

Tree-D State of the Art Report

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1 Introduction

For more than twenty years there has been a continuing growth in the 3Dprinting industry [1]. In recent years the research and development regarding 3D-printing materials has increased. The main interest for this project is 3Dprinting techniques and materials. This project is a collaboration with Innventia, a world-leading Swedish material research institute. Their role in the project is to provide insight as well as the material to be used in the final prototype. This section will present the background to the project as well as the problem formulation.

1.1 Background

Today 3D printing/Additive Manufacturing (AM) techniques use materials such as liquid, solid and powder polymers; powder metals; and ceramics [2]. AMrefers to creating layer-by-layer three-dimensional objects using computer-aided design [3]. The major advances from using AM are the time and cost reduction, as well as the possibility to create almost any shape that could be difficult to machine. The most common material of use is although synthetic polymers, more known as plastics. Conventional plastic, which is mainly used, is made from petrochemical synthetic polymers. These are extensively used in a wide range of applications, e.g. for packaging, automobile, construction etc. [4].

There are several environmental issues related to the use of plastics in our products. Plastic pollution is a huge problem in the world today. It is globally distributed across all oceans due to its properties of buoyancy and durability [5]. According to Rochman et al., synthetic polymers in the ocean should be regarded as hazardous waste [6], due to the absorption of toxicants to plastic while traveling through the environment [7]. Plastic pollution impacts and endangers marine fauna, seabirds, and marine reptiles. According to Eriksen et al., in 2014, an estimation of at least 5.25 trillion plastic particles weighing 268,940 tons was floating in the sea [8]. This is far from the only issue related to the use of plastics. As described by Amin et al. [9], "The low degradability behaviour of plastics is an important environmental problem. The end-use of plastic creates waste-disposal problems as these plastics do not readily or naturally degrade and gives severe effect when plastic-waste requires more time to break down".

There is however an other alternative being presented as a high promising solution to the environmental problem of conventional non-biodegradable plastics. The solution, emerging from the bio-polymer industries advance in this field, is biodegradable plastics [9]. Biodegradation can be defined as the process in which organic substances are broken down by microorganisms such as bacteria, fungi and algae. Biodegradable plastics are derived from starch-based tubers such as sugar cane, cassava, tacca, and corn [10]. In Sweden up to this point, bio based plastics have not yet been created from the forest [11]. There is an initiative by SEKAB called Locally Produced Plastics to research whether it is possible or not to produce a biodegradable plastic from the Swedish forest. They have found that the demand for the finished product as well as the technology to create the material is already in place, but can not be implemented due to the lack of political incentives to facilitate investment decisions [11].

Sweden has a large wood industry as well as being a world-leading exporter

of paper, pulp and sawn timber [12]. Being able to make AM using wood material would have many environmental advantages to using plastics, especially conventional plastic. Innventia is developing ideas and concepts to make wood and wood component materials of choice in 3D processing. To reach further in the innovation process a better understanding of how to produce and process wood derived based components are needed. Fundamentally it is important to better understand the flow dynamics during the filament extrusion and how wood derived entities, e.g. fibres, fibre fragments or other solid components, mix, disperse and influence process ability and filament quality. Innventia has developed a pre-mixed filament containing 70 % of the biodegradable plastic Polyactic Acids (*PLA*) and 30 % of wood fibres.

1.2 Problem Formulation

Is it possible to make a machine that can create 3D-objects using free form AM using a material that are made from 100 percent wood material (Wood fibres, hemicellulose and lignin).

2 The Material

A big part of this project has been to understand the characteristics of the materials which will be used in the final product. This is a mechatronic project and even though the aim in this report has been to find the *State of the Art* among the many methods, there has also been a big emphasis on analyzing how the materials behave. This was important in order to not exclude any possible solutions. One special aim has been to understand the materials in order to get the most environmentally friendly outcome. The whole idea with the Tree-D printer is to reduce plastic use and use sustainable materials like natural fibres instead. Therefore material research has been important.

2.1 PLA

The filament material developed by Innventia contains the biodegradable plastic PLA. The material will be explained further in section 2.3. PLA are not new polymers [13]. PLA is a synthetic polymer derived from natural monomers [14]. Interest in the manufacture of an aliphatic polyester from lactic acids was pioneered by Carothers in 1932 [15]. At that time the product was of a low molecular weight, as well as having poor mechanical properties [13]. After further work on the product, DuPont patented it in 1954 [16]. It was not until 1972 that high-strength, biocompatible fibres, which are copolymers of lactic and glycolic acids, were introduced by Ethnicon. Their application was restricted until the late 1980s, when advances in the bacterial fermentation of D-glucose obtained from corn made it considerably cheaper to obtain lactic acid [13]. The PLA used in Innventias developed filament material is derived from corn in the USA, which is then manufactured in Japan before transported to Sweden.

2.2 Natural fibres

Natural fibres have been used in various works on the applications as composite materials. For example pineapple, sisal [17], coconut coir [18], jute, palm, cotton, rice husk, and bamboo have been used in various tests where tensile strength and young modulus have been tested. [19] In this project wood fibres are of interest, and since wood itself is a composite, it has been useful to examine the methods used for the different natural fibres mentioned above. Natural fibres are gaining more interest as composite materials as they have many attractive properties, such as low weight, higher stiffness and lower cost. [17] Also, environmentally friendly composites have been a hot topic recently due to the increasing environmental awareness. [20]

The natural fibres which have been interesting for application in this project are mainly spruce, pine and birch, since these are the most common trees in the Swedish wood industry. [21]

The fibres are treated differently, depending on what tests will be done. The fibres can for example be woven, as specified by Jones [22]. This method is further discussed in Section 3.4.

The applications of natural fibres are plenty, and are currently used as reinforcements in composite materials for automotive, aerospace, marine, sporting goods and electronic industries [23]

2.3 Cellulose-PLA Super Material

This material has been extra interesting to investigate, since it originates from Innventia which are stakeholders in this project. Innventia has investigated certain demands on materials, such as transparency for food packaging. The solution they have found is to mix PLA and cellulose fibres, which makes the material's characteristics depend on the heat and force with which it is pressed. [24] By doing this, only one material is needed for the packaging, instead of paper for the box and plastics for the transparent window. If the PLA originates from local producers, and the super material is recycled after use, this is highly environmentally friendly. Bags made of the Cellulose-PLA material has also been shown to be lighter than traditional paper bags in the same size.

2.4 Lignin

All natural cellulosic fibres contain lignin [23] which is a polymer in the cell walls. The structure of lignin is very complex, and the amount differs between different plants and different kinds of woods. Some examples of the composition can be seen in Table 1.

Type of bio fibre	Source	Cellulose	Lignin
Wood	Hardwood	43-47%	16-24%
Wood	Softwood	40-44%	25-31%

Table 1: Composition of lignin and cellulose between different types of wood [23]

Lignin has been interesting in this research, since it is a component of wood and also already is used in 3D-printing filaments. Therefore the possibility to use lignin as the binding substance in the final material in this project might be a possible solution to regain the characteristics of wood.

2.5 Commingled Material

Commingling is a process where two or more materials are getting physically mixed or mingled to a uniformly distributed blend. Most often the term is used when a matrix material, usually plastic, is getting reinforced with fibres. These composite materials are intended to give better material properties than the components separately. The commingled material can have different shape, texture and property depending on the basic materials, blending ratio, the commingling techique used and so on. The materials are suitable for different applications and can give a wide range of properties depending on how these are processed. Usually the commingling is made by extrusion or compounding, but there are other methods as well [25].

Innventia has developed a commingled material composing of PLA and wood fibers, where the fibres are kept intact and the fibre content is up to 40%. This composite has a paper or fluffy cotton pad like texture and is produced by wet forming preferably followed by hot molding [24] [26].

Comfil, a Danish company, uses its own commingling process. The COMFIL® technology is done through commingling the fibres at filament level with compressed air [27].

2.6 Paper

The making of paper is an interesting subject to research about for this project because paper mainly consists of wood and to understand how different methods of manufacturing can alter the papers material properties. Paper can also be used for 3D printing using methods called SDL or LOM which will be discussed further in section 3.5

Wood mainly consist of three components, cellulose, lignin and hemicellulose. When making the pulp for paper manufacturing lignin is often removed to allow the fibres to be separated easily. The pulp can be made from 4 different processes, chemical, semi-chemical, chemi-mechanical and mechanical pulping. They are in order of increasing mechanical energy used and decreasing chemicals used to separate the fibres. The more chemicals used the more lignin is removed from the pulp. Thus, mechanical pulping does not remove any of the lignin. The strength of a paper can be measured from two characteristics, the strength of the individual fibres and the strength of the bonding between the individual fibres. The strength of the individual fibres is dependent of the angle of the micro fibrils that the fibres consist of. The angle of the fibrils is different in different types of wood. Paper fibres are held together by hydrogen bonding from the hydroxyl groups in cellulose and hemicellulose. As mentioned before chemical pulping decrease the amount of lignin in the paper, this is to make it stronger because ligning interfers negatively with the hydrogen bonding between the fibres [28].

3 State Of The Art

This section will present all relevant scientific report on 3D printing processes and AM. It covers Extrusion, AM with Fibres, Spray Up Technique, Weaving Fibres and Paper 3D-Cutting.

3.1 Extrusion

Extrusion is interesting to research for this project because a lot of methods of 3D-printing are depending on some kind of extrusion. This section is about the classic use of extruders and what can be learnt from them.

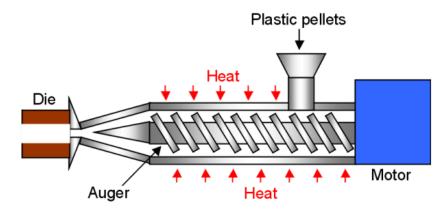


Figure 1: How extrusion works [29]

Extrusion is a process that is mostly used to create objects with a fixed cross-sectional profile. One or more materials are feed into the extruder and then the materials are pushed forward and mixed by one or two big screws. At the same time the materials are heated by the shear friction from the screws and with help from external heating. When the material is mixed and no longer solid the screw will push it through the iron die with the desired cross-section. This is possible because "During extrusion, Compressive and shear, but not tensile, forces are developed in the stock, thus allowing the material to be heavily deformed without fracturing" [30] which means that you can make very complex cross-sections without the risk of the material breaking. "The main purposes of the heaters are to melt the polymer that remains in the barrel at cold startup, to assist in forming the initial melt, and to "trim" the barrel temperatures for specific purposes such as improving feed rate." [31] which makes the shape of the screw important depending on what kind of material that are being processed.

The kind of extrusion that is interesting in this project is where plastic and wood fibres are put into the extruder and made into pellets that are used for injection molding. This process is used by Innventia to try to make use of wood in new areas and therefore very interesting for this project. The problem with putting wood fibres through an extruder is that it will crush the fibres and the material will lose of the benefits with the fibres.

3.2 Additive Manufacturing with fibres

There has been little research regarding 3D-printing with wood fibre material. Therefore it is essential to also study 3D-printing with other types of fibres such as carbon fibre as the structure of the material is similar.

Fused filament fabrication (FFF) with fibre reinforced plastic is the most common way to 3D-print fibre materials. There is a large amount of different fiber reinforced material commercial available. In 2000 Zhong et al. [32] showed that FFF printing with glass fibre reinforced plastic filament increased the mechanical strength of the final part compared to a non-reinforced plastic part. Furthermore Zhong et al.[32] observed that the fiberglass reduced shrinkage of the 3D-printed part. Because of the stiffness of fibres, increasing the fibre content severely effect the brittleness of the filament. A glass fibre content of more than 25 % was not possible to use with a FFF 3D-printer. Filaments for FFF are manufactured using extrusion as mentioned in 3.3. Increasing the fiber content in the plastic mixture fed to the extruder results in shorter fibers in the filament produced[33].

With the aim to increase the fiber length in the 3D-printed part a new technique called Continuous Filament Fabrication (CFF) has been developed. It is based on a FFF machine with a few modifications. The biggest difference with FFF is that the polymer and the fiber is not bonded together before entering the print head. A few variations of the technique exists.

Matsuzaki et al. [34] have adapted a FFF printer for use with CFF. The printer head have two inlets, one for the thermoplastic filament and one for a fibre wire, in this case they use unidirectional twisted jute yarn and unidirectional carbon fiber tow. A motor driven drive gear push the filament into the nozzle. The fibre wire is fed to the nozzle inlet and impregnated with the melting plastic as shown in Figure 2. The fiber wire is pulled by the plastic filament, no extra driving mechanism is needed. It is possible to orient the direction of the fibers by adjusting the path of the nozzle. The length of the fiber is intact in the printed part. The Tensile strength of both the jute and carbon reinforced specimen was increased compared to pure PLA which was used as binding material. Furthermore Matsuzaki et al. [34] states that the carbon reinforced specimen had superior tensile strength compared to fiber reinforced plastic part made by AM. Another benefit of this method is that no pre-treatment of the fiber is necessary, which makes it possible to use a vast range of different fiber material. Matsuzaki et al. [34] assumes that it is possible to have a fiber volume of up to 50 % with this method

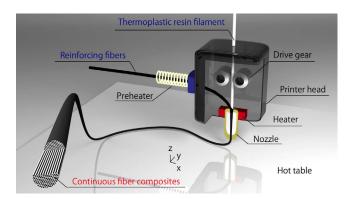


Figure 2: CFF process with one nozzle [34]

Mark Two is a Commercial off-the-shelf 3D-printer that uses CFF. It uses a nylon based polymer which is possible to reinforce with glass fiber, carbon fiber or Kevlar. The printer head has a nozzle for the nylon filament as well as a special nozzle for the fiber tow. The fiber tow is pre-coated with Nylon. The layer adhesion binds the nylon coated fiber tow with the nylon matrix on the print bed [35].

3.3 Spray Up

The Spray Up-process is an additive process method. A chopper gun is fed with fibres that are cut off and sprayed to a surface simultaneously with resin. The mixed material is then rolled with a roller in order to get rid of air on the surface. [36] The process is usually done manually by a operator who therefore control the thickness. The main advantage with the process is that the equipment is portable and on-site fabrication can be made.

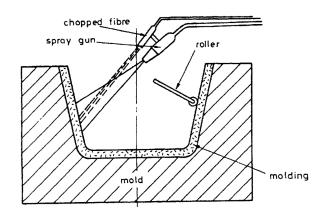


Figure 3: The spray-up process [36]

3.4 Weaving Wood Fibres

In the methods described in Section 3.2 and 3.3, the direction of the fibres are important. For Mark Two, the fibres has to be spun into a yarn like material. In cotton textiles, cotton fibres are woven into a thread, which then is used to weave a fabric. When textiles are made, the fibres has to be at least 5mm long, and we know both pine and spruce have fibres which are up to 6mm long. [21] In *Mechanical Properties of Woven Banana Fibre Reinforced Epoxy Composites* [19] banana fibres are used to reinforce an epoxy material. Stress tests and young modulus tests are done on the material and the results showed that the characteristics of the material were good when it came to how much force it could take.

In *Mechanics of Composite Materials* [22] the characteristics of fibre-reinforced composites are discussed. When the fibres are woven perpendicularly to each other the material becomes stronger than when the fibres are woven in 45 $^{\circ}$ angle.

3.5 Paper 3D-printing

This method consists of gluing papers together and cutting out a preferred shape using either a laser or a blade. The process is as follows. First a piece of paper is applied to the workplace, then the layer is cut according to the model from the user, glue is then applied to this layer and ultimately a new sheet of paper is applied on top and pressed against the previous layer to bond the two papers together. There are two different, yet very similar, methods to do this, *Selective deposition lamination*, *SDL* and *Laminated object manufacturing*, *LOM*. The difference between these two methods is that *LOM* applies glue uniformly to the whole layer while *SDL* deposit more glue to the area that will become the part and less to the surrounding area that is the support structure. *LOM* uses a laser while *SDL* uses a blade, usually a tungsten carbide blade, to cut the edges of the part.[37]

4 Status Report

The project work so far has been focused on researching StateoftheArt. A couple of 3D-printing tests have been done with existing materials on the market. Based on the SOTA-research it has been decided to work further with CFF. A meeting is set with Innventia in order to discuss the possibilities of how to make fibres into a thread. For machinery it is decided to order a Iprusa3, but depending on the delivery time the order can be made in the autumn.

5 Discussion

The researched methods and materials above all have their advantages and disadvantages. Some can be combined, and some cannot, and this gives us endlessly of possibilities for a project. However, the aim in this project has been to increase the ratio of wood fibres in AM without loosing the characteristics that wood has. The critical functions that were found were:

- To mix wood fibres with binding substance
- To add as much wood as possible
- To control the directions of wood fibers, in x and y direction
- To keep long fibers intact
- To be able to maintain shape after print

The research was set against these critical functions to be able to evaluate them and find a solution for this project.

The use of a biodegradable plastic in the material is an alternative with good prospects. The current use of PLA in the filament, due to the long transportation of both raw material and finished product, is not a sustainable solution to the problem. Since this project has a limited time frame and funding, using PLA for this prototype could be a temporary option. If the initiative and technology Locally Produced Plastics, using wood material from Sweden to create a bio-based plastic, could be funded and realized in the future, it could be an interesting solution to use as a binding substance in AM, and something to research further on.

Natural fibres are already in use as reinforcement in many different composites. The fibres then serve as the matrix, and are an environmentally friendly option in many products. However, the epoxies and "glue" materials are often made of plastics or other materials which are not broken down. *PLA* seems to be one of the better options.

The multimaterial approach which Innventia apply is new thinking and high tech. *PLA* and wood-based fibres have properties that complement each other in many ways, but there are still problems when it comes to recycling. Also, in this project we do not need some of the characteristics that the super material has, and therefore it is redundant.

Different natural fibres have been used to reinforce composites in the methods described above. In this project the aim is to print a material which has characteristics as close to wood as possible, and with fibres from Swedish trees. One idea is to use cellulose fibres with lignin as glue, to recreate the characteristics of wood.

The commingled material is difficult to use for AM because of the material texture. To be able to use the material for AM, for example 3D printing, a filament is being created, through extrusion, but the drawback is that the fibres are getting damaged during the process.

The problem with extrusion is that when you put long fibres through an extruder the fibres is crushed by the screw/screws inside the extruder and make it almost pointless to use wood fibres in the first place since they no longer add any strength to the material and make it a lot more expensive to produce without that much gain in strength and material properties.

3D-printing using FFF limits the benefits of a reinforced fiber composite. Wood fibre filaments that are available have no or very little improvement of the mechanical properties compared to the binding plastic. It is restrained both by the printing technique and manufacturing process of the filament.

However CFF adapts the FFF technique in order to use fibre composites. As described in Section 3.2 it can be highly customized in order to work with different fibres and binding materials. As it is possible to modify an existing FFF printer, much of the fundamental parts of 3D-printer is already done. Thus it allows us to focus on the binding and extrusion of the wood material. Although parameters such as printing speed and resolution will be low. It is not of great importance as the critical functions described regarding material and mechanical properties seems feasible to achieve with this method.

The Spray-up method is manual, for this project a more mechatronic solution is more suitable.

The method of weaving fibres, which is described in Section 3.4, provide us with the possibility to give the fibres a direction. However, it might be difficult to weave fibres as short as the ones we have in Swedish trees. Still, the knowledge from this area will be used in the project since fibre direction is crucial and we can learn a lot from this technique.

The disadvantage of using SDL or LOM is that it creates a lot of waste and also that it is using paper that needs a lot of energy to be produced. The pulp and paper industry is the fifth largest energy consumers and accounts for four percent of the world's energy use [38]. This method will therefore not be used in this project.

5.1 The Result

After the research in this *State Of The Art* report was done a concept had to be chosen. This was done by evaluation of the possible outcomes by combining the good qualities from different methods described above.

As a result, the CFF was chosen, but adapted to fit our needs. From this technique we get the possibility to choose direction of the fibres, and also to maintain their length. This could mean that we can increase the amount of wood fibres (compared to SOTA materials). For this concept we need wood fibres woven into a thread, for which we need Innventia's help. As binding substance we hope to use lignin, as it is a part of natural wood. If absolutely needed, PLA will be used together with the lignin.

Finally, we also think this could be a project where we can adapt our mechatronic knowledge and make a really good product.

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