



Prediction of Plastic Fragments in Recycled Paper Using Near-Infrared Spectroscopy

Prediktion av plastfragment i återvunnet papper med hjälp av nära infraröd spektroskopi

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Abstract

Sustainability has gained a lot of attention in the field of research. Researchers and consumers both prioritize sustainability and environmental issues over previously dominant materials, such as plastic. Packaging and disposable items that used to be made of plastic have largely been replaced with paper. Unfortunately, paper does not perform as well as plastic regarding barrier properties against grease, oxygen, or water vapor. Barrier properties are an important factor when choosing packaging material for food, among other things, as they help maintain the shelf life of the product. In order to improve the properties of the paper packaging and expand its use, the paper is coated with a polymer. However, the polymer contributes to challenges in the recycling of the products as some of the polymer attaches to the fibers, causing difficulties in the separation of each material. Small fragments of plastic may end up in the material streams and the recycled pulp due to the existing challenges in completely removing plastic from cellulosic substrates during recycling.

This thesis analyzes the possibilities of identifying and classifying plastic fragments of polyethylene (PE) and polyvinyl alcohol (PVOH) in recycled paper sheets using near-infrared spectroscopy together with multivariate data analysis. The purpose of the work is to develop models that can identify possible residues that may appear in recycled products from various industries.

Paper sheets of two different grammages and six different compositions of recycled fiber and virgin fiber were created and scanned by NIR, with and without plastic film under the sheets. The scans were used to develop classification models to identify and categorize scans not included in the calibration data set. The performance of the models was tested by applying them to images of sheets of paper with plastic fragments of different sizes and different type underneath.

The results indicated potential in the method. The prediction of the paper sheets with a lower grammage was mostly correct, whereas the classification of polyethylene showed the best performance. There was some noise in the prediction of the plastic fragments, regardless of the grammage of the paper. The noise may be due to a wide variation in the calibration data set since it consisted of paper sheets of six different compositions. A large part of the noise was incorrectly classified as polyvinyl alcohol, which can be due to differences in the manufacturing process of the plastic films. The conclusion of the thesis is that it is feasible to identify and categorize plastic fragments of polyethylene and polyvinyl alcohol in recycled paper sheets with a certain margin of error. It can be stated that the method shows promise, but further research and development in the field is required to build models that can be applied to a wider range of samples.

Keywords: Near-infrared spectroscopy, paper packaging, barrier materials, recycling, multivariate data analysis

Sammanfattning

Hållbarhet har idag stor uppmärksamhet inom forskningsvärlden. Både forskare och konsumenter prioriterar hållbarhet och miljöfrågor framför material som tidigare dominerat, plast. Förpackningar och engångsartiklar som tidigare brukade vara av plast har i stor utsträckning ersatts med papper. Tyvärr presterar papper inte lika bra som plast när det gäller barriäregenskaper mot exempelvis fett, syre eller fukt. Barriäregenskaper är en viktig faktor vid val av förpackningsmaterial till bland annat livsmedel då det hjälper till att bibehålla produktens livslängd. För att förbättra pappersförpackningens egenskaper och öka dess användningsområden bstryker man pappret med någon form av polymer. Polymeren bidrar dock till svårigheter vid återvinning av produkten då vissa av fibrerna binder till polymeren och orsakar svårigheter vid separationen av materialen. De hinder som finns med den totala avlägsningen av plast från cellulosasubstrat vid återvinning av materialet kan orsaka att små plastfragment följer med i materialströmmarna och den återvunna pappersmassan.

Detta examensarbete analyserar möjligheterna att identifiera och klassificera plastfragment av polyeten (PE) och polyvinyl alkohol (PVOH) i återvunna pappersark med hjälp av nära infraröd spektroskopi tillsammans med multivariat dataanalys. Syftet med arbetet är att utveckla modeller som hjälper till att uppmärksamma rester av material som kan framkomma hos olika aktörers återvunna produkter.

Pappersark med två olika ytvikter och sex olika sammansättningar returfiber kontra nyfiber tillverkades och skannades spektroskopiskt, med och utan plastfilm under arken. Dessa skanningar användes till att utveckla klassificeringsmodeller för att identifiera och kategorisera skanningar utanför kalibreringsdatasetet. Modellernas prestanda kontrollerades genom att applicera dem på skanningar av pappersark med plastfragment, av olika storlek och olika typ, under.

Resultatet tydde på potential hos metoden. Prediktionen hos arken med låg ytvikt var övervägande korrekt, där kategoriseringen av polyeten visade bäst prestanda. Det framkom en del brus hos predikteringen av plastfragmenten, oberoende av papprets ytvikt. Detta misstänks bero på att kalibreringsdatasetet innehöll data med för mycket variation, då pappersarken bestod av sex olika sammansättningar. En stor del av bruset kategoriserades felaktigt som polyvinyl alkohol, vilket kan grundas i att tillverkningen av de olika plastfilmerna skiljde sig åt. Slutsatsen av examensarbetet är att det är möjligt att identifiera och klassificera plastfragment av polyeten och polyvinylalkohol i återvunna pappersark, med viss felmarginal. Det som kan konstateras är att metoden har potential, men det krävs ytterligare forskning och utveckling inom området för att bygga modeller som kan appliceras på ett större urval av prover.

Nyckelord: Nära-infraröd spektroskopi, pappersförpackningar, barriärmaterial, återvinning, multivariat dataanalys

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1 Introduction

In present era, it is hard to avoid the subject of sustainability, and I want to believe that the majority of us aspire towards living more sustainably. Unfortunately, despite their negative environmental effects, we stubbornly refuse to replace some things we rely on because they simplify our lives. Many of these necessities are made of plastic derived from petroleum, a material that is not sustainable in several aspects. Due to its beneficial properties and low cost, plastic is a material widely utilized. One of the several uses for plastic is in consumer packaging and disposable items. In Sweden, many disposable plastic products have been replaced with fiber-based ones, which have a significantly lower environmental impact, as a result of regulations. However, paper is rarely used alone in the packaging industry as the properties of paper make the material difficult to apply in the field. In order to optimize the qualities of paper packaging, plastic (such as polyethylene and polyvinyl alcohol) is usually applied as a barrier material. Although there has been progress in the study of bio-based plastics, there is still more to be done in terms of e.g., cost, availability, and properties. The barrier material used for cellulose-based packaging and disposable items is primarily petroleum-based plastic.

The sustainability of paper as a material is due to the fact that it is biobased, renewable, and easy to recycle, as well as the high yield of the product when recovered. However, as previously stated, fiber-based packaging is composed of both fibers and plastic, which complicates the recyclability of the product. The separation of plastics and fibers in the recycling process can be challenging, allowing plastic fragments from barrier materials to follow the recovered mass. Due to increased awareness of environmental and sustainability challenges, industries and businesses are continually subjected to stricter regulations from the government. It is just a matter of time before plastic particles in recycled paper become strictly prohibited, so developing methods for identifying these plastic fragments in paper is essential.

1.1 Aims and Objectives

The aim of this thesis was to investigate whether it is possible to identify polyethylene (PE) and polyvinyl alcohol (PVOH) under paper sheets of varying composition. The identification is done by the development of different models to predict and classify plastic fragments of different sizes. In this project, near-infrared (NIR) spectroscopy together with multivariate data analysis are utilized. The objective of the project is to identify and reduce barrier fragments in recycled pulp streams. Research questions to be answered by this project are:

- Can near-infrared spectroscopy accurately predict the presence of plastic fragments in recycled paper?
- How do the type and size of plastic fragments in recycled paper affect the near-infrared spectroscopic prediction accuracy?
- How do the composition and thickness of the paper affect the near-infrared spectroscopic prediction of plastic fragments?

2 Theoretical background

2.1 Consumer packaging

The term "consumer packaging" refers to a wide range of packaging. Simply put, it applies to most of the packages that consumers encounter when exploring the aisles of a store. These packages enclose anything from cosmetics and perfume bottles to food (Finlands Förpackningsåtervinning RINKI Ab, 2019). Maintaining the quality of the product and preventing contamination during transit and storage depend heavily on the packaging. Furthermore, it contributes to preserving and extending the shelf life of the contents. The packaging of a product is important for logistical reasons as well. The responsibility of maintaining the product safe and secure during different types of transportation journeys is one of the roles of the packaging. One aspect that many consumers overlook is how effective the packaging is in conveying information about and marketing the product (Oloyede et al, 2021).

Different materials are used to utilize the packaging depending on the contents, with plastic being one of the most common. When selecting a material, a variety of factors are considered, including its permeability properties, cost, and transparency. The packaging industry accounts for approximately 26% of total polymer consumption worldwide, making it the biggest application of plastic materials. This is due to the unique properties that polymer-based materials provide in their field. Plastics have excellent barrier properties and are lightweight, which are both desirable characteristics in most packaging. In addition, it is a low-cost material, which makes it even more appealing (Ibrahim et al, 2022).

2.1.1 Cellulose-based consumer packaging

Aside from plastics, paper is the most common choice of material for packaging. Paper is a low-cost option, similar to plastics; however, unlike plastics, paper is a sustainable one. Unfortunately, when it comes to permeability and strength, paper does not compare favorably to plastics (Ibrahim et al, 2022). Paper and board are sensitive to gas and liquid since the paper fibers that link together to create a network have voids in them. When paper packaging is exposed to moisture and liquid, the material absorbs it resulting in a loss of mechanical and physical properties. To improve the permeability and liquid resistance of the paper packaging, a coating is often added (Tryding & Söremark, 2007). The coating can consist of both aluminum and polymers, whereas polyolefins (PE, PP) and PET are some common types used in the packaging industry (Feldman, 2001). Extrusion coating, dispersion coating, and lamination are methods used for providing a barrier to paper and cardboard (Tryding & Söremark, 2007). Extrusion coating uses thermoplastic polymers as barrier materials since the polymer melts onto the surface of the substrate (Moody & Needles, 2004) while when dispersion coating, a water-soluble barrier material is applied on the substrate, using methods like spraying, dipping or roller coating depending on the properties of the involved materials (Tryding & Söremark, 2007). The latter alternative is said to be more sustainable regarding biodegradability and repulping (Bollström et al, 2013).

There are two categories that divide cellulose-based packaging depending on its purpose: flexible packaging and rigid packaging. The material and end-product differ for the categories; the flexible packaging is made of paper and is used for applications such as paper bags and

sacks. The rigid packaging includes folding cartons and corrugated boxes, that are composed of paper board and corrugated board, respectively (Tryding & Söremark, 2007).

Liquid packaging board (LPB) is a common type of rigid packaging that is made for liquid contents, such as juice and milk boxes. These are generally made of 80% paperboard that has been coated with thin layers of aluminum and plastic, 5% and 15% of the total weight of the packaging, respectively (Khan et al, 2023) (The Alliance for Beverage Cartons and the Environment, 2022).

2.1.1.1 Barrier coating

Barrier coating, a fundamental technique for paper packaging, involves coating a substrate with a layer of material. Depending on the intended use, the coating can be applied to either one or both sides of the substrate. This technique makes it feasible to replace numerous polymer-based packaging and disposables with cellulose-based ones while maintaining their high performance. Barrier coatings are usually made of polymers, which help protect the contents from contaminants like oxygen, moisture, fat, and grease since cellulose-based materials fail to do so on their own (Tryding & Söremark, 2007). Polymers are compounds formed by a long chain of numerous repeating units that make up plastics (Science History Institute , n.d.).

Aluminum can also be used as a barrier coating, especially when the content is sensitive to light and oxygen. Aside from improving the barrier properties, the coating also improves the aesthetics of the product (Tryding & Söremark, 2007).

2.1.1.1.1 Polyethylene (PE)

Polyethylene (also called polythene), a synthetic polymer, is one of the most widely manufactured polymers overall. The plastic comes in various forms, most of which have a simple structure with a chemical formula $(C_2H_4)_n$ (Basmage & Hashmi, 2020). Low-density polyethylene (LDPE) is the most common form of the plastic. LDPE is inexpensive, strong, and has effective barrier properties that protect against moisture, grease, and oil. As the plastic has no odor or taste impact, it is also permitted for contact with food. The plastic can be reshaped when heated, which means that it is a thermoplastic (Enqvist, 2009). High-density polyethylene (HDPE) is, as well as LDPE, widely used in food packaging (Tyagi et al, 2021) and are both the most commonly used polymers when extrusion coating (Tryding & Söremark, 2007). HDPE has better gas barrier properties and is resistant to higher temperatures. In order to optimize their properties, LDPE is most commonly applied inside the packaging while HDPE is applied on the outside of paper packaging (Tyagi et al, 2021).

2.1.1.1.2 Polyvinyl alcohol (PVOH)

Polyvinyl alcohol $((C_2H_4O)_n)$ is also a synthetic polymer, but unlike PE, it loses its barrier and mechanical characteristics when exposed to moisture. Since it is water-soluble, it can result in total breakdown. PVOH works well as an oxygen barrier and is often combined with other packaging materials when used as a coating (Tyagi et al, 2021).

2.1.1.2 Sustainability aspects

The known definition of sustainable development is “development that meets the needs of the present without compromising the ability of future generations to meet their own needs”

(United Nations General Assembly 1987, Emas 2015). To raise awareness of the issues we confront, e.g., climate change and overconsumption, the topic of sustainability is a current topic in both schools, companies, and everyday life.

There is a lot of ongoing research in the packaging industry, trying to achieve good barrier materials without compromising on the sustainability aspect. Yook et al (2020) studied the possibility of applying different types of cellulose nanofibers (CNF) as barrier coatings and evaluated their properties as such. They concluded that it is necessary to further study the subject with a focus on the barrier properties against water vapor and oxygen (Yook et al, 2020). Until scientists come up with packaging solutions that are both efficient and sustainable, we will have to remain with our present methods.

Ideally, we should utilize cellulose-based packaging, without additional fossil-based or limited materials, for all kinds of purposes. However, the lack of barrier properties of paper and board would result in waste and loss of food and other products. From a sustainability point of view, the plastic barrier is required, as long as the materials are managed properly.

Paper packaging is also environmentally beneficial in other ways. Paper and cardboard are lightweight materials; thus their transportation is more sustainable because of their form and weight, which favor stacking (Skogsindustrierna, 2019).

As the primary function of a package is often to protect the content in various ways, the packages are most likely disposed after the content has been consumed. Paper is said to be a sustainable option for packaging materials, not only because the raw material is renewable, but also because of its recyclability, which is relevant only if the waste is managed properly. When discussing waste management, there are four fundamental principles: reduce, reuse, recycle and proper disposal. The first three mentioned are the most environmentally desirable but some waste cannot be put in either of the mentioned boxes, therefore, it is of importance to properly dispose it (Reduce, Reuse and Recycle, n.d.). The energy used to produce paper and cardboard from recycled pulp is 15% lower than when using non-recycled raw materials (Ragn-Sells, n.d.). Thus, we not only reuse our resources but also do so using less energy.

2.1.1.2.1 Microplastics

Microplastics, described as plastic pieces that are less than 5 millimeters in size, are produced intentionally or as a result of the fragmentation of larger plastics, which causes significant harm to the environment. In order to fully understand their sources as well as their sinks, it is crucial to keep track of microplastic quantities in the environment. Yet, the majority of microplastic quantification techniques depend on visual identification, which has a high rate of misidentification due to human errors and mistakes (Prata et al, 2019). What has been established is that microplastics are currently contaminating every ecosystem on Earth. Given the increasing concerns about these particles, there is a rise in studies investigating their impact on organisms and their presence in the environment (Prata et al, 2019).

A variety of methods are used to assess the ability to identify microplastics. Elert et al (2017) investigated four different techniques—Raman microscopy, FTIR microscopy, liquid chromatography, and thermal desorption and extraction (TED-GC-MS)—for the detection of a

known quantity of PE, PP, PET, and PS. In the results, factors like measurement duration, procedure handling, detection limits and sample preparation requirements were compared. The correct method should be selected considering whether the goal is to quantify the amount of plastic particles in the specimen or simply to make an accurate estimation of the sample's contamination with polymers. The combined use of multiple methods should be taken into consideration in order to gather complete data regarding microplastics in environmental samples (Elert et al, 2017).

There are major problems with plastic in the oceans, as there are more than 150 million tons of plastic pieces floating around. Larger plastic pieces gradually disintegrate into smaller ones, which then turn into a growing amount of microplastics. Due to the extremely long degradation time of plastics, even after the production of new plastic has stopped, the amount of microplastic will continue to rise. The knowledge of how individuals and the environment are affected by microplastics is minimal today, meaning that studies are necessary to understand the long-term effects of the particles (Naturvårdsverket, n.d.).

2.1.1.3 Recyclability of paper-based consumer packaging and disposables

Over the years, consumers have become more conscious of the environment and the issues they confront. Recycling as much as possible is a simple approach to living sustainably by encouraging a circular economy and a circular material stream. In 2021, 85% of all the paper packaging that entered the Swedish market was recycled, compared to around 62% of the total amount of packaging (Swedish Environmental Protection Agency, 2022). The most easily recyclable materials used for packaging are paperboard and corrugated board (Tyagi et al, 2021). Eckhart (2021) investigated the recyclability of carton and carton board by analyzing the properties of recycled fibers after increasing cycles. It was concluded that the fibers could be recycled at least 25 times without complications and without predictions of a limiting trend (Eckhart, 2021).

The main applications of recycled fibers are newspaper, tissue, and packaging. Corrugated boards and newspapers are almost entirely made of recycled fibers, while only around 35% of the folding boxes consist of recycled fibers. Folding boxes can be used as food packaging; however, these need to be safe and hygienic, which is hard to guarantee if made of recycled fibers (Eckhart, 2021). The separated aluminum and plastics can be used in applications that are not related to food or medicine (Mäki-Tulokas, 2021).

The process of recycling depends on the type of paper and product. Since cellulose-based packaging and disposables most likely consist of more than one layer of material, it complicates recyclability. Besides plastic and paper, these packages often contain non-renewable substrates such as inks and metals. Therefore, the recycling process includes many steps, one of the most difficult being the separation of the fibers and the barrier coating (Tyagi et al, 2021). Many polymers, e.g., PE, are extrusion coated on the board, which results in a mix of some of the fibers and the polymer (Al-Gharrawi et al, 2021).

For fiber-based packaging, there are typically four steps in the recycling process: repulping, screening, cleaning, and papermaking. Repulping is a process intended to divide paper into its fibers along with additional components such as fillers, inks, and coatings. The removal of

impurities and contaminants from the fibers is done by screening, which is based on the distinction between fiber and non-fiber components. The screening process includes coarse and fine screening that differ mainly by the size of the screening holes, which in turn affects the particle size of the contaminants separated. The fiber slurry can then be put through hydrocyclones to separate fibers from contaminants by using density as a tool. The resultant pulp is then combined with additives and used to manufacture recycled paper (4evergreen, 2022).

To determine the recyclability of materials and products derived primarily from paper and board, a process at laboratory scale is done to simulate the industrial stages in conventional paper and board recycling plants. In this process, different parameters are measured to easily assess how recyclable a product is, one of which is the level of disruption materials' fragmentation which is evaluated by the quantity of flake content through fine screening after repulping and coarse screening, according to TAPPI T275 sp18 (Cepi European Paper, 2020). There is a current issue with plastic rejects at large volume recycling papermills as a result of the recycling of fiber-based packaging with multi-material constructions. Additionally, valuable cellulose fibers that are attached to the plastic are being carried along, resulting in a reduced fiber yield (Kay et al, 2021). The complete separation of plastic from paper during recycling is a hot topic in both academic and industrial research to prevent the plastic from ending up in the material flow (Tyagi et al, 2021). One of the studies related to the subject is done by Al-Gharrawi et al (2021) who added cellulose nanofibers (CNF) as a layer between the PE and the fibers, partly to analyze if the properties of the packaging improved but also to investigate how the recyclability of the fibers changed. The addition of NFC improved the fiber recovery by reducing the number of fibers integrated into the PE-film (Al-Gharrawi et al, 2021).

2.2 Near Infrared (NIR) Hyperspectral imaging

The NIR spectroscopic method uses the near-infrared spectrum, which includes wavelengths between 780 and 2500nm. The method is divided into absorption spectroscopy since it measures how much near-infrared radiation a specific compound or solution absorbs. In this way, different chemical compositions can be determined. What separates NIR spectroscopy from other spectroscopical methods is that it has an impact on the vibrational motion of the molecules rather than exciting the electrons within the atoms. The vibrational motion is specific depending on chemical composition, which is useful when determining each compound (tec5USA, 2022). At the NIR region, the chemical bonds that can be analyzed are C-H, O-H, S-H and N-H (Pasquini, 2003).

The spectra received by NIRS contain broad absorption bands that provide a wide abundance of information, which can make the analysis of the samples challenging (Pasquini, 2003) (de Oliveira et al, 2018). However, the full potential of the technique for identification, discrimination, classification, and quantification applications has been achieved due to the employment of multivariable techniques for data processing (Araujo-Andrade et al, 2021).

As seen in *figure 1*, the NIR instrument at RISE consists of a camera detector that is connected to the spectrograph and has a changeable objective lens attached to it. The samples, moving forward on a conveyor belt, are placed on a tray and exposed to halogen light by two ramps

while the camera gathers and records the diffuse reflectance. The scanner includes a changeable objective lens, meaning that the NIR camera might be modified to scan at varying resolutions, depending on the application. If working with very small objects, a micro-lens allows scanning of samples at a high spatial resolution (30 μ m); however, the field of view will be narrow. The changeable objective lens results in different fields of view (Grahm & Yassin, 2022), whereas for this project, the lens with a maximum spatial resolution capability of 0,48 mm was used, resulting in a field of view of 150 mm.

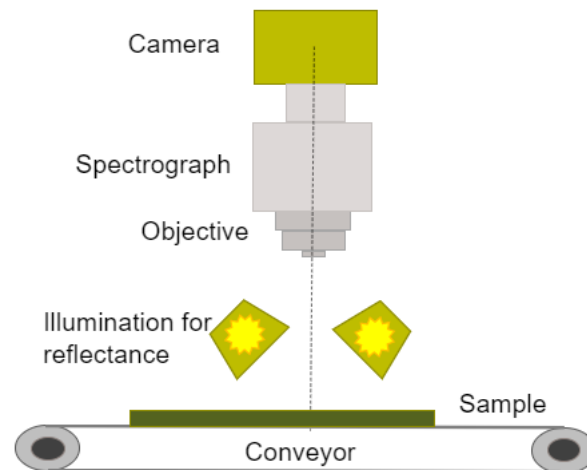


Figure 1: NIR-scanner (Grahm & Yassin, 2022).

The use of NIR spectroscopy is growing in several industrial applications, paper and pulp amongst all. Studies including NIR spectroscopy are not only in the field of paper and pulp but also in applications regarding food, pharmaceutical and agriculture (Araujo-Andrade et al, 2021). Since the technology provides information-rich spectra, together with multivariate data analysis, it has been used to study the quantity of various compounds in different systems in an easy, fast, and environmentally friendly way (Marquez et al, 2023). The possibility to install the NIR spectrometer on-line in the industry, makes the technology an efficient way to apply in processes (Brink et al, 2010).

Brink et al (2010) used NIR to predict the amount of aspen content in unbleached pulp via an on-line NIR spectrometer prototype (Brink et al, 2010). In a different study, Marquez et al (2023) developed a method using multivariate analysis to determine the pentosan contents of bleached and unbleached wood pulp by using NIR spectroscopy. Measuring the quantity of pentosan in cellulose pulp is important in the paper industry since it indicates the amount of hemicellulose remaining in the pulp (Marquez et al, 2023).

The analysis of plastics using NIR spectroscopy has evolved significantly and is utilized to detect many common polymers such as PET, PE and PP. Due to the possibilities in the field, the technology is used for sorting plastics in order to recycle correctly, which is widely employed for scientific, industrial, and governmental objectives (Araujo-Andrade et al, 2021). The technology is used commercially in the recycling industry due to its efficiency in terms of

time and range of plastics. Additionally, because it is a non-destructive technique, the plastic can be recycled following the analysis. The sorting process involves identifying the absorption bands of various polymers and separating them based on that information. By measuring the absorption of infrared radiation at various wavelengths and then comparing the results to a standard, the purity is ascertained (Solid Scanner, 2022).

Given that NIR spectroscopy has been successfully applied in the paper and plastics fields, respectively, there should be promise in combining the two industries and applying the procedure to identify plastic fragments in paper and boards.

3 Method

3.1 Sample preparation

3.1.1 Paper sheets

Four batches containing pulp were prepared. The fibers used in the preparation were Imperial Anchor® (bleached softwood kraft paper grade pulp) from Iggesund Bruk and recycled fibers from DS Smith. The dry matter content of the fibers was approximately 90%. There were two batches for each fiber, each containing 33,3g of fiber. The fibers were soaked, as seen in *figure 2*, for four hours, according to ISO 5263-1:2004.



Figure 2: Four batches of fibers during the soaking process, SW stands for softwood and RF stands for recycled fibers.

After the soaking process, the fibers were separated and diluted with 2 L of water to get disintegrated at 30 000 revolutions for 10 minutes, according to ISO 5263-1:2004. The pulp was diluted up to approximately 20 L of water.

The concentration was determined by letting the fibers filter and dry according to ISO 4119. The filters were weighed before and after the filtration. The batches were stored in a refrigerator overnight.

Twelve different paper sheets were made in the sheet form (*figure 3*) based on *table I*, according to ISO 5269-1. Each sheet was created into triplicates, resulting in a total of 36 sheets.

Table I: Recipe for the making of paper sheets where SW stands for softwood and RF stands for recycled fibers.

| Sheet | Grammage (g/m ²) | SW | | | RF | | |
|-------|------------------------------|------------|--------------------------|---------------------|------------|--------------------------|---------------------|
| | | Dosage (%) | Pulp concentration (g/L) | Required volume (g) | Dosage (%) | Pulp concentration (g/L) | Required volume (g) |
| 1 | 60 | 100 | 3,37 | 452 | 0 | 3,23 | 0 |
| 2 | 60 | 80 | 3,37 | 361 | 20 | 3,23 | 110 |
| 3 | 60 | 60 | 3,37 | 271 | 40 | 3,23 | 220 |
| 4 | 60 | 40 | 3,37 | 181 | 60 | 3,23 | 330 |
| 5 | 60 | 20 | 3,37 | 90 | 80 | 3,23 | 439 |
| 6 | 60 | 0 | 3,37 | 0 | 100 | 3,23 | 549 |
| 7 | 30 | 100 | 3,37 | 226 | 0 | 3,23 | 0 |

| | | | | | | | |
|----|----|----|------|-----|-----|------|-----|
| 8 | 30 | 80 | 3,37 | 181 | 20 | 3,23 | 55 |
| 9 | 30 | 60 | 3,37 | 136 | 40 | 3,23 | 110 |
| 10 | 30 | 40 | 3,37 | 90 | 60 | 3,23 | 165 |
| 11 | 30 | 20 | 3,37 | 45 | 80 | 3,23 | 220 |
| 12 | 30 | 0 | 3,37 | 0 | 100 | 3,23 | 275 |

After each sheet was made, a couching equipment was used to press the sheet, *figure 3*, for 20 seconds. The sheets were pressed twice in the sheet press with a force of 0,395 MPa, the first press lasting 5 minutes and the second press lasting 2 minutes, according to ISO 5269-1. The sheets were dried overnight.

The dried sheets were all punched to measure $150\text{mm} \times 150\text{mm}$. All the paper sheets were tested for thickness and weight, according to ISO 534-2011. The grammage of the sheets was calculated according to ISO 536-2020.



Figure 3: (1) sheet form, (2) couching equipment, (3) sheet press, (4) sheet drier.

3.1.2 Polymer films

Five commercial PE films were punched to a $150\text{mm} \times 150\text{mm}$ dimension. The weight and thickness of the PE film were noted.

For the PVOH films, a solution of 255,0 g of water and 45,0 g PVOH (Kuraray Poval™ 15-99) was prepared. The solution was stirred under heat (90°C - 95°C) for approximately 4 hours, till the PVOH got dissolved. By drop casting, PVOH films were made in various attempts to achieve the desired results. The attempts consisted of using two different types of petri dishes: a disposable one made of polystyrene and a reusable one made of PTFE. Different amounts of PVOH solution were used to make the films, until the desired thickness was achieved. An attempt to dry the film in the oven under heat (55°C - 105°C) was made resulting in a film with visible differences in texture. The method that gave the desired results was diluting 0,6 g of PVOH solution with water to a total of approximately 15 g. The solution was stirred under heat

(70°C) for approximately 2 minutes (till the solution was homogenous). The solution was cast on a reusable petri dish with a diameter of 12cm and made of PTFE and placed on an even surface to evaporate for approximately 24 hours. The petri dishes were covered with a lid, but a gap was left to allow the solution to evaporate, *figure 4*.



Figure 4: PVOH solution left until dry.

Fragments were cut out of each polymer film, in approximately following sizes: $1,0 \times 1,0 \text{ cm}$, $0,5 \times 0,5 \text{ cm}$, $0,2 \times 0,2 \text{ cm}$.

3.2 NIR Spectroscopy

The samples were placed on the conveyor belt to get their NIR spectra measured by using the UmBio Inspector (UmBio AB), *figure 5*. Each paper sheet was measured with and without each plastic underneath. To reduce disturbances through the edges of the sheets, plastic strips were placed on each edge, as seen in *figure 6*. The sheets were also placed on top of each other to obtain a wide range of data. The fragments were placed under the sheets, whereas the type of plastic and location were noted, and images were taken. After the measurements, all the data were uploaded to the software program for analysis.

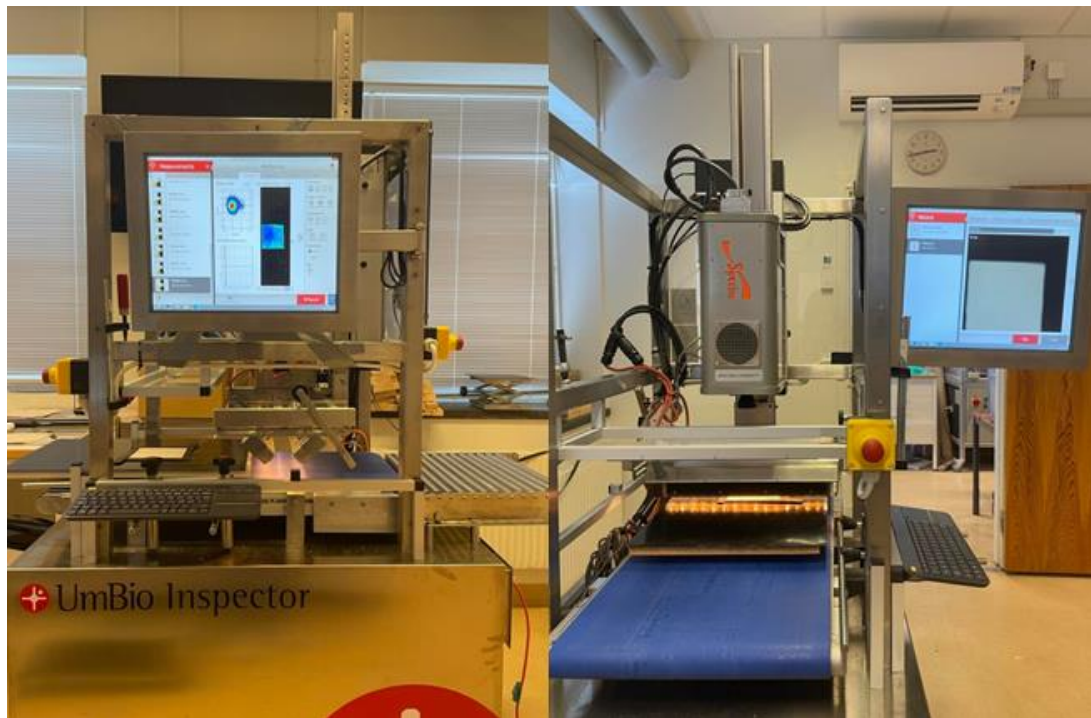


Figure 5: UmBio Inspector.

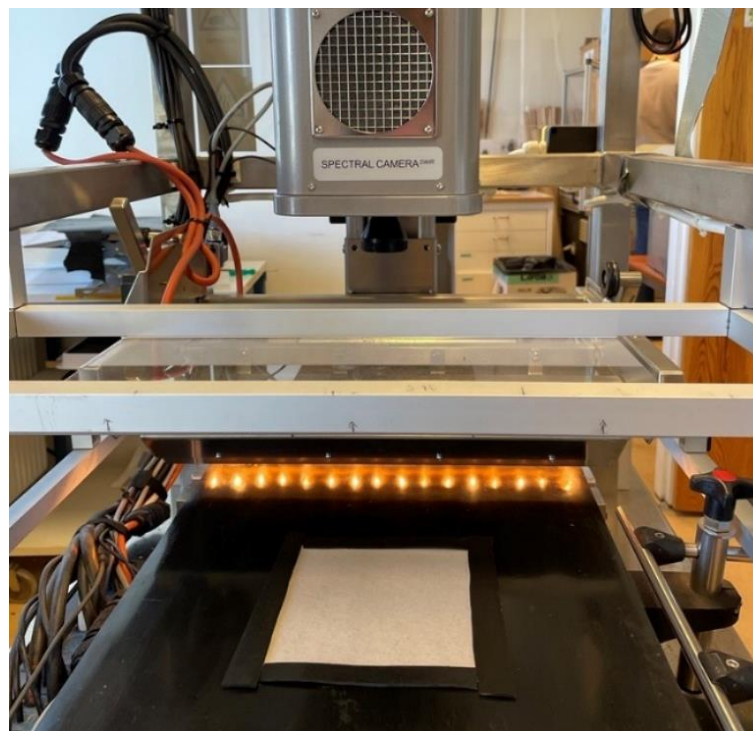


Figure 6: One of the paper sheets during measurement.

3.3 Model building

The software program breeze, by Prediktera was used to capture the images and do the analysis of the scanned samples. Models of PCA (principal component analysis) and PLS-DA (partial least squares discriminant analysis) were built to analyze the data.

Principal component analysis (PCA) is a multivariate data analysis method that helps reduce the dimensionality of data with many variables while still maintaining as much information as possible. The technique is based on converting the original, partially correlated variables to a smaller number of new, uncorrelated variables known as principal components. Principal components are used to observe and interpret correlations that exist among the original variables. By examining similarities between the principal components, you might be able to interpret and illustrate a lot of the original information (Abrahamsson, 2013).

Partial least squares discriminant analysis (PLS-DA) is a variant of partial least squares (PLS) regression that is used to discover the correlation between two matrices (X, Y). When the Y-matrix is a categorical variable, PLS-DA is the technique used (Grahm & Yassin, 2022). By providing a good amount and quality of data, the model is trained to predict each sample/pixel. When the model includes a good number of samples, with an equivalent balance between the samples of each category, the data set is divided into train and test sets. By doing this, the model is trained using some of the train set to see how well it applies to a different set, the test set (Barkved, 2022).



Figure 7: The division of a data set into a train set and a test set.

4 Results and Discussion

4.1 Model calculation

In this study, PCA models were created to eliminate the pixels that belong to the background of the samples and only include relevant information (spectra). PLS-DA models were also created in order to determine if the NIR spectra could distinguish between samples that contain plastic and those that do not.

4.1.1 PLS-DA-models

The first approach was to build two PLS-DA models that were meant to distinguish between the following categories:

- Sample content: non-plastic or plastic
- Type of plastic: PE or PVOH

When building a PLS-DA-model, it is important to include a balance of variables in the data set. With that said, both PLS-DA models included an equal number of samples from each category. Each data set is also divided into test and train sets of a random pick of samples, whereas the test set is 1/3 of the samples and the train set is the rest, which equals 2/3.

4.1.1.1 Paper versus plastic

The PLS-DA model predicting paper versus plastic was done using samples that included paper of different compositions of recycled fibers and softwood fibers with a plastic film underneath and the same paper sheets only.

Grahn & Yassin (2022) developed PLS-DA models to detect wood species and their defects using NIRS. Their approach was to build different models using data from the average spectra and data from representative spectra (of 10 random pixels), respectively. In general, they achieved better results from the models based on the average spectra in comparison with the models based on representative spectra (Grahn & Yassin, 2022). It would be ideal if the same approach were used in this project, however due to time limitations, the models were developed by using data from average spectra of a manually chosen area of each sample since Grahn & Yassin yielded best results from the approach but also to cut down on noise.

4.1.1.1.1 Paper of a variety in thickness versus plastic

Since a variety in thickness ($\sim 60 \mu\text{m}$ – $\sim 360 \mu\text{m}$) of the paper sheets were measured, these were included in the model.

Table II: The statistical parameters for the PLS-DA-model of paper versus plastic.

| | <i>Paper</i> | <i>Plastic</i> | |
|--------------------------|--------------|----------------|-----------|
| <i>Number of samples</i> | 24 | 24 | |
| <i>Type of plastic</i> | | <i>PVOH</i> | <i>PE</i> |
| <i>Number of samples</i> | | 12 | 12 |
| <i>Train set</i> | 16 | 16 | |
| <i>Test set</i> | 8 | 4 | 4 |
| <i>Total</i> | 48 | | |
| | | | |

| | |
|---------|---------|
| R^2 | 0,90831 |
| Q^2 | 0,7673 |
| $RMSEP$ | 0,28499 |

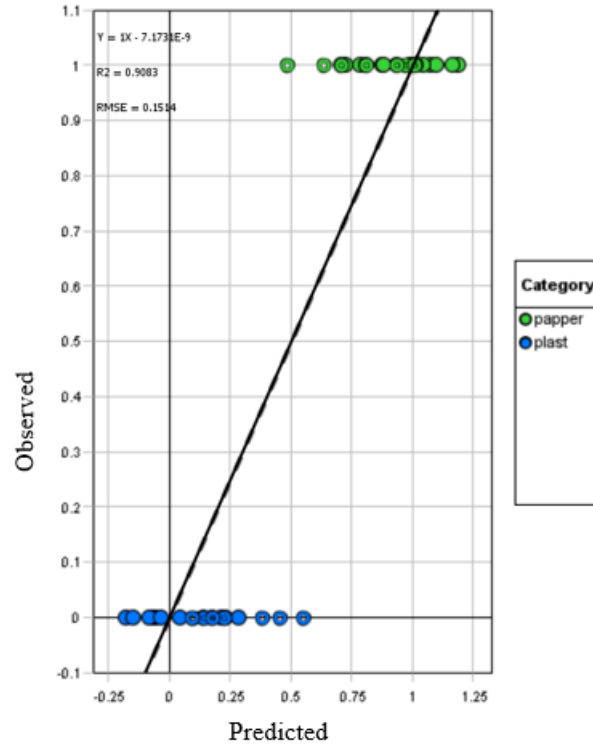


Figure 8: The predicted (x-axis) versus the observed (y-axis) plastic and paper, the points with a hole inside them being test set and the rest being train set.

In *table II*, there are some statistical variables that describe the model. The "goodness to fit", R^2 , explains how well the model reflects the variance within the x-variable. The model's ability to predict data that are not included in the calibration data set is shown by the "goodness to predict", Q^2 . Both variables vary between 0 and 1, with a greater number indicating a stronger performance of the model. $RMSEP$, "root mean square error of prediction", describes the uncertainty of the prediction when the model is applied to samples not included in the calibration set. The $RMSEP$ is desired to be as low as possible (Grahn & Yassin, 2022). The values shown in *table II* indicate that the model built should perform well when applied to new samples. *Figure 8* implies that some of the predicted samples overlap, which is part of the reason for the $RMSEP$ value. When studying a graph of this kind, a good separation between the two different colored points, thus the categories, is the desired outcome. The points with different colors should not overlap on the x-axis.

To include a large amount of thickness in the paper substrate, I had to layer these since I only created paper sheets of two grammages, $\sim 30 \text{ g/m}^2$ and $\sim 60 \text{ g/m}^2$. When layering materials, there is a risk that the light bounces between the layers, causing disturbances in the

measurements. Due to this, I decided to build two similar PLS-DA models to categorize paper versus plastic but only include samples with paper substrates of one layer. I also decided to divide the paper sheets of different thicknesses into two different models to clarify how the thickness of the paper sheets affects the models by comparing them.

4.1.1.1.2 Paper of $\sim 30\text{g/m}^2$ versus plastic

A PLS-DA model was built to classify between paper of $\sim 30\text{g/m}^2$ with a plastic film underneath and paper of 30g/m^2 only. To improve the quality of this model, I included more relevant data, as seen in *table III*:

Table III: The statistical parameters for the PLS-DA-model of paper $\sim 30\text{ g/m}^2$ versus plastic.

| The statistical parameters for the PLS DA model of paper = 50 g/m ² versus plastic | | | |
|---|---------|---------|----|
| | Paper | Plastic | |
| Number of samples | 30 | 30 | |
| Type of plastic | | PVOH | PE |
| Number of samples | | 15 | 15 |
| Train set | 20 | 20 | |
| Test set | 10 | 5 | 5 |
| Total | 60 | | |
| | | | |
| R2 | 0,90359 | | |
| Q2 | 0,88211 | | |
| RMSEP | 0,22613 | | |

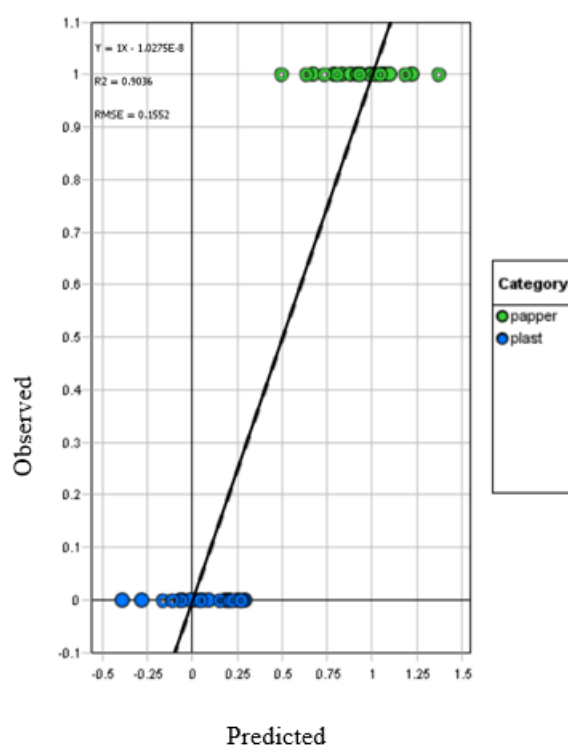


Figure 9: The predicted (x-axis) versus the observed (y-axis) plastic and paper of 30g/m^2 , the points with a hole inside them being test set and the rest being train set.

The statistical parameters shown in *table III*, R^2 and Q^2 , indicate that the model of paper 30g/m² versus plastic has a stronger performance regarding the prediction of samples not included in the calibration set, in comparison with the model of paper versus plastic. The separation between the predicted categories is complete, as shown in *figure 9*, indicating a strong model. This may be because a larger number of samples were included in the second model. The samples included in the better model did not differ from one another in terms of thickness, meaning that all the samples shared this characteristic and therefore contributed to the quality of the model.

4.1.1.1.3 Paper of ~60g/m² versus plastic

A PLS-DA-model similar to the previous one was built, except the paper sheets were of ~60g/m². Comparing the statistical parameters of the models in *tables IV* and *III*, we can see that they are alike, especially the values of R^2 and $RMSEP$. The Q^2 -value of the model made for paper of ~30g/m² versus plastic is slightly greater than for the model for paper of ~60g/m², implying that the model for paper of ~30g/m² is better at predicting samples not included in the calibration set.

Table IV: The statistical parameters for the PLS-DA-model of paper ~60 g/m² versus plastic.

| | <i>Paper</i> | <i>Plastic</i> | |
|--------------------------|--------------|----------------|-----------|
| <i>Number of samples</i> | 30 | 30 | |
| <i>Type of plastic</i> | | <i>PVOH</i> | <i>PE</i> |
| <i>Number of samples</i> | | 15 | 15 |
| <i>Train set</i> | 20 | 20 | |
| <i>Test set</i> | 10 | 5 | 5 |
| <i>Total</i> | 60 | | |
| | | | |
| <i>R2</i> | 0,90537 | | |
| <i>Q2</i> | 0,85577 | | |
| <i>RMSEP</i> | 0,22118 | | |

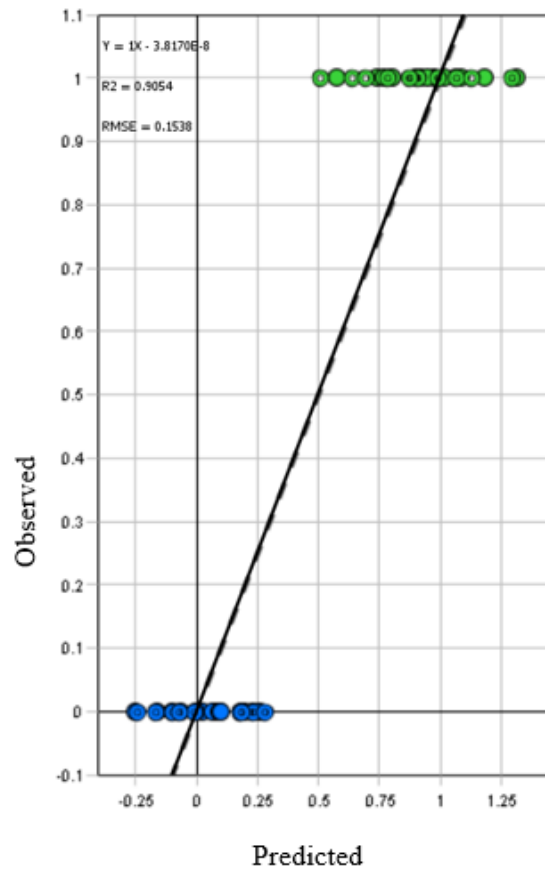


Figure 10: The predicted (x-axis) versus the observed (y-axis) plastic and paper of 60g/m², the points with a hole inside them being test set and the rest being train set.

4.1.1.2 PE versus PVOH

The PLS-DA models for classifying PE or PVOH were developed on the same principle as the latter models for predicting paper or plastic, meaning that the thickness of the paper sheets did not vary in each model. The models consisted of samples of paper with PE-film or PVOH-film underneath and were trained to distinguish between the different types of plastic. As with the previous models, only average spectra from a manually chosen area were included.

4.1.1.2.1 PE versus PVOH under paper of ~30g/m²

To classify and predict plastic fragments under paper sheets of ~30g/m², a PLS-DA model was built containing the average spectra of 36 samples. The statistical parameters, shown in *table V*, imply that the model is very strong and capable of predicting samples not included in the calibration set. A complete separation in the categorization is shown in *figure 11*, which is expected after analyzing the statistical parameters. The points representing PE are slightly more spread out than the points representing PVOH, but none of the points overlap, which is most important for a good classification model.

Table V: Statistical parameters for the PLS-DA-model of PE versus PVOH under paper of $\sim 30\text{g/m}^2$.

| | <i>PE</i> | <i>PVOH</i> |
|--------------------------|-----------|-------------|
| <i>Number of samples</i> | 18 | 18 |
| <i>Train set</i> | 12 | 12 |
| <i>Test set</i> | 6 | 6 |
| <i>Total</i> | 36 | |
| <i>R2</i> | 0,97889 | |
| <i>Q2</i> | 0,97075 | |
| <i>RMSEP</i> | 0,05252 | |

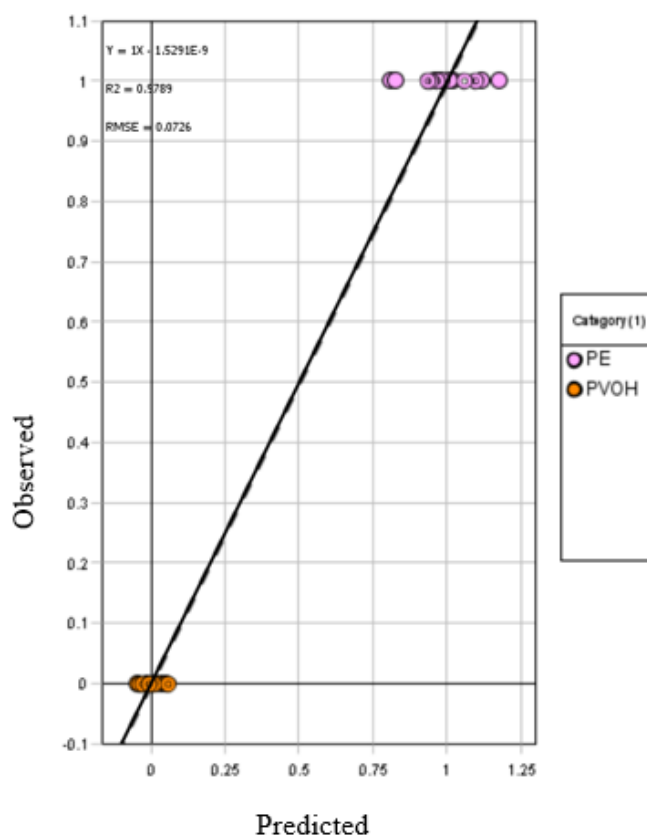


Figure 11: The predicted (x-axis) versus the observed (y-axis) PE and PVOH under paper of 30g/m^2 , the points with a hole inside them being test set and the rest being train set.

4.1.1.2.2 PE versus PVOH under paper of $\sim 60\text{g/m}^2$

A classification model to predict and classify plastics under paper of $\sim 60\text{g/m}^2$ was built containing the average spectra of 36 samples. Just like the previous model, the statistical parameters for this model, shown in table VI, state that the model is strong and has a good ability to predict samples not included in the calibration set. The error of prediction is somewhat

higher for this model, which can be explained by the distribution of the points in *figure 12*. The points do not overlap, but they are more spread out than in the previous model, which is increasing the *RMSEP* value.

Table VI: Statistical parameters for the PLS-DA-model of PE versus PVOH under paper of $\sim 60\text{g/m}^2$.

| | <i>PE</i> | <i>PVOH</i> |
|--------------------------|-----------|-------------|
| <i>Number of samples</i> | 18 | 18 |
| <i>Train set</i> | 12 | 12 |
| <i>Test set</i> | 6 | 6 |
| <i>Total</i> | 36 | |
| | | |
| <i>R2</i> | 0,98051 | |
| <i>Q2</i> | 0,9593 | |
| <i>RMSEP</i> | 0,13017 | |

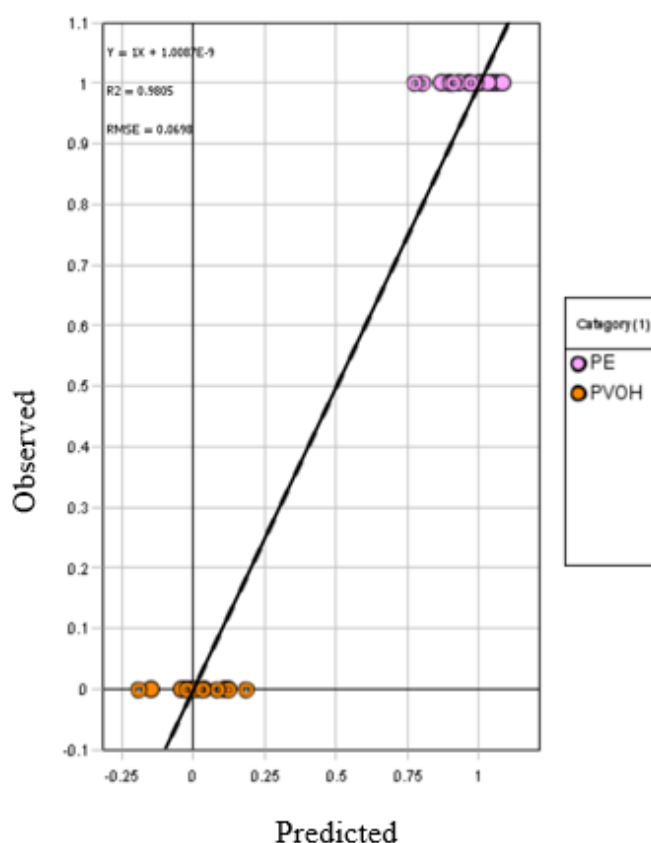


Figure 12: The predicted (x-axis) versus the observed (y-axis) PE and PVOH under paper of 60g/m^2 , the points with a hole inside them being test set and the rest being train set.

4.1.1.3 Prediction of plastic fragments under paper sheets

When the models are developed, it is time to apply them to unknown samples to get a better understanding of their performance. To evaluate the models, we apply them to samples of paper sheets with small plastic fragments underneath.

The application of the “paper versus plastic” models was done by two different strategies. The first strategy was to use the models to predict each pixel within the scanned area. The second strategy was to divide each sample into grids; the majority of the class predicted inside the grid determined the class of that particular grid. As for the classification of PE and PVOH, a different approach was used. The models were only applied to those areas (with a minimum of 50 pixels) that were predicted as plastic in the first classification, i.e., the classification of paper versus plastic. In *figure 13*, the order of the models is shown in a pipeline.

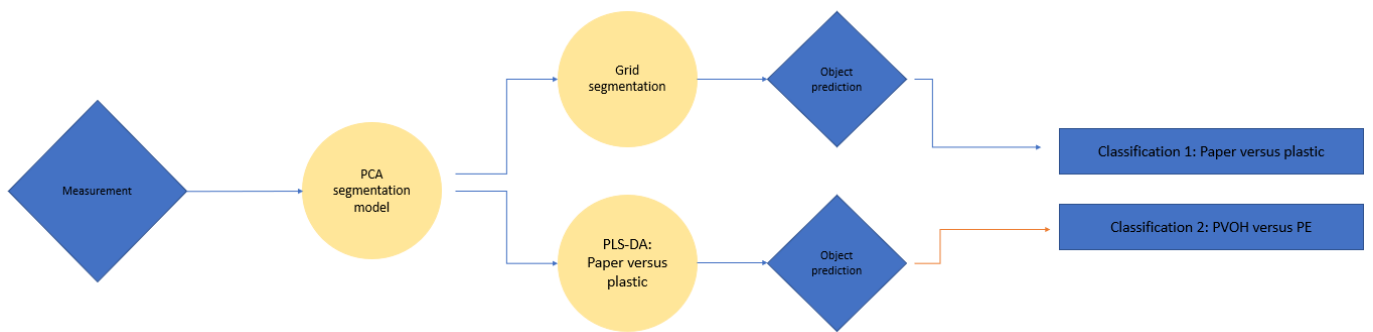


Figure 13: Prediction workflow.

Figure 14 illustrates the predictions on a paper sheet of 30g/m² with the composition of 100% recycled fibers, two PE- and two PVOH- fragments with an approximate size of 1 × 1 cm² were placed under the sheet. The approximate placement of the fragments is within the drawn circle in the pictures, whereas the two upper fragments are of PVOH, and the two lower ones are of PE. In the middle of the paper sheet, there is a plastic stripe, which was placed to make it easier to separate the different types of plastic. To the left, the grid-based prediction is shown, and, in the middle, the pixel-based prediction is shown. To the right, the prediction of PVOH and PE is shown, which was only in the pixels classified as plastic. The grid-based prediction is accurate, but it is a simplification of the pixel-based one, since it reduces a lot of the noise that is shown in the middle picture. The picture to the right is also a correct prediction of the classes; however we can see some noise in the prediction of PVOH. The noise can be caused by a few reasons whereas the *RMSEP* value, corresponding to 0,22613 in the paper versus plastic model, can be one of them. With that said, a certain amount of noise is expected in the predictions.

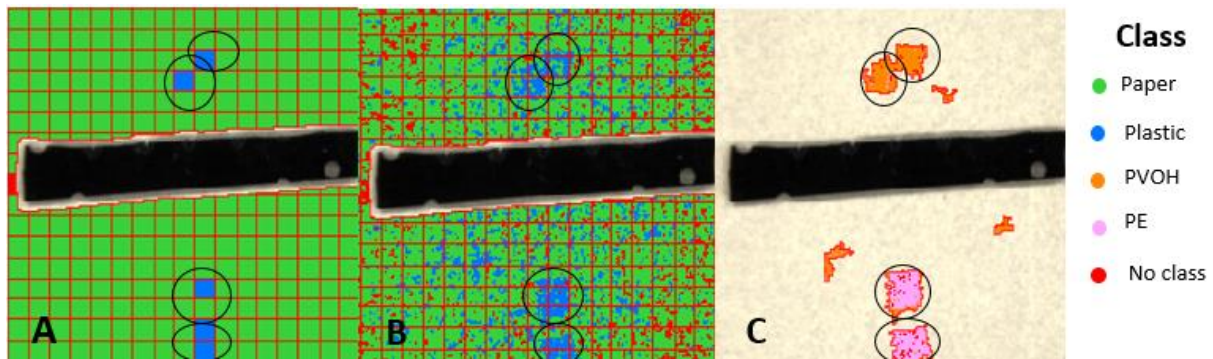


Figure 14: Prediction on paper sheet of 30g/m^2 with the composition of 100% recycled fibers and four plastic fragments of size $1 \times 1 \text{ cm}^2$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic on pixel-level (C) classification of PVOH and PE.

In figure 15, the conditions are similar to the previous prediction, but the size of the fragments is halved, corresponding to an approximate size of $0.5 \times 0.5 \text{ cm}^2$. The prediction of plastic is less accurate, as seen in the pixel-prediction but also in the grid-prediction. The blue pixels, corresponding to pixels of plastic, are less intense, causing the grid-prediction to be less accurate. The solution could be to adjust the grid size, or perhaps to modify the prediction cut off, which is the number of blue pixels necessary for the grid to be predicted as plastic. There is still a lot of noise scattered around the actual plastic fragments. In the classification of PVOH and PE, there is an accurate prediction, however some of the noise in the pixel-prediction is classified as PVOH. It is important to keep in mind that the noise seen in image C is caused by the PLS-DA-model that discriminates between paper and plastic, giving the PLS-DA model that discriminates between PVOH and PE an inaccurate representation.

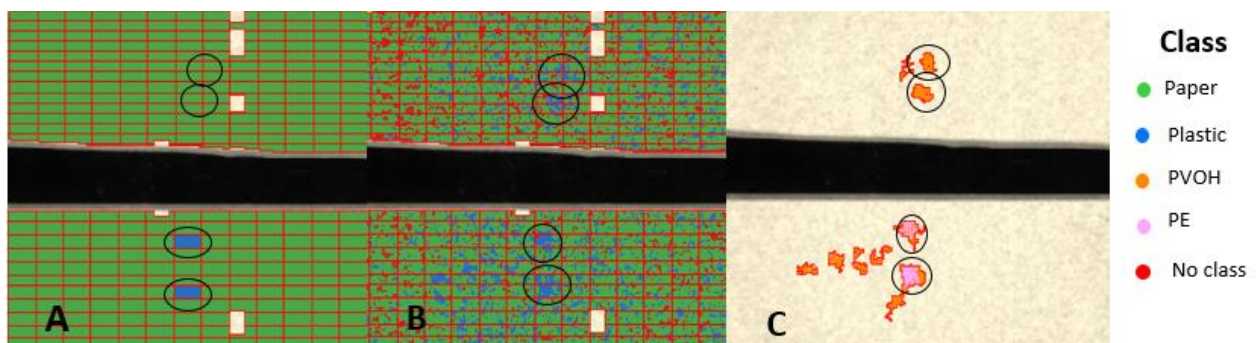


Figure 15: Prediction on paper sheet of 30g/m^2 with the composition of 100% recycled fibers and four plastic fragments of size $0.5 \times 0.5 \text{ cm}^2$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic on pixel-level (C) classification of PVOH and PE.

In this project, three different sizes of plastic fragments were evaluated. In figure 16, four plastic fragments with an approximate size of $0.2 \times 0.2 \text{ cm}^2$ were placed under the paper sheet. Apart from the size of the fragments, the conditions are the same as in the previous images, in figures 14 and 15. In the images shown in figure 16, we can state that the prediction is worse than the

prediction of the larger plastic fragments. The pixel-prediction contains a lot of noise and inaccurate classification causing inaccuracies in the grid-prediction as well. As for the classification between PVOH and PE, the model is capable of distinguishing between PVOH and PE, as seen to the right in *figure 16*, however a lot of noise is predicted as PVOH. It is difficult to determine what the large amount of noise could be due to; my suspicions are that the models get confused when different compositions are involved in the data set.

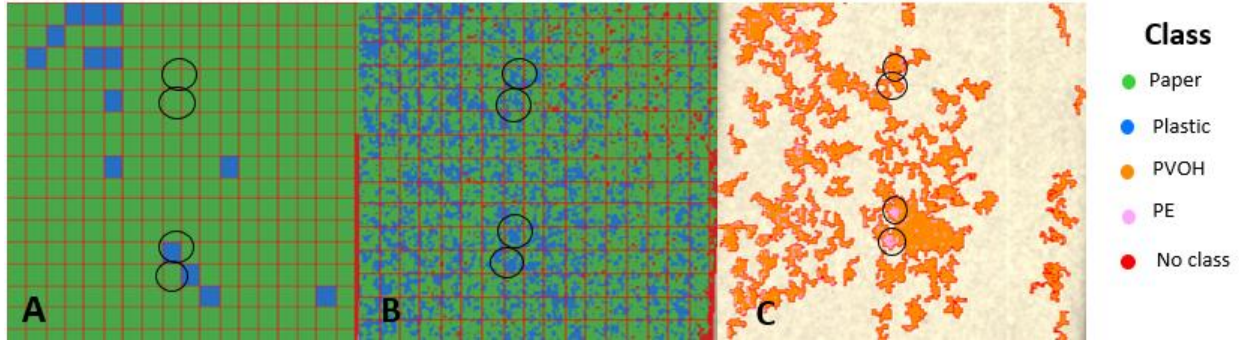


Figure 16: Prediction on paper sheet of 30g/m² with the composition of 100% recycled fibers and four plastic fragments of size 0.2x0.2cm² under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic on pixel-level (C) classification of PVOH and PE.

Figure 17 illustrates the prediction of four plastic fragments of size 0.5x0.5cm² under a paper sheet of 30g/m² with the composition of 80% recycled fibers and 20% softwood fibers. Analyzing the pixel prediction, we can see that the plastic fragments are there, especially the PE-fragments, but as well as for previous predictions, noise is involved in the measurements. Even though the pixel predictions are somewhat correct, it is not enough to predict correctly at grid-level. As for the classification of PVOH and PE, it was partially correct. Only one of the PVOH fragments is accurately predicted, and some noise is predicted as PVOH, whereas both PE fragments are correctly predicted.

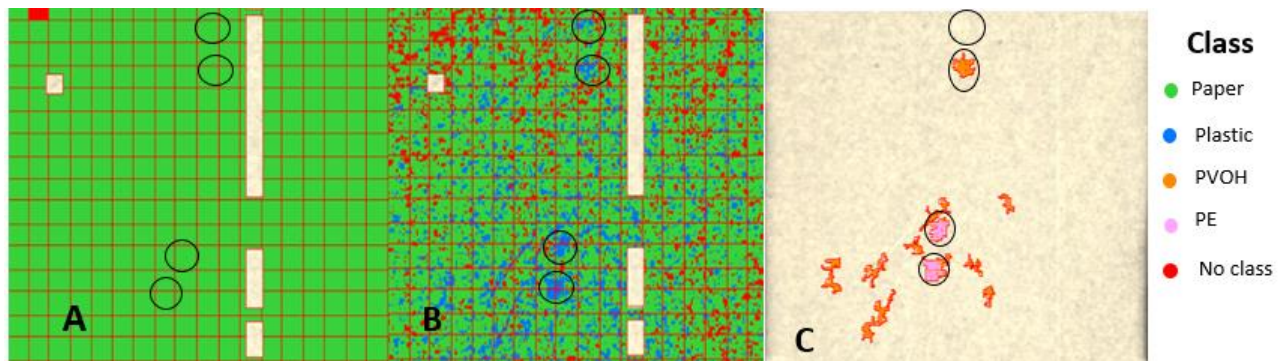


Figure 17: Prediction on paper sheet of 30g/m² with the composition of 80% recycled fibers and 20% softwood fibers and four plastic fragments of size 0.5x0.5cm² under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic on pixel-level (C) classification of PVOH and PE.

In *figure 18*, the conditions are the same as in *figure 17* apart from the plastic fragment size which is $0.2 \times 0.2 \text{ cm}^2$. The prediction is similar to the previous figure; however, the plastic fragments are less accurately predicted. In the pixel-prediction, the PE-fragments contribute to some blue pixels while the PVOH-fragments only leave traces. Despite this, none of the plastic fragments are predicted enough to be shown on grid-level. However, the PE-fragments are correctly classified in image (C), while none of the pixels are correctly predicted as PVOH.

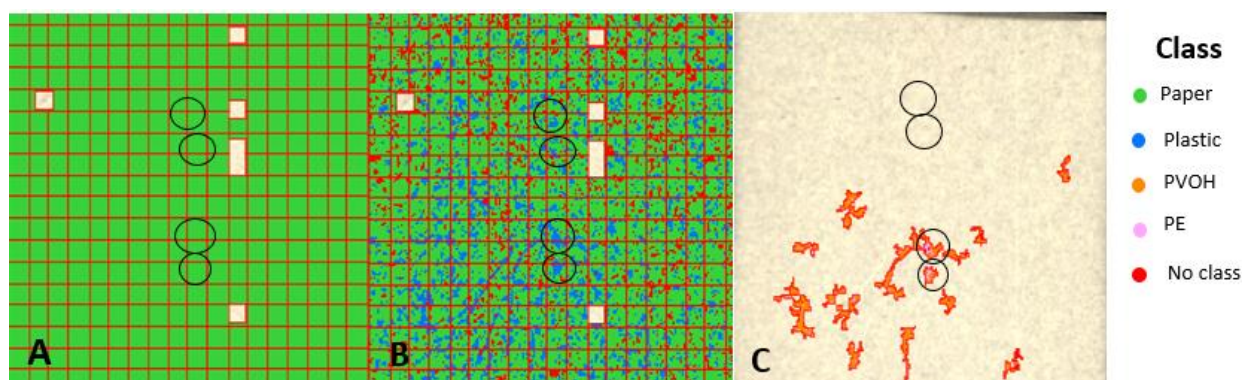


Figure 18: Prediction on paper sheet of 30 g/m^2 with the composition of 80% recycled fibers and 20% softwood fibers and four plastic fragments of size $0.2 \times 0.2 \text{ cm}^2$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE.

In previous figures, the plastic fragments were predicted, in different amounts. With that said, near-infrared spectroscopy is able to predict the presence of plastic fragments in paper, however, the prediction is not flawless. The size of the plastic fragments plays an important role in the prediction since the noise increases with decreasing fragment size, making the prediction less accurate. The previous statement is reasonable since the measurements were made with a spatial resolution of 0.48 mm . With that said, the results given from the images containing plastic fragments of size $0.2 \times 0.2 \text{ cm}^2$ are better than expected. As for the kind of plastic, the classification of PE is significantly more accurate and the largest part of the noise in the measurements is predicted as PVOH. This does not only apply to the classification between PVOH and PE (image C in the figures) but also to the classification of paper and plastic. Comparing the PVOH-fragments to the PE-fragments, which contribute to numerous blue pixels and thus blue grids, the PVOH-fragments are less intensely predicted as plastic. As NIRS reacts differently depending on molecular vibrations, the composition of the plastics plays a big part in the classification. Both polymers have C-H bonds, but PVOH has an O-H bond as well, which differs between the polymers (Basmage & Hashmi, 2020) (Tyagi, Salem, Hubbe, & Pal, 2021) and contributes to the classification of the models.

Zheng et al. (2017) used NIRS together with a Fisher discriminant model to classify different unknown plastics. The predictive ability of the model for classifying unknown plastics was high. The plastics analyzed in the project were PS, PP, ABS, PE, PET, and PVC (Zheng et al, 2017). Similar to this project, Zheng et al. classified PE among all and had a good outcome. What differentiates the project from this one is the construction of a different type of model as well as the fact that the plastics were predicted by themselves, without any paper involved. The

prediction of PE is successful in both projects; however, it would be interesting to try different methods, such as a Fisher discriminant model, for the classification of PVOH fragments since the models built in this project occasionally had a hard time discriminating them.

When doing the measurements, the PE-film used was commercial, unlike the PVOH-film, which was man-made and not completely even throughout the surface. This may have an impact on the inaccurate prediction of PVOH and further research should be done with a commercial PVOH-film to rule out this source of error. As for the composition of the paper sheets, it is hard to tell if it has an impact. The predictions on paper of 100% recycled fibers included every plastic fragment of both types while the predictions on paper of 80% recycled fibers and 20% softwood fibers failed to classify the PVOH-fragments. However the latter prediction included less noise which is also valuable. It is impossible to say for sure if the composition was responsible for the noise reduction. What can be ascertained is that all images contain noise, which I suspect is due to the fact that the models built are confused when predicting, because of the large amount of data of paper with varying composition in the the data set.

In *figure 19*, the models are applied on a paper sheet of 60g/m^2 with a composition of 100% recycled fibers, and four plastic fragments of size 1cm^2 are placed under the paper sheet. In each image, a lot of noise is present and it is difficult to estimate the placement of the plastic fragments. Comparing the images with previous predictions on paper of 30g/m^2 , a significantly higher number of pixels are unidentified. Even though the models have high values of R^2 and Q^2 , the prediction is incorrect.

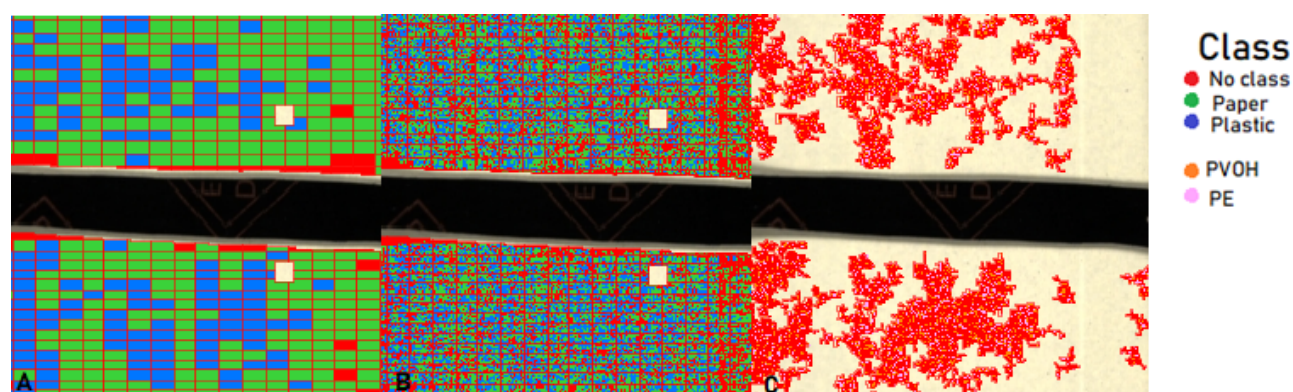


Figure 19: Prediction on paper sheet of 60g/m^2 with the composition of 100% recycled fibers and four plastic fragments of size 1cm^2 under. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE.

In order to draw conclusions, we need to analyze more samples and make comparisons between them. *Figure 20* illustrates predictions on paper sheets under same conditions as the previous figure, however the composition is of 80% recycled fibers and 20% softwood fibers. As well as previous predictions, these images contain a lot of noise. The grid-prediction is less noisy due to less noise in the pixel-prediction, however, the placement of the plastic fragments is still hard to estimate. In the pixel-prediction as well as the classification of PVOH and PE, unidentified pixels appear.

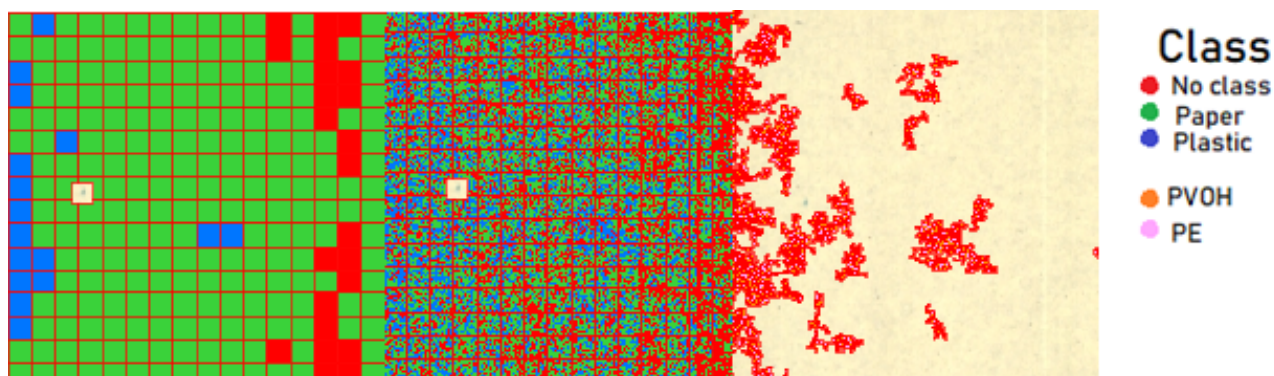


Figure 20: Prediction on paper sheet of 60g/m² with the composition of 80% recycled fibers and 20% softwood fibers and four plastic fragments of size 1x1cm² under. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE.

The application of the models built for classifying plastic fragments under recycled paper of 60g/m² is difficult. Besides the lack of prediction of the plastic fragments, a lot of noise and unidentified pixels appear in the images. Because of this, the models need to be worked on by including more data and also experimenting with the pipe line. Due to a lack of time, I did not get the opportunity to continue optimizing my models. I suggest that future research build models using a larger data set and more samples that are similar. I would include paper sheets with one sort of composition only since I believe that, at times, the model was confused and therefore responded with a significant amount of unidentified pixels. A different approach would be to add classifications for each composition of paper and not include all compositions in one classification, as done in this project. However, one should expect some unidentified pixels as this may be a result of the *RMSEP* value, the recurring unidentified pixels may be defective ones.

The classifications of PVOH and PE differ in quality. As for the paper sheets of 30g/m², the fragments of PE were predicted accurately with a negligible amount of noise while the fragments of PVOH were difficult to predict, especially when the smaller fragments were put under the paper sheet. This may be due to the fact that fewer rays reach the substance since the correlation between paper and plastic decreases when studying smaller fragments. The majority of the noise that appeared in the images was predicted as PVOH. The PLS-DA-model of PE versus PVOH for paper of 30g/m² showed good properties and the *RMSEP* value of 0.05252 does not indicate that a large amount of noise will occur, however, this is a good example of the need for applying the models to get a good understanding of their performance. To get better predictions of PVOH, the model needs to be optimized by including more data and only building models with paper sheets of one composition to minimize the risk of confusion. The plastic films used in the models should be manufactured by the same principle, either both being commercially made or both being man-made, to obtain predictions that can be fairly compared. An interesting aspect for future research would be to analyze different thicknesses of the plastic films, to gain a better understanding of the prediction and its relation to the thickness of the plastic film.

These models are made to be used in industrial applications in order to reduce the amount of plastic fragments that appear in paper recycling streams. In reality, the fragments may be randomly placed throughout different depths of the paper, which makes the models not fully applicable in the industry. To increase the applicability of the method, samples that are similar to those in reality should be added to the data set by including the plastic fragments in the making of the paper sheets. In this way, the plastic fragments would be spread out in different depths of the paper, however, it would be more difficult to control the placement of the plastics as well as the type of plastic.

5 Conclusion

Four different PLS-DA models were devised in order to identify barrier fragments in recycled pulp streams. The method aimed to spectroscopically distinguish between paper (of different grammage and thickness) and plastic, as well as between polyvinyl alcohol and polyethylene.

- Although the statistical parameters obtained from each model showed good qualities, the performance of the models was not flawless. The models made for paper sheets of a grammage 30g/m^2 could predict the presence of plastic fragments in recycled paper of 30g/m^2 , with a margin of error.
- The type and size of plastic fragments have an impact on the prediction accuracy as the smaller fragments caused a greater amount of noise but also less accuracy in the prediction. The polyethylene-fragments were predicted and classified correctly, even when analyzing samples with small fragments while the prediction and classification of the polyvinyl alcohol-fragments had more errors.
- The prediction differed greatly when using thicker sheets as it became worse with a significantly higher amount of noise. As for the impact of the composition, it's hard to draw any conclusions since the prediction included noise caused by an unknown source. It can be concluded that there is a possibility that the various paper sheet compositions affected the appearance of noise by confusing the model, thus a more detailed model for the paper substrate would be recommended for an industrial application.

The method developed in this project has potential, but further research and development in the field are required to build models that can be applied to a larger range of samples. Future research ideas include developing individual models for the different paper compositions and improving the models by expanding the data set. The result may be used to understand how the composition of the paper sheets affects the prediction of the plastic fragments. Additionally, the production of the plastic films should be similar to rule out the possibility that the manufacturing of the plastic fragments has an impact on the classification of the different types of plastic. An interesting aspect of this project would be to try changing the plastic types to investigate if the results obtained would be better. Another interesting further development of the project would be to include the plastic fragments in the pulp when creating the sheets, making them more similar to industrial cases.

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Appendix I

Table VII: Mean values of the properties of the paper sheets and the plastic films

| Sheet | Area (m ²) | Thickness (mm) | Mass (g) | Grammage (g/m ²) | Density (g/μm ³) |
|------------------|---------------------------|-------------------|-------------|---------------------------------|---------------------------------|
| 1 | 0,0225 | 116,96 | 1,43 | 63,47 | 0,542 |
| 2 | 0,0225 | 118,35 | 1,45 | 64,49 | 0,544 |
| 3 | 0,0225 | 117,60 | 1,43 | 63,60 | 0,540 |
| 4 | 0,0225 | 117,95 | 1,45 | 64,28 | 0,544 |
| 5 | 0,0225 | 118,09 | 1,43 | 63,64 | 0,538 |
| 6 | 0,0225 | 120,15 | 1,46 | 64,81 | 0,539 |
| 7 | 0,0225 | 62,87 | 0,71 | 31,33 | 0,498 |
| 8 | 0,0225 | 64,27 | 0,70 | 31,02 | 0,482 |
| 9 | 0,0225 | 64,35 | 0,68 | 30,19 | 0,469 |
| 10 | 0,0225 | 67,71 | 0,69 | 30,61 | 0,452 |
| 11 | 0,0225 | 68,89 | 0,68 | 30,25 | 0,439 |
| 12 | 0,0225 | 67,87 | 0,70 | 31,14 | 0,458 |
| PE-film | 0,0225 | 13,12 | 0,255 | 11,33 | 0,864 |
| PVOH-film | ~0,0144 | 11,56 | - | - | - |

Appendix II

Predictions on paper sheets of 30g/m^2 .

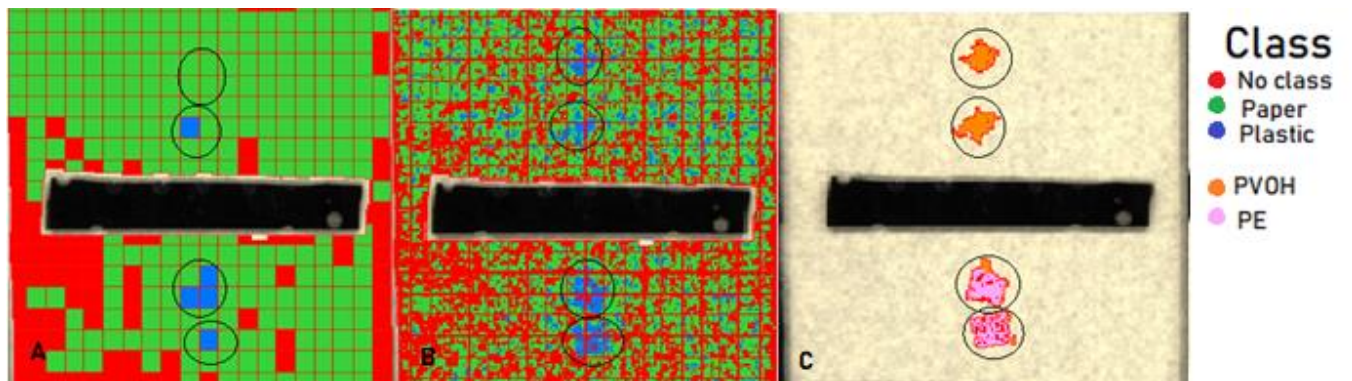


Figure 21: Prediction on paper sheet of 30g/m^2 with the composition of 100% softwood fibers and four plastic fragments of size $1\text{cm} \times 1\text{cm}$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE

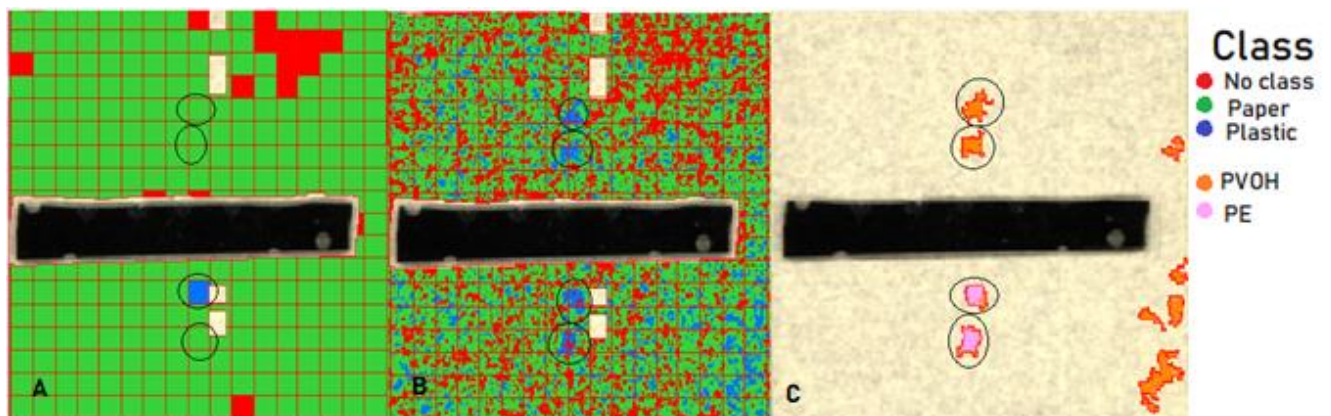


Figure 22: Prediction on paper sheet of 30g/m^2 with the composition of 100% softwood fibers and four plastic fragments of size $0.5\text{cm} \times 0.5\text{cm}$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE

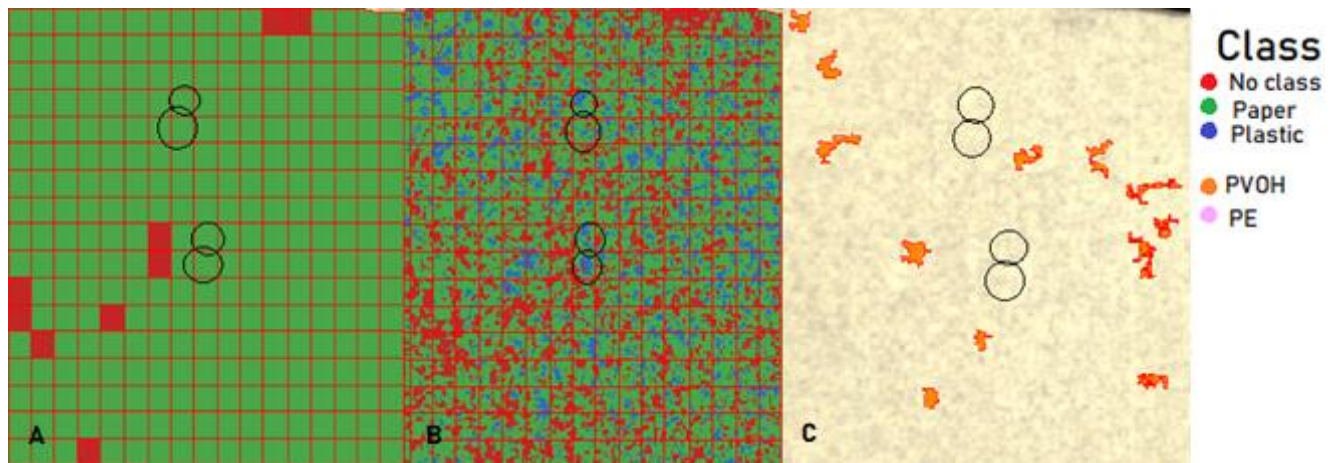


Figure 23: Prediction on paper sheet of 30g/m^2 with the composition of 100% softwood fibers and four plastic fragments of size $0.2 \times 0.2\text{cm}^2$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE

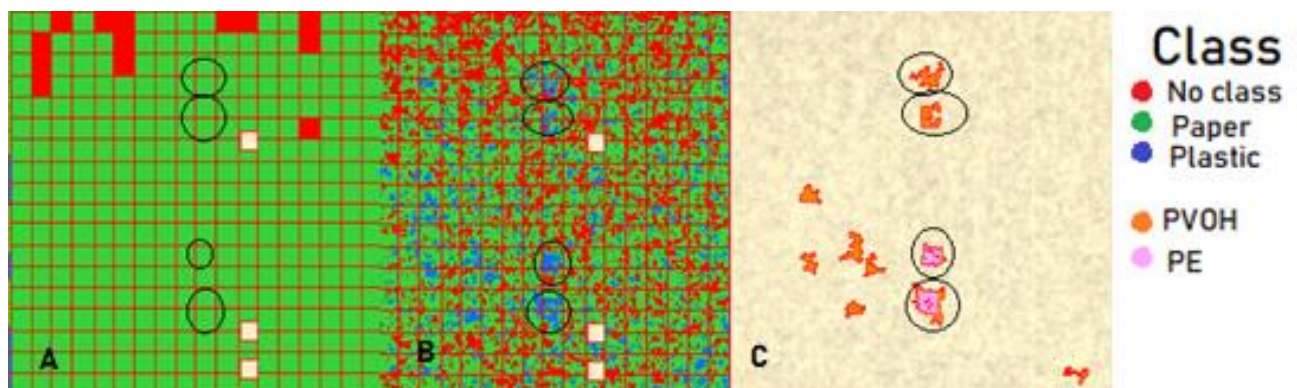


Figure 24: Prediction on paper sheet of 30g/m^2 with the composition of 20% recycled fibers and 80% softwood fibers and four plastic fragments of size $0.5 \times 0.5\text{cm}^2$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE

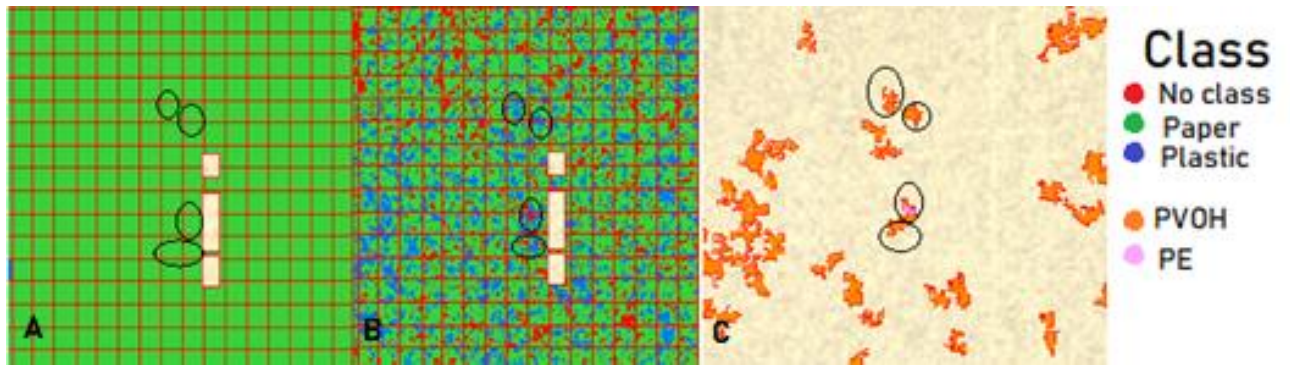


Figure 25: Prediction on paper sheet of 30g/m^2 with the composition of 20% recycled fibers and 80% softwood fibers and four plastic fragments of size $0.2 \times 0.2\text{cm}^2$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE

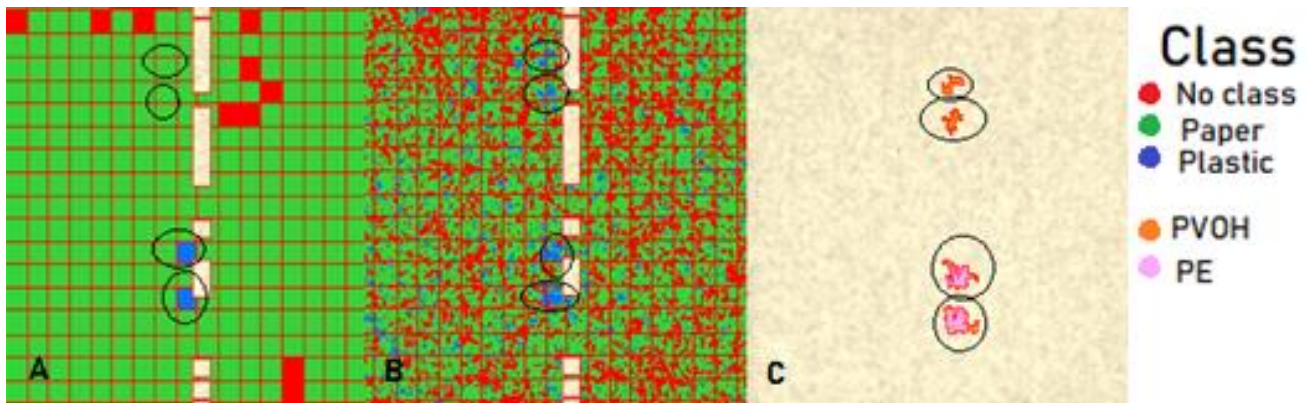


Figure 26: Prediction on paper sheet of 30g/m^2 with the composition of 40% recycled fibers and 60% softwood fibers and four plastic fragments of size $0.5 \times 0.5\text{cm}^2$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE

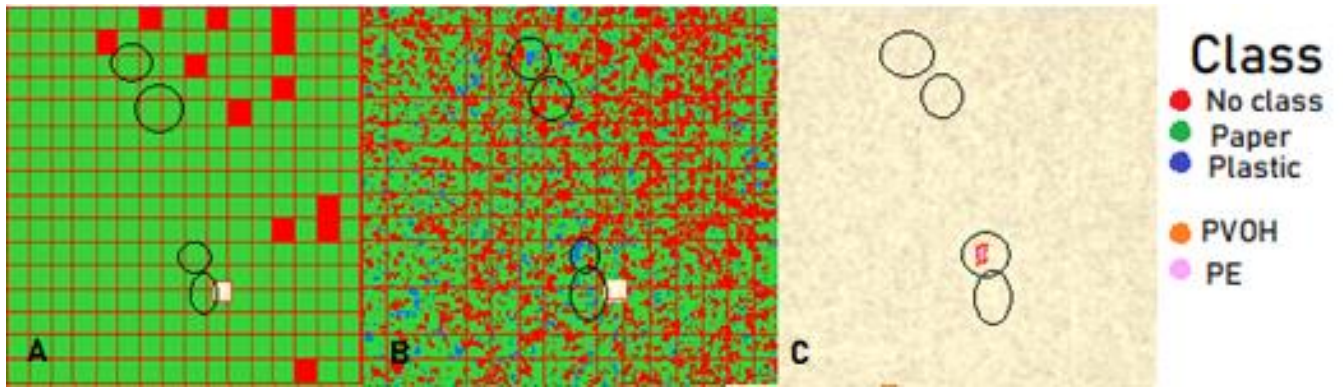


Figure 27: Prediction on paper sheet of 30g/m^2 with the composition of 40% recycled fibers and 60% softwood fibers and four plastic fragments of size $0.2 \times 0.2\text{cm}^2$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE

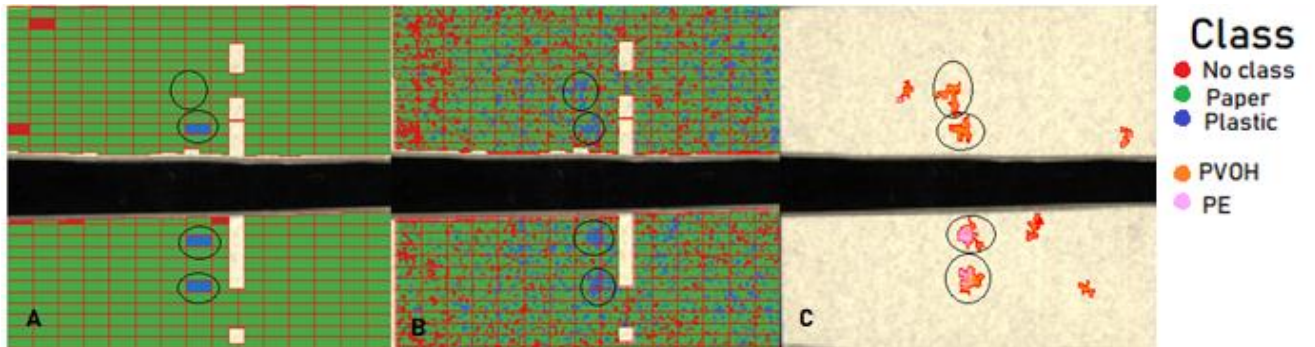


Figure 28: Prediction on paper sheet of 30g/m^2 with the composition of 60% recycled fibers and 40% softwood fibers and four plastic fragments of size $0.5 \times 0.5\text{cm}^2$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE

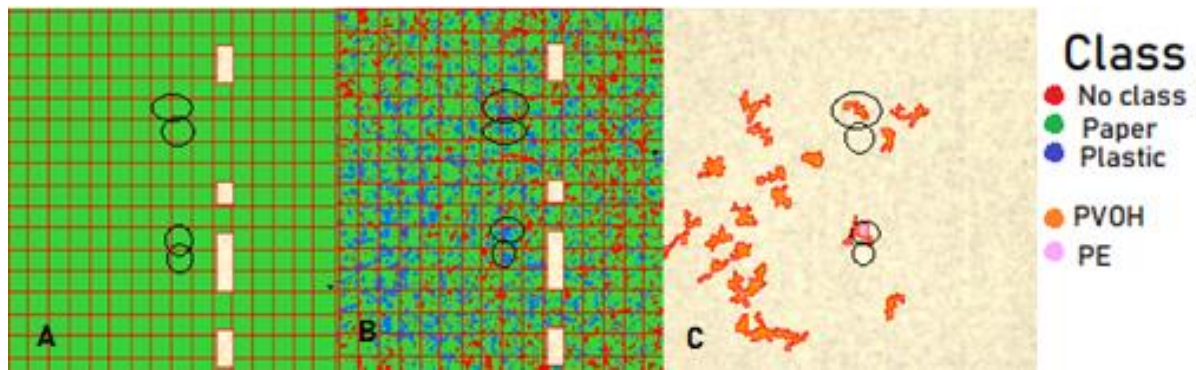


Figure 29: Prediction on paper sheet of 30g/m^2 with the composition of 60% recycled fibers and 40% softwood fibers and four plastic fragments of size $0.2 \times 0.2\text{cm}^2$ under. The circles illustrate an approximation of the actual placement of the fragments, the two upper ones are PVOH and the two lower ones are PE. (A) classification of paper and plastic on grid-level, (B) classification of paper and plastic pixel-level (C) classification of PVOH and PE