. These criteria are homogeneity, viscosity and bonding or adhesivity. These criteria proved to be important in a later stadium when the material would be printed with a 3D printer.

Homogeneity
Material samples are tested on homogeneity. This is done through checking if the material behaves like one material, or liquids with dissolved lignin and cellulose appear to react independently from each other. This is done with a visual check, where the surface of the material is checked for bundles of fibers laying in fluid instead of a relatively solid paste where fibers and fluid can be distinguished. A second check is done via moving the petri dish with the sample in it. The petri dish should be tilted in an angle of 20 degrees. If cellulose fibers stay at the position where they were, and the fluid, acetone with dissolved lignin, moves away from the fibers, the mix is interpreted as being inhomogeneous. The score for this test is based on the two tests described. The material could score for each test a + or -. A score ++ would mean that the material sample behaved well during both tests where a score -- means that a material behaved bad during both tests.

Viscosity
Each material sample is tested on viscosity. Viscosity, also known as treaclyness, determined by slow flowing or vast flowing, is a physical material property which indicates to what extend a fluid or gas offers resistance to shear stress. For each material sample an indication low, medium or high is given. A low viscosity means that a material flows like water and is very fluid. A high viscosity means a material sample is not fluid and behaves like a paste and can resist some shear force. The medium score is used for material samples which can resist some shear-force, but also flow like a fluid. These samples do not behave like a dense paste. Figures 81 and 82 show figures of the viscosity tests which were done. In figure 81, a plate is put on a material sample. This plate is moved in a horizontal direction with a constant speed. The force needed to do this, gives us an idea about the viscosity of the material. Figure 82 shows an image of a sample with a completed test.

Bonding or adhesivity
Each material sample is tested for the criteria of adhesiveness. This determines if a material is able to stick to another material. This criteria is needed to generate cohesion between print layers once the material is printed. This criteria is checked via the test which is shown in the pictures below. The material is held between two fingers which will move away from each other. If the material has an adhesive character it will stick to the fingers. The longer the material spans between the fingers, the better the score. Scores are --, -, +, ++. Figures 83 t/m 87 show the process.
Based on the data which is presented on the previous pages some material samples were selected to test for machine printing. This was done based on the tests, but also based on the notes. Some notes described that the material couldn’t be printed by hand, but might be possible to print with a machine. The most important criteria for selecting the samples is the way how the material behaves in case of printing with a syringe. Material sample 9, 10, 11, 16 and 20 showed good results in printing with a syringe.

Figure 88 shows a print sample of material sample 9. This was printed with a 20 ml syringe. The visual print quality remained relatively constant, although extruding this material needed a lot of force. This might lead to problems with machine printing.

Figure 90 shows a hand printed sample of material sample 11. The quality of this sample seems to be lower compared to sample number 10 and 9. The reason for this decline in quality can be found in the amount of fluids and the amount of cellulose which was put in the mix. The amount of cellulose, which was double compared to sample 10, clearly caused more difficulties in extruding resulting in a lower homogeneity and print quality. Also the amount of fluid was relative high resulting in a non consistent flow.
Figure 91 shows a print sample of material sample 16. The print quality was good since the material was easy to extrude. However, shrinkage turned up during drying since the material had a relatively low cellulose content and relatively high lignin content. That caused less ability to deal with shear-force once printed, resulting in the non planar edges of the print. The viscosity of this sample was higher compared to sample 11 but comparable with sample 10.

Figure 92 shows a print-sample of material sample number 20. This sample was extruded with a 20 ml syringe. This sample has a relative high amount of lignin in the mix in order to see if that would benefit the printing process. The content of cellulose is higher compared to sample 16 which results in a more stable behavior after printing. The stable behavior can be seen as better shear force resistance resulting in less shrinkage after printing compared with sample 16.

**Testing material sample with microscope**

After these samples were extruded and have dried, sample 9 was taken to do further investigation on the material itself. In order to do so, a microscope was used to zoom in on the material.

Figure 93: sample 9, material in petri dish studied.
Figure 93 shows a picture of material sample 9. In this view, fibers and dissolved lignin can clearly be seen. Although the amount of lignin seems a bit high, based on this picture, a careful conclusion can be drawn that the lignin is nicely bonded to the fiber. As in the literature review of the previous chapter, as in wood, lignin can be found in cellulose cell-walls and in between cellulose cells (Bowyers, Shmulsky & Haygreen, 2007). In this view the lignin seems to bind to the cell-walls again, so it can function as a “natural” glue between the cellulose fibers. However, more research is needed on the exact effect in order to give a more reliable explanation of the situation showed in figure 93.

Figure 94 shows a hand printed sample which was studied underneath a microscope. This sample was taken from the middle of a longer print sample and was taken away by tearing it from the original sample. The picture shows how the cellulose fibers are placed within the printed material. As can be seen, the cellulose fibers are nicely distributed in the cross-section of the material. If we study the side elevation of the material, it can be noticed that fibers seem to be aligned with the print direction. This results in anisotropic material properties, what can be found in wood and gives wood its distinctive properties compared to materials like steel or concrete. A explanation about how this happened can be found within one of the projects which was elaborated in the chapter with reference projects. A hydrodynamical process which takes place in the tip of the syringe caused a rotation of the fibers within the fluid. The material is being pressed into a smaller flow channel, which forces the fibers to align with the print direction. After this has happened, the fibers can’t rotate back, since the viscosity of the material is too low for the fibers to move. Carefully we can conclude that the material seems to have anisotropic properties.

Figure 94: sample 9, material which was hand printed studied with a microscope.

Figure 95: material sample showing aligned fibers

Figure 96: material sample, clearly showing aligned fiber on the material surface.

Figure 95 and 96 show more material samples which have been checked for anisotropic properties. These gave a clear indication that the material might have anisotropic properties. However, in order to do a more detailed explanation, more research should be done on printed material samples.
Testing material for water resistance

An interesting question to determine the field of application of the material is; how does the material behave in wet conditions? In order to answer this question, a simple test was done to see what will happen with the material in water.

A petri dish was taken and a small material sample of sample number 9 was put in it. In the dish, some water was put, and the petri dish with water and the material sample was put aside. After one full week, 168 hours, the material sample was compared with the original material.

![Figure 97: water test: on the left material sample of batch 9. On the right material sample of batch 9 after being welled in water for 168 hours.]

Figure 97 shows the material after 168 hours (right side). If we compare the welled sample with the original material (left), we can conclude that the color has changed. This can be explained because of the water. The size of the wet sample didn’t change significantly compared to the original sample. This means that the material does not expand a lot when it gets exposed to water. Also the stiffness of the material seems to be similar compared to the original material from the batch. Most important conclusion which can be taken from this test is that the material does not fall apart when it gets wet. However, the material has still a biological background, which still means that the material might be vulnerable for decay in wet conditions. However, more research is needed to determine the exact material properties and durability.
Conclusions

The focus of this chapter was to find a way to use wood in order to create a material that could be printed. Therefore basic components of wood were used and combined in several tests to find out how the materials behaved. This was done with the goal of this thesis in mind; "Exploring the opportunities of the use of wood in an additive manufacturing process". The conclusions of this step in the process are summarized as follows:

Can wood be used in an additive manufacturing process?
Based on the test which have been done, and the materials which have been used, a strong case has been built to answer this question positively. All the material samples were made of cellulose and lignin, the main components present in wood. The materials were combined in a paste like material which could be extruded with a medical syringe. In order to be able to create this material, a process had to be designed which explains the steps in production of the material. Using the right quantities of the material seems to be key, and is therefore really important. Once this has been done properly, a material, based on wood can really be extruded by hand. Next step in the process would be to check if this material can also be extruded with a 3d printer. The next chapter will elaborate on this question.

Material properties
Based on the tests which were done in this chapter, some material properties became clear. For every material sample, a test was done on the test criteria homogeneity, viscosity and adhesiveness. Besides these tests, on a few samples tests were done on waterproofness, and material properties like fiber orientation within the samples. These test gave some interesting results. The materials which were extruded with a syringe showed signs of anisotropic material properties. This was concluded after microscopic views of the material where studied. In these views, cellulose fibers within the material appeared to be aligned with the print direction of the material. Furthermore, waterproofness was tested as well. Hydrodynamic processes in the tip of the syringe make this happen. A possible explanation for this process can be that the material is being pressed into a smaller flow channel, what forces the fibers to align with the print direction. After this has happened, the fibers can’t rotate back, since the viscosity of the material is too low for the fibers to move. However, more research is needed to proof this. Furthermore, waterproofness was tested as well. The main conclusion from that test was that the material does not fall apart in case it gets wet.

Suggestions for further investigation
The results of this chapter defined a first hint about how to create a material out of wood which can be printed. However, the tests reached a certain limit and therefore more research needs to be done to get more accurate data about the material. The tests in this chapter were done at chamber temperature and therefore are not investigated temperature-wise. An interesting development would be the use of temperature in this print process. Lignin has a melting temperature of about 150 degrees and therefore will start cross-linking once the molecules are heated. This means that larger molecules of lignin are formed as result of combination of normal lignin molecules. Another interesting field of research would be to check if other sources of cellulose and lignin can be used as well. This research only used kraft lignin, as described before. However there are other pulp processes used in the paper industry. The residual material from those processes will probably also contain lignin, which is slightly different concerning chemical structure. Also more research is needed with different sizes of cellulose, in order to see if it is possible to improve the homogeneity of the print mix. The material mix in general could probably be more optimized and therefore more research is needed in order to find the optimal material mix.
Prototype phase

Part 7
This chapter will focus on the printing of material samples which were elaborated in the previous chapter. The aim of this research is to explore the opportunities of the use of wood in an additive manufacturing process. In this chapter we will try to answer the main question of this master thesis; can wood be used in an additive manufacturing process? This will be done through printing the material with a 3d printer. Also a design will be made which will be printed to show the potential of the material. Furthermore, potential fields of application will be elaborated and finally, the print results will be elaborated in order to see how the materials behaved in a 3d print process.

3D printing
In the previous chapter the process is explained and materials are designed. However in order to be able to print the materials which were made in the previous chapter a 3d printer is needed. Since the material, and the process of making it, and the material properties are different compared to the materials currently being printed, a new print process has to be designed or an already existing one has to be adapted. Additional to that, printer-settings have to be discovered in order to find out what works best in combination with the material.

Looking at part 2 of this thesis, the closest print method is FDM printing. However, most FDM printing is based on temperature, something that is not a part of the process which has been developed in the previous chapter. Therefore most of the printers which were available, and were used for printing plastics, were not suitable for the job. However, FDM printing can be done in combination with a clay extruder. A clay extruder is mostly used to extrude ceramics or clay. The material clay shows some interesting similarities looking at viscosity and therefore this method might be the key to success.

In order to test this, a homemade printer was redesigned in order to fit a medical syringe. Figure 98 shows the printer and Figure 99 and 100 show the details about the syringe is mounted on the printer. Although this is not a clay extruder, this was the first test-set-up to see if we could print the material with a machine.
The details about the printer are as follows;

The printer is controlled via a minitronics board obtained via Reprapworld.nl. The machine has stepper motors of the type NEMA 17 with a holding torque value of 4.08 kg.cm. The stepper motors were also acquired from Reprapworld.nl. The resolution is 1,8 deg per step with 1/16th microstepping. The bed is a standard bed without heating. The maximum print size is limited to 180 x 120 x 60 mm.

For material sample 20 the following settings were used;

Layer height = 0,5 mm  
Shell thickness  =3 mm  
print speed =5mm/s  
Printing temperature =0 degrees  
Support type =none  
Platform adhesion type =none  
Filament diameter = 29 mm (inner diameter syringe)  
Flow = 600%  
Nozzle size =1,6 mm

**First print**
The first print was made of the material of sample 20. The print settings which are described on this page were not known and had to be discovered. In order to do so a print test based on a 38 mm cross-sectional circle was done. Based on the printer-settings described hereafter, the first print was made.

Layer height = 2 mm  
Shell thickness =3 mm  
Print speed =5mm/s  
Printing temperature =0 degrees  
Support type =none  
Platform adhesion type =none  
Filament diameter = 29 mm (inner diameter syringe)  
Flow = 800%  
Nozzle size =1,6 mm

Figure 101 shows the first print which was made with a 3d printer. As described before, material sample 20 was used for the print.

As can be seen, the printer extruded too much material resulting in an inconsistent shape. The printer settings had to be adjusted and a new test had to be done.

For the next test, the settings were changed as described underneath.

Layer height = 0,2 mm  
Shell thickness =3 mm  
Print speed =5mm/s  
Printing temperature =0 degrees  
Support type =none  
Platform adhesion type =none  
Filament diameter = 29 mm (inner diameter syringe)  
Flow = 200%  
Nozzle size =1,6 mm

**Figure 101: First print made with 3d printer.**

Based on a 38 mm circle a print was made. Problems with extrusion meant that the print had to be stopped.

**Figure 102: print test 2**
The second test had to be stopped because, as the sample shows, the flow rate of the material was not high enough. This influenced the print badly. Later on the problem was found in the print material itself. Figure 102 shows the print which was made.

The third print was based on the following settings:

- **Layer height**: 0.5 mm
- **Shell thickness**: 3 mm
- **Prints speed**: 5 mm/s
- **Printing temperature**: 0 degrees
- **Support type**: none
- **Platform adhesion type**: none
- **Filament diameter**: 29 mm (inner diameter syringe)
- **Flow**: 500%
- **Nozzle size**: 1.6 mm

In this test a triangular shape was printed with sharp corners. By doing this test, we could check if the material flow was constant enough or that it caused the problems regarding the shape of the print. The experiment for this print was designed to point out the problem. In case the material flow is not constant enough, the corners will move towards the center of the triangle. If this happens, we know the material flow is not constant enough and problems are related to the homogeneity of the material. The print settings were as following:

- **Layer height**: 0.5 mm
- **Shell thickness**: 3 mm
- **Prints speed**: 5 mm/s
- **Printing temperature**: 0 degrees
- **Support type**: none
- **Platform adhesion type**: none
- **Filament diameter**: 29 mm (inner diameter syringe)
- **Flow**: 500%
- **Nozzle size**: 1.6 mm

Based on the fourth print some interesting details showed up. The corners of the triangle moved towards the center and the triangle ended up having a conical shape. The reason for this had to be the material. Figure 104 shows the print-sample in side view and figure 105 shows the material in top view.
During the printing process flow problems were detected resulting in the conical shape of the triangle. The reason for the flow problems could be the material itself, but during the process it seemed that the stepper motors of the printer were not powerful enough to press down the plunger of the syringe. This could be solved by changing the viscosity of the material, or using a different system for extruding. Based on these findings, new tests need to be done with a printer equipped with a clay extruder. The reason why a clay extruder would be better is the propulsion of the material within the extruder. This is based on air pressure in combination with a screw which pushes the material forwards towards the nozzle.

Figure 105: fourth print made with 3d printer. Top view

Design
Based on the findings so far a design is made for an object that could be printed. This object should express the benefits of using the fiber reinforced lignin instead of wood. Also the prototype should express the feasibility of 3d printing with the material fiber reinforced lignin.

Benefits
The fiber reinforced lignin can be shaped with a 3d printing process. The result of this technology is the minimum amount of waste, compared with conventional production technologies such as milling. The materials which are needed to produce the fiber reinforced lignin material, are residual wood, or paper in combination with the product lignin, which is a residual product as well. The fiber reinforced lignin material is in fact based on materials which are considered to be waste. Using these materials will benefit mankind and adds value to our sustainable economy.

Another potential benefit is the fact that fibers can be aligned to the print direction. This might be beneficial in certain designs where complex curves and organic shapes are made out of wood. In such situations wood seems to be less suitable since wood fibers are not aligned anymore, resulting in less strength since fiber will be cut. Since cellulose as a material is suitable for dealing with tensional and compressional forces, both situations might be possible. However, since the little amount of fibers in the material (up to 11% (sample 10)) the best solution for now would be to use the material in compression.

Disadvantage
Wood is loved for the looks it has. Fiber reinforced lignin doesn’t have these looks for now. Research is needed to see if woodgrain structure can be mimicked with the material fiber reinforced lignin.
Design
The design is based on an already existing design in Germany. The tree shaped columns are designed for the airport of Stuttgart. However, as can be seen in figure 106 and 107, bending moments will show up in the pipe members, since the forces from the upper tier don’t form an equilibrium in the direction of the lower tier.

In the design which is made for this thesis, a node was designed with an equilibrium in forces. This will result in no bending moment in the node and therefore less material can be used to acquire the same strength. The tree shaped column is showed in figure 109.

Figure 106: Tree shaped columns at airport Stuttgart.

Figure 107: Schematization of tree shaped column.

Figure 108 shows the forces in the structure when a normal force is applied on the tubes. As a result of the forces and the angles of the pipes a bending moment will show up in the lower tier. The nodes which are placed at the lower tier have to deal with these bending moments and therefore more material is needed.

Figure 108: Forces in tree shaped structure. Blue = bending moment; red = resulting force.

In the design which is made for this thesis, a node was designed with an equilibrium in forces. This will result in no bending moment in the node and therefore less material can be used to acquire the same strength. The tree shaped column is showed in figure 109.

Figure 109: Isometric view of tree shaped column.

Figure 109 shows drawings of the column. The node is shown in figure 110.

Figure 110: Drawing of node.
The design is made in rhino in combination with grasshopper. Figure 111 shows the grasshopper code which was used to build the tree shaped column. The code is created with the plugin rabbit, which is designed for making nature inspired branching columns. The plugin does this with Lindenmayer systems, also called L-systems. The system is a vector based tool which is able to generate forms with the help of the Turtle component. The turtle has as input a step size which depends on the length of each element, an angle increment, which determines the angle of the vector based on a 3x3 rotation matrix. A starting point as input is needed to determine the starting point and its plane. The last function of the turtle is used to generate tubes based on the length and direction of the vectors.

The input of the L-system are mainly letters from the alphabet and punctuation marks. These letters have a meaning as well as the punctuation marks. for example the punctuation mark ‘/’ is used to determine the angle between the branches in the YZ plane. In between the[B] input in the L-system, four / marks are used. In the turtle an angle of 30 degrees is mentioned, which results in 4 x 30 degrees in between the branches. This results in an angle of 120 degrees, creating exactly 3 branches on the first and second tier.
Figure 112: Drawing of treebranching column with detail drawing of node.
In order to be able to produce this node with conventional methods out of wood, lots of material need to be wasted. Also, fibers within the cross section will not ideally be aligned according to the direction of the force. Being able to print such parts, will save lots of material but can also benefit the structural properties of the node.

In order to print this node, An UR5 (universal robots robot arm 5), shown in figure 113, is combined with a Luthum clay extruder which is shown in figure 114. The UR5 is controlled via Hal robotics, a plugin for grasshopper. The clay extruder is controlled via a stepper motor driver on a homemade printer. By doing this, the extruder has a manual switch to turn it on and off. This setup was chosen because of the lack of time during my thesis process. The clay extruder can be controlled via the software pronterface. With this setup, both the ur5 and the clay extruder can be controlled separately, which makes it possible to make simple prints. More time would be needed to install the luthum extruder and ur5 robot arm in order to be fully functional.

**Applications**

Besides the application chosen in the previous design, applications can be found in furniture, toys, or other consumer goods, like for example bio-degradable flower pots. Figure 115 and 116 show a stool which was designed. The complex and intertwined pipes can be printed, and therefore be made of wood. Again, material is saved if we compare it with this design made with conventional processes.
8. Conclusions and recommendations

This research emphasized the potentials of using wood in an additive manufacturing process. The main question of this thesis is as follows: Can wood be used in an additive manufacturing process?

Throughout this study, we have proven that the basic components of wood can be combined in a material that can be 3D printed. With this technology, a new step in the direction of a sustainable economy is taken and new design options, based on fiber reinforced lignin have become possible.

The potential of this material has been partially demonstrated. Many more options regarding material mixes and types of cellulose and lignin are available on the market. The amounts of available materials is huge, as described in chapter 3 & 4, and dissipation of those resources would be a shame. Further research should be done to get a better understanding of the optimal mix of the materials for 3D printing, also depending on the field of application.

Material
For this research the materials cellulose, type Skogcell 90Z bleached kraft WFBR, and lignin retrieved from the kraft process, type Indulin AT, has been combined into a print material. According to the data which was generated in chapter 6, the best results were made with the following parameters. The amount of cellulose in the mix was found to deliver the best results in the range between 7.25% and 11%. One sample (11) had a cellulose percentage of 20% resulting in more difficulties with extrusion compared to the other samples (9,10,16,20). The best ratio’s of fluid were found in between the 33 and 60%. This ratio was important because it directly influenced the viscosity of the material. The amount of acetone applied in the fluid mix was found best between 38.1% and 73.68%. According to the theory these values have reasonably high dissolving capacity of lignin in combination with water. Combining the lignin with acetone was the right step, resulting in a binder between the cellulose and lignin, but also resulting in water resistant properties. The looks of the fiber reinforced lignin can’t be compared with wood, and therefore the created material should be used differently. The aesthetics of the material for example, showed to be dark brown. The reason for this color can be found in the color of the input material lignin. The powder has a dark color as result of the kraft process it has been in. However, some woods species have dark brown colors for example wenge, which is often used for furnitures. Also the wood pattern is missing in the printed material. The looks of the material are like that of mdf, but with a darker color. In order to get a higher aesthetic quality, research is needed to see how the aesthetics of the fiber reinforced lignin can be improved. Adding a pigment to the mix might be the solution to solve this color problem.

Another point of attention would be the fiber size which is applied in the mixes. The fibers used in this research are produced with a sieve of 2 mm squared. However, smaller fiber sizes might be beneficial for the printing process and might solve flow problems during printing. On the other hand, if fibers are less long, it might have an negative effect on the strength of the printed material. The fibers of hardwood are known to be shorter compared to the ones from softwoods. This part was not covered in this research but might be a next step in research.

The materials used in the process originate from softwood trees. However, there are many more resources of cellulose available, which all could potentially fit this print material. Also on this topic more research is needed in order to learn about the different resources and their effect on the strength, printability and aesthetics of the resulting material.

Printing
Printing the material seemed to be more complex than estimated. The main problem was found in the available resources. Finding the right print equipment took quite some time and resulted in some delay. Also the transition from hand printed with a syringe to printing with a machine was a complex step, since flow problems emerged or the printer didn’t have enough power to extrude the material.

Also temperature was not taken into account. Temperature might change the drying time of printed samples but can possibly also be used to mimic a woodgrain pattern on the material. More research is needed on the print process to improve the aesthetics of the fiber reinforced lignin after printing.
Another result from this research is that fiber alignment might be possible with fiber reinforced lignin. This would give the material a big advantage in certain applications since the strength of an object can be optimized just by aligning the fibers to the direction where they are needed. In order to do so, more research is needed to get a better understanding of the fluid dynamic process in the printing nozzle and the effects on the alignment of the fibers.

**Process**
During this thesis a new process for creation of the fiber reinforced lignin had to be designed. Based on the outcome of the research, the process works relative well and is easy to implement. However, more research is needed to see if the process can be optimized. For example time management, and after development, the switch to an industrial process.

**Tests**
The test which were done during this research gave a good qualitative understanding of the material. The first test showed if the materials were homogenous enough. The second test was used to investigate the viscosity of the material and the third test was used to check the adhesive force of the material. The quality of these tests was just enough to get a understanding of the material, but would be difficult to repeat. This results in limitations and more accurate testing is needed in order to get a better understanding of the material behaviour. Accurate test on homogeneity, viscosity and stickyness should be done in such a way it is repeatable and quantitative. Also tests which give more information on the strength of the material should be done.

**Recommendations**
This thesis focused on a small subset of the available opportunities. In order to get a better understanding of the material, more research is needed on material properties. How does the material behave in wet conditions overtime, what is the tensile and compressive strength of the material and how does it deal with decay? This thesis was ment to explore the opportunities of using wood in additive manufacturing, which means it is just the beginning of a long learning process.

Also material-wise a lot of new things could be investigated. Cellulose is one of world's most available materials and can be retrieved from many plant species. Also lignin can be found in many sorts. Research is needed to investigate how other sorts of cellulose and lignin can be used in a 3d print process. Also different sizes of cellulose should be tested in research in the future. This research only used one size, what might have caused problems with extruding.

**Alternative Applications**
As this study focussed mainly on the use of wood in a additive manufacturing process. The research produced some first results which showed that wood can be used for additive manufacturing. Additive manufacturing is basically extruding material with a 3d printer. Most of these machines use small nozzles. But the material might also be suitable to use in normal extrusion processes. Maybe panels can be produced, or certain profiles can be made with extrusion technologies. The material could potentially be applied in various processes suitable to produce products for a wide variety of industries.
Personal reflection

Thomas Liebrand

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Reflection

How is your graduation topic positioned in the studio?
The topic of this master thesis is the use of wood in a 3D printing technique. The master study Building technology teaches people how to design, build and maintain building related products, buildings or even civil projects such as bridges. One of the main pillars within these study topics is sustainability and it is worldwide more and more seen as a common standard. Tools such as computational design skills and 3D printing are an upcoming technique and are more and more used to design and build something. Where additive manufacturing was seen as a tool only for prototyping, nowadays it is used for manufacturing of small product batches. Compared to traditional production methods, additive manufacturing is a process with a low amount of material spill and is therefore considered to be sustainable. Where traditional methods remove material to shape a product, additive manufacturing only adds the necessary material to get to the final desired shape. As mentioned before, this master thesis is about the use of wood within an additive manufacturing process. Within woodworking, enormous amounts of material are spilled and have to be down-cycled or used as cheap fuel. This is mainly to do with the shaping methods, traditional ones, that only remove material to shape a product. In order to deliver a new method of woodworking, this research has investigated whether there is a more sustainable method of woodworking possible. The main goal is therefore to explore the possibilities of the use of wood in additive manufacturing processes.

How did the research approach work out (and why or why not)? And did it lead to the results you aimed for? (SWOT of the method)

According to the literature study, the use of solely wood in a 3D printing process was not done before. However some commercial parties claimed to be able to print wood, with a wood filament and a FDM printer. The filaments contain about 40% of wood fibers and a plastic polymer. This product is claimed to be a wood-like product but it behaves differently and therefore can’t be seen as wood. The goal of this master thesis is to see if wood, which mainly consists of cellulose and lignin, can be used in a 3D printing process. Therefore research into these materials was needed and also existing methods of additive manufacturing where studied. This was done with the aim to design a new bio based material used for additive manufacturing with an existing AM process or even a new process. The basic knowledge of AM was studied with books, containing scientific articles and research articles itself. In order to get more knowledge about the chemical part of the research a specialist from Wageningen university was contacted. Also materials where obtained via Wageningen university. Looking back at the research approach of this thesis, the process worked out well. Since multiple disciplines were needed which were not part of the knowledge I had at the time, and resources where also a problem to get. The most important information and materials were obtained via Wageningen university and the researcher from Wageningen university. Being able to get the information and material really opened doors and made me able to start testing. Since I had no clue where to start and what to do with the cellulose and materials, a session was planed with the researcher from the university of Wageningen. Together we brainstormed about he possible ways to convert the material into an extrudable material. As result of these meetings I had enough input to start a testing process and actually make material samples. The theory sessions helped me define the path I took. Concluding, the research approach worked out for this thesis, mainly because the path which I had to take was not known yet and therefore it has a high level of trial and error methodology, which was framed by the knowledge obtained during the sessions.

How are research and design related?
The design of a new material is done based on certain knowledge. However the output of the design was not sure yet and therefore multiple iteration steps were required in order to get to a reasonably good result. Therefore, the output of the design was needed in order to do the next step in design, and the output of design also delivered knowledge.
For example the knowledge about the process to apply, material results and characteristics of the tested material. Because the design phase lead to knowledge, the relation between research and design can be stated by the method of research by design.

Societal impact

To what extent are the results applicable in practice?

The results of this master thesis can be seen as one of the first steps towards the use of wood in an additive manufacturing process. This means that the results of the master thesis will contain mainly concepts and a prototype for the proof of concept. This means the process and material are not market ready and need to be developed before it can be used on an industrial scale. However, once the technique is developed and applicable on an industrial scale, society can benefit from it, and the technique can be used to build building related products based on a bio composite which is sustainable and efficiently produced.

To what extent has the projected innovation been achieved?

The ultimate goal would be to be able to print wood. However, wood is one of nature’s most complex materials and it is the result of a growing organism. Wood has some important characteristics which make wood special and loved by humans. For example the aesthetics, odor, natural origin, chemical composition and structural behavior. The results of this master thesis have achieved some of the projected innovations. The result of the research so far is that we can apply a bio based (read wood-based) material in a 3d printing process. This means a material can be printed which is based on the chemical constituents of wood. This also means that the material is fully originated from nature. Additional to that, there are some signs that the research have resulted in the ability to print an anisotropic material. This has to do with the fiber orientation within the wood, and fibers within the printed material look like they have a similar way of fiber alignment. However, characteristics such as odor, aesthetics and structural behavior have not been compared and the printed material is not comparable for now. Future research might result in a solution for these existing problems and might be able to solve these problems.

Does the project contribute to sustainable development?

The results of this master thesis describe a process and result of a innovative and sustainable technology used to produce a wood based bio composite. The techniques could be used to make building related materials, and the methodology should save materials which are spilled nowadays.

What is the impact of your project on sustainability (people, planet, profit/prosperity)?

The project potentially unleashes a new route to infinite bio based material which could be used in a additive manufacturing process, but maybe also different industry related products. This means that materials such as wood potentially could be used more efficiently without the need to down-cycle or burn it.

What is the relation between the project and the wider social context?

The technology might result in technologies which are more sustainable compared to current technologies. Sustainable technologies will lead to a better world for all people living on it.

How does the project affects architecture / the built environment?

The results of this master thesis can potentially result in new materials and techniques used to shape and build the built environment. Therefore it could lead to innovation which will contribute in a sustainable way and therefore help people live in a more healthy way.
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Figure 18: Schematic of Selective Laser Sintering (polymers) process source: Hopkinson, Hague & Dickens (2006)
Figure 19: Three-Dimensional printing process source: Hopkinson, Hague & Dickens (2006)
Figure 20: Fused metal deposition system process source: Hopkinson, Hague & Dickens (2006)
Figure 21: Selective masking sintering process source: Hopkinson, Hague & Dickens (2006)
Figure 22: Fused Deposition Modelling process source: Hopkinson, Hague & Dickens (2006)
Figure 23: Sheet Stacking Technologies process source: Hopkinson, Hague & Dickens (2006)
Figure 24: Overview of paper pulping process. Source: (Bajpai, 2015)
Figure 25: Overview of cellulose molecule Source: (Bowyers, Shmulsky & Haygreen, 2007)
Figure 26: Lignin building block, part of a lignin molecule Source:
Figure 27: Lignin building block, part of a lignin molecule Source:
Figure 28: Lignin molecule as can been found in wood. Source: (Bowyers, Shmulsky & Haygreen, 2007)
Figure 29: Graph showing effect of acetone concentration on lignin solubility at 23 celcius degrees source: (Bajpai, 2015)
Figure 30: The separation process of lignin from black liquor based on a ANDRITZ lignin recovery process which is suitable for kraft pulp processes. Source: Pehu-Lehtonen (2017)
Figure 31: Image of process used by Henke & Treml (2012) source: Henke & Treml (2012)
Figure 32: Image of material created by Henke & Treml (2012). Cone is produced with gypsum as binder material. Source: Henke & Treml (2012)

Figure 33: Inner structure of cone with bonded with cement (top) and gypsum (down). Source: Henke & Treml (2012)

Figure 34: Image of nozzle and fluid dynamic process explained. Source: Håkansson et al., (2014)

Figure 35: Step 1: A CAD model is needed of the design which has to be build. In this case a hammer.

Figure 36: Step 2: Hammer is divided into slices with the same thickness as the material, in this case paper. The colour is printed onto the paper in order to give it some esthetics.

Figure 37: Step 3: All sheets are put in the machine, which is now ready to start producing.

Figure 38: Step 4: The machine takes a sheet and puts adhesives on it. At the location of the hammer a lot, next to it, on the excess material, less.

Figure 39: Step 5: The machine takes a new slide and puts it on top of the previous layer.

Figure 40: Step 6: The machine presses the layers together.

Figure 41: Step 7: The machine starts cutting so the object can be removed easily from the excess material. Now the process can repeat from step 4 till 7, until the process is finished.

Figure 42: Step 8: Now the machine is finished, ‘a pack of paper’ is ready to be unpacked.

Figure 43: Step 9: Now the printed object can be separated from the excess material.

Figure 44: Step 10: Once finished, the product is ready to use.

Figure 45: Used tools and equipment for research.

Figure 46: Sheet of Skogcell 90Z bleached kraft WFBR

Figure 47: Chips of Skogcell 90Z bleached kraft WFBR with approximate size of 25 mm x 15 mm

Figure 48: Laboratory beaker filled with cellulose chips and water acetone solution according to prescribed amounts. Beaker is capped to prevent evaporation of the acetone.

Figure 49: Flowchart of experiment steps, the red ellipse shows the step where the first problems emerged.

Figure 50: From chips to shredded fibers

Figure 51: Microscopic view of shredded fibers

Figure 52: Illustration of mixing process, where cellulose inclusions appear to happen.

Figure 53: Adjusted flowchart of experiment steps, New step for shredding the cellulose chips was implemented. Next problem showed up at the stage where the red ellipse is positioned.

Figure 54: Adjusted flowchart of experiment steps, This process was used to create the material for the test prints.

Figure 55: Basic dry ingredients of material

Figure 56: Mixed dry materials with little demi water.

Figure 57: Product of the process, fiber reinforced lignin, ready to be tested.

Figure 58: Product of the process, fiber reinforced lignin draped in a petri dish

Figure 59: Medical syringe filled with material ready to be extruded.

Figure 60: Table with test data

Figure 61: Picture sample 1

Figure 62: Picture sample 2

Figure 63: Picture sample 3

Figure 64: Picture sample 4

Figure 65: Picture sample 5

Figure 66: Picture sample 6

Figure 67: Picture sample 7
Figure 68: Picture sample 8
Figure 69: Picture sample 9
Figure 70: Picture sample 10
Figure 71: Picture sample 11
Figure 72: Picture sample 12
Figure 73: Picture sample 13
Figure 74: Picture sample 14
Figure 75: Picture sample 15
Figure 76: Picture sample 16
Figure 77: Picture sample 17
Figure 78: Picture sample 18
Figure 79: Picture sample 19
Figure 80: Picture sample 20
Figure 81: start viscosity test
Figure 82: sample with finished viscosity test.
Figure 83: step 1 adhesivity test
Figure 84: step 2 adhesivity test
Figure 85: step 3 adhesivity test
Figure 86: step 4 adhesivity test
Figure 87: step 5 adhesivity test
Figure 88: sample 9, printed with a 20 ml syringe
Figure 89: sample 10, printed with a 20 ml syringe
Figure 90: sample 11, printed with a 20 ml syringe
Figure 91: sample 16, printed with a 20 ml syringe
Figure 92: sample 20, printed with a 20 ml syringe
Figure 93: sample 9, material in petri dish studied.
Figure 94: sample 9, material which was hand printed studied with a microscope.
Figure 95: material sample showing aligned fibers
Figure 96: material sample, clearly showing aligned fiber on the material surface.
Figure 97: water test: on the left material sample of batch 9. On the right material sample of batch 9 after being well in water for 168 hours.
Figure 98: printer which was used for printing the first samples
Figure 99: fitting for syringe
Figure 100: printer installed with syringe
Figure 101: First print made with 3d printer.
Figure 102: print test 2 Based on a 38 mm circle a print was made. Problems with extrusion made that the print had to be stopped.
Figure 103: third print made with 3d printer.
Figure 104: fourth print made with 3d printer.
Figure 105: fourth print made with 3d printer. Top view
Figure 106: Tree shaped columns at airport Stuttgart.
Figure 107: Schematization of tree shaped column.
Figure 108: Forces in tree shaped structure. Blue = bending moment red = resulting force.
Figure 109: Isometric view of tree shaped column.
Figure 110: Drawing of node.
Figure 111: Grasshopper code of tree branching column.
Figure 112: Drawing of tree branching column with detail drawing of node.
Figure 113: UR5 robot arm
Figure 114: Luthum clay extruder.
Figure 115 & 116: Stool designed for showing potentials