Developing Edible Barcodes from Laser Cutting Biodegradable Materials

By

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Abstract

The counterfeiting of food and pharmaceutical products is slowly becoming a very dangerous threat for the global healthcare. In developing countries 10% of the medicines in circulation are already counterfeit. Current anticounterfeiting methods do not offer the right protection for the customers. Most of the anticounterfeiting methods are on the outside packaging, where they can be easily counterfeited or replaced. Furthermore, research has already shown that new anticounterfeiting methods are possible. These new methods are no longer on the outside packaging but on or in the product. Although these new anticounterfeiting methods show promising results, they have difficulty complying to the strict rules of regulatory organisations. This research focuses on developing edible barcodes by laser cutting different biodegradable/food grade materials. Besides the laser cutting this research also focuses on the formation and the properties of glycerol-alginate sheets. These sheets are made by mixing the glycerol and alginate under high temperature. After the sheets are produced, they are put in contact with water and their behaviour is observed. Overall, this research provides an insight that it is possible to develop such barcodes from laser cutting biodegradable/food grade materials, although much research on glycerol-alginate sheets still needs to be done.

Contents

1

Introduction

Globalization makes it possible to distribute an increasing number of medical products, like medicine and vaccines, around the world. As a result, more people around the world get access to the healthcare they require. However, there is an increasing amount of counterfeit medical products in circulation. According to the World Health Organization (WHO) 10% of the medical products are counterfeits or substandard. Simultaneously 42% of the recorded cases of medicine counterfeiting came from the Sub-Saharan African region.[1] Not only in the pharmaceutical sector counterfeiting is a large problem, but counterfeiting is also in the food sector becoming more apparent. In 2008, 300000 babies were affected by milk powder that was contaminated with melamine.[2]

The current method of authentication is not sufficient anymore. The counterfeiters can repackage the food or medicine products with high accuracy[3]. For better product authentication, methods have to be developed to ensure product identification. Methods using isotopes[4], DNA particles[5], microparticles[6], nanoparticles[7], [8] and Radio-Frequency Identification (RFID) tags[9] have been discussed in literature and some of these methods have also been commercialized (Safetracers™[10], Taaneh™[11], Trutag™[12]and SmartTrac[13]). The reason that these products are not applicable on a worldwide scale is due to the requirement of a special reader or special method to authenticate the product. Making the identification of the product for the users of the food or medical product more difficult.

To ensure that pharmaceutical or food products are difficult to counterfeit, authentication tags have to be placed in the product. If authentication tags are placed in the product, the tags must not harm the user. Therefore, the placed tags should be biodegradable. Several studies have shown results in developing biodegradable authentication tags or films.[14]–[16]On the tags a QR or barcode is imprinted. The customer can scan such code with a clip-on microscope[17] The QR or barcode contains the information regarding the contents and origin of the product. The microparticle created by Rehor et al.[14] not only contained a barcode, but also has two sensors built in. The first one is a pH-sensor that indicates when the product is contaminated with micro-organisms. The second one is a temperature sensor that causes a deformation to the tag due to a change in temperature. Unfortunately the chemistry used in to produce the tags is free-radical polymerization[14], [15] which has chemistry that is difficult to define. Because the tags will be placed in edibles, the chemistry has to be defined otherwise the tags cannot be placed in the product.

To further develop this technology, new methods to transform biodegradable raw materials into authentication tags have to be researched. Research has to focus on simple and definable chemistry or on alternative methods that require no chemistry at all. Methods that look promising
are tablet pressing or wet pressing powders to manipulate their shape, using alginate hydrogels to produce edible barcodes or laser cutting/etching dry sheets made out of biodegradable materials, for example dried seaweed also known as nori, to create barcodes.
The work described in this thesis, focuses primarily on laser cutting and testing of different materials and can be used as a framework for further research. The aim in the future is to provide a working method that is applicable on a global scale, especially in the food and pharmaceutical industry.

2

Theoretical Background

The theoretical background presented in this chapter will first focus on polymers and their variations. After that it will focus on the specific polymer used in the work presented. The last part will focus on the basic of lasers and their application. This information can be useful to understand the work presented in this thesis.

2.1 Polymers

A polymer is a substance that is composed out of macromolecules which in turn are composed out of monomers. These monomers can be linked together via several chemical or physical mechanisms.[18] Polymers can be sorted by skeletal structure, molecular structure and variety (Figure 1).

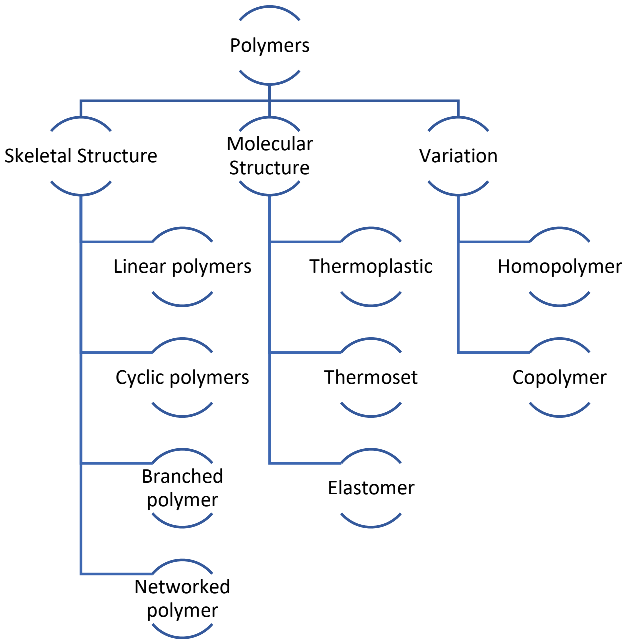

Figure 1: The different ways polymers can be sorted[19]

The several subclasses displayed in Figure 1 , determine the structure and the behaviour of the polymer. If the polymer is linear skeletal structure it is a chain with two ends. Besides the linear structure there are several non-linear structures. *Cyclic polymers* have no end chain and show different behaviour compared to their linear counterpart.[18] When the linear polymer has side chains or branches bonded to the main chain it is called a *Branched polymer*. When these branched polymers connect with each other and form a 3D structure, it results in a *Networked polymer*.[18]

Besides skeletal structure polymers can also be classified by their molecular structure. They are separated in three main groups *thermoplastics*, *elastomers* and *thermosets*.

Thermoplastics are linear or branched polymers that can melt when they are heated and can be moulded in every shape possible. They can be further separated into crystalline polymers and amorphous polymers. Generally, these polymers are semi-crystalline with both crystalline and amorphous regions.[18]

Elastomers are rubbery polymers that are networked with each other. They have the ability to easily stretch 3x to 10x their original size and can easily return to normal shape when the applied stress is released.[18]

Thermosets are often rigid materials and consist out of network polymers with a high degree of crosslinking. Thermosets degrade under the application of heat rather than melt like the thermoplastics.

Likewise, a polymer can be a homopolymer or a copolymer. A homopolymer consist out of the same monomer an example for a homopolymer is polyethylene. A copolymer consists out of multiple monomers. There are several sorts of copolymers defined by their specific arrangement. [18].

Block copolymers are linear copolymers in which the repeat unit only is in so called blocks of the same monomer. *Grafted copolymers* are branched polymers in which the branches differ from monomers than the main chain.[18]

When polymers are made by living organisms they are called biopolymers. However not all biopolymers that are produced are biodegradable or biocompatible. These two terms don’t have the same meaning. Biocompatibility means the ability for a substance to be in contact with a living system without producing an adverse effect.[20] Biodegradability is defined as the ability for a substance to be degraded by a living system.[20]

2.2 Alginate

2.2.2 Introduction

Alginate is typically extracted from brown algae (Phaneophyceae) by treating it with an aqueous alkali solution, mostly NaOH. After filtrating the extract, Na-Cl or Ca-Cl2 is used to precipitate the alginate[21].

2.2.1 Structure of Alginate

The structure of alginate consist of linear copolymers, with blocks of (1,4)-linked β-d-mannuronate (M) and α-l- guluronate (G). [21] The blocks are composed out of G residues (GGGGGG), M residues (MMMMMM) and alternating G and M residues (GMGMGM) (Figure 2)

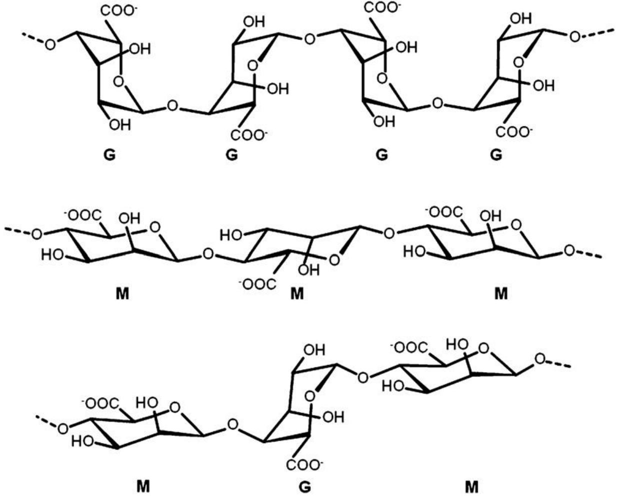

Figure 2: The different blocks that can exist in alginate

2.3 Laser

2.3.1 Introduction

The word Laser is actually an acronym that describes the working of a laser.[22] The meaning of this acronym is **L**ight **A**mplification by **S**timulated **E**mission of **R**adiation.

There are three parts in a laser to form a beam of light: a lasing or gain medium, an optical resonator and a pump source.[22] The pump source is used to excite the atoms in the lasing medium. The lasing medium normally consist out of solid, liquid or gas. These excited particles will send out photons, these photons will oscillate in the optical resonator to form a concentrated beam of light.

This beam of light emitted by a laser can be categorized by single wavelength, which results in a single colour of light beam also known as monochromaticity, same phase position also known as coherency and low divergence. These characterizations make the laser special and suitable for its different applications.

2.3.2 Types of Laser

There are several types of lasers. Solid state lasers, gaseous state laser, liquid state laser, chemical laser and semiconductor laser.[22] All these lasers have different lasing mediums, different pump sources to excite the atoms and different wavelengths that are emitted. (Table 1).

Table 1: Different types of lasers and their properties

Type of Lasers	Lasing Medium	Excitation	Wavelength
Solid State Laser	Solid (e.g. Ruby)	Absorption of pumped photons	Infrared (~1064nm)
Gaseous State Laser	Gas (e.g. CO2)	Transitions between vibration and rotation states of bonds	Infrared (~1000nm)
Liquid State Laser/dye laser	Liquid (e.g. Rhodamine)	Flash lamps	Ranging from Ultraviolet to infrared (~100-1000nm)
Chemical Laser	Chemical substances (e.g. COIL)	Exothermic chemical reactions	Infrared
Semiconductor Laser/Diode laser	Semiconductor (e.g. InGaN)	Electric pumping	Ranging from visible to infrared (~405-100nm)

2.3.3 Properties of Lasers

There are 3 main properties that make a laser so unique and efficient. That are monochromaticity, coherency and directionality.

Normal light emits a broad range of wavelengths. A Laser only emits a very range of wavelength and is therefore considered as a single wavelength radiation (Figure 3) . [22] This effect is called *monochromaticity*. With monochromaticity, a high intensity energy is achievable because all the energy is concentrated in this one wavelength.[22] The monochromaticity origins from 2 conditions. The first is that the emission of photons by excited atoms has well defined energy levels. The second condition is that the generation of the beam involves bouncing the atoms between two mirrors resulting in a narrower spectrum of light.

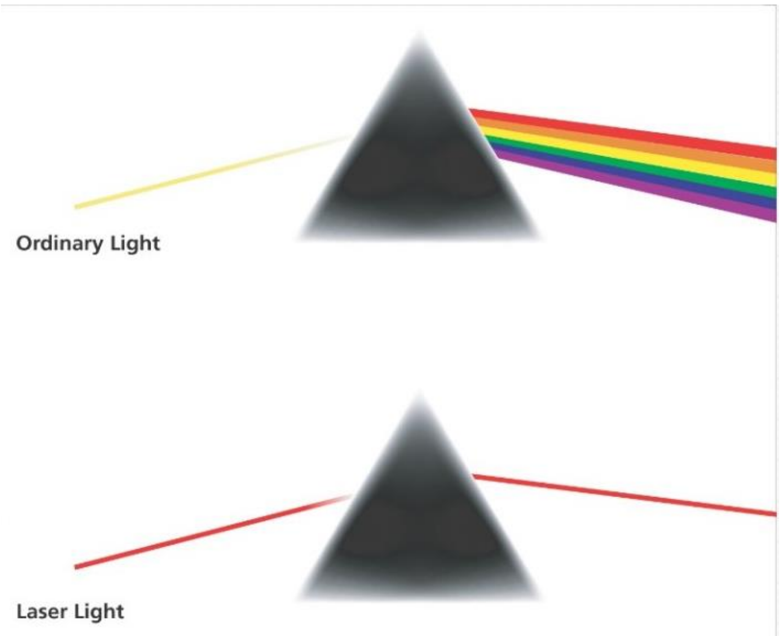

Figure 3: The difference between the diffraction of normal light compared to the light of a laser

Coherency is another special property of a laser. This means that all the waves in a laser are of the same phase. They have both temporal and spatial coherence.[22] Spatial coherence is that the amplitude of a wave at any point in space and at any time can be predicted. For temporal coherence, there is a correlation between waves at any time in the light beam while spatial coherence has a correlation at both any time and any space (Figure 4). Because of this coherency, two waves that join together will produce a magnified wave that is stronger.

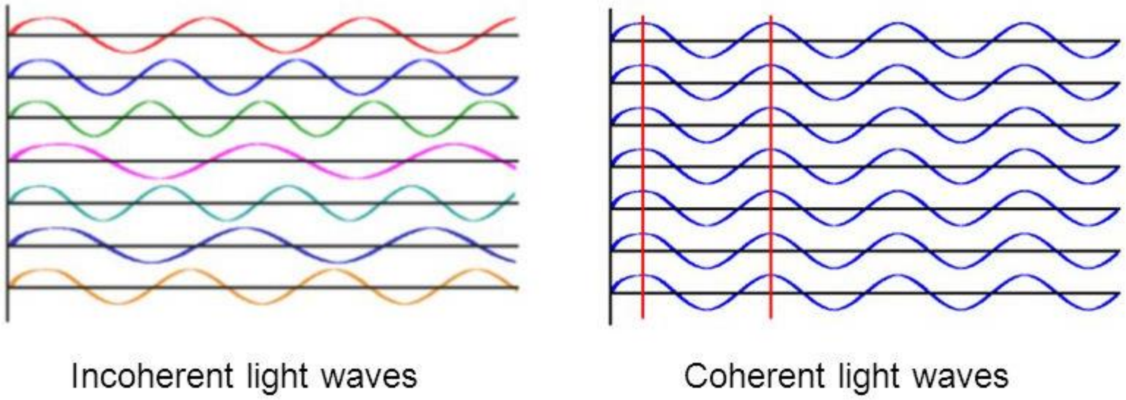

Figure 4: The difference between incoherent and coherent light waves

Directionality is the last important property of a laser. It describes the limited divergence of a laser beam. Usually, light is emitted in every direction by the irregular emission causing the photons to spread out very quickly. Lasers are able to minimalize the spread of photons. Because of this, lasers do also not lose their intensity with distance.[22]

2.3.4 *Interactions between Laser and Material*

Because of the unique properties of the laser such high directionality and high power. It is possible to process several places in materials in a very detailed way without altering the material around it. The interaction between the laser and the material depends on several factors such as the laser wavelength, the beam spot size, power etc., but it also depends on the material properties such as reflectivity, and the thermal conductivity property of the material.[23]

When the laser beams comes in contact with the surface of the materials, two things happen: Coupling or absorption of the laser beams by the material and heating or melting of the material.[22] The beams that hit the surface will either be reflected or absorbed by the material depending on the properties of the material. The beams that are absorbed are now coupled with the material. When the atoms of the materials have absorbed enough energy, the atoms will be excited. The atoms will now be in a higher energy state and will collide with each other. The collisions between the excited atoms will result in heat, which will heat up the material. How the heat is distributed throughout the material depends on the properties of that material. In laser cutting the laser will mostly be used to remove the material in form of liquid or vapour.

2.4 Thesis Objective

The work in this thesis is inspired by earlier work done by Pavithra Vasanth Bailey[19] and research from Rehor et al. [14]. The work mainly focused on taking an alternative route by mainly focusing on the laser cutting of different biodegradable materials.

The thesis objective was to check if it was possible to create barcodes out of biodegradable materials by laser cutting these materials. To check if this is possible, several different materials need to be researched. For this thesis, three different materials were researched. Nori paper made out of dried sea weed. Rice paper made out of dried rice residues and Glycerol-Alginate sheets.

3

Materials & Methods

In this section the materials used in the experiments and the procedures of the experiments are explained.

3.1 Materials

Sodium alginate (Na-Alg) was supplied from Sigma Aldrich (CAS number 9005-38-3). The purity of the sodium alginate is food grade. The glycerol had at least the purity of food grade. The nori and rice paper sheets were bought from the Albert Heijn. The water used in all the experiments was deionized water from an in-house dispenser at atmospheric pressure.

3.2 Determining dissolving rate of Glycerol-Alginate films

In the following sub-sections, the method of determining the dissolving rate of Glycerol-Alginate films will be explained. First the method of preparing the Glycerol-Alginate films and after that the actual experiment will be explained.

3.2.1 Preparation of Alginate solutions

The Na-Alg was added in DI water. The solutions were covered with a parafilm and stirred on a hot plate at 70 °C for approximately 20 minutes and / or until the Na-Alg was completely dissolved. The solution was cooled down. The necessary amount of Na-Alg was added to obtain the desired Na-Alg concentrations of 1%, 1.9%, 3%, 5.1% and 8% w/w respectively.

3.2.2 Preparation of Glycerol-Alginate films

The glycerol and the six different Na-Alg solutions were mixed in falcon tubes manually. Different amounts of glycerol were added to obtain different mixtures. See **Appendix A** for all the different final mixtures. The falcon tubes were centrifuged to remove the air bubbles and create a homogenous mixture. The solutions were poured on petri dishes and dried in an oven at 38 °C for 24 hours.

3.2.3 Dissolving Glycerol-Alginate films

Sample 13 and 18 a small piece was cut and was put in a 15cm petri dish. The petri dish was placed under a Vividia Digital microscope[24]. 5 ml DI water was added to the petri dish to submerge the small sample in water. Pictures were taken by the microscope every minute for 1 hour long.

3.3 Laser cutting nori/rice/Gly-Alg sheets

In the following sub-sections, the laser cutting of sheets made out of different materials is done.

3.3.1 Laser cutting with NEJE DK-BL laser

The first laser used to cut the materials was the NEJE DK-BL laser.[25] This laser has a power of 1.5 Watt and the source of the laser is a Nichia diode with a wavelength at 405nm. Different sheets of materials were placed beneath the laser and a basic shape of barcode was used to cut out of the material (Figure 5) and to test if the laser could successfully cut the shape out of the materials.

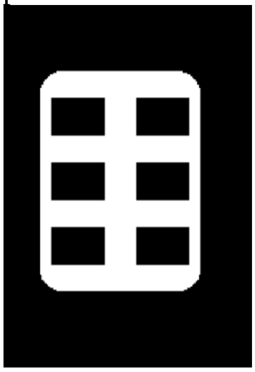

Figure 5: The shape that was used to cut the sheets

3.3.2 *Laser cutting with Epilog Zing Laser*

After the first experiments a larger laser was used. The laser used in these experiments is the Epilog Zing[26]. This laser has a max power of 40 Watt and the source of the laser is CO2 with a wavelength at 10.6 µm. It can be set in different settings, so it is able for processing different materials.
The Glycerol-Alginate sheets were put underneath the laser and were cut and engraved with the barcode of the site erallab.com which was created by https://www.the-qrcode-generator.com. (Figure 6).

Figure 6: QR-code that was used for engraving

4

Results & Discussion

This section focuses on discussing the results that were obtained from the different materials in forming the different Gly-Alg sheets and dissolving some in water. Furthermore, will the results of laser cutting the different biodegradable sheets with the two lasers be discussed.

4.1 Gly-Alg sheets

The different sheets were successfully formed. By increasing the alginate concentrations, there is also a change made in the colour of the sheets. The colour change of the sheets is due to the natural colour of alginate[27], [28] and because the concentration of alginate increases the colour of the sheets will likely more and more match the colour of the alginate. Furthermore, increasing the glycerol concentration of the sheets resulted in more sticky and more soft sheets.[27], [28]. The higher the concentration of glycerol, the more the sheets adopted the stickiness of the glycerol.

Unfortunately, there has not been time to further investigate if all the water is dissolved after drying or if there still remains some water inside the sheets. There is no certainty that all the glycerol stays in the sheets or that some glycerol just like the water also evaporates when the solutions is being dried.

4.2 Dissolving Gly-Alg sheets

After the successful formation of the sheets the sheets with 3% w/w alginate, 0% w/w glycerol and with 3% w/w alginate, 3.1% w/w glycerol are formed. They are put back into DI water and their dissolving behaviour is closely watched.

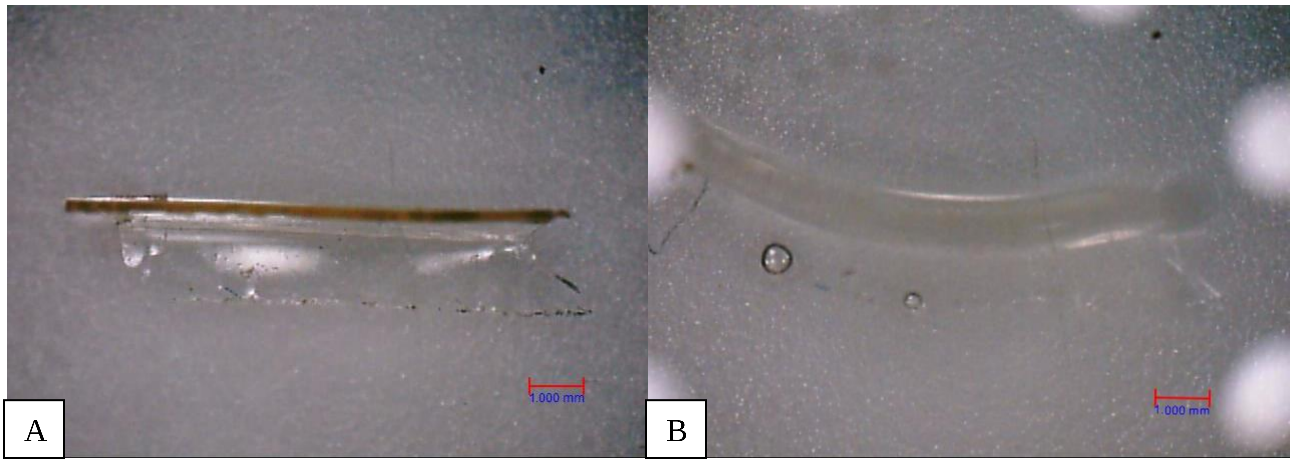

Figure 7 A is a piece of the sheet containing 3% alginate and 3% glycerol just before the 5ml DI water was added; B is a piece of the sheet containing 3% alginate and 3% glycerol 2 minutes after the 5ml DI water was added;

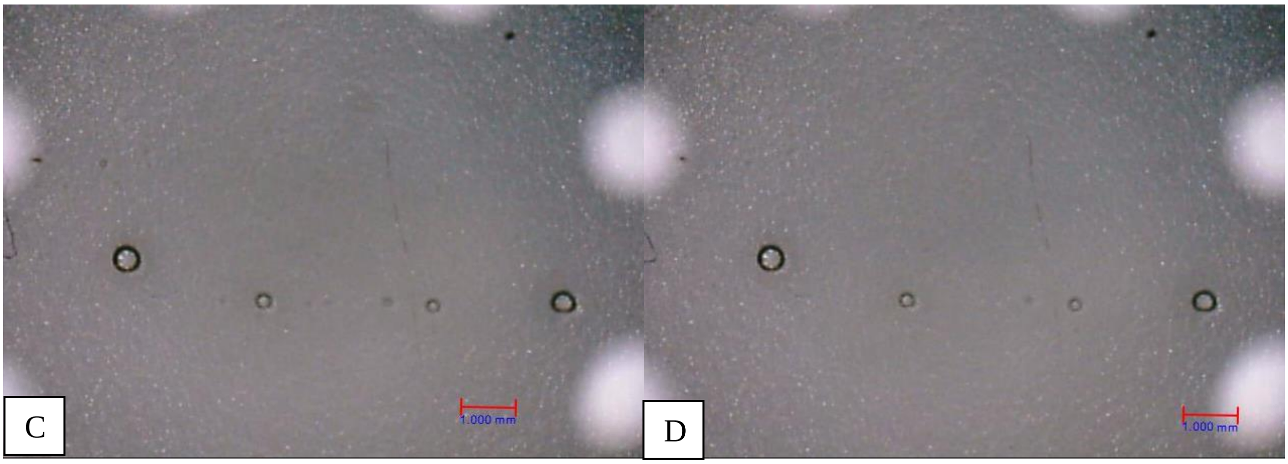

Figure 8: C is a piece of the sheet containing 3% alginate and 3% glycerol 10 minutes after the 5ml DI water was added; D is a piece of the sheet containing 3% alginate and 3% glycerol 60 minutes after the 5ml DI water was added.

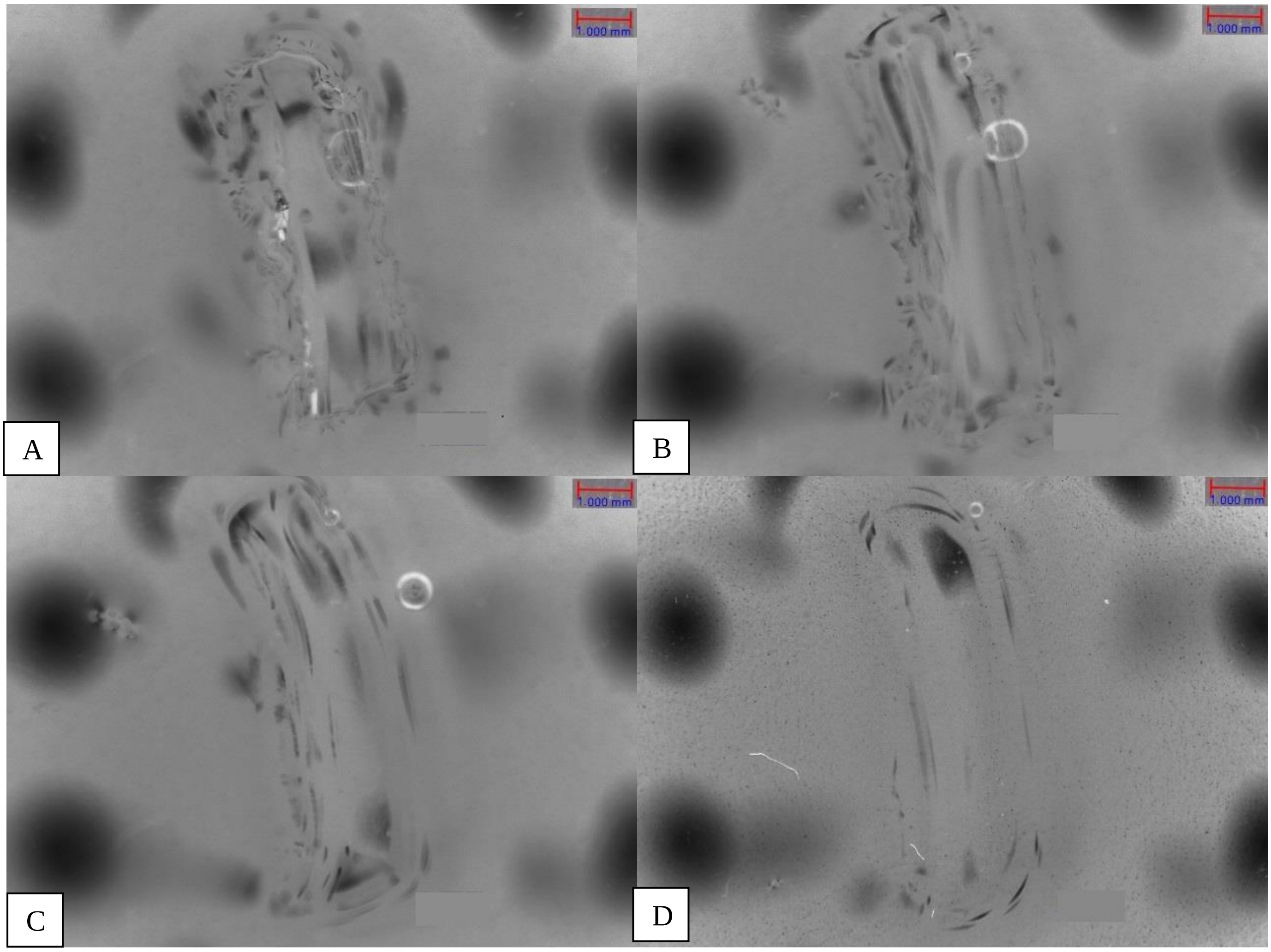

Figure 9: A is a piece of the sheet containing 3% alginate and 0% glycerol just after the 5ml DI water was added; B is a piece of the sheet containing 3% alginate and 0% glycerol 5 minutes after the 5ml DI water was added; C is a piece of the sheet containing 3% alginate and 0% glycerol 20 minutes after the 5ml DI water was added; D is a piece of the sheet containing 3% alginate and 0% glycerol 60 minutes after the 5ml DI water was added.

Comparing the results (Figure 7, Figure 8, Figure 9), the conclusion can be made that the sheet containing the higher percentage of glycerol dissolves faster into the water than the sheet that contains no glycerol at all. However, Gao et al. have reported that adding glycerol to alginate sheets will improve the homogeneity of the alginate in the film[28]. Consequently, when these films are put back in water the alginate is more dispersed in the film so therefore the film will more easily dissolve than the sheets where no glycerol is added at all because here the alginate is more concentrated on one spot and will take longer to fully dissolve.

One note is that the size of both the samples does not 100% match. This has influence on the results and a better way of taking the same size samples needs to be considered. One option can be a biopsy stamp to ensure that every sample has the same size.

Because there is still a lot unknown about the Gly-Alg sheets, further tests have to be done to improve our knowledge of these sheets and fully understand what the properties are of the material and what happens when you increase or decrease the alginate or glycerol concentration of the sheets.

4.3 Laser cutting biodegradable nori/rice/Gly-Alg Sheets

For the first laser cut experiment, the nori and rice sheets are put under the NEJE DK-BL laser. The nori sheets can easily be cut by the laser into the preferred shape. The rice paper on the other hand, is very difficult to cut in the preferred shape. The sheet has to be coloured black before it is put underneath the laser, because the rice paper is much more reflective compared to the nori paper. Because the rice paper is much more reflective, it will absorb less photons meaning the laser will have a much lower impact.[22] After the rice paper is blacked out, it is easier to cut through. However, the result is still not optimal. There are parts the laser has burnt through but also parts the laser is unable to vaporize the rice paper sheet.

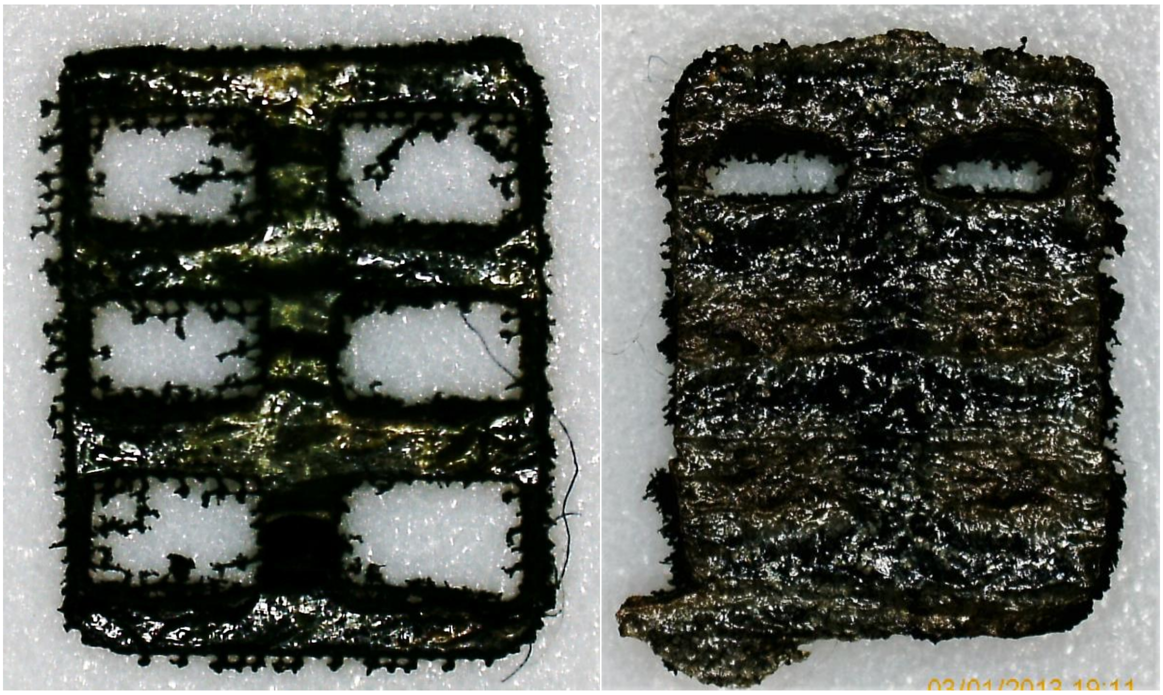

Figure 10: Nori and Rice sheets cut by NEJE DK-BL laser

For the second experiment an advanced laser of the science centre in Delft is used.[26] This laser can easily cut through the nori, rice and Gly-Alg sheets. This laser has more power and uses a laser with a longer wavelength. Because light with a smaller wavelength carries more energy than light with a larger wavelength[22], it is expected that the power or the dot size of the laser has an important role to play in cutting the sheets.

The power of the Epilog laser can reach 40 Watt but only up to 4 Watt is used. The higher power is more suited for metals or big plastic materials. The power of the NEJE laser is only 1.5 Watt. The power difference can be an explanation why the Epilog laser could cut through the material and the NEJE laser could not. Furthermore, the NEJE laser needs to be focused manually with special glasses on, in comparison to the automated focusing of the Epilog laser. This means the NEJE laser can be out of focus and can have a bigger spot size. This also influences the laser, because now the 1.5 Watt is divided over a larger area on the material and the material will not be heated as easily.

The Gly-Alg sheets that are engraved underneath the Epilog laser clearly show that the laser successfully vaporized parts of the sheet. The part that is vaporized by the laser feels rough in comparison to the non-vaporized part and after inspection under the microscope the conclusion can be made that part of the material is successfully vaporized as can be seen by the black lines (Figure 11).[22]

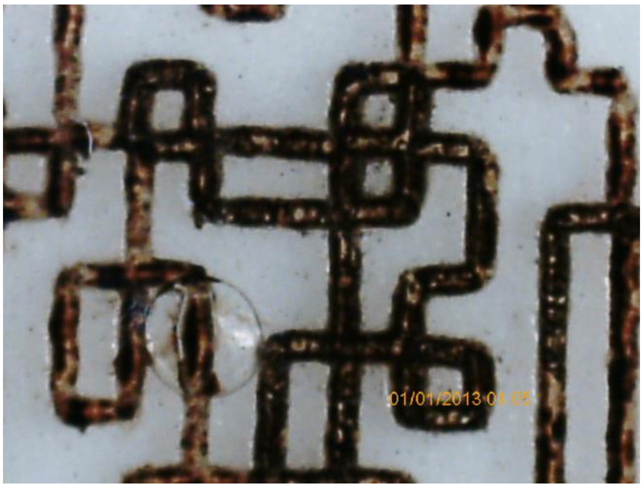

Figure 11: Sheet of 3% w/w Glycerol and 3% w/w alginate after it was engraved by the Epilog laser

5

Conclusions and Recommendations

5.1 Conclusions

This work is aimed to obtain edible barcodes that are cut from biodegradable sheets with a laser cutter. We find that nori paper can be used for cutting even by low-grade cheap lasers like the NEJE DK-BL. Other materials, such as the glycerol-alginate and the rice paper, have a difficult time being cut because of the low power outage of the laser and the reflectivity of the materials causing the wavelengths of the laser to not be fully absorbed.

A different laser , the Epsilon zing laser from the science centre, is used for further research. This laser successfully cut through all the three materials and also successfully engraved the glycerol-alginate sheets. This is verified underneath a microscope

The glycerol-alginate sheets are successfully produced. Several observations are made on first glance from varying the glycerol and alginate. The colour of the sheets changes with the concentration of alginate. The higher the concentration of alginate is, the yellower the sheets get. Another observation made is that by increasing the glycerol concentrations in the sheets, the texture in the sheets also changes. The higher the concentration of glycerol the more soft and stickier the sheets get.

Furthermore, after dissolving two sheets into DI water. The amount of glycerol in the sheets seems to influence the rate in which they dissolve. Because the sheet with the higher amount of glycerol dissolves much faster than the sheet that had no amount of alginate. A possible answer can be that the glycerol homogenates the alginate over the sheet which makes it easier to dissolve. More testing should be done to further support this theory, because now there is a lack of experimental data.

5.2 Recommendations

- More research needs to be done into all the properties of the glycerol-alginate sheets.[1] Following from this the optimal combination of glycerol and alginate can be found. Also an easier method of taking samples from the sheets should be devised for more accurate research results
- Humidity chamber can be used to check, how the nori paper and the glycerol-alginate sheets respond to different relative humidity. This concept can be worked out further to add an sensor part to the barcode.
- The impact of engraving on the sheets can also be something that can be looked into how does the engraving really change the structure of the sheets.

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[1] Gabrielle is for her Master Thesis already looking at this.

Appendix A

The several Gly-Alg sheets made out of different w/w% of alginate and glycerol.

Table 2: Different samples of Gly-Alg sheets

Sample	Glycerol	Water	Alginate
1	0.0%	99.0%	1.0%
2	0.3%	98.7%	1.0%
3	0.5%	98.5%	1.0%
4	1.0%	98.0%	1.0%
5	2.1%	96.9%	1.0%
6	3.0%	96.0%	1.0%
7	0.0%	98.1%	1.9%
8	0.3%	97.8%	1.9%
9	0.6%	97.5%	1.9%
10	1.0%	97.0%	1.9%
11	2.0%	96.1%	1.9%
12	3.2%	94.8%	1.9%
13	0.0%	97.0%	3.0%
14	0.2%	96.7%	3.0%
15	0.7%	96.3%	3.0%
16	1.0%	96.0%	3.0%
17	2.0%	95.0%	3.0%
18	3.1%	93.9%	3.0%
19	0.0%	94.9%	5.1%
20	0.3%	94.7%	5.1%
21	0.7%	94.2%	5.1%
22	1.1%	93.9%	5.1%
23	2.1%	92.8%	5.1%
24	3.7%	91.2%	5.1%
25	0.0%	91.8%	8.2%
26	0.4%	91.4%	8.2%
27	0.7%	91.1%	8.2%
28	1.1%	90.7%	8.2%
29	2.9%	88.8%	8.2%
30	3.8%	88.0%	8.2%

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