TAMPEREEN TEKNILLINEN YLIOPISTO
TAMPERE UNIVERSITY OF TECHNOLOGY

MARIANNE HURNANEN

HEAT SEALING PAPER WITH POLYMER FILM

Master's thesis

Examiners: Jurkka Kuusipalo and Sanna Auvinen
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ABSTRACT

MARIANNE HURNANEN: Heat sealing paper with polymer film
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This thesis concentrated on studying heat sealing paper with polymer film by hot-bar sealing. The objectives included learning more about the relationship of the materials and how they behave in different heat sealing conditions. Also some methods were tested out for heat sealed samples to find out if they would provide useful information because one interest was to find new methods for testing.

Theory part introduces flexible paper-polymer films packages that are opened by peeling. Also the heat sealing process and the method to measure seal strength have been presented. Adhesion theories that are the most applicable considering heat sealing have been introduced.

The research part of the thesis included more comprehensive matrix study about the influence of sealing time, temperature and pressure to seal strength and peel characteristics of sealed materials. Smaller studies concentrated to the effect of peel angle. Profilometer was used to measure surface roughness of both paper and polymer film parts of heat sealed samples that had been peeled open. The angle method was tested to find out if it would give additional information about the seal edge of the samples since it was originally developed for polymer-polymer samples. For fibre amount index measurements samples are usually peeled open by hand but it was tested if using an instrument for that would give smaller standard deviations of fibre amount index.

It was found that sealing temperature affects seal properties greatly. When certain level of pressure is applied, it doesn't affect seal strength notably anymore if it's increased. Correlation between seal strength and fibre amount index was found not to be straight forward. Results suggested that pressure effects on the peel characteristics so that high pressure possibly makes the paper surface more compact and molten polymer doesn't flow into valleys and voids of paper's surface thus giving smaller fibre amount index. With profilometer it was possible to obtain differences to the surface roughness for samples heat sealed with different parameter and the images showed clearly the fibres that were pointing out of paper surface and attached to polymer film. When comparing manual peeling to using an instrument it was found that with the instrument fibre amount index levels were higher but standard deviations were not smaller.

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Tämä työ keskittyi paperin ja polymeerifilmin kuumasaumaukseen kuumapalasaumauslaitteella. Tavoitteena oli saada lisää tietoa materiaaleista ja siitä miten ne käyttäytyvät erilaisissa kuumasaumausolosuhteissa. Lisäksi muutamia tutkimusmenetelmiä testattiin kuumasaumattujen näytteiden tutkimukseen, jotta saataisiin selville tuottaisivatko ne hyödyllistä tietoa. Yksi kiinnostuksen kohde olikin löytää uusia sopivia testausmenetelmiä.

Teoriaosuudessa esitellään joustavia paperi-polymeerifilmipakkauksia, jotka avautuvat peelautuvasti. Lisäksi on selitetty kuumasaumausprosessi ja saumojen lujuuksien mittausmetodi. Kuumasaumauksessa parhaiten sovellettavissa olevat adheesioteoriat ovat myös selitetty.

Kokeellinen osa sisälsi laajemman matriisi-tutkimuksen saumausajan, -lämpötilan ja paineen vaikutuksesta saumanlujuuteen ja kuiturepeämään. Suppeammat kokeet keskittyivät peelauskulman vaikutuksiin ja eri materiaalien käyttäytymisen tutkimiseen. Profilometritutkimuksilla mitattiin paperin ja polymeerifilmin pinnankarheuksia näytteistä, jotka olivat ensiksi kuumasaumattu ja sen jälkeen avattu. The angle -metodilla testattiin saataisiinko sillä mielenkiintoista tietoa paperi-polymeerifilminäytteiden sauman reunoista. Alun perin tämä metodi on kehitetty polymeeri-polymeeri näytteille. Kuiturepeämää tutkittaessa näytteet revittiin normaalisti auki käsin. Jotta saatiin selville vaikuttaako käsin avaaminen kuiturepeämän keskihajontoihin, käytettiin laitetta avaamaan saumat.

Kokeiden perusteella todettiin, että saumauslämpötila vaikuttaa muodostuneiden saumojen ominaisuuksiin huomattavasti. Kun tarpeeksi korkeaa saumauspainetta on käytetty kuumasaumauksessa, ei sen nostaminen enää vaikuta suuresti saumanlujuuteen. Tulokset viittasivat siihen, että saumauspaine vaikuttaa kuiturepeämään siten, että suuri paine mahdollisesti puristaa paperin pinnan tiiviimmäksi, jolloin sula polymeeri ei pääse virtaamaan paperin pinnanmuotoihin ja kuiturepeämä jää pienemmäksi. Profilometrilla saatiin eroja pinnankarheuksiin näytteiden välille, jotka olivat kuumasaumattu eri parametreilla ja saadut mikroskooppikuvat paljastivat kuituja, jotka osoittivat ulos

paperista ja kuituja jotka olivat kiinnittyneinä polymeerikalvon pintaan. Kun vertailtiin näytteiden auki repimistä käsin ja laitteella todettiin, että laitteella saatiin suurempi kuiturepeämä, mutta keskihajonnat eivät olleet pienemmät.

PREFACE

This work was done at Tampere University of Technology's Paper Converting and Packaging Technology Research unit.

I would like to thank my professor Jurkka Kuusipalo for trusting me with this opportunity and for his help during the project. Also big thank you belongs to Sanna Auvinen for helping with all practicalities and always making time for me. Malin Kraft, thank you for advising me and for the helpful and inspirational conversations. I would also like to thank all other staff at Tampere University of Technology that have in any way helped me with this work, especially Hilkka Koivuniemi-Mäkinen who was always very helpful in the laboratory. Thank you for Tampereen teknillisen yliopiston tukisäätiö sr for the funding of the project.

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Marianne Hurnanen

CONTENTS

LIST OF SYMBOLS AND ABBREVIATIONS

ASTM	American Society for Testing and Materials
CPP	cast polypropylene
DSC	differential scanning calorimetry
EVA	poly(ethylene-vinyl acetate)
FT-IR	Fourier transform infrared spectroscopy
HDPE	high-density polyethylene
JIS	Japanese Industrial Standard
LDPE	low-density polyethylene
LLDPE	linear low-density polyethylene
PE	polyethylene
PET	polyethylene terephthalate
PVC	poly(vinyl chloride)
SEM	scanning electron microscope
TUT	Tampere University of Technology
b	the width of the bonded area
F	peel force
F_p	peak force
G	bonding strength
Sa	average height of selected area
Sz	maximum height of selected area
T_g	glass transition temperature
T_m	melting temperature
γ_{LV}	surface free energy of the fluid material in equilibrium with its vapour
γ_{SL}	interfacial free energy between the solid and liquid material
γ_{SV}	interfacial free energy of the solid material in equilibrium with a fluid vapour
θ	contact angle

1. INTRODUCTION

Package is a vital part in assuring the safety of a product. It protects during transportation and it keeps the product isolated from environment. Examples of these environmental factors are contamination with bacteria, toxins, oxygen and moisture. On the other hand package should maintain the atmosphere inside the package if for example some protective gas has been used. What else is expected from a package is ease of use and low cost. [1, p. 18]

One way to form a package is by heat sealing technology. For example in Japan every person every day uses over ten heat seal packed products [1, p.1]. Heat sealing is a method where two materials are attached to each other by heat and pressure for a certain time. Generally these materials that are sealed are thermoplastic polymers but other materials such as paper can be heat sealed with thermoplastic polymer as well. Packages closed by this method are used for example pre-heated and sterilized foods, baby and family care products, injectable and oral medicines, snacks, toiletries, electronic components etc. [1, p. 2].

In this study the focus was on heat sealing polymer film with paper using hot-bar heat sealer. The opening system of these structures is wanted to be peelable meaning that the paper and polymer film separates from each other when the seal is opened. Loose fibres that might be separating from the paper surface in the opening process are not desirable.

Majority of available literature concentrates on heat sealing thermoplastic polymer with thermoplastic polymer. That is why some of the theory presented in this work concerns also sealing of polymer with polymer. Why paper is used in this type of packages is because for example the item inside can be sterilized after sealing the package, paper is sustainable material with low costs and can be easily disposed [2].

In the theory part paper and polymer film based flexible packages are introduced. It covers the hot-bar heat sealing technique and explains how the seal strength of a package is measured. Different sealing parameters (time, temperature and pressure) have different kind of effects to seal strength and this has been given some consideration in theory part as well. Adhesion theories that are the most applicable regarding heat sealing have been introduced and then a closer look has been taken into paper/polymer laminate adhesion.

This study examines the effects of sealing temperature, time and pressure to seal strength and also to peel characteristics of paper-polymer film structures. The prior studies concerning this had mostly been done to polymer-polymer structures. The effect of peel angle

was tested for different paper and polymer film combination to see how this affects seal strength. Other interests were finding out if profilometer would be suitable method for studying the surfaces of heat sealed and then peeled open paper and polymer film surfaces. The angle method of Hishinuma which was originally created for heat sealed polymer-polymer structures was tested for these samples in hope that it would provide some interesting information [1]. The samples for peel characteristics were generally peeled open manually but some samples were prepared with an instrument to find out if this affects the standard deviation of fibre amount index at all.

This work had on its background some longer term goals. This study was not even thought to be able to solve everything completely but rather help taking steps forward. One of these goals was obtaining understanding in general what happens during heat sealing process and learn more about the relationship of the materials. Another goal was to decrease the standard deviation of fibre amount index or figure out if there is a better way to measure it than what the existing way is.

2. PAPER AND POLYMER FILM BASED FLEXIBLE PACKAGES

Flexible packages that have paper as the other component are combined with other materials such as for example plastics and aluminium foil. Because of all the possible material combinations these flexible packages offer wide scale of different properties that are for example heat sealability, barrier properties and printability. From packaging types these packages are the fastest growing application and some examples of the packages are sachets, pouches and bags made on form. [3, p. 277-278]

Paper based packages are used for example in medical packaging and one such important application is combining paper with polymer films or laminates to create peel pouches. These pouches are closed by heat sealing or adhesive coat can also be used. These types of packages are often used for sterile disposable medical devices that are terminally sterilised. Plastic film sealed to paper can be flat or shaped by heat forming. These peel pouches are used the most with articles such as for example syringes, needles, catheters, gloves and dressings - articles that are used in large volumes. [2, 4, p.110] An example of this type of package is shown in Figure 1.

Figure 1. *A package with peel open system by Arjowiggins-Healthcare [5].*

Paper-polymer packages have some important features. One of them is that the packed item can be sterilised after it is sealed in its package. This is possible because paper is porous material. Sterilisation can be done with steam in an autoclave, some other form of steam sterilisation, ethylene oxide gas or gamma radiation. After sealing and sterilisation the package has to retain its microbiological barrier. Controlling the maximum pore size of the paper affects to this. When opening the package, it must peel open which means that the paper and polymer film separate from each other so that the paper does not tear

in a way that loose fibres appear. If there would be loose fibres they could end up for example to a wound site and cause irritation of tissue or other problems. Furthermore once the package has been opened it should not be possible to reseal it. [4, p. 109-111]

These paper-plastic peel pouches have other advantages in addition to sterilisation of the already packed item. They have relatively low cost and they fit for high-volume or small-run devise packages. Material wise they can be manufactured of variety of choices. Forming of the package can be prefabricated or formed in-line. Ease of use of this package is enhanced by visibility of the product and with an easy opening system. The pouch can also be printed with product information and instructions. [2]

Some disadvantages come from that they are not suitable neither for high-profile devices nor products with a high mass. These peel pouches also have low capabilities for dynamic protection. Highly irregular shaped devices can't be packed in these or kits or multicomponent devises. [2]

Paper was the sole option for this type of medical packaging material until Tyvek ® was introduced to the market. Nevertheless, paper still has a considerable role in medical device industry. It has some properties that maintain it as a feasible packaging material. These are sustainability, cost, disposability, suitable sterilisation methods, possibility to be combined with other materials, versatility, peelability and range of grades. It also has some limiting factors such as low tear and puncture resistance, dimensional stability, moisture sensitivity and aging is limited under certain environmental conditions. [2]

2.1 Direct seal paper

Direct seal paper is a kraft paper type that can be sealed directly to non-corona-treated polyethylene [4, p. 112]. It used to be so that direct seals weren't as strong as seals done with heat seal coated papers or when trying to get higher seal strengths it led to excessive tearing of fibres from the paper surface. However, nowadays second-generation direct seal packaging papers reach the requirements regarding seal strength and peel cleanliness. Why this direct seal technique is desirable is because it offers benefits such as lower costs, maximised porosity and no potential interactions with coatings and the packed product. [6] In addition, it offers environmentally friendlier option compared to adhesive coated materials [7].

Paper is material that is formed from short fibres. Usually during the fabrication the fibres orient in the machine direction. They are not completely flat but have some z-plane orientation as well and their formation could be compared to roof shingles. This orientation of paper affects its peelability and that is why it is good to give some consideration to the machine direction of paper and the peeling direction of the ready packages when designing the package. The direction of the peel should be in the machine direction and the z-orientation of fibres should be away from the peeling direction. Designing the package

like this doesn't guarantee fibre-free peel but it helps to avoid delamination through the fibre layers leading to total paper tear. [8, p.68] However, the newest papers can be made in the way that the fibres' alignment with machine direction is prevented thus making the peel cleaner [6].

A very light weighted coating (sizing) can be applied to paper surface to modify its properties. This coating improves seal strength and also gives very clean and undirectional peel. [6]

The cleanliness of the peeled seals used to be evaluated subjectively but some progress has been made and the peeled seals can be for example scanned and the images of them are digitally compared to references and based on that the level of fibre amount index is determined. [6]

2.2 Polymer films in heat sealing

In the case of flexible medical packaging generality of them have been constructed so that they have at least one part made from plastic film. Using polymer film creates some favourable properties for the pouch such as for example visibility of the product, puncture resistance, sealability and peelability. [9]

Polymer film is partly melted when it is heat sealed. This breaks its original crystal structure and possible orientations which means that mechanical properties are altered. This is one reason why multilayer polymer films are favoured because then only the adhesive layer is partially melted but the structural layers stay unaffected. [10, p. 38] Pinholes can be problematic with one layer films in heat sealing because then the seal will not be complete. Use of multiple-layer films prevents this and their use has become popular. In addition, using more than one layer in film makes it possible to tailor properties such as for example mechanical strength, formability and barrier properties. [1, p. 3, 6].

With medical device packaging the most commonly used material is lamination of polyester and polyethylene. One typical example of the film consists of oriented polyester film with a thickness of 0.0127 mm which is adhesively laminated to PE with a low-to-medium density with film thickness of 0.038-0.051 mm. Usually the PE is modified with poly(ethylene-vinyl acetate) (EVA) to get better sealability. [9]

2.2.1 Low-density and linear low-density polyethylene

Different polymers have quite different heat sealing characteristics. These depend on such properties as molecular weight, degree of crystallinity, melting temperature and overall composition. [3, p.262] Because in this study's experimental part the sealing layer of the used materials has been polyethylene some of the properties of different grades of polyethylene are presented here.

Different grades of polyethylene (PE) are primarily classified based on density. Difference in density between low-density (LDPE) and high-density polyethylene (HDPE) is due to their molecular chain structures. Compared to HDPE, LDPE has several more fairly long branches from the main chain. These side branches prevent molecules from packing together as tightly as in HDPE. Difference between linear polyethylene grades and LDPE is that linear polyethylene grades have more branches from the main chain but they are shorter than in LDPE. [11, p. 15-18] Lower density usually means lower crystallinity for polyethylenes and also lower melting temperature [3, p. 262]. Some properties of LDPE, LLDPE and HDPE films are gathered in Table 1.

LDPE and LLDPE films are the most common ones used in packaging applications. Both of them have hazy appearance and as a material they are soft and flexible. If they are compared with each other when having equal thickness and density LLDPE has greater impact strength, tensile strength, puncture resistance and elongation. LDPE seals at lower temperature, has a wider temperature range where it seals and has better hot tack than LLDPE. Long-chain branching affects greatly to these properties of LDPE. [10, p. 242] The higher melt flow rate polymer has the lower its melt viscosity is and also the average molecular weight is lower. Usually lower melt index means higher seal strength but also the minimum sealing temperature is then higher. [3, p. 262]

Table 1. *Typical properties of polyethylene films [10, p. 243, 11, p. 19, 12, p. 152-179].*

Property	Polymer		
	LDPE	LLDPE	HDPE
Glass transition temperature, T_g [°C]	-120	-120	-120
Melting temperature, T_m [°C]	105-115	122-124	128-138
Density [g/cm^3]	0.915-0.940	0.915-0.935	0.94-0.97
Tensile strength [MPa]	8-31	20-45	17-45
Tensile modulus [GPa]	0.2-0.5	−	0.6-1.1
Degree of crystallinity [%]	40-50	−	60-80
Melt viscosity [kPas], shear rate = 0 s^{-1}	54.5 (150 °C)	25.5 (150 °C)	

3. HEAT SEALING

Heat sealing is a method where two materials are sealed together by heating them while applying pressure on them for some certain time. Ordinarily the materials that heat sealing was used for were thermoplastics which can be heated 20-100 °C above their melting temperature and then cooled down, what improves material's complete sealing. [1, p. 2]

There are different methods in heat sealing and these are for example hot-bar sealing, impulse heating, hot air blast heating and ultrasonic heating [1, p. 30-34]. Here only hot-bar sealing has been introduced because it is the used method in this study.

3.1 Hot-bar sealing

Hot-bar sealing or by another name heat jaw sealing is the most used heat sealing method [1, 13]. The very basic idea of hot-bar sealing is to attach two heated materials by pressing them together. Heat conducts from the jaws' surfaces to the materials and melts them. Cooling is done after heating and it finishes the bond. [1, p. 6] With the conventional method cooling is done after the heating jaws are removed, so there isn't any pressure applied on the seal during it. This may sometimes cause reopening of the seal when the jaws are opened. As a solution for this there's a variation where a cooling tool is used. The tool is pressed on the seal after the heated jaw is removed. [13]

In Figure 2 is shown more detailed picture of the method. Heat jaws are heating blocks that have a built-in heat source and a temperature sensor. The arrows in the image depict the movement of the jaws. For maintaining low temperature distribution in the heating block there is a heating tube between the sealing surface of the block and the heat source. This way the heating distribution at the surface can be kept within 0.2 °C tolerance. [1, p. 31]

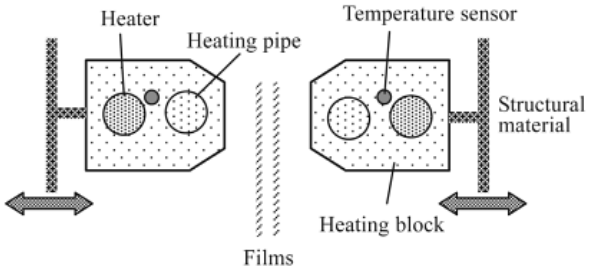

Figure 2. *The basic idea of hot-bar sealing [1, p. 31].*

For preventing overheating and to decrease the fluctuation of the set and actual temperature the sensor is placed next to the heating source [1, p. 31].

This method is based on the conduction of heat from the heating blocks to the material. That of course limits how thick the sealed materials can be. Either just one of the blocks can be heated or both of them. Heating both of them reduces needed sealing time. [13]

3.2 Peel and tear seal

There are two different ways a heat sealed seal can break. These failure modes are interfacial/pseudo-adhesion which is also called peel seal and the other one is melt/cohesive adhesion also called tear seal. They are presented in Figure 3 where tensile testing is applied to the films and causes the failure. In the case of peel seal the layers delaminate from one another whereas with tear seal the failure happens close to the heat sealed area but not in the interface of the materials. [1, p. 6-8]

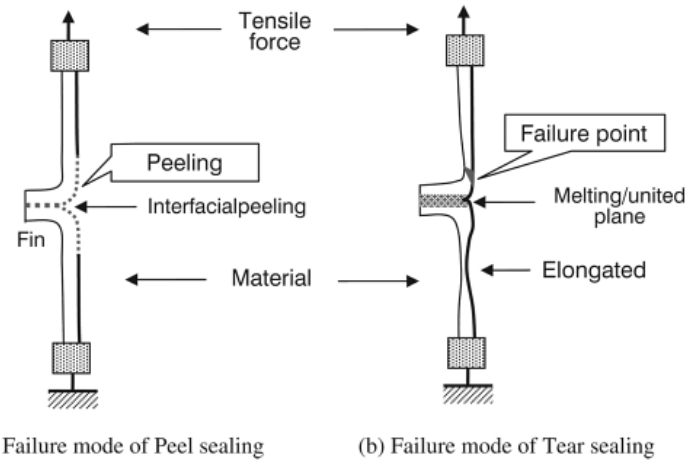

Figure 3. *Failure modes for peel seal and tear seal [1, p. 9].*

When heat sealed materials are polymers and the failure mode is peel seal, polymer molecules have not yet diffused entirely therefore the interface of the two films has not disappeared. Whereas, in the case of tear seal polymer molecules have diffused well and the interface of the films has vanished. If a tensile strength that is higher than the strength of the used polymer film is applied on the film, plastic behaviour occurs. [1, p. 8-9]

What type of failure mode a seal will have, depends on the sealing temperature. In Figure 4 tensile strength of a seal is presented as a function of melting surface temperature. The melting surface temperature means the actual temperature of the melting material during the sealing and not the temperature of the sealing bars. The region for peel seal to occur is with lower temperatures than for tear seal. Depending on materials the peel seal zone width and temperature differ. [1, p. 6] With high temperatures the melt viscosity of polymer decreases so much that excessive deformation can happen which leads to decrease in seal strength [3, p. 260].

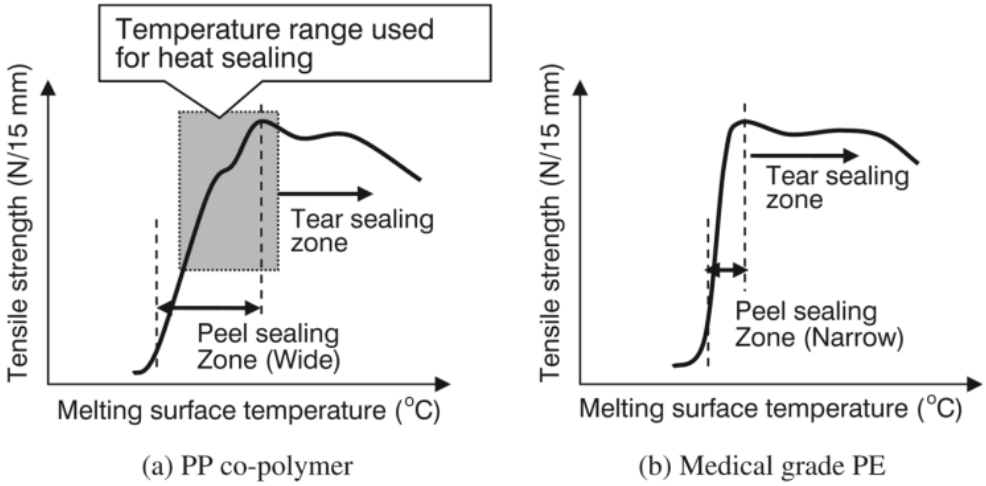

Figure 4.*Change of tensile strength as a function of melting surface temperature [1, p. 8].*

3.3 Determining seal strength of a package

Packages have important role in delivering the content in such a condition that they are safe to use for the application they were meant for. There has to be high confidence that the items have stayed in sterilized condition in the package through the supply chain if sterile condition is demanded. International and domestic regulatory agencies follow the design and development of packages more carefully nowadays than earlier. There has been emphasises on standardising the development of packages and therefore there exists standards that describe how to for example test some qualities of medical packages. [2] Here we concentrate on seal strength of a package because it is essential part of this study.

Package seal strength gives fundamental information about manufacturing process of a package. It is used in process validation and process control. Packaging seal strength refers to strength needed to separate two components of a package from each other and it is expressed as force per unit width. The American Society for Testing and Materials' (ASTM) standard ASTM F88-00, "Standard Test Method for Seal Strength of Flexible Barrier Materials" describes the method for measuring seal strength. This standard is industry's definitive technique to characterise seal strength. [2, 14]

This method defines seal strength of a certain width of some point of the seal. Hence, it doesn't tell about the seal continuity of a whole package. In the test a 25 mm wide strip is clamped from each end to a tensile strength testing instrument. In this test the force is applied perpendicular to the heat sealed line. Peel angle can be 90° or 180°. The 90° peel angle test can be done with or without support. [2] Figure 5 demonstrates these setup options. The testing equipment usually gives curve where there is force versus displacement. An example of this curve is shown in Figure 6. Many times the maximum seal force is the most important data acquired from the test but sometimes the average force for

opening the seal is more important [14]. In the case of peeling polymer from paper this curve is susceptible for the surface quality of the paper. As demonstrated in the image some spots that have low adhesion can be easily noted from the force differences. [15, p. 265]

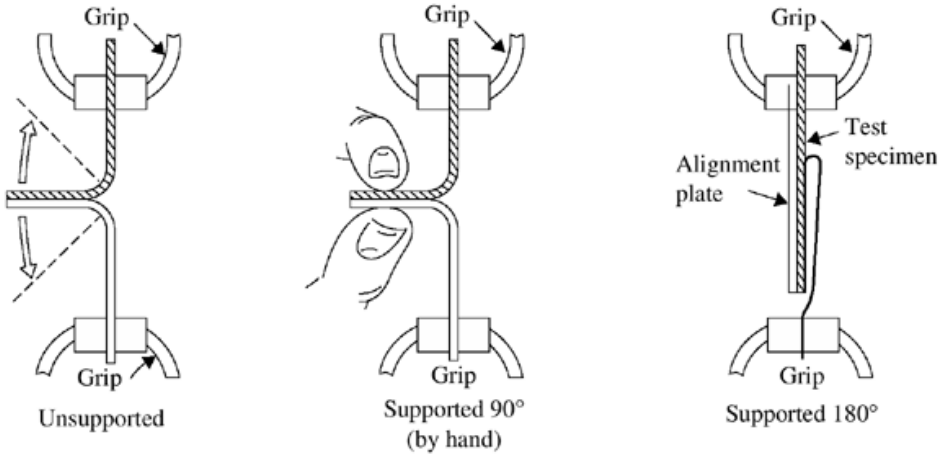

Figure 5. *Tail holding methods for seal strength test [14].*

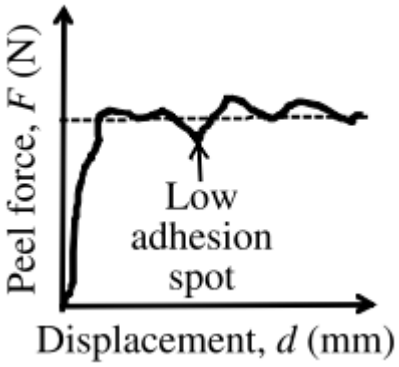

Figure 6. *An example of a peel curve obtained from a peel test [15, p. 564].*

When using this testing method terms peel rate and grip separation rate should not be mixed. If in the test parting of the grips translates fully into peeling the seal, the grip separation length x cm is only 0.5x cm because of the advance of the failure line in the seal. In this case the peel rate of the seal is actually ½ of the grip separation rate. [14]

3.3.1 The angle method

According to Hishinuma polyballs form during heat sealing if too high pressure is used and the polymer film is in liquid state. Polymer is forced out from the sides from under the sealing bars and this polymer that is along the side of the seal is called polyball. Because some of the sealing material has flowed out of the seal area it makes the formed seal weaker than what it would be if a polyball wouldn't occur. These polyballs cause microscale jaggedness and notches to appear on this area if stresses are applied on it.

From the notch a pinhole forms easily because of the stresses' concentration on the notch and later a crack gets its start from the pinhole and can lead to the failure of the package. [1, p. 73-74, p. 103]

Polyball's size is about 30-50 µm. With the standard for testing heat sealed films the sample width has been defined to be 15-25.4 mm (ASTM F88-00 and JIS, Japanese Industrial Standard, Z 0238) and the force is directed perpendicularly to the heat sealed line. With this method it is difficult to distinguish peel and tear seal in the range of 30-50 µm. The diagram in Figure 7 presents analytical model for seal strength testing. The "wave line" presents the edge of the heat seal and it is also the part where the load is applied first when doing the peel test. On the tensile testing diagram the edge of the seal shows at the start as the parts marked with (1) and (2). The diagram also shows some lower adhesion spots (3) and (4) which can be for example air bubbles and foams at the interface. From Figure 7 can be seen that the edge of the seal, where the possible polyball is, is really small part of the diagram so it is not possible to tell by using it if there are polyballs present or not. [1, p. 104-105]

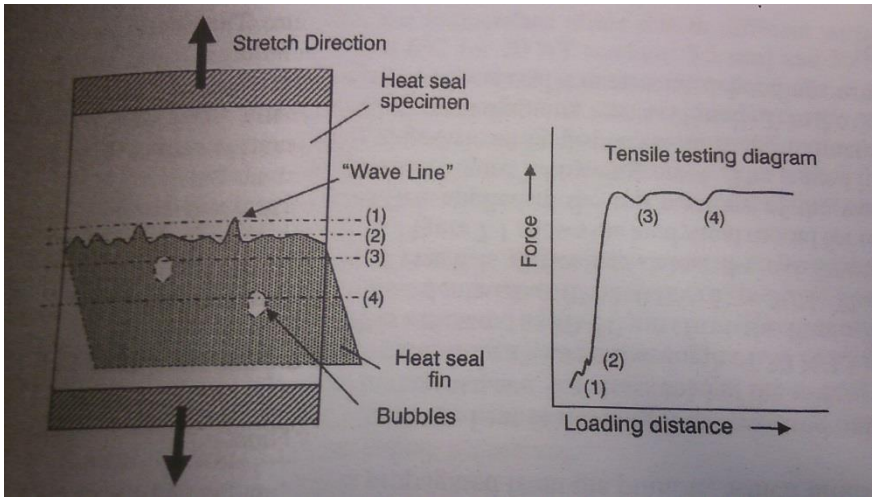

Figure 7. *Analytical model for seal strength testing [1, p. 105].*

The angle method has been created by Kazuo Hishinuma and it can be used for optimizing sealing conditions. In it the heat seal line isn't parallel to the tearing line like in ASTM F88-00 standard, but instead it is in 30-45 degree angle. With this set up the stresses are concentrated on the heat sealed edge. [1, p. 106-107]. The setting of this test method is presented in Figure 8.

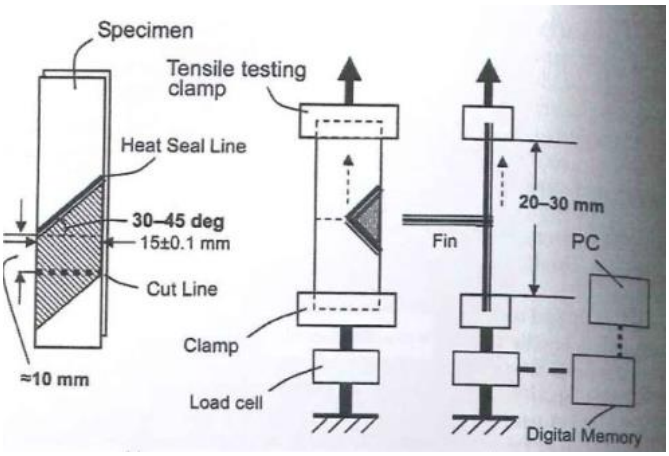

Figure 8. *Principle of the Angle Method [1, p.108].*

With peel seal in question the peeling of the seal starts from one point. The area of the seal during peeling increases linearly till it reaches the whole length of the sample. During peeling of this part where the debonding area increases linearly also tensile load grows linearly. After reaching the part where the whole width of the sample is sealed tensile load reaches plateau value. The plateau value should match with load applied to a sample prepared according to the JIS standard. In case of tear seal the sample fails or when composites are used the films become delaminate due to polyball and the stage of the tear seal on the heat sealed edge line. Because of the failure or delamination seal strength decreases radically. [1, p. 107-108]

3.4 Effect of time, temperature and pressure in heat sealing

In heat sealing variable parameters are sealing temperature, time and pressure. As mentioned earlier the seal failure mode depends on sealing temperature but these other parameters have some effect too. For example with higher sealing temperature a shorter sealing time might be needed than with lower sealing temperature. The parameter range where acceptable seals are obtained is important factor in manufacturing [16]. For example with a wider sealing temperature range the unintended changes in processing conditions, such as fluctuation of sealing bar temperature, will not lead to unacceptable seal characteristics as easily as with materials that have narrower sealing range. [16, 17, p. 1337]

In the study of Dixon et al. medical grade Tyvek® which was coated with a water-based adhesive was bonded with PE/PET film. It was found that from the variable parameters pressure had minimal influence on maximum peel strength but with low temperatures and short sealing time peel strength was sensitive to pressure. Other observations were that minimum peel strength was sensitive to too high temperature and long sealing time. When high temperature and long sealing time were used it produced irregular peel trace on load-extension curve with peaks and troughs. [16]

The study of Najarzadeh et al. dealt with heat sealing monolayer linear low-density polyethylene film. They also found that there was a strong relation between seal strength and sealing time and temperature. Whereas, sealing pressure wasn't as notable as them provided that the films have adequate contact between them. It was pointed out that temperature and time influence seal strength in the same way since longer time lets bigger amount of heat reach film interface where it changes the film surface from crystalline to partially molten and in the end to completely molten. Pressure affects seal strength in a different way than sealing temperature and time. Its purpose is to bring the materials to be sealed into a close contact at molecular scale. [17, p. 1337-1339]

Aithani et al. studied also the processing parameters of heat sealing. They found out that heat sealing samples with temperature near the fusion point though below melting point produced the highest seal strengths. The fusion point is at the temperature of inflection point and on the time-temperature curve on it the second derivative changes from negative to positive values. The idea of the inflection point is based on a change in heat flow rate as the polymer film starts to melt. [18, p. 247-252]

In the case of LDPE a peel seal occurred until interface temperature of 112 °C and the highest seal strength was observed in this interface temperature at 112 °C and 110 °C. Those temperatures were in the vicinity of the fusion temperature of LDPE. At higher temperatures mixture of the peel and tear failure modes were obtained. This behaviour of the fusion temperature being in the proximity of the fusion temperature was observed with other polymer films studied which were made from high-density polyethylene (HDPE), LLDPE and cast polypropylene (CPP). The effect of sealing pressure was found to be limited as in the earlier studies mentioned in this chapter. It was found that the sealing time did not affect seal strength after the interface of the materials to be sealed reached the set sealing temperatures. [18, p. 256-259]

3.5 Peel rate and peel angle

In Figure 9 a) is shown peel force as a function of peel distance for paper/adhesive laminates. The graph has two curves from which the other presents a typical curve when interfacial failure happens and the other when paper failure occurs. These curves have different peel rates: 100 mm/min and 400 mm/min. With the lower rate happens interfacial failure and the curve is noisy but roughly constant. Whereas, the other curve first has a maximum peak (the peak force F_p) and then falls low to a steady value which corresponds to delamination of paper. On the tape there is at least one layer of fibres after peeling. This paper failure usually starts from a weaker area on the contact line after which the area expands and merges so that the whole layer will be peeled on the tape. Customarily the engineering polymers that are used for coating paper have such strong bulk strength that their cohesive failure is rare. [15, p. 568]

Numerous experimentations have shown that the peak force F_p is the key property when studying interactions between paper and polymer. Based on this discovery a new method to analyse peel data was made. In it the peel behaviour of paper/polymer combination is presented by drawing the log peak peel force as function of log peel rate. This gives general peel curve which has rate-dependent interfacial failure domain and rate-independent paper failure domain (Figure 9 b). [15, p. 568]

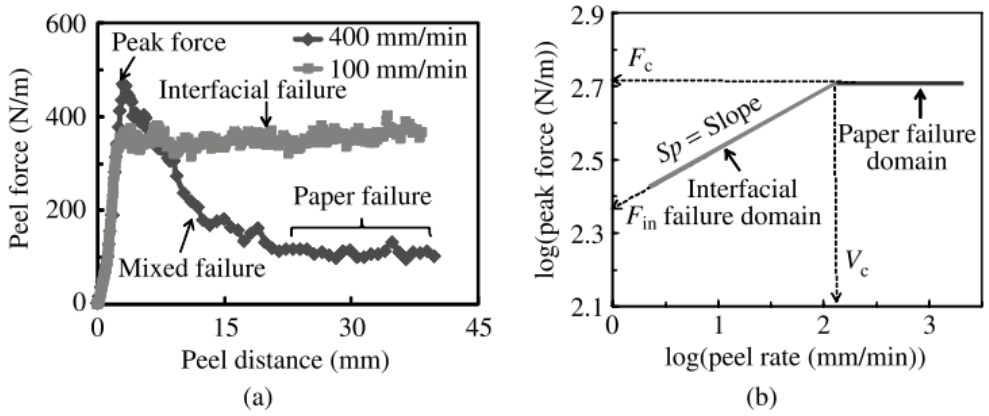

Figure 9. *A) Peel force as a function of peel distance and typical failure modes for paper/adhesive laminate, b) generalized peeling map as log peak force as a function of log peel rate. [15, p. 568]*

Peel angle affects this generalized peel curves by moving it vertically. The properties of the polymer adhesive have an effect on the slope of interfacial failure domain but not considerably on the paper failure domain. The direction of peeling influences on delamination of paper: paper delaminates easier when it's peeled to the fibre orientation direction. For both of these peeling directions the maximum peak force was discovered to be same which implies that it's a direction independent parameter. [15, p. 568-569]. What has to be noted here is that the research above was done to polymer film-paper laminate structures and not for heat sealed ones, which was the research target in this study.

Seal strength between polymer and paper is strongly determined by peel angle. In the case of general peel test where peeling is done incrementally the force can be obtained from the energy balance approach as:

$$G = \frac{F}{b}(1 - \cos\theta), \tag{1}$$

Where G is the energy release rate but it is often used as bonding strength with peel test, b is the width of the bonded area and F is peel force. [15, p. 565] Because $\cos 90° = 0$ and $\cos 180° = -1$ for the 90 degree peel test the equation (1) can be derived into form

$$G = \frac{F}{b}, \tag{2}$$

and for the 180° peel test

$$G = \frac{F}{b}2. \quad (3)$$

The bonding strength should be same for samples that are prepared in same conditions and with same sealing parameters. This means that the peel force of samples peeled in 90° should have two times the force than the samples peeled in 180° angle.

3.6 Critical points in heat sealing

While pressure is necessary to reduce the distance between heat sealed materials to obtain intermolecular bonding too high pressure can cause problems. High pressure can push the melted polymer away from the heat sealed region and cause formation of polyballs. [1, p. 23] Also this will lead to reduced film thickness on the seal area which will in turn lead to lower seal strength [3, p. 264]. Experiments have showed that appropriate pressure range is 0.08-0.2 MPa when heat sealing polymer with polymer. Lower pressure than that will result in loss of thermal conduct and create insecure adhesion. Higher pressure than 0.2 MPa is found to create polyballs. [1, p. 23]

With hot-bar and wire sealers silicon rubber pads or PTFE-coated glass fibre coverings are often used. If these are not cleaned or replaced regularly it can lead to uneven sealing pressure. [3, p. 264]

If excessive sealing temperature is used it can lead to denaturation of polymer. It means that the polymer undergoes depolymerisation and volatile contents evaporate. Depolymerization happens when radicals react due to heating and covalent bonds are created with hydrogen and oxygen in polymer chains. These reactions shorten polymer chains, which lowers polymer's elasticity and increases its brittleness. [1, p. 74]

4. ADHESION

By definition adhesion is a state where two objects are held together by very close interfacial contact through what mechanical forces can be transferred. Practical adhesion is usually connected to the force that is needed to break this bond between materials. [19, p. 14] There are several adhesion theories that describe the phenomena in different ways. Each of these theories is important in different applications. Adsorption theory is however considered to be the most likely relevant in most of the cases. [20, p. 4]

The main adhesion theories are adsorption, electrostatic, diffusion and mechanical interlocking theories. Adsorption theory states that mobile phase's macromolecules are adsorbed onto a substrate where forces from stronger chemical bonds to weak dispersion forces hold them in place. According to the electrostatic theory there exists transfer of charges between the surfaces and thus they are held together by electrostatic forces. [20, p. 5] Polymers are insulators by nature so this electrostatic theory is difficult to apply to adhesives [21, p. 9]. In diffusion theory macromolecules of the mobile phase diffuse to the substrate. Here the interface of the two materials is eliminated. In the fourth theory which is about mechanical interlocking other phase flows into the substrate's surface irregularities. After this mobile phase is hardened and it is attached to the surface because of the shapes, hence, a keying action occurs. [20, p. 5] In addition to these theories there is also one about non-adhesion called the weak boundary theory. [20, p. 4; 21, p. 4]

4.1 Adsorption theory

As stated earlier, in adsorption theory a mobile phase's macromolecules are absorbed on substrate and surface forces are created between them [20, p. 4; 22]. These attracting forces are usually secondary or van der Waals forces. One precondition for the forces to develop is that the surfaces have to be in close contact with each other and they cannot be more than 5 angstroms apart. [22]

Contact between an adhesive and a substrate is called wetting. To obtain good wetting the adhesive should flow into the irregularities (valleys, crevices, voids etc.) of the surface. In case of a poor wetting the adhesive bridges over these irregularities and there is less actual contact area between the materials. [22]

Wetting can be measured with contact angle measurements where a droplet is dropped on to a surface and the droplet's contact angle θ is determined [22, 23]. This droplet on the surface either spreads or beads up. If it beads up its contact angle can be determined from three-phase contact line from solid-liquid interface to the liquid-vapour interface. [24] This can be described by the Young's equation:

$$\gamma_{LV} \cos\theta = \gamma_{SV} - \gamma_{SL}, \quad (4)$$

Where θ = contact angle

γ_{LV} = surface free energy of the fluid material in equilibrium with its vapour

γ_{SV} = interfacial free energy of the solid material in equilibrium with a fluid vapour

γ_{SL} = interfacial free energy between the solid and liquid material. [22, 24]

According to a general definition an ideal surface is wettable when the surface angle is less than 90° and nonwettable when the angle is greater than 90° [23]. Wetting is perfect if contact angle is zero [20, p. 5]. In Figure 10 are demonstrated these droplets on wettable and partially wettable surfaces.

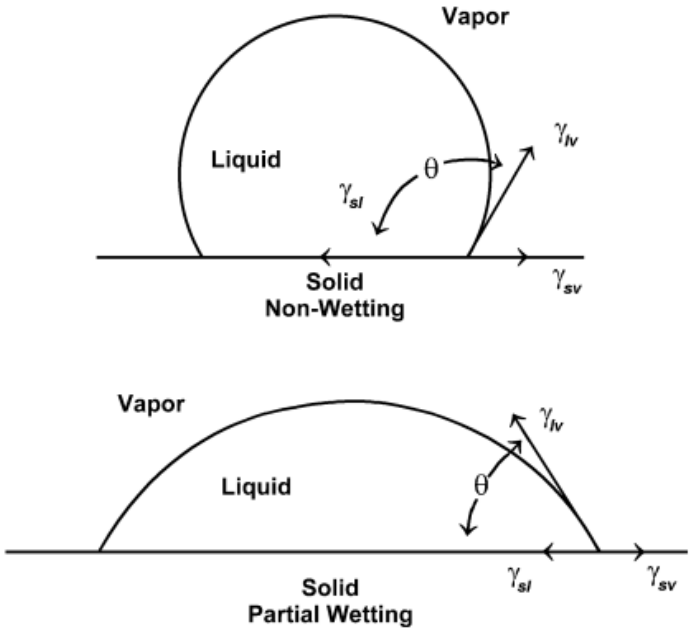

Figure 10. *Droplets on nonwettable and partially wettable surfaces [25, p. 38].*

When separating interfaces in reversible process work is needed. [24, p. 43]. This work equals magnitude of W_a and that is why it is called the work of adhesion and it is defined to be energy change per area as a result of eliminating two bare surfaces and forming of an interface:

$$W_a = \gamma_{sv} + \gamma_{lv} - \gamma_{sl}. \text{ [15, p. 560; 24, p. 43]} \quad (5)$$

When Young's equation and equation (5) are combined it gives Young-Dupree equation:

$$W_a = \gamma_{lv}(1 + \cos\theta). \quad (6)$$

According to it good wetting is attained when W_{12} is higher than zero. [25, p. 38]

Good wetting occurs if substrate has a high γ_{SV} and the adhesive has a low γ_{LV}. For example polymers that have low surface free energy easily wet metals that have high free surface energy. But if polymeric coating or substrate has a low surface energy it is not easily wetted by other materials and hence they are good for applications needing non-stick and passive surface. [22]

4.2 Mechanical interlocking

Solid material always has peaks and valleys on its surface and it is never completely smooth. In mechanical interlocking theory adhesive, meaning the mobile phase, fills these pores, holes, crevices and other irregularities of the substrate. After this the adhesive hardens and is thus mechanically attached to the substrate (Figure 11). For this type of attachment to work properly the adhesive has to penetrate to the pores and other shapes of the surface so that no air is trapped at the interface. [22] If voids are left between materials it leads to trapped air bubbles which allow gathering of moisture. This moisture will in time lead to a decrease of adhesion. [26, p. 38]

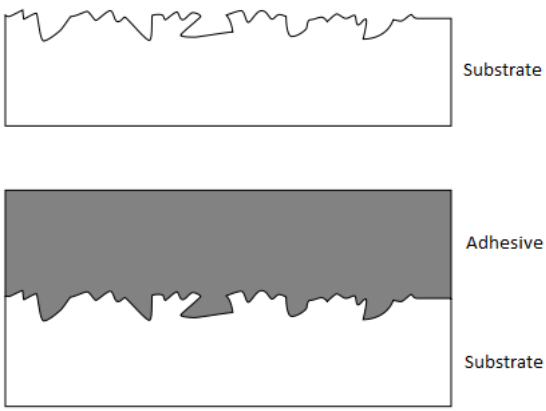

Figure 11. *Schematic illustration of mechanical interlocking [26, p. 38].*

The rougher the surface of the substrate is the more there is contact area for the adhesive and the substrate. If there exist interfacial or intermolecular attractions that have effect on adhesion then increase of contact area also increases the total energy of surface interaction. [22]

4.3 The weak boundary layer theory

The weak boundary layer theory suggests that often when bonding looks to have failed at the interface of materials in reality there is a cohesive rapture of a weak boundary layer [22]. This theory could explain why the calculated bond strength is not same as in the case of actual failure [19, p. 22].

A weak boundary layer at polymer interface can be a result of migration of additives, contaminants or excessive treatments done to the surface which cause a low-molecular-weight layer by breaking polymer chain structures. Also if air stays trapped between materials in bonding it can react with them and create weak boundary layer. On the surface of a paper a weak boundary layer can be created because of fibres that are loosely bonded. [19, p. 22]

4.4 Diffusion theory

Diffusion theory regards primarily polymeric materials [22]. According to the theory when polymers are in contact with pressure applied and heated to high enough temperature they can interdiffuse. This means that chain segments from the two polymer surfaces will interpenetrate thus eventually eliminating the initial boundary between them. [20 p. 5; 21, p. 9] Adhesion is created from this polymer chains' movement across the interface into the other surface [22].

This will only happen if the polymer chains are mobile hence the temperature has to be higher than glass transition temperature [21, p. 9]. Diffusion will occur when the two surfaces are from same polymer but in the case where they are of different material the occurrence will depend on chemical compatibility of the two materials which means that they have to be mutually soluble. [20, p. 9; 22]

Diffusion theory is relevant only in limited number of applications. This is because it is quite uncommon for the adherent and adhesive to be soluble. Mainly this theory is applicable when solvent or heat sealing thermoplastic polymers. [22]

4.5 Paper/polymer laminate adhesion

Conventionally surface energy and surface chemistry properties such as contact angle, composition and acid-base functional groups explain paper adhesion. With equations (5) can be calculated the thermodynamic work of adhesion between paper and polymer adhesive layer. There γ_{SV} would stand for surface energy of the paper, γ_{LV} for surface energy of the polymer adhesive and γ_{SL} for the interfacial energy of the paper and the adhesive. This value for work of adhesion does not predict the practical adhesion but it gives the ideal adhesion which is dependent on surface chemistry. This ideal adhesion refers to making of the bond whereas practical adhesion refers to mechanical energy that is needed to separate the bonded materials. [15, p. 566-567]

This distinction between adhesion and practical adhesion points out, that when paper and polymer are separated from each other the failure does not necessarily happen at the interface. If that is the case then the interfacial forces like van der Waals and acid-base interactions that take part in forming the adhesion bond are not the primary concern anymore. Because paper is a porous material that has high surface energy the adhesion bond

between it and polymer is quite easily created. Other characteristic of paper is that it has layered network structure which is prone to tearing and delamination. Because of this the failure of paper/polymer laminates usually does not happen at the interface. [15, p. 567]

Fibre tear refers to residual cellulose matter from paper which is attached to the other side of the package or is as free particles when opening a package. For instance in the case of flexible paper/polymer film pouch when it is torn open some fibres of the paper can stay attached to the polymer film. As pointed out earlier these residual or free fibres are not wanted for example in medical packaging. [7]

Oni et. al studied the mechanism of fibre tear by heat sealing different papers with multi-layer polymer films. They concluded that to adhesion of direct seal papers with polymer film affects both the mechanical and chemical interaction mechanisms. In the study was found that excessive fibre tear occurred in sample combinations when on the surface of the film there were no imprints of paper fibres when imaged with scanning electron microscope (SEM). This proposes that mechanical interlocking is the underlying reason for fibre tear. In the opening of the seals this excessive interlocking of polymer into the paper structure causes fracturing and breaking up the paper cellulose fibres. The level of interlocking is determined by physical structure of the paper cellulose, paper surface's chemical modification and also by the polymer sealant film's composition and molecular structure. Their results also suggest that fibre tear happens above some certain seal strength value for a specific paper/film combination. [7]

5. RESEARCH MATERIALS AND METHODS

Most of the tests were done at TUT's Paper Converting and Packaging Technology Research unit but some were done at the product laboratory of BillerudKorsnäs AB at Skärblacka. All the fibre amount index measurements were done at Skärblacka. Tests regarding seal strength that were done at Skärblacka were to see how their results compare with the sample preparing and testing equipment at TUT.

5.1 Objectives of the research

One of the objectives in this research was to learn more about the relationship of the heat sealed materials and what is happening during the sealing process. It was wanted to learn how different sealing conditions affect seal strength and fibre amount indexes. That is why a more comprehensive matrix-study was done where the effect of sealing time, temperature and pressure were studied.

Also one interest was to find good ways to measure peel characteristics and evaluate the current method. Methods that had not earlier been used for studying the sealed materials were tested to find out if they would provide valuable information. Profilometer was tested to study the surface of the paper and polymer film parts of the seals. In the angle method the load-time curve for seal strength was obtained in a different way than in the ASTM F88-00 Standard Method for Seal Strength of Flexible Barrier Materials. Its purpose was to provide information about the edge of the seal and the failure mode of the seal.

All in all this study is just a part of long term research for getting better understanding of heat sealing process and developing better ways to study and measure parameters and factors connected to it. And so this work's goal is not to solve everything but rather help taking steps forward in the process.

5.2 Used Materials

There were two different types of papers in this study which were called Kraft paper A and Kraft paper B. Kraft paper A was chosen because it does not have optimal peeling characteristics, whereas Kraft paper B's peel characteristics are better.

Also two different multi-layer polymer films were used and these were called Film A and Film B. Kraft paper A was always tested with Film A and Kraft paper B with Film B. For Film A the structure of the film wasn't known exactly other than the sealing layer was known to be PE and the outer layer PET.

5.3 Equipment and test methods

The most important devices used in this study were the heat sealing equipment and the instrument for testing seal strength. Other methods for studying heat sealed materials were profilometer and an optical microscope. In addition, the characteristics of the polymer films were studied with optical contact angle and surface tension meter, Fourier transform infrared spectroscopy and differential scanning calorimeter (DSC).

5.3.1 Heat sealing equipment

In this work the heat sealing at TUT was done with KOPP SPGE 20 (Figure 12). In it the upper sealing bar was smooth 10 mm wide and 100 mm long metal bar and polytetrafluoroethylene (PTFE) coated. As a lower sealing bar a bar with a silicone rubber insert was used. Samples were sealed so that the polymer film was against the silicon rubber insert and the paper to the metal sealing bar.

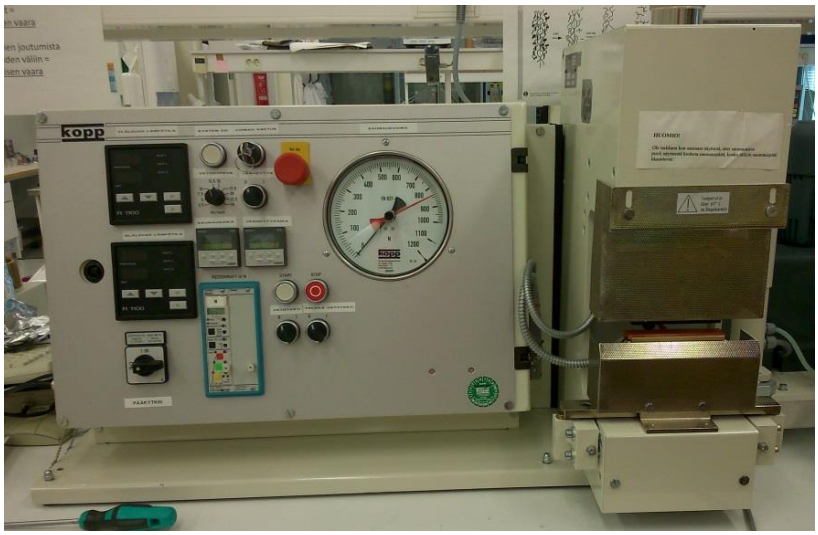

Figure 12. *Heat bar sealing equipment at TUT.*

Temperature range of the equipment is from 0 °C to 300 °C but in practise the minimum temperature is the room temperature. These sealing bars can be heated separately and in this study the sealing bar with silicone rubber insert wasn't heated. The pressure range of KOPP is 0-1000 N. Also the sealing time can be set. The parameters used in the tests varied depending on the tests and they are described later in more detail. The equipment was set in laboratory where there were no set standard conditions.

Even though the pressure in the equipment is in newtons it is converted into pascals when mentioned in text. One pascal equals 1 newton per square meter: 1 pascal = 1 Pa= 1 N/m^2 [27]. In most of the cases the sealing bar is 10 mm x 100 mm = 1000 mm^2 = 0.001 m^2. So if for example 500 N is changed into pascals it equals:

$$\frac{500\ N}{0.001\ m^2} = 500000\ Pa = 0.5\ MPa.$$

5.3.2 Seal strength measurement

The instrument used for studying seal strength was Hounsfield which is a material testing machine. Seal strength measurements are done according to the ASTM standard F 88-00 Standard Method for Seal Strength of Flexible Barrier Materials. The basic idea of it has been presented earlier. These tests were done in a conditioned room in a temperature of 23 °C and with relative humidity 50 %. Grip separation rate depended on the test and it was either 300 mm/min or 150 mm/min so that the seals peeled open in different angles would have same peel rate. Sample width was usually 25 mm but also 10 mm and 15 mm were used for the angle method tests. For some study only samples cut from the middle of the seal were used but for others also sample strips cut from left and right sides were used. In Figure 13 is shown how the samples are cut. Machine direction of the paper was parallel to the seal. In the equipment the paper part of the sample was clamped down and the polymer film up. Used peel angles were 90° and 180°.

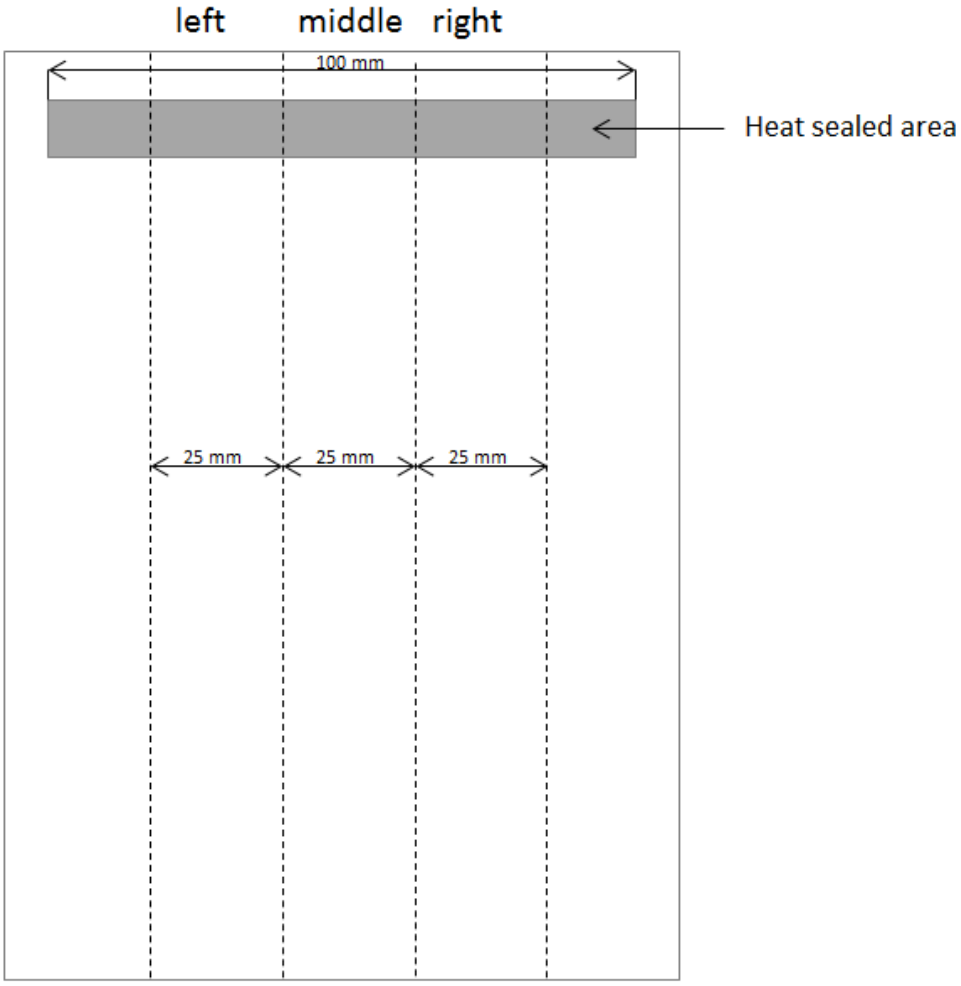

Figure 13. *Schematic image how samples are cut for seal strength measurements.*

What was measured was the average seal strength from a load-time curve. This means that the beginning and the end of the recorded curves have not been taken into calculations. This is because at those points the curve often has peaks in force.

Hounsfield was also used for the angle method measurements. In it the used peel angle was 180° and the samples were supported even if in the description of the method they were originally done unsupportedly. This was to eliminate causes of variations in the test. For these tests the paper was first cut in 45 degree angle and after that the polymer film was sealed along this edge of the paper. This was done so that the paper's machine direction would be the same when peeling the samples open as in the other peel tests.

5.3.3 Profilometer

The surface texture of the paper and plastic films were studied with Alicona InfiniteFocus G5 (Figure 14). It is non-contact, optical profilometer and its measurements are based on Focus-Variation. Two different kinds of measurements were done where in the first one

only the surfaces of paper parts were studied. In the second one also the polymer film parts of the samples were scanned.

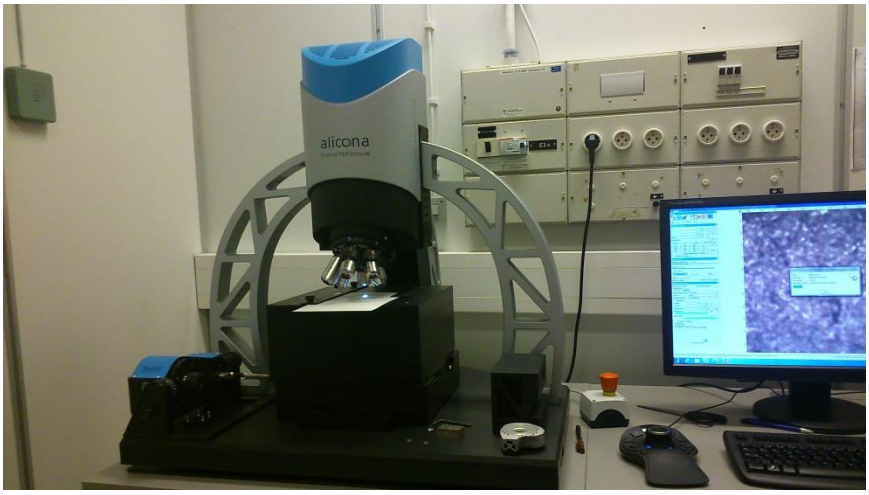

Figure 14. *Alicona InfiniteFocuse G5 which was used for the surface roughness measurements.*

The samples in both studies were from Kraft paper A which had been heat sealed with Film A with varying parameters. The polymer film had been peeled of the paper with either grip separation rate of 300 mm/min or 500 mm/min with an angle of 90 or 180 degrees. The actual peel rate of the seal is half of the grip separation rate for the samples peeled in 180 degree. In the case of 90 degree peel angle the grip separation rate is also the actual peel rate of the seal. That is why when a sample has been torn in 180 degree angle with a speed of 300 mm/min the corresponding 90 degree sample has been torn with grip separation rate of 150 mm/min. That way the results from different peel angles can be compared with each other.

The measurements for the papers were done with 5x objective. One scanned area was 2.81 mm x 2.81 mm by size from which the calculations for average height of selected area were done. This average height of selected area portraits the surface roughness so that the bigger the value is the more the surface has variations in it, hence the bigger it is the rougher it is. The conditions for the samples concerning the study for only paper samples for the sealing (sealing temperature, sealing pressure and sealing time) and tearing (peel rate and peel angle) of the seals were the following:

- Sample 1: 180 °C, 0.7 MPa, 3.0 s, 300 mm/min, 90°
- Sample 2: 180 °C, 0.7 MPa, 3.0 s, 500 mm/min, 90°
- Sample 3: 160 °C, 0.5 MPa, 1.5 s, 300 mm/min, 180°
- Sample 4: 160 °C, 0.5 MPa, 1.5 s, 300 mm/min, 90°
- Sample 5: 160 °C, 0.5 MPa, 1.5 s, 500 mm/min, 180°.

In addition, unsealed paper was measured as a reference.

From each sample three different seals were studied and from each seal four to six 2.81 mm x 2.81 mm areas were scanned. The difference in the number of scans was to limit the work amount because at first it wasn't sure if this research method would give any interesting information. Before the program calculated the average surface height some editing had to be done for the images and this included such as filling possible holes if the profilometer did not see every spot of the sample from some reason and doing plane correction. The calculation program of the profilometer implemented standards ASME B46.1-2002; Assessment Surface Topography (Blunt/Jiang 2003); Characterisation of Roughness (Stout 2000); ISO 25178 Areal –Part 2; ISO 1278-1:2003.

The second part of profilometer study included also polymer samples in addition to paper samples. The main interest was to find out if and how the optical profilometer can see the transparent polymer samples. The study was conducted so that the areas to be studied with profilometer were marked on the paper and polymer film by drawing a square of about 4 mm x 4 mm. Before the samples were heat sealed the areas were measured for average height of selected area. Heat sealing of the samples was done so that the squares on paper and polymer were placed on top of each other. After sealing and peeling open the samples the same areas that were studied before were scanned again with profilometer to see the change.

Only two different conditions (sealing temperature, sealing pressure, sealing time, peel rate and peel angle) for preparing the samples were studied and they were the following:

- 180 °C, 0.7 MPa, 3.0 s, 300 mm/min, 90°
- 160 °C, 0.5 MPa, 1.5 s, 300 mm/min, 90°.

From both of them three different seals were examined and from each seal five 2.81 mm x 2.81 mm areas were scanned.

5.3.4 Fibre amount index measurements

Fibre amount index measurements were done by BillerudKorsnäs Ab at Skärblacka. For these measurements sealed samples were torn along the seal along the machine direction of the paper. The seals of these samples were torn open in two different ways: manually and with a materials testing machine. Manually done tearing was done at Skärblacka. Tearing has always been started from the left side of the sample.

The instrument used to tear samples open for fibre amount index tests was mechanical testing machine Instron 8800 which is servohydraulic. It was located in non-conditioned room. The used grip separation rate was 3 000 mm/min which equals 50 mm/s. Hence, the actual peel rate was 25 mm/s (1500 mm/min) which is notably higher than the peel rate that was used with Hounsfield. This difference in the peel rate was why Instron was used because Hounsfield could not reach as high peel rate. The idea was to simulate the

manual way of tearing the samples open. It was evaluated at Skärblacka that the manual tearing speed is about 15 000 mm/min which equals 250 mm/s. This 250 mm/s tearing speed was not possible to reach with available instruments. Why an instrument was used for tearing the samples open instead of doing it manually was to find out if the standard deviation of fibre amount index would be smaller when compared to manually torn samples. It was thought that by using an instrument there are fewer variables than in the manual tearing.

With Instron the polymer film was attached to the grip down and the paper up. This is contrary to the attachment in Hounsfield. The reason for this was that the lower piston was the moving grip in Instron whereas with Hounsfield it is the upper one. The total distance between the grips of Instron was about 13 cm before starting the peeling.

The evaluation of fibre amount index was done by scanning the area of the samples and then evaluating the fibre material from it. This scanning is done always separately for left and right sides of the samples. The scanned fibre material is separated into small and large fibre fragments from which the large fragment surface areas are used to determine fibre amount index.

5.3.5 Other research methods

The optical contact angle and surface tension meter that was used in this study was KSV CAM 200. A droplet of liquid was dropped on the studied surface with the instrument. After that an image was taken from the droplet and from it the contact angle was possible to determine. Two liquids for measuring the contact angle were used and those were water and ethylene glycol. With them the surface energy of the polymer film was calculated with Wu's method.

Optical microscope Axioskop 40 was used for determining the thicknesses of different polymer layers in the polymer film. Also the cross sections of the seals were studied with it to see how they looked like.

For differential scanning calorimetry (DSC) measurements Netzsch DSC 204 F1 was used. These measurements were done for both polymer films that were used in tests. These measurements were done to find out the melting points of different materials in the films and also to confirm and find out from what polymers they consist of. Two heating and cooling cycles were done to remove the processing history of the films first.

Fourier transform infrared spectroscopy (FT-IR) measurements were done with Bruker Optics Tensor 27. These measurements were also done to identify and confirm the materials of the polymer films. Both surfaces of the films, sealing surface and outside surface, were run with the instrument as well as the spectrum through the whole film.

6. RESULTS AND ANALYSIS

In this chapter are presented the results from the tests and measurements done for this study.

6.1 Material characterization of Film A

Because the composition of Film A multilayer plastic film was unknown some analyses were done for it. It was thought that knowing the characteristics of this film helps with understanding its behaviour during the heat sealing.

6.1.1 Film A's polymer layers

In Figure 15 is a typical cross section image taken from the Film A. It consists of six layers which are different polymer and adhering layers. In the image the bottom layer is PE layer that is against the paper when heat sealing and the top layer is PET. These were confirmed by doing FT-IR. FT-IR spectrums are presented in Appendix 1. In Table 2 are results for calculated average layer thicknesses that are determined from five different cross section images.

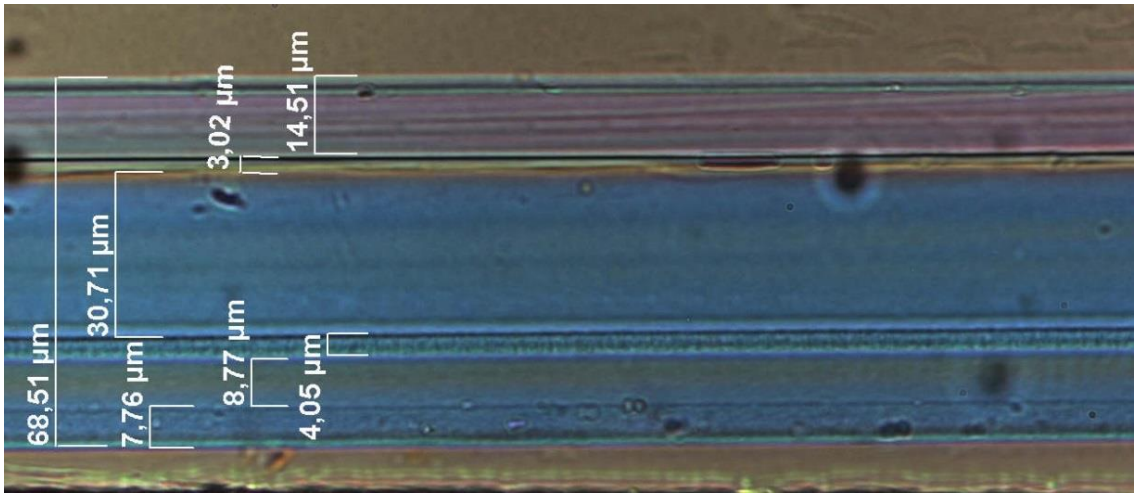

Figure 15. *Cross section image of Film A. The bottom layer is the sealing layer.*

Based on the appearance of the film layers and their low thicknesses second and fourth layers are adhering layers for attaching the different polymers together.

Table 2. *Average layer thicknesses of the Film A.*

Layer	Thickness [µm]
1st: top layer PET	13.5
2nd:	2.3
3rd	30.6
4th:	3.9
5th	9.4
6th: sealing layer, PE	7.0
Total thickness	66.4

DSC curve for the film is presented in Figure 16. On it there are only two peaks, though the first peak is wide and has three separate peaks on it at temperatures 109 °C, 118 °C and 124 °C. Since the only other peak on the curve is at 248 °C, which is the PET surface layer, this film probably has several different PE layers [12, p. 386]. Based on the melting temperatures they are most likely to be LLDPE and LDPE layers but there is no telling which layer is which in the structure [12, p.152-179]. The thinner adhering layers of the film are likely to be some polyethylene based so they also contribute to this first peak on the curve.

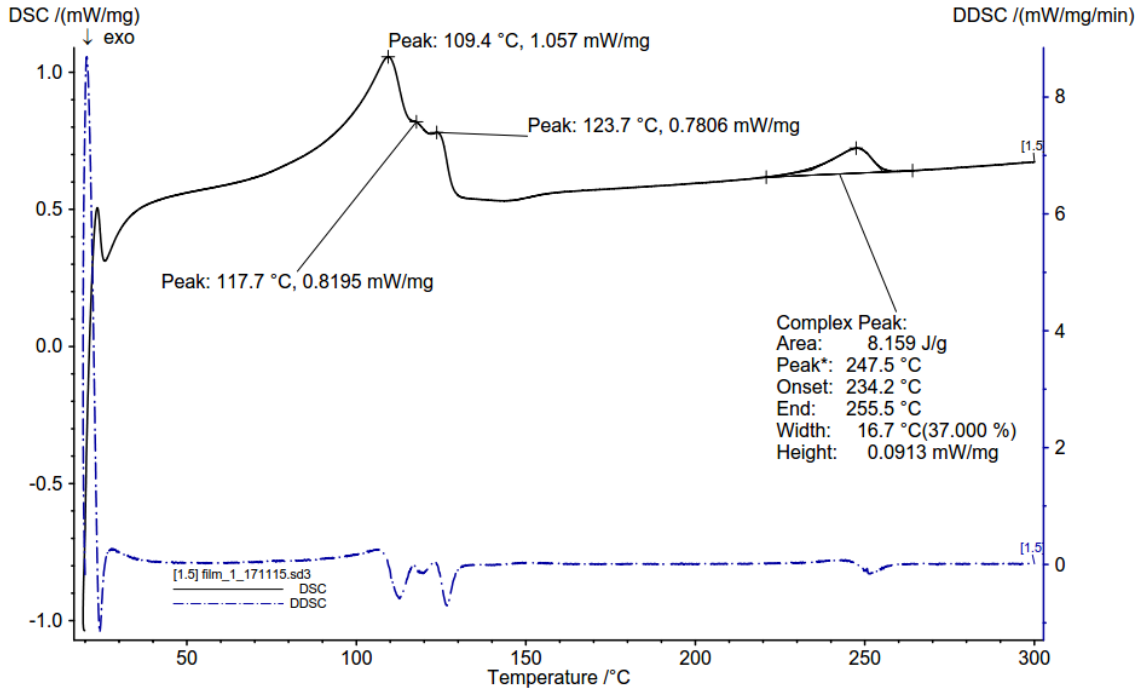

Figure 16. *DSC curve for Film A.*

6.1.2 Surface energy

Surface energies of Film A were studied from films of three different ages. One of them was named old which means that it is from a lot that isn't in use anymore in the production. The second sample was taken from a film that is used now and the last one was from film that hadn't been taken into use yet. The surface energies were measured in normal conditions (23 °C, 50 RH). Two different liquids' contact angles were measured. Those liquids were water and ethylene glycol. Why two different liquids were used is because they have differing surface energies so based on the results the surface energies can be calculated with Wu's method. The results are gathered in Table 3 where also the water contact angles are shown.

Table 3. *Surface energies of different aged Film A's.*

Film	Average contact angle of water, left [°]	Average contact angle of water, right [°]	Surface energy [mN/m]
Date: 13.08.01 – Old	102.4	102.4	19.9
Date: 14.11.03 – Used Now	102.4	102.5	20.3
Date: 14.12.14 – New (Not opened before)	105.4	105.5	18.4

It can be seen form the results that the surface energy of the non-opened film differs the most from the other ones and old and now used films' energies are closer to one another. These differences are very small and can be just a result from the measuring practise. For the further studies regarding Film A "date: 14.12.14 – New (Not opened before)" has been used. Also this new film's surface energy was measured after receiving it at TUT. These results are shown in Table 4. The first surface energy measurement was done right after opening the package.

Table 4. *Tracking the surface energy of the Film A, Date: 14.12.14 – New (Not opened before).*

Date of the measurement	Average contact angle of water, left [°]	Average contact angle of water, left [°]	Surface energy [mN/m]
1st July 2015	106.0	105.9	18.7
	103.7	103.5	18.3
9th July 2015	103.0	103.5	18.7
23rd July 2015	102.4	102.4	18.9
	103.2	103.2	18.3

Because the different aged film's surface energies are so close to each other it can be reasoned that as time passes the films' surface properties do not change. In Figure 17 are presented different surface energies for a polyethylene with different kind of surface treatments. Because the surface energy of the film used in this study is lower than any of the ones with treatment in the figure it can be concluded that it hasn't very likely been treated.

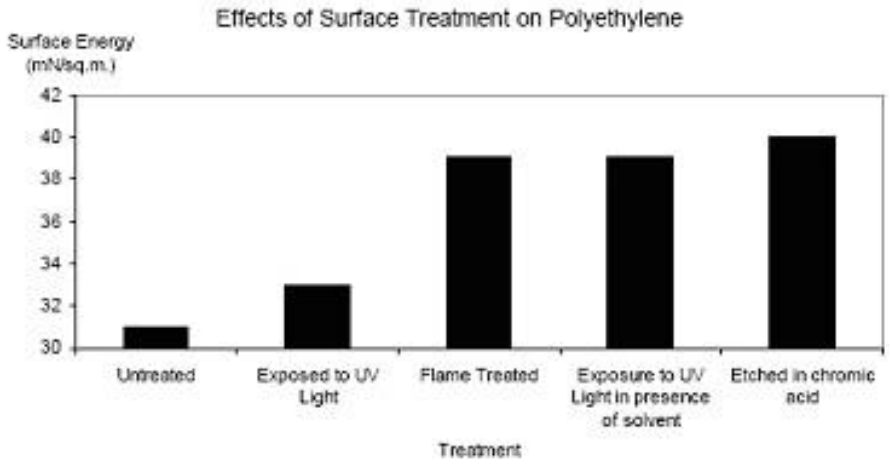

Figure 17. *Effects of surface treatments on polyethylene's surface energy.[28].*

It isn't expected that the surface energies of the films change as the time goes by since the differences between old, now used and not opened before films were so small. Also there were not really any changes when tracking the water contact angles and surface energies of the new film (Table 4). That is why the surface energies were not tracked for a longer time period.

6.2 Material characterization of Film B

The structure of Film B was better known beforehand than the structure of Film A. Still its cross section images were studied to confirm its composition and also DSC and FT-IR were run for it. The water contact angles were measured to find out if some surface treatments had been done for the film.

In Figure 18 is presented a typical cross section image of the film. Otherwise there seemed to be the layers corresponding to the given structure but the adhering layer between LLDPE (sealing layer) and PA layer was not seen. Sometimes adhering layers are difficult to see, so it might still be there even if it does not show in the image.

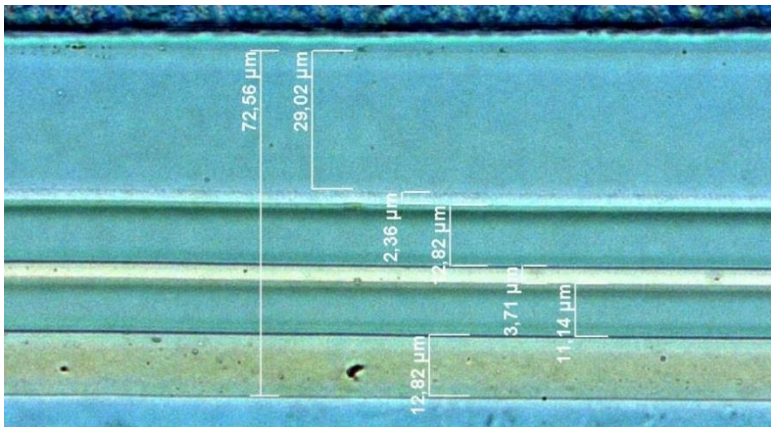

Figure 18. *Cross section image of Film B. The lowest layer is the sealing layer.*

The average layer thicknesses are presented in Table 5. They are calculated based on measurements from seven different images. It seemed that the total thickness of the film was always higher than the 70 µm. As expected, adhering layers were much thinner than polymer layers.

Table 5. *The average layer thicknesses of Film B.*

Layer	**Thickness [µm]**
1st : PP	29.6
2nd : adhering layer	2.9
3rd: PA	12.7
4th: adhering layer	3.7
5th: PA	11.0
6th: LLDPE	13.3
Total thickness	73.8

FT-IR measurement confirmed the sealing layer to be PE and the top layer to be PP. FT-IR spectrums are presented in Appendix 1. DSC curve for the film is shown in Figure 19. On it there are three higher and clearer peaks to be seen. The first one is at temperature range from 109 °C to 124 °C. It indicates the melting temperature for polyethylene [12, p.152-179]. What makes this peak wide is that the adhering layers are polyethylene based and thus contributing to this peak. The second peak is at 155-170 °C and this is the peak for polypropylene [12, p. 480]. This correlates well with the fact that 180 °C for 3.0 s seemed to be too harsh conditions for heat sealing this film because then the outer surface of the film started to melt which is not desirable regarding finished package. The last peak is at 207-224 °C and it indicates the melting temperature of polyamide [12, p. 210-226].

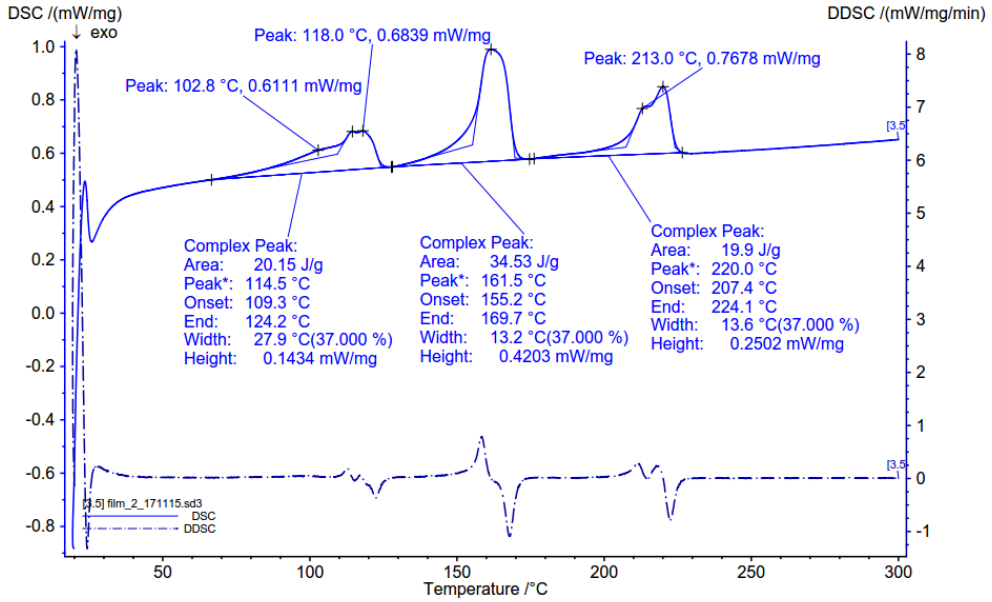

Figure 19. *DSC curve for Film B.*

The water contact angles for Film B were measured and they are gathered in Table 6. They are really close to the water contact angles of Film A (Table 4) with maximum difference of about 6 degrees. In the case of Film B it is know that the sealing layer is LLDPE. The water contact angle for LLDPE without any surface treatments at 20 °C is 99.1 degrees [12, p. 179]. Maximum difference between this theoretical contact angle value and the measured values for the film are about 2 degrees, meaning those values are very close to each other. It can be concluded that this film has not been surface treated either.

Table 6. *Water contact angle of Film B.*

Sample	Average contact angle of water, left [°]	Average contact angle of water, right [°]
1	100.1	99.8
2	101.0	100.8

6.3 Comparison tests between TUT and Skärblacka

For finding out how comparable the results obtained at TUT are with the results gotten at Skärblacka some comparison tests were done. The objective was to find out if the results differ and if they do, how much and is the factor responsible for the difference the heat sealing instrument or the equipment for measuring seal strength. Kraft paper A and Film A were used in this study.

Three different sealing parameter combinations were used for the measurements which were the following:

- 180 °C, 0.7 MPa, 3.0 s
- 160 °C, 0.7 MPa, 3.0 s
- 160 °C, 0.5 MPa, 1.5 s

The samples were torn with grip separation rate 300 mm/min which means 150 mm/min peel rate. Peel angle was 180°. Both at TUT and Skärblacka the samples were sealed and then 5 samples of each parameter combinations were sent from TUT to Skärblacka and vice versa. In addition, samples using the same parameters were prepared for fibre amount index tests. Those tests were all done at Skärblacka.

At TUT measurements were done only from middle of the seal but at Skärblacka also measurements from right and left side of the seals were done. From these test some of the results measured at TUT of samples that were also sealed at TUT with parameters 160 °C, 0.5 MPa, 1.5 s and 160 °C, 0.7 MPa, 3 s had to be rejected because polymer film parts of the samples were sliding in the grip of the measuring device. In both of these cases there were three samples instead of five to be used in the calculations for seal strength. For the rest of the samples the equipment was fixed so that it didn't happen anymore.

In Figure 20 is drawn a diagram of the seal strength measurements done at TUT and Skärblacka. What is really clear from it is that the seal strength measured with Skärblacka's instrument are significantly higher than the ones done with TUT's equipment. The percentage differences of seal strength are calculated and gathered in Table 7.

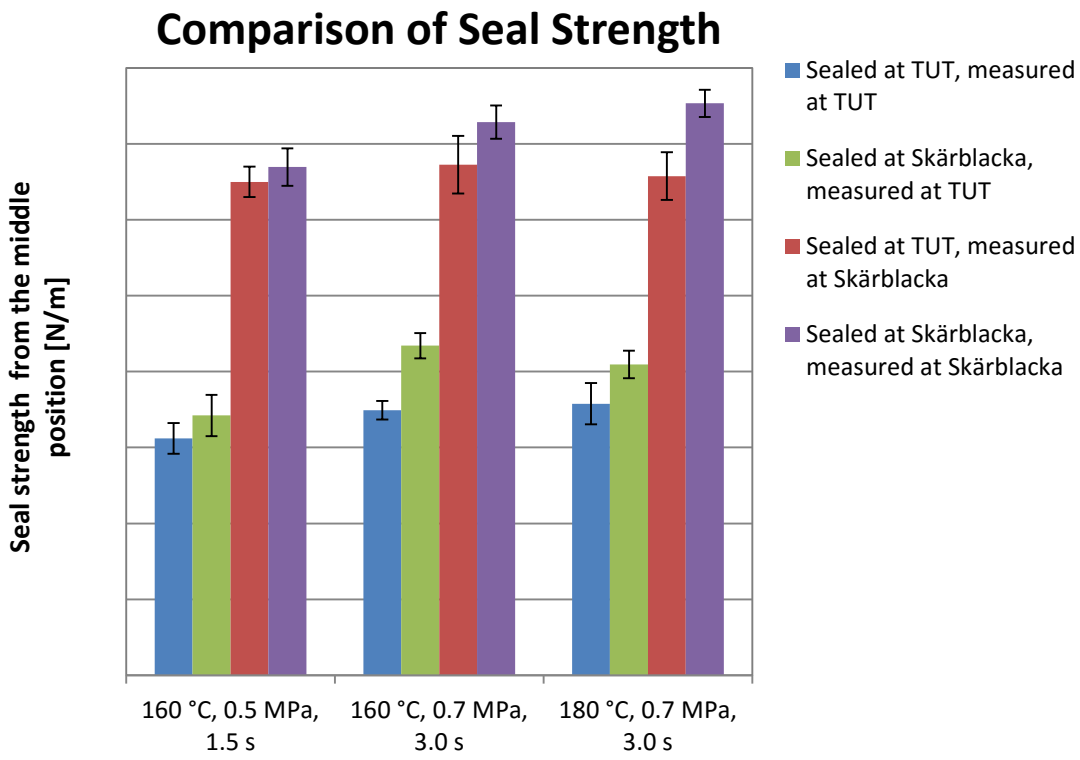

Figure 20. *Seal strength measurements done at TUT for the comparison between TUT and Skärblacka.*

Samples that were sealed at different places but their seal strength measured at same place do not give big difference in seal strength. Though it has to be pointed out that Skärblacka sealing equipment always gave stronger seals when compared samples that were measured at the same place but prepared at different places. Based on this a conclusion can be drawn that the sealing equipment at TUT and Skärblacka give quite similar seals and the biggest differences in the results aren't due to sealing equipment but due to the seal strength measurements.

Table 7. *Difference in seal strength between TUT and Skärblacka presented as how many per cents TUT's results are from the ones obtained at Skärblacka.*

Sealing place and sealing parameters	TUT's results as % from Skärblacka's results
TUT, 160 °C, 0.5 MPa, 1.5 s	48.0
TUT, 160 °C, 0.7 MPa, 3.0 s	51.9
TUT, 180 °C, 0.7 MPa, 3.0 s	54.4
Skärblacka, 160 °C, 0.5 MPa, 1.5 s	51.1
Skärblacka, 160 °C, 0.7 MPa, 3.0 s	59.6
Skärblacka, 180 °C, 0.7 MPa, 3.0 s	54.3

Since at Skärblacka also the different positions (left, middle and right) of the seals were measured some other observations were made too. Standard deviations of the seal strength results were higher at the middle position for samples sealed at TUT and the samples prepared at Skärblacka have higher standard deviations at the sides.

These seal strength measurements done at TUT from samples prepared at TUT can be compared with later results from matrix-study which was done at TUT using same materials. From Figures 29 and 30 can be noted that on those later measurements notably higher seal strengths have been obtained for samples that have been heat sealed with same parameters. There is maximum of 80 % raise and minimum of 40 % raise on seal strength when these results from Figure 20 are compared to matrix-study's results. All the figures having seal strength as a function of temperature are comparable with each other because same scale has been used in them. One thing that was corrected to the later tests was that a support was made for the heat sealing equipment so that the seals would be straighter and more securely parallel to the machine direction of the paper. One thing that could have had some effect on the difference between TUT's and Skärblacka's results are operator's skills.

As mentioned earlier all the fibre amount index samples were prepared at Skärblacka and the results from them are presented in Figure 21. Fibre amount index of the samples sealed at Skärblacka gave higher results than the ones sealed at TUT. This correlated with their higher seal strength and was to be expected.

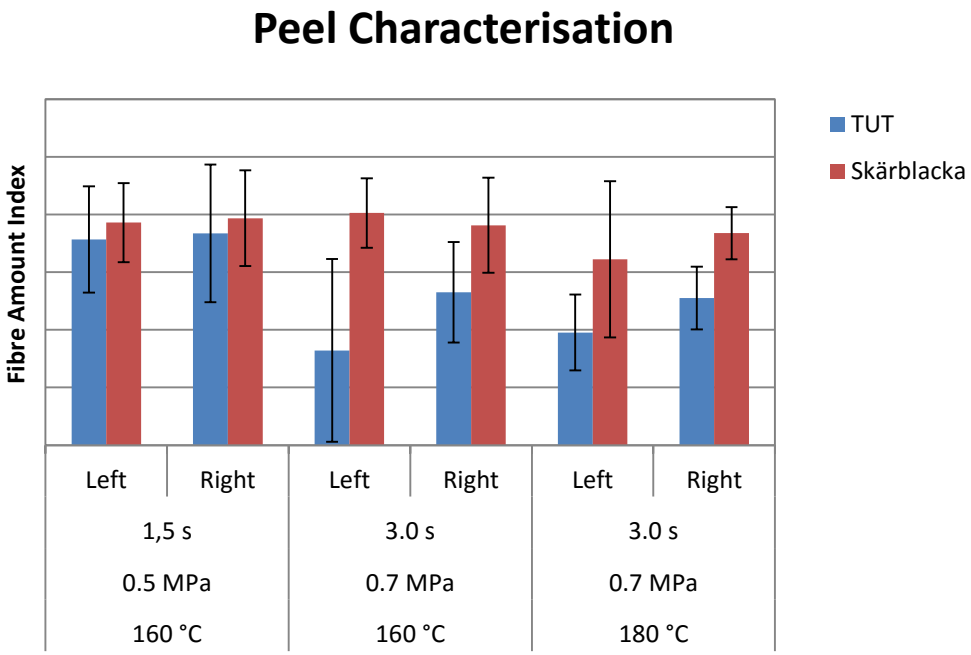

Figure 21. *Fibre amount indexes and their standard deviations.*

Fibre amount index is bigger for the right side in 5 cases out six, though, in one of them the results are almost equal. From all the standard deviation for fibre amount index (TUT and Skärblacka) 67 % of the cases it is bigger for the right side. In general the standard deviations for samples prepared at TUT had bigger standard deviations. What could create difference to fibre amount index on different sides (left and right) is that the samples are torn from left to right.

6.4 Study with Kraft paper A and Film A

The purpose of this study was to determine how peel angle affects to seal strength before starting more comprehensive matrix study with these same materials which were Kraft paper A and Film A. In addition cross section images from the seals were studied with optical microscope and fibre amount indexes were measured. This study was done with four different sealing parameter combinations. These parameters were the following:

- 130 °C, 0.5 MPa, 1.0 s
- 160 °C, 0.5 MPa, 1.5 s
- 160 °C, 0.5 MPa, 0.5 s
- 180 °C, 0.7 MPa, 3.0 s

Because three seconds seemed quite long sealing time it was wanted to find out what shorter time would give as a result. Also 180 °C and even 160 °C seemed quite high sealing temperature for PE so lower temperature was tested as well. The first plan was to use 120 °C for one of the samples instead of 130 °C. This was changed because the samples sealed with 120 °C opened on their own after a little moment from the sealing probably due to the polymer film's tendency to roll. Four parallel samples were measured. Results for seal strength are presented in Figure 22.

With the samples prepared with parameters 160 °C, 0.5 MPa, 1.5 s and 160 °C, 0.5 MPa, 0.5 s the only variable is time. From Figure 22 can be seen that they have difference in seal strength in the middle position for 180° peel angle so that the seal strength for 1.5 s is 2.8 times the seal strength for 0.5 s samples. Based on this sealing time is important variable in heat sealing because it affects seal strength notably. Then again if we compare the samples sealed with 160 °C, 0.5 MPa, 1.5 s and 180 °C, 0.7 MPa, 3.0 s with both peel angles the differences in seal strength are small even though the other samples have higher sealing temperature, sealing pressure and longer sealing time. This could suggest that above some certain sealing pressure and sealing time there is no significant change in seal strength anymore by increasing them.

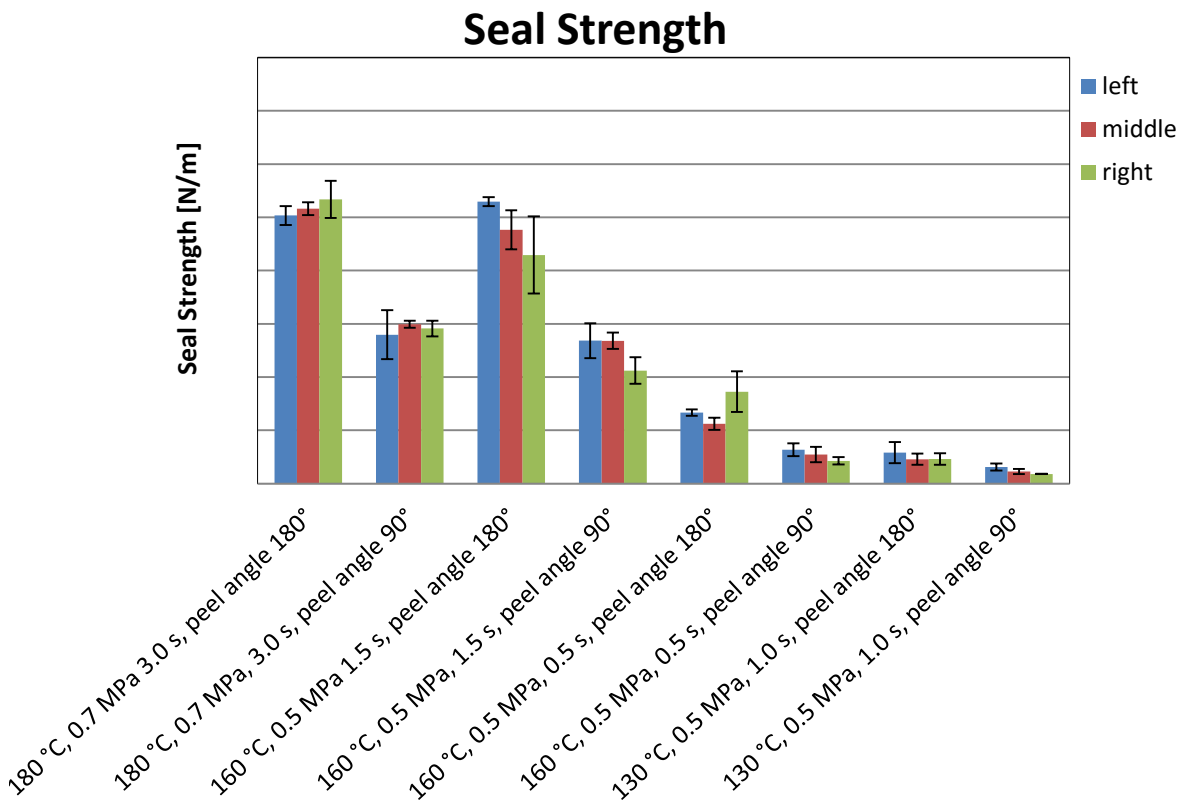

Figure 22. *Seal strength results for Kraft paper A and Film A.*

From Figure 22 can be seen that the sealing strength is higher for the samples peeled with 180° angle than with 90° angle. In Table 8 has been calculated how many per cents the seal strengths obtained with 90° peel angle are from the ones obtained with 180° peel angle. Most of these values are in the range of 40 to 60 % with only 3 exceptions. These results do not really follow the equation (1) since the result should be the opposite: peel test done in 90 degree angle should give twice the seal strength of the 180 degree peel test.

Table 8. *The percentages how much is the peel strength of the sample peeled in 90° peel angle from the one peeled in 180° angle.*

Sample	Left [%]	Middle [%]	Right [%]
130 °C, 0.5 MPa, 1.0 s	60.5	50.0	39.1
160 °C, 0.5 MPa, 0.5 s	47.8	48.9	24.8
160 °C, 0.5 MPa, 1.5	50.7	56.2	49.5
180 °C, 0.7 MPa, 3.0 s	55.6	58.0	54.6

What else can be noted from table 8 is that the lowest percentage is always on the right side of the seal while the highest is in the middle except in the case of samples sealed in 130 °C. Samples sealed in 130 °C seem to give most varying results in this comparison because all of the positions (left, middle and right) give such a varying result from each other.

Because these tests are measured with an instrument having many things done manually there are some factors that can cause deviation to the results such as are the samples set exactly the same way in the grips. Furthermore, with the 90 degree peel test the sample is attached to this wheel that rotates around its axis and this wheel is not completely balanced (Figure 23). This means that after attaching the sample on it the wheel is directing little pulling force to the seal. This is especially problematic in the case of samples that have low seal strength because the wheel might start pulling the seal open already before starting the measurement if not being really careful with the setting of the sample. That is why with the sealing condition 130 °C, 0.5 MPa and 1.0 s some of the peel test samples done at 90° peel angle had to be rejected since the seal started to open before starting the test.

Figure 23. *The setting for peel test done in 90° peel angle.*

It is very likely that the temperature and maybe the pressure as well are not even throughout the sealing bar. With these measurements seal strength has been taken from different positions of the samples and these should be given closer consideration now. From Figure 22 can be seen right away that the results are not even between different positions. What can be noted though is that the position from same sample series that is the most abnormal from seal strength value usually also has the highest standard deviation which points out that there can be one measured sample that has had from some reason much higher or lower seal strength than the other samples of the series.

In Figure 4 was shown the shape of seal strength-temperature curve when heat sealing PE. Also in Matrix-study are these curves obtained for these specific materials used in this study for different sealing conditions in Figures 29 and 30. From these images can be seen that first seal strength usually increases quite rapidly after what it reaches plateau and with really high temperatures seal strength starts to decrease. If the sealing bar has unevenness in the temperature it could mean that if for example the right side is hotter than the left side of the sealing bar, the right side could give higher seal strength if the sealing temperature is at the part where seal strength is rising strongly. Then again if the sealing temperature is so high that it is at the area where the sealing strength starts to decrease it could be that then the side of the bar that has higher temperature would give lower seal strength.

Figure 29 shows that the sealing conditions 180 °C, 0.7 MPa and 3.0 s is on an area on the seal strength-temperature curve where seal strength is quite even. In figure 22 seal strengths with these conditions are not completely even. One sample in the results for right side samples peeled in 180 degrees is notably higher than the other ones and if this sample was ignored seal strength would be at the same level with the middle position seal strength. The same situation is with the samples tested at 90 degree peel angle, the left and right positions have both one low results and if they were ignored all the positions would have very same level seal strength. In this case the results would be even like expected.

On the seal strength-temperature curve in Figure 30 sealing condition 160 °C, 0.5 MPa and 1.5 s is at a part of the curve where there is slight increase of seal strength. When considering seal strength results from these sealing conditions for different positions even if some differing samples are taken into consideration it seems that the left side has higher seal strength than the right one. Here it might be possible to think that the sealing bar is hotter from the left side, though then again the increase of seal strength on matrix-curve is so slight that it should not show too much between the different positions.

Sealing conditions 160 °C, 0.5 s MPa, 0.5 s and 130 °C, 0.5 MPa, 1.0 s are not directly comparable to the matrix study. But considering sealing temperatures, times and seal strength it is most likely that they both are on the seal strength-temperature curve at a part where seal strength is on the rise. From Figure 22 can be seen that in most of the cases the left side has higher seal strength than right side. There is just one exception to this, the samples sealed at 160 °C, 0.5 MPa, 0.5 s and peeled open at 180 degree angle. One of the right side samples has notably higher seal strength than the other but even if that one would be ignored the average seal strength would still be higher than for the left side samples. As a conclusion these samples suggest as well that the left side could have higher temperature than the right side.

Even if these results might suggest that the right side of the sealing bar is warmer than the left there are only four parallel samples. This means that based on this it is not possible

to make definite conclusions but these result support the idea that the left side of the sealing bar might be heating more and depending the sealing temperature it affect the seal strength of different sample positions in different ways.

Then again if pressure is also an influential factor here it can make the equation quite complicated. Let's imagine that sample is heat sealed at a temperature that is on the seal strength-temperature curve at the beginning part where the sealing strength is rising rapidly. Here quite small changes in temperature could lead to big differences in seal strength. If for example sealing bar's right side has bit higher temperature than the left side but if at the same time pressure on the right side is lower than from the left side these factors would have opposite impact to seal strength.

In Figure 24 are results from fibre amount index scans for the samples. As can be seen from the graph samples with sealing parameters of 160 °C, 0.5 MPa and 1.5 s have the highest fibre amount index. This is an unexpected result because fibre amount index was expected to correlate with seal strength. Therefore, the highest fibre amount index would have been expected to be given by samples sealed with conditions 180 °C, 0.7 MPa and 3.0 s. What is also quite unexpected is that the difference in fibre amount index for these samples is quite big while the difference in seal strength was not.

The effect of sealing time is clearly seen on the results for fibre amount index as well as it was with sealing strength. The samples sealed at 160 °C have only difference in sealing time. 0.5 s sealing time results in much lower fibre amount index than 1.5 s sealing time does. This means that sealing time has also notable effect on fibre amount index when the change is from 0.5 s to 1.5 s.

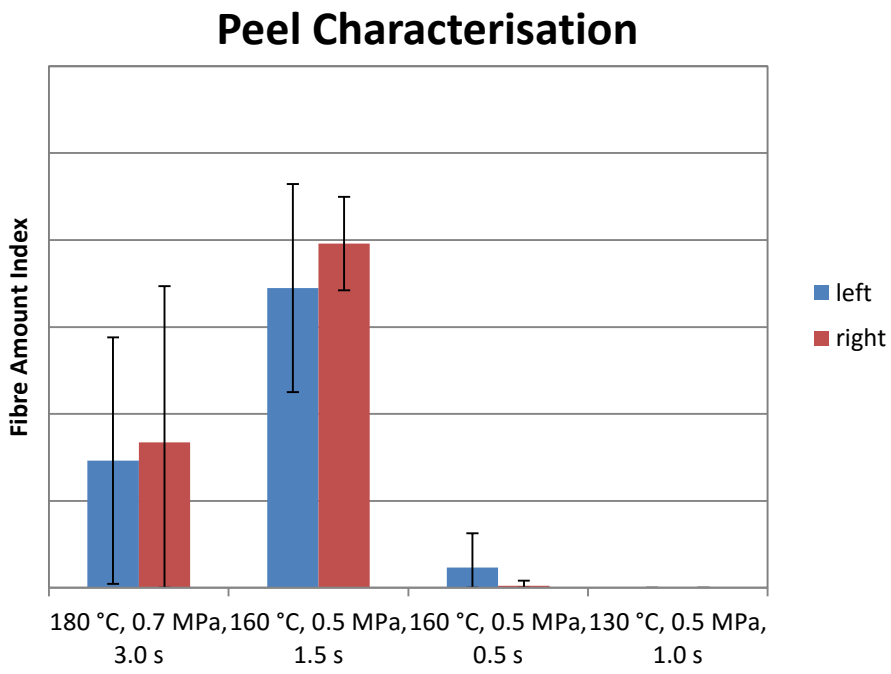

Figure 24. *Results of fibre amount index tests.*

From examining cross section images of the seals prepared with parameters 130 °C, 0.5 MPa and 1.0 s the seal looked good. PE layer of the film was intact from all the places. This is not surprising when compared with the fibre amount index result of 0 for these samples. In these cases the film has not yet melted completely and penetrated to the paper.

With the parameters 160 °C, 0.5 MPa, 0.5 s the PE layer was from most of the part uniform but it had some parts were it looked as if the PE layer was pressed or had spread into the paper and the adhering layer, that is closer to the sealing surface, looks as if it has little bending on it (Figure 25). This could be due to a pore on the paper. There are some parts where there are some particles in the film so that it is surrounded by PE. These particles look as if they could be transvers fibres on the paper (Figure 26). The polymer film has formed well around the particle which means that PE of the film seems to shape well along the paper surface.

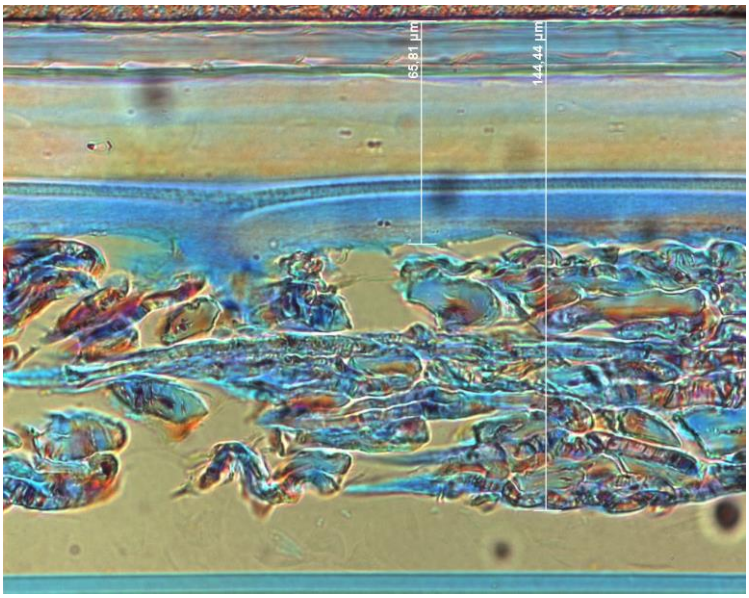

Figure 25. *Crossection image from a sample sealed at 160 °C with pressure of 0.5 MPa and with a sealing time of 0.5 s. The seal has a part where the PE layer isn't uniform possibly because of a pore in the paper.*

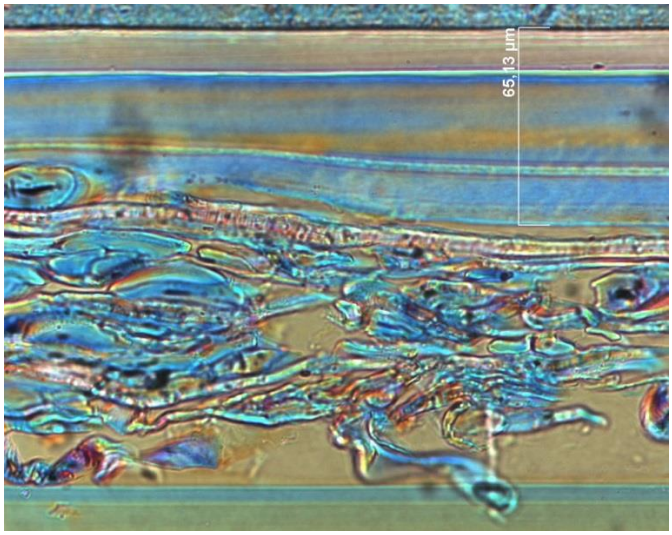

Figure 26. *Cross section image of a sample sealed with parameters 160 °C, 0.5 MPa, 0.5 s having a particle on the paper surface on the very left of the image.*

With sealing parameters 160 °C, 0.5 MPa, 1.5 s the first PE layer is from most of the part identifiable, though there are some parts where the PE layer looked as if it was spreading into the paper (Figure 27). In the cross section images of the seals prepared with 180 °C, 0.7 MPa and 3.0 s the sealing PE layer was always visible and it was also quite even from most of the part. From some parts the thickness of the lowest PE layer was changing a bit but parts where it would have spread into the paper were not noticed. In Figure 28 is shown image taken from a sample sealed with parameters 180 °C, 0.7 MPa and 3.0 s.

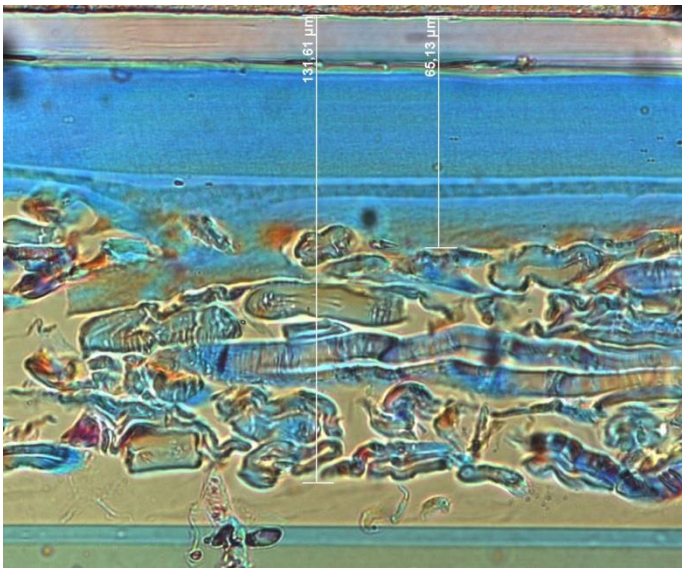

Figure 27. *Seal prepared with parameters 160 °C, 0.5 MPa, 1.5 s.*

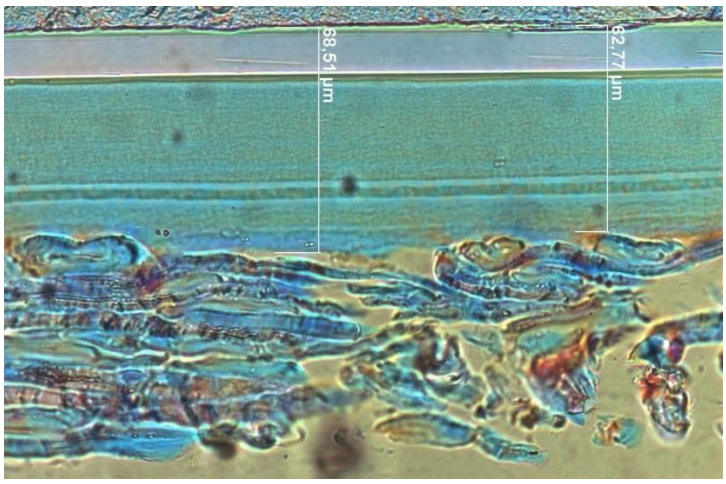

Figure 28 *Seal prepared with parameters 180 °C, 0.7 MPa, 3.0 s.*

The microscope images from samples sealed in 180 °C correspond to the fibre amount index results. These samples' polymer film's PE layer looked more even on the microscope than the samples prepared with parameters 160 °C, 0.5 MPa, 1.5 s. Regarding the microscope images it is logical that the fibre amount index for them is lower, though, it does not explain the behaviour of seal strength.

6.5 Matrix-study

In the matrix-study the purpose was to examine the effects of sealing parameters: sealing temperature, time and pressure. Used materials in this study were Kraft paper A and Film A. The first test series with sealing time 3.0 s and sealing pressure of 0.7 MPa had more temperature points than the other series to get a general idea of the behavior of seal strength. For the later sample series it was thought to be not necessary to do that many measurements.

In Figure 29 are presented the results of variable the sealing time. Sealing pressure was kept constant at 0.7 MPa while sealing times 0.5 s, 1.5 s and 3.0 s were used with varying temperatures. With sealing time of 3.0 s paper and polymer film started to seal together at 120 °C. They also sealed at 120 °C with sealing time of 1.5 s but the loads recorded from most of the measurements were very close to a zero or even below zero with two exceptions from which the other one was more than ten times compared to the lower results. The shapes of the curves made it difficult to evaluate what parts of the curves should have been taken into calculating seal strength so it was not determined for this temperature. One explanation for the big differences in seal strength could be that the sealing bar's temperature has had some fluctuation during sealing and the temperature is on the part of PE's seal strength vs temperature curve where seal strength starts to rise drastically meaning that little changes in temperature can lead to notable changes in seal strength.

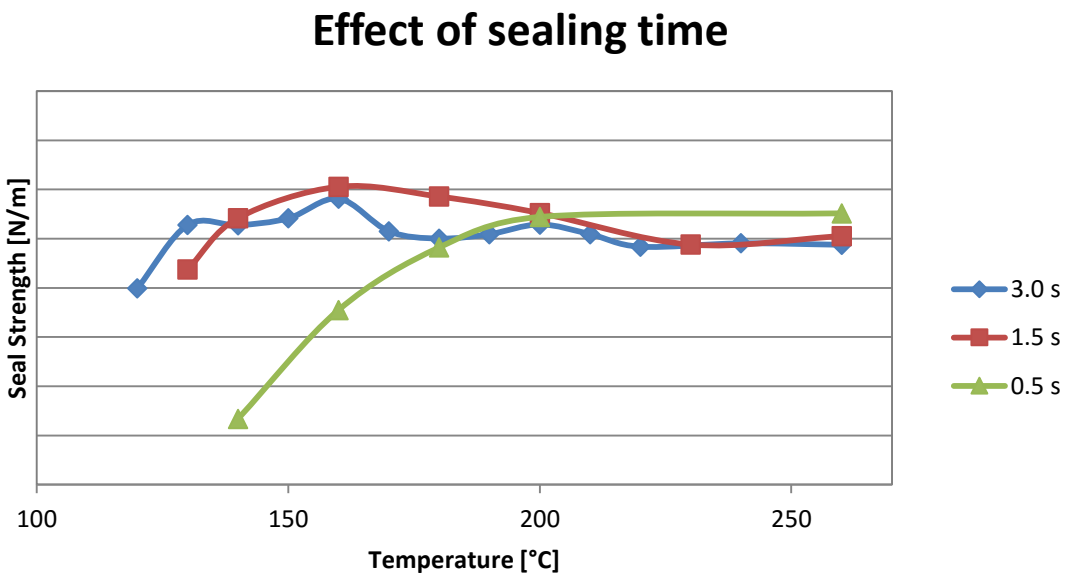

Figure 29. *Effect of sealing time to seal strength. Sealing pressure was 0,7 MPa with every sample.*

With sealing time of 0.5 s there was some sticking between paper and polymer film at 130 °C but it was so weak that the seals opened when only removing the samples form the sealing equipment. This made 140 °C the temperature where the sealing started properly. DSC gave melting temperature for the PE to be in range of 105- 125 °C. If long sealing time is not used, higher temperature than the melting temperature of PE has to be used for heat sealing. What has to be remembered is that what here has been referred as the sealing temperature is actually the temperature of the sealing bar and not the temperature of the melting polymer film in the seal. Because heat sealing here is done so that only the sealing bar in contact with paper is warmed the heat has to conduct through the paper. Logically with longer sealing time sealing starts with lower sealing temperatures

because there is more time for the heat to transfer to the polymer and it will more likely reach the temperature of the sealing bar and melt.

Based on Figure 29 and the information when the materials started to seal with one another the seal strength versus temperature curves with 3.0 s and 1.5 s sealing time are quite steep from the beginning of the curve. The curve of 0.5 s sealing time has much more gradual rise. This indicates that it is too short time for the PE layer to reach the temperature of the sealing bar. Even if the beginnings of the three curves differ from one another their sealing strength reaches same level at 200 °C.

The curves for 1.5 s and 3.0 s sealing time are quite close to each other. At many test points seal strength for the 1.5 s sealing time curve is actually higher than for the 3.0 s one. This could suggest that after the sealing time is long enough for the polymer film to melt and reach the sealing bar's temperature a longer sealing time than that does not increase seal strength anymore. But if sealing time is too short as in the case of 0.5 s curve the polymer film probably doesn't melt properly until at temperatures fairly higher its melting temperature. This idea was suggested in study of Aithani et al. presented in theory.

In Figure 30 is the graph from the tests done with different pressures. For all the tests sealing time was kept at 1.5 s and three different sealing pressures were used which were 0.2 MPa, 0.5 MPa and 0.7 MPa. With 0.7 MPa and 0.5 MPa sealing strength is quite the same but when using 0.2 MPa it is much lower especially with lower temperatures. Though, at higher temperatures the samples sealed with 0.2 MPa sealing pressure reached the level of the samples sealed with higher pressures. Here can be seen well that with high enough pressure there is not much effect on seal strength since the curves for 0.5 MPa and 0.7 MPa are so close to each other.

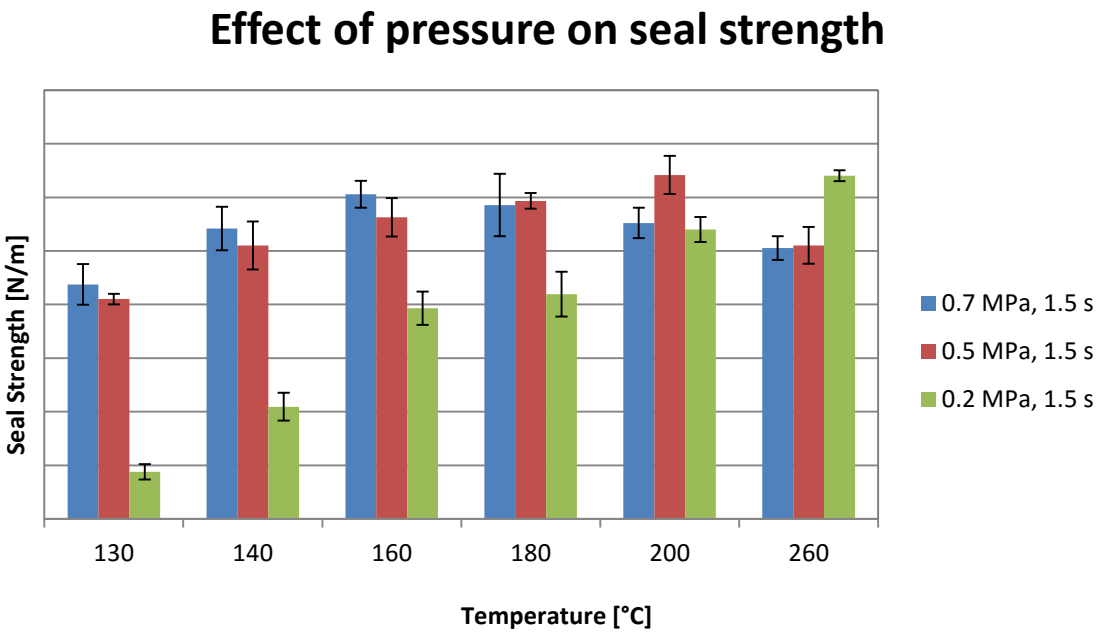

Figure 30. *The effect of sealing pressure on seal strength.*

In Figure 31 is presented a graph regarding standard deviations for seal strength. For the y-axis has been calculated how much in percentage standard deviations are from seal strength. From it can be seen that otherwise they are nearly all under 10 % except at low and high sealing temperatures. It is logical that the standard deviation percentage is wider at lower temperatures because there seal strength is low meaning that even small changes can cause big standard deviation percentages. Since most of the standard deviation percentages are under 10 % they are at an acceptable level. There was not found correlation with standard deviation of seal strength and seal strength levels.

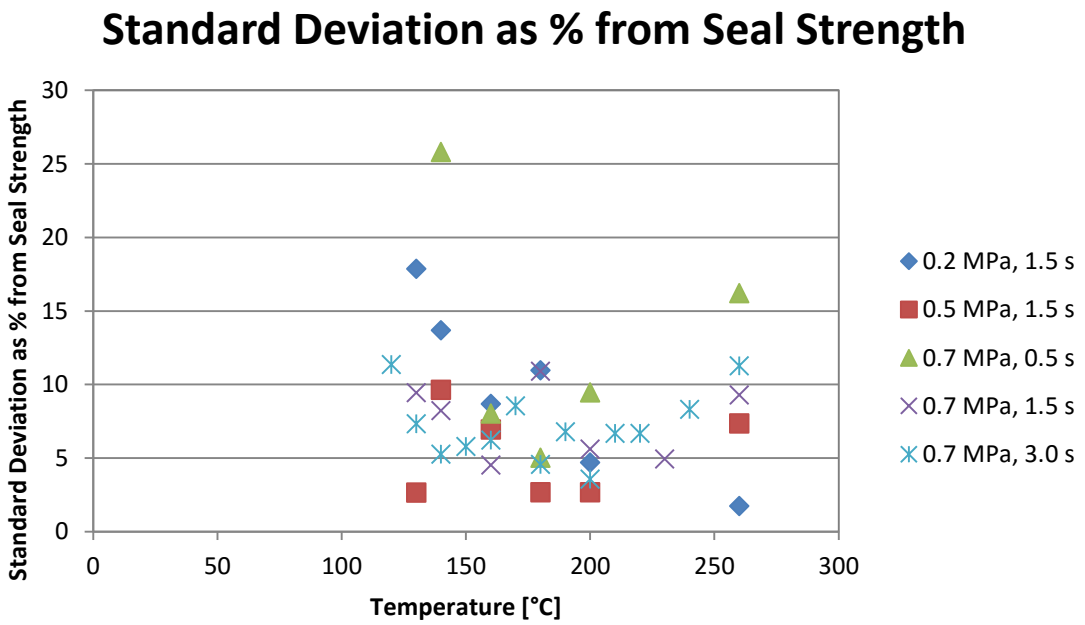

Figure 31. *Standard deviations as percentage from seal strength.*

Fibre amount index was measured for the samples as well. They were done separately for left and right sides and the results are presented in Figures 32 and 33.

When comparing left and right sides of fibre amount indexes it can be noted that the samples sealed with 0.2 MPa and 1.5 s correlate with each other quite well, as well as do the samples sealed with 0.5 MPa and 1.5 s. In their case the beginning of the curves are similar but from the end the left side decreases while the right side increases. Samples sealed with 0.7 MPa and 1.5 s have almost identical fibre amount index at low temperatures but then the curves differ. The right side continues rising after which it starts to descend, whereas, the left side has more fluctuating curve after the beginning's rise.

Correlation is also good for samples sealed at 0.7 MPa and 0.5 s. The increase of fibre amount index for the right side starts with a bit higher temperature than for the right side but at 200 °C right side passes left side and has higher fibre amount index from that on. Samples sealed with 0.7 MPa and 3.0 s have somewhat good correlation between left and right side. Both curves rise higher from the beginning and then go down. From the middle part both of them have fluctuation, though this fluctuation seems to be at different phase because when left side has a peak the right side has a valley and vice versa. From the end of the curves both of them are rising.

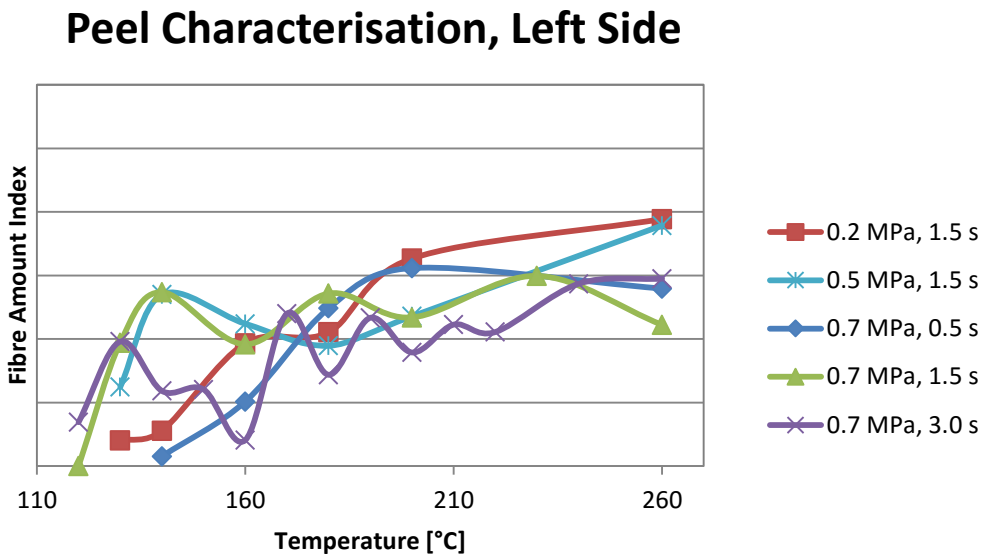

Figure 32. *Fibre amount index results for left side of the samples.*

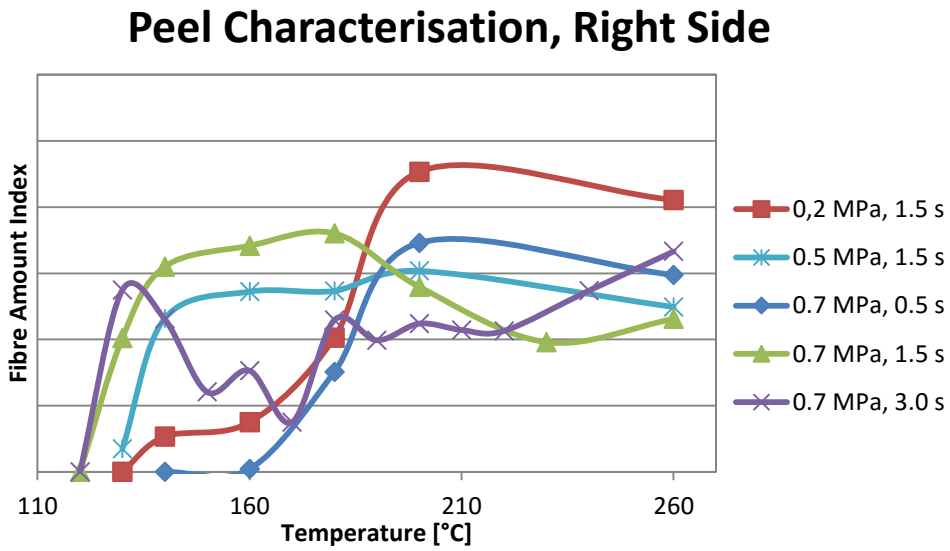

Figure 33. *Fibre amount index results for right side of the samples.*

There was no clear trend in left side giving always higher results for fibre amount index or the other way around. Right side's samples gave slightly more often higher results: 57.9 % of the samples had higher fibre amount index on right side when the mean values of right and left sides were compared. This is still so close to 50 % that it is not significant difference. When calculating the standard deviations for fibre amount index on 52.6 % of the cases the standard deviation is bigger on the right side of the sample than on the left side. Because the samples are always torn from left to right it could have affected to fibre amount index results but it was not evident based on these results. Also if the sealing bar is heating unevenly it could affect fibre amount index too but this effect would be harder

to notice since depending on the temperature the effect can be increasing or decreasing for one side.

Because there was no clear trend in the fibre amount index between left and right side and their correlations were quite good the individual results from both sides are calculated together for average fibre amount index and their curves are drawn in Figure 34 to see how they look when put together. Samples having the sealing conditions of 0.2 MPa, 1.5 s and 0.7 MPa, 0.5 s fibre amount index results correlate the best with seal strength when comparing figures for seal strength (Figure 29 and Figure 30) with fibre amount index curves (Figure 34) and also when considering the highest values of fibre amount index and seal strength.

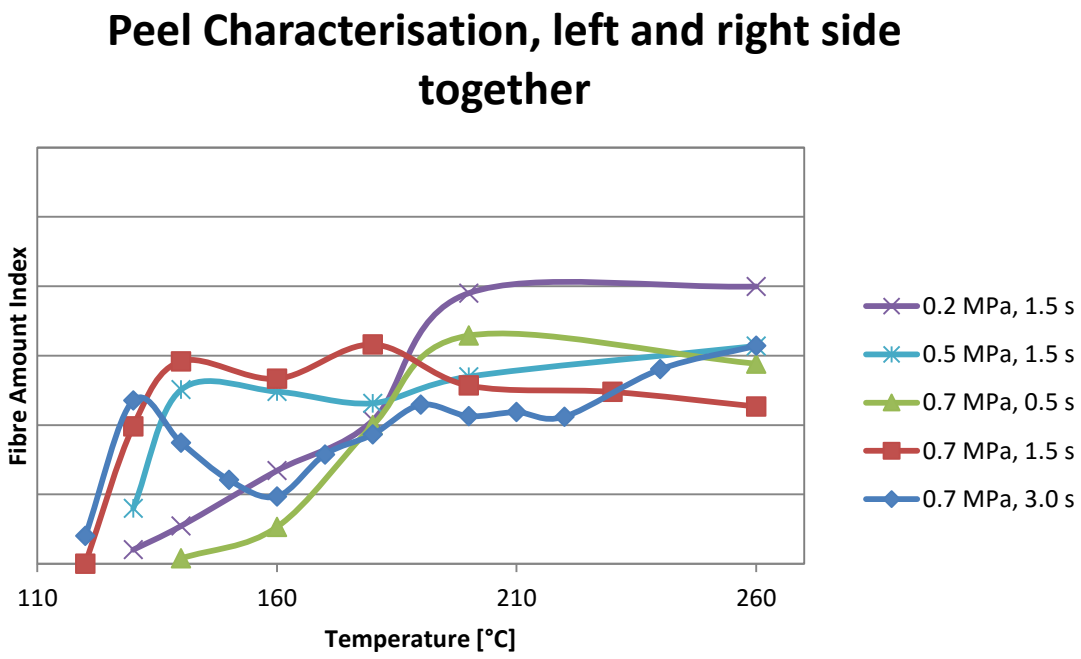

Figure 34. *Fibre amount index results when left and right sides' values are put together.*

Fibre amount index of samples sealed with 0.5 MPa, 1.5 s and 0.7 MPa, 1.5 s correlate with seal strength somewhat well. If comparing the curves for samples sealed with 0.7 MPa and 3.0 s they don't correlate well at all. From temperatures 140 °C to 170 °C seal strength is quite high but fibre amount index curve has a drop there. Because of the results it seems that these sealing parameters would be good since high seal strength but low fibre amount index is favourable combination.

Interesting question here is why the fibre amount index suddenly descends in the case of 0.7 MPa and 3.0 s curve but then rises back up. Also 0.7 MPa and 1.5 s curve has a little drop on fibre amount index at 160 °C. Even on the 0.5 MPa and 1.5 s curve seal strength is very slightly lower at 180 °C than on the adjacent measured temperatures. But since on

these two curves there is only one point where the fibre amount index is lower it could be just some sort of fluctuation and not the same kind of drop as on 0.7 MPa and 3.0 s curve.

When the results for seal strength at 200 °C with varying sealing times (Figure 29 and Figure 30) are studied it can be noticed that they are very close to each other in value except for the samples sealed with 0.5 MPa and 1.5 s. If this varying result is not taken into consideration, other results for seal strength are very close to each other. Then when considering the corresponding values for fibre amount index at 200 °C (Figure 32 and 33) some of the values have rather big difference when considering the scale of fibre amount index. This and the drop on fibre amount index curve of samples sealed with 0.7 MPa and 3.0 s suggest that the value of fibre amount index does not correlate with seal strength only.

An interesting observation is also that the highest fibre amount index values get the samples sealed at 200 °C and 260 °C with 0.2 MPa pressure and sealing time of 1.5 s. Though those samples do have quite high seal strength also, however some other samples have that high seal strength too but not as high fibre amount index.

These observations bring up the thought that pressure affects fibre amount index differently than seal strength. It could be that high pressure somehow presses the paper structure so that it is smoother form the surface and the polymer does not penetrate into it and the fibre amount index is low. Since the samples sealed with 0.2 MPa have the highest fibre amount index from all results at high temperatures it could be thought that the lower pressure doesn't smoothen the paper and the polymer gets to flow freer into it causing higher fibre amount index by mechanical interlocking.

If the curves having different pressure but the same sealing time of 1.5 s are compared it can be noticed that with pressure of 0.2 MPa the fibre amount index is lowest till 180 °C but then with higher temperatures it has the highest fibre amount index. The fibre amount index for curve of 0.7 MPa pressure has the highest fibre amount index till 180 °C but from 200 °C on ward it has the lowest. The curve for 0.5 MPa sets between these other curves and looks to be little bit closer to the 0.7 MPa curve than 0.2 MPa one. Maybe with lower pressure when the temperature is also low the heat doesn't conduct to the polymer so well as with higher pressures and it only properly starts to melt at temperatures fairly past the melting point. And when the temperature is high enough to melt the polymer even cause of the poor contact the polymer has more pores and other cavities to flow into and get attached to fibres.

Correlations for fibre amount index's standard deviations are not found. If they are compared with seal strength there is no clear trend and the distributions of standard deviations for certain seal strengths are widely spread. Same applies if standard deviations are compared with their corresponding fibre amount indexes. Graphs for these are presented in Appendix 2.

Like explained earlier fibre amount index is calculated based on scanning the polymer film surfaces. Large fibre fragments are separated from smaller ones and only the large ones are used for calculating the index. Some consideration was given to the total fibre material that was scanned from the polymer films in case if this would correlate better with seal strength than fibre amount index.

With gentler sealing conditions (1.5 s, 0.2 MPa and 0.5 s, 0.7 MPa) this connection seemed to be somewhat linear- as fibre material amount grows so does seal strength. Sealing conditions 0.5 MPa, 1.5 s and 0.7 MPa, 1.5 s have rather more level total fibre material amount than ascending even if seal strength changes. In the case of 0.7 MPa and 3.0 s sealing conditions the results do not correlate well with seal strength. The connection between seal strength and total fibre material does not seem any better than the connection between seal strength and fibre amount index.

From examining optical microscope images it can be noted that PE has formed really well around the shapes of the paper surface, for example if there is some particle on the paper surface PE has surrounded it. Only at the lowest sealing temperatures PE has not spread into the paper at all. The samples sealed with pressure 0.2 MPa are the only ones that don't have any spreading of PE at 140 °C. Though it has to be stated that even if PE has spread from few parts, otherwise the film looks even and the seal looks good.

Often where PE has spread to the paper there can be seen a notch on adhering layer. This probably indicates that when PE has spread for example into some pore on the paper surface it has pulled the other film structures towards the pore. With higher temperatures adhering layer shows to be wider from some parts. Possibly it has started to melt and spread partly.

260 °C is the only temperature where on the cross section images adhering layer can be seen to be against paper meaning that PE layer has moved aside. An example of this is shown in Figure 35.

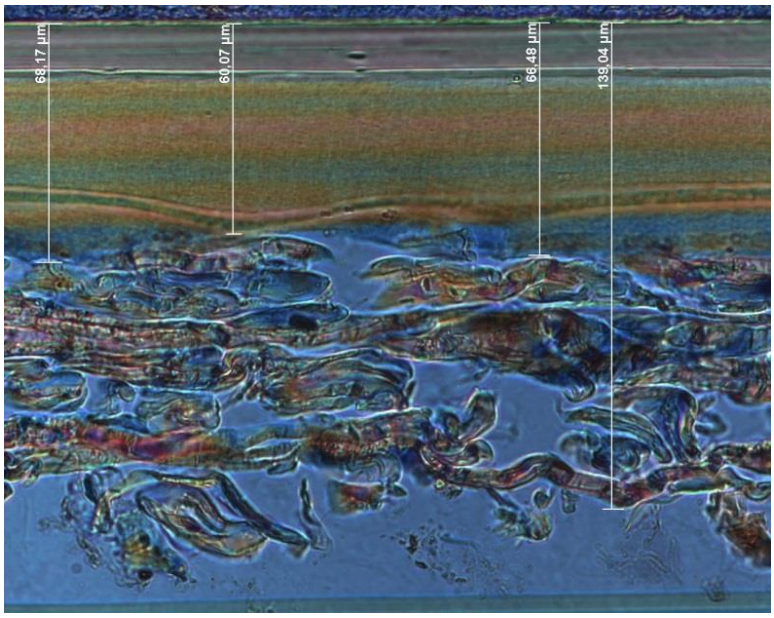

Figure 35. *Cross section image of sample sealed at 260 °C, for 1.5 s with 0.2 MPa pressure. PE layer has been partly pushed aside and adhering layer is against the paper.*

When comparing microscope images to fibre amount index results there doesn't seem to be too straight forward connection with them. For example with some samples PE is spreading into the paper but fibre amount index is still low, also in some cases the seals of different temperatures look same but the fibre amount index values have significant differences.

6.6 Study with Kraft paper B and Film B

Film B was heat sealed with Kraft paper B. This study for new materials consisted of four different heat sealing parameter combinations which were the following:

- 130 °C, 0.5 MPa and 1.0 s
- 160 °C, 0.5 MPa and 0.5 s
- 160 °C, 0.5 MPa and 1.5 s
- 160 °C, 0.7 MPa and 3.0 s

Heat sealing at 180 °C, 0.7 MPa and 3.0 s which the company had used for these materials as well in some earlier measurements was left out of this study because with those parameters the outer PP layer started to melt. This was predictable from the DSC results.

In Figure 36 are gathered the results from seal strength measurements. It can be noted that the 90 degree peel angle gave in this case usually higher result for seal strength than what does the 180 degree peel angle. This is the opposite when compared the case with Kraft paper A and Film A. Why do these different material combinations give opposite results is an interesting question. These results clearly don't follow the equation (1) even though

at least here the 90 degree peel angle gives higher results like this equation suggested it would.

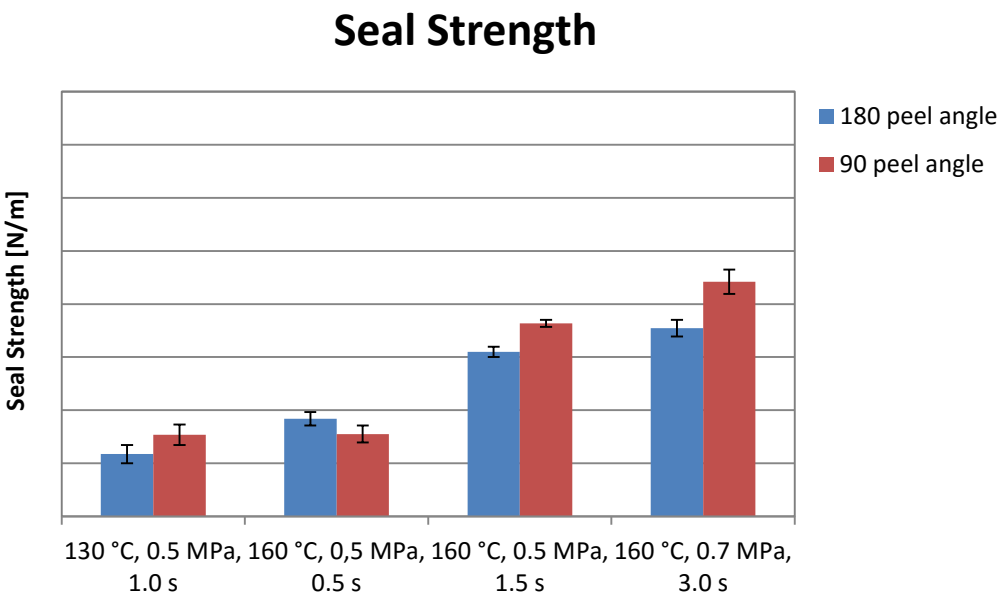

Figure 36. *Results from seal strength measurements for Film B.*

The question here is that why these different materials behave so differently. Maybe the properties of polymer films differ from each other so much that it creates this difference. The properties of the films were studied a bit and it was found out that the Film B has higher tensile strength and tensile stiffness.

Fibre amount index was also measured for these materials. The results are shown in Figure 37. Here in three cases out of four fibre amount index is bigger for the right. It can be noted that the fibre amount index values are much lower here than what they are for kraft paper A and Film A. This kraft paper B has better peel characteristics than kraft paper A, meaning that not as much fibres get loose from the surface of the paper when peeling the seal open. Standard deviation values for fibre amount index are big for many of the sample series. This is because in most of the cases majority of the fibre amount index scans had given zero for fibre amount index but then there was one or two samples that gave divergent result and thus causing high standard deviations.

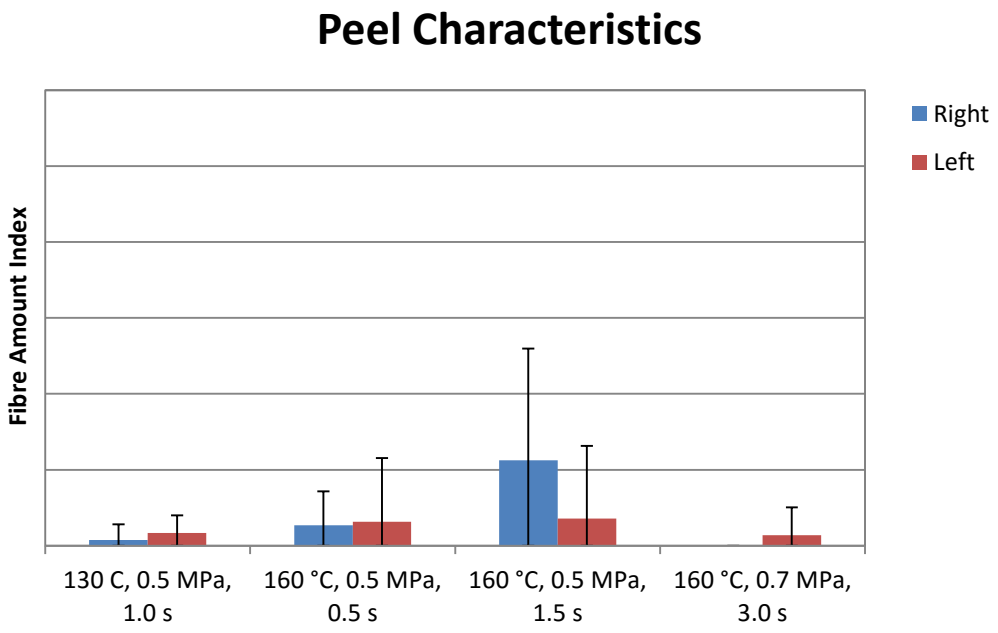

Figure 37. *Results from fibre amount index tests.*

Cross sections of the samples were studied with optical microscope. The samples sealed at 130 °C PE seems to be formed well around the surface irregularities of paper and the seals looked good. From the studied seals there are only two spots where PE layer looks to be going a bit deeper into the paper.

In the case of samples heat sealed with parameters 160 °C, 0.5 MPa and 0.5 s seals look quite good and PE layer is also quite even. With these samples there was one part where PE had moved aside from between paper surface and PA. Samples sealed with parameters 160 °C, 0.5 MPa and 1.5 s looked similar to the ones that had shorter sealing time but otherwise same heat sealing conditions.

With sealing parameters 160 °C, 0.7 MPa and 3.0 s there is the most variety in the appearance of the cross section images. There are parts where the seal looks good, PE layer is even and has not penetrated to the paper. But then in some sample there are parts where the PE layer has disappeared and the paper is against PA layer (Figure 38). In another sample PE has not disappeared completely but is notably thinner than from the other parts. Also PE layer has spread into the paper in few samples.

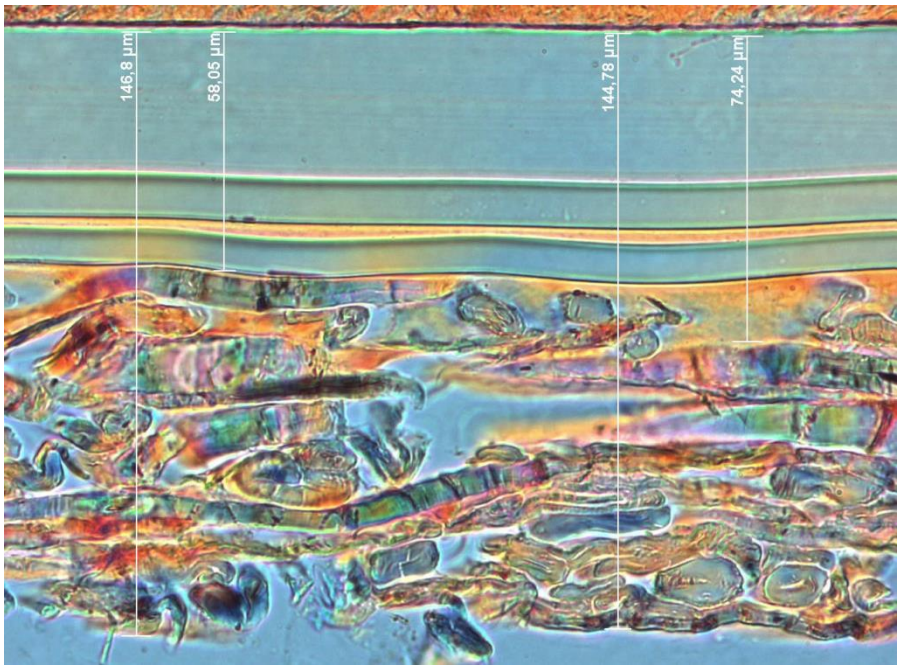

Figure 38. Cross section image of samples sealed with 160 °C, 0.7 MPa and 3.0 s. PE has partly moved aside from between paper and PA.

6.7 Comparison between manual and mechanical peeling method for fibre amount index

In this study an instrument was used for peeling samples for fibre amount index measurements. Purpose was to simulate manual tearing with an instrument while eliminating possible variable factors that are present with the manual method. One of those is that the tearing speed by hand is not constant but most likely at the beginning it is slower and then it speeds up towards the end. In addition, the speed is different for different samples because it is impossible to do the peeling exactly the same way for every sample manually especially if the person doing the tests changes. With an instrument the speed is constant during measurement and same for all the samples. Hence the measurements done this way seem more reliable than the ones done manually.

Manual way was evaluated to have still much higher peel rate than it was possible to use with Instron, the used testing instrument. Another fact that should be noted is that when tearing by hand the polymer film can be held from quite close to the spot where the seal starts. But because of the setting of the instrument there is about 11 cm of unsealed polymer film between the grip and the spot where the seal starts to tear open.

Kraft paper A and Film A were used in this study. The grip separation rate was 50 mm/s which equals to 3000 mm/min. This in turn means 1500 mm/min peel rate of the seal.

In Figure 39 are presented the fibre amount index results. It can be seen that in three cases out of four the fibre amount index is higher for the right side. These results can be compared with Figure 24 where same materials and heat sealing parameters have been used. The only difference is the peeling method of the samples.

First it can be noted that the fibre amount index levels are different. Samples peeled manually have lower fibre amount index. Actually when manually peeled some of the samples (130 °C, 0.5 MPa and 160 °C, 0.5 MPa, 0.5 s) have zero or close to zero fibre amount index. In these cases also the standard deviations are smaller than for the samples peeled with Instron.

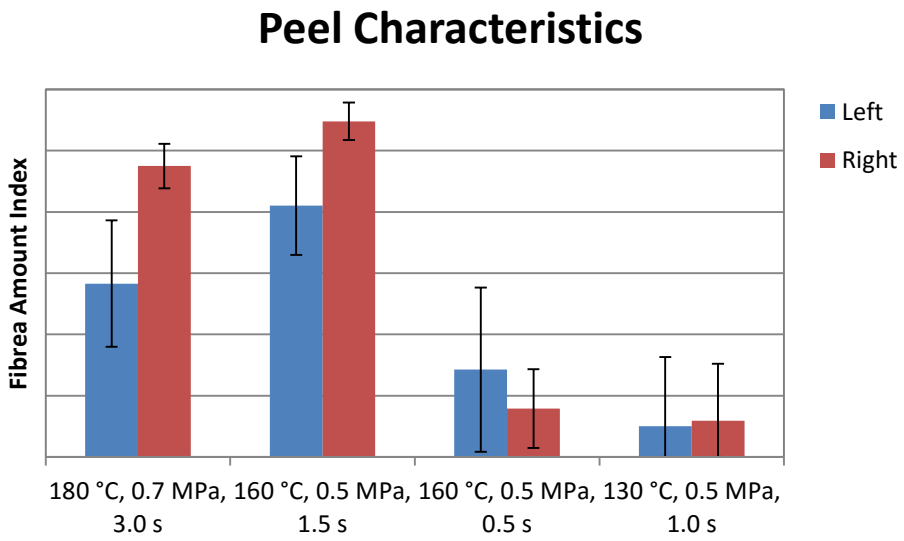

Figure 39. *Fibre amount indexes obtained from Instron measurements.*

If the two other heat sealing conditions (180 °C, 0.7 MPa, 3 s and 160 °C, 0.5 MPa, 1.5 s) are studied they have notably higher level of fibre amount index when compared to the other conditions. As mentioned already the samples peeled with Instron have higher fibre amount index and the biggest rise is 184 % and the smallest 20 %. Standard deviations are bigger with manual peeling method.

These results suggest that fibre amount index is higher when peeled with Instron. Differing factor between these tests is of course the peeling method but also the peel rate. Even if Instron can peel the samples fairly faster than Hounsfield that was used in some other tests the difference between Instron's and manual way's speed is still notable. In the study done with profilometer (chapter 6.8) it was suggested that the peel rate might have some effect at least to the surface roughness of paper. Also in theory part it was suggested that with lower peel rate interfacial failure happens and with higher paper failure occurs. Therefore, it is possible that this difference of speed might cause the difference in fibre amount index. What comes to the standard deviations it seems that when fibre amount index is small then standard deviation is small for manual method but with higher fibre

amount index standard deviation is higher for the manual method than when used an instrument.

The problem here is that can these results be directly compared with each other since the levels of fibre amount index are so different. Because of this in Figure 40 has been taken a different approach to the subject. A graph having standard deviation as a percentage from fibre amount index as fibre amount index's function has been presented in Figure 40. Sealing conditions are not separated in this graph to keep it easier to read. All the points from matrix-study are included in the graph so that there are more points to compare with.

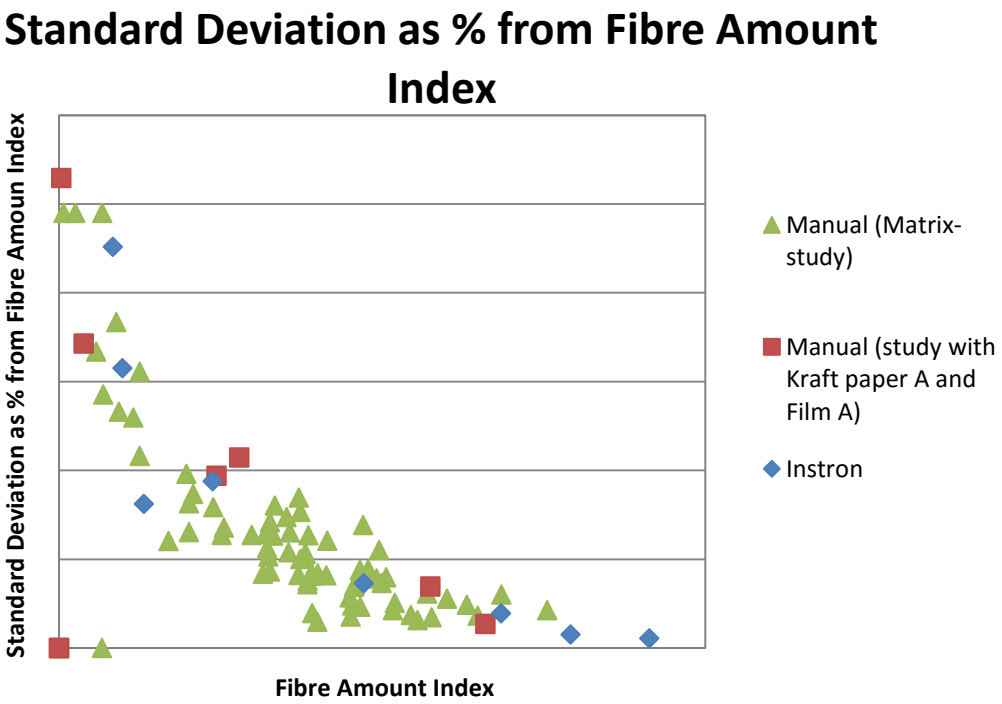

Figure 40. *Comparison between different peeling methods for standard deviation as percentage from fibre amount index.*

The idea is that if standard deviation would be smaller for Instron samples also when the standard deviation is calculated as a percentage from fibre amount index this value should be lower too. From Figure 40 can be seen that the points of Instron's samples are not positioned lower than the other test's point. With high fibre amount index they are quite low but there aren't any other points to compare them with. This graph suggests that there isn't really change in the standard deviations weather the tests were done manually or with a testing machine. In Appendix 3 has been presented figure where standard deviations are drawn as a function of fibre amount index and also this image shows that the standard deviations aren't lower if the samples are peeled with Instron other than when the fibre amount index is high the standard deviations are low.

6.8 Profilometer measurements for paper

In Table 9 are gathered the surface roughness measurements done with profilometer for Kraft paper A that was sealed with Film A. What was measured was average height of selected area, *Sa*, and also standard deviations were calculated from the results. This *Sa* value is used as a measurement for surface roughness.

Table 9. *Results for average height of selected area of paper samples that have been heat sealed with differing parameters and also peeled open with differing parameters.*

Sample	Sa [µm]	Standard deviation
Unsealed paper	5.7	1.3
Sample 1: 180 °C, 0.7 MPa, 3.0 s, 150 mm/min, 90°	9.3	1.5
Sample 2: 180 °C, 0.7 MPa, 3.0 s, 250 mm/min, 90°	8.7	1.3
Sample 3: 160 °C, 0.5 MPa, 1.5 s, 150 mm/min, 180°	9.1	1.4
Sample 4: 160 °C, 0.5 MPa, 1.5 s, 150 mm/min, 90°	7.9	1.6
Sample 5: 160 °C, 0.5 MPa, 1.5 s, 250 mm/min, 180°	8.5	1.5

In Figures 41, 42 and 43 are areas that were scanned with profilometer. In the first image of them is a paper surface that has not been heat sealed so in other words nothing has been done to it and so the surface is quite smooth. Some fibres can even be seen on the topography image going flat across the surface. The two following pictures' papers have been heat sealed with polymer film which had been peeled off before the measurements. The sealing parameters for the samples are the ones shown in Table 9.

From these images it is obvious that when the seal is torn open the paper surface gets broken so that some fibres are pointing out and the unsealed paper surface is much smoother when compared to the others. The results in Table 9 support this since the surface area roughness is notably smaller for unsealed paper than for the other samples. Although, it is possible that formation of paper affects to the results also. This means that the paper density per area has some changes so other parts have more fibres than others. [29] This could possibly affect surface roughness of paper when measured with profilometer.

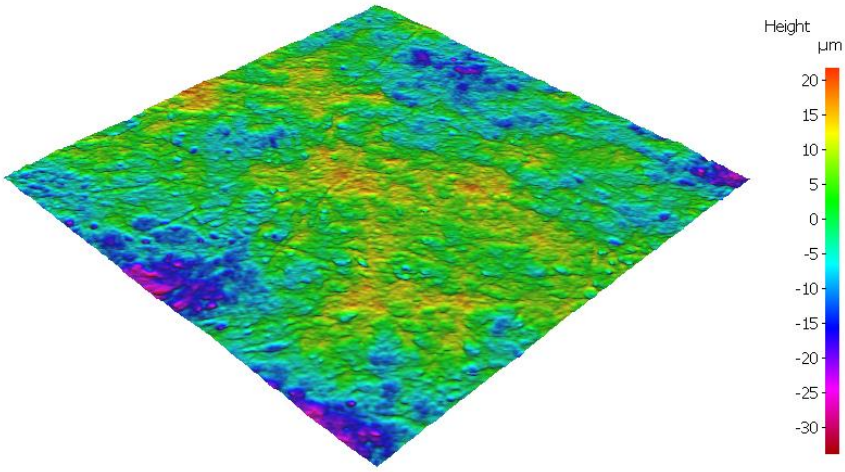

Figure 41. *Topography image of the surface of unsealed paper from 2.81 mm x 2.81 mm area.*

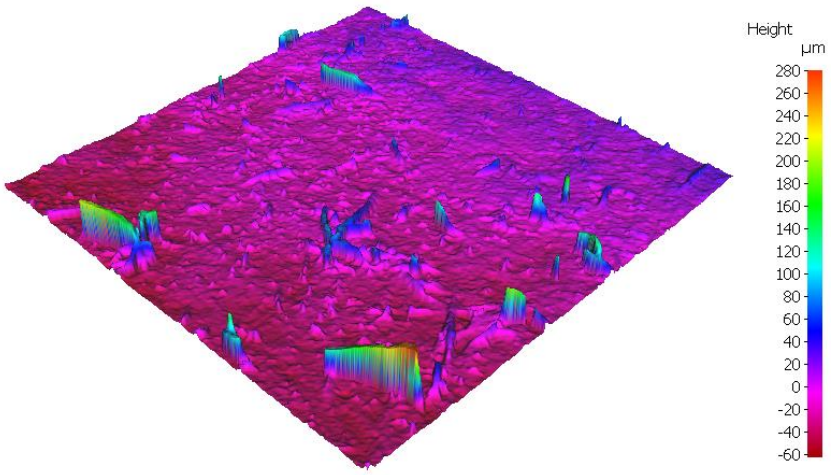

Figure 42. *Topography image of sample 1 from 2.81 mm x 2.81 mm area.*

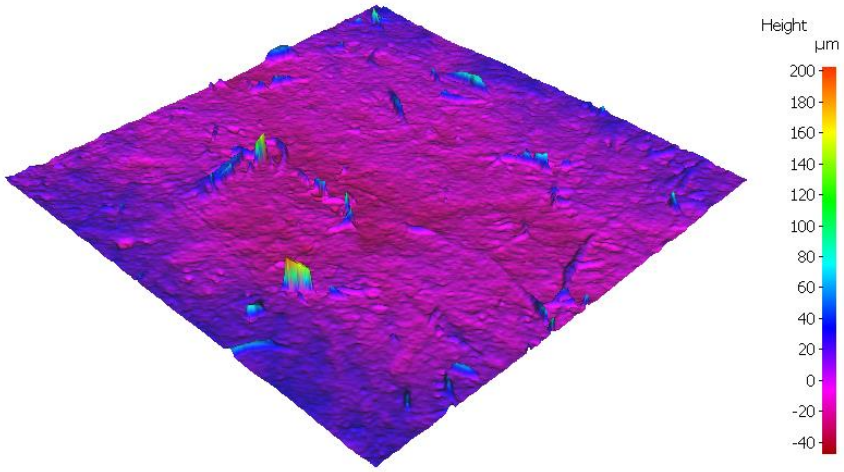

Figure 43. *Topography image of sample 4 from 2.81 mm x 2.81 mm area.*

When looking closer to the Figures 42 and 43 it can be noted that there are some fibres sticking out from the paper which indicate possible loose fibres. Those fibres also make *Sa* value higher. Sample 1 has the highest *Sa* value and Figure 42 shows that the surface of the sample is quite uneven with lot of spikes. The sample in Figure 43 has some fibres sticking out as well but not as much as in the image of sample 1. Sample 4's average surface height is the closest to the untorn paper.

Since this study with profilometer was not very extensive, no definite conclusions can be made from it. Also, it should be noted that most of the results are inside each other's standard deviations. In two cases, when comparing samples which only difference is the peel rate, with a higher peel rate, 250 mm/min, the average surface height is lower than with slower peel rate of 150 mm/min. Samples 3 and 4 have the only difference in the peel angle and the results show that with 90° the average surface height is smaller than with 180°. Based on this both peel rate and angle have an effect on how cleanly the seal peels open. Samples 1 and 4 have the same peel angle and grip separation rate but they differ in the sealing conditions. Sample 1 has been sealed in rougher conditions (180 °C, 0.7 MPa) than sample 4 (160 °C, 0.5 MPa) and the value for average height of selected area, *Sa*, is higher for sample 1 which should when thinking logically indicates that there is more tearing of fibres and fibres poking out of the paper than with gentler sealing parameters. But if these results are compared with fibre amount index results from matrix-study then it is noted that the fibre amount index for the harsher conditions is lower than for the gentler conditions. These results are not necessarily straight comparable because the peel angles are different. These profilometer measurements have been done in 90 degree peel angle but the fibre amount index samples have been torn by hand in 180 degree peel angle which also means that their peel rate has been much faster.

6.9 Profilometer measurements for polymer film

In this study not only the paper parts of the samples were scanned but polymer film parts as well. Materials in this study were Kraft paper A and Film A. The studied samples had the following sealing conditions:

- Samples 1, 2 and 3: 160 °C, 0.5 MPa, 1.5 s
- Samples 4, 5 and 6 : 180 °C, 0.7 MPa, 3.0 s

These sealing conditions were chosen because in the earlier profilometer study they had the highest and lowest values for *Sa*. Peel angle for these samples were 90 degrees and the grip separation rate 300 mm/min which also was the actual peel separation rate. This means that it was faster peel rate compared to the earlier profilometer measurements. The results are presented in Table 10 and Table 11. From one sample five measurements were done from different parts of the sealed area. Exceptions are samples 7 and 8 because they are actually just single measurements made from unsealed polymer film and their purpose was to test and improve the measurement procedure with profilometer.

Table 10. *Results from profilometer measurements done for polymer film.*

Samples	Average Sa [µm], before sealing	Standard Deviation	Average Sa [µm], after sealing	Standard Deviation
1	8.4	1.9	10.3	2.3
2	7.2	0.8	10.4	2.2
3	7.8	1.0	13.5	3.3
4	7.9	1.3	7.5	1.2
5	9.5	1.5	6.4	0.5
6	8.8	1.4	6.7	0.2
7	2.4			
8	2.2			

When looking at the average *Sa* for polymer film before sealing (Table 10) it can be noticed that there is quite a lot of fluctuation in them. For example the corresponding results from paper surface (Table 11) are much closer to each other. What caused this variation in the case of polymer film is that the optical profilometer probably saw the sample table through the film to some extent. Hence, the results should not be compared to the results after sealing. The used polymer film is commercial film so the results should be more even if the surface of it was seen properly in profilometer images. Samples 7 and

8 are measurements from single areas on unsealed polymer film and they were measured later than the other samples to test what kind of effect there is if polarizer is used when taking the image. Because they are single samples instead of a series there are only the *Sa* values for them. From the results it can be seen that the *Sa* is much smaller for them than for the other samples. This would suggest that here the profilometer does not see the sample table as much as in the earlier measurements. Also these results for samples 7 and 8 have only 0.2 µm difference between them but since they are only two samples it does not prove that with this way of doing the measurements the variety in *Sa* would necessarily be smaller, though it is very possible and likely.

In the case of polymer film samples after sealing this seeing sample table through polymer film is not a problem. This is because when the seal is torn open the polymer part isn't transparent anymore since there is some substance attached to the seal part of the polymer film, so there are no problems to get the surface to show on profilometer images.

In Figure 44 and Figure 45 can be seen the difference in appearance of the films when it has not been heat sealed and after the sealing and peeling the seal open. The black and white images are two dimensional live view images where the texture of the material shows but not the topography, whereas, the colour images show the topography of the samples. In Figure 44 are the images taken from sample 8 since it is thought that in these images the polymer surface topography will show more correctly than in the images that have been taken without polarizer. The unsealed film has peaks and valleys and it resembles little bit of surface of an orange peel. Based on the images, after heat sealing the texture of paper appears on the surface of polymer film even if there are no actual fibres remaining on the polymer surface at that part. Some fibres were seen on the polymer film also during the measurements. In Figure 45 there is one fibre and it shows as a darker line that starts from left, curves little bit up and then goes down. This image demonstrates how well the fibres that have gotten attached to the film show in profilometer images. These fibres were even more obvious when using the profilometer. When looking at the topography image of this one fibre than can be seen well in the live view image, it can be noticed that there are parts where the fibre vanishes in the topography image. This suggests that the fibre is partly covered by polymer. This would mean that in the heat sealing process polymer has melted and surrounded the fibre partly. Thus when the sample has been torn open the fibre has been better attached to the polymer film than to the paper and it has gotten torn off from the paper.

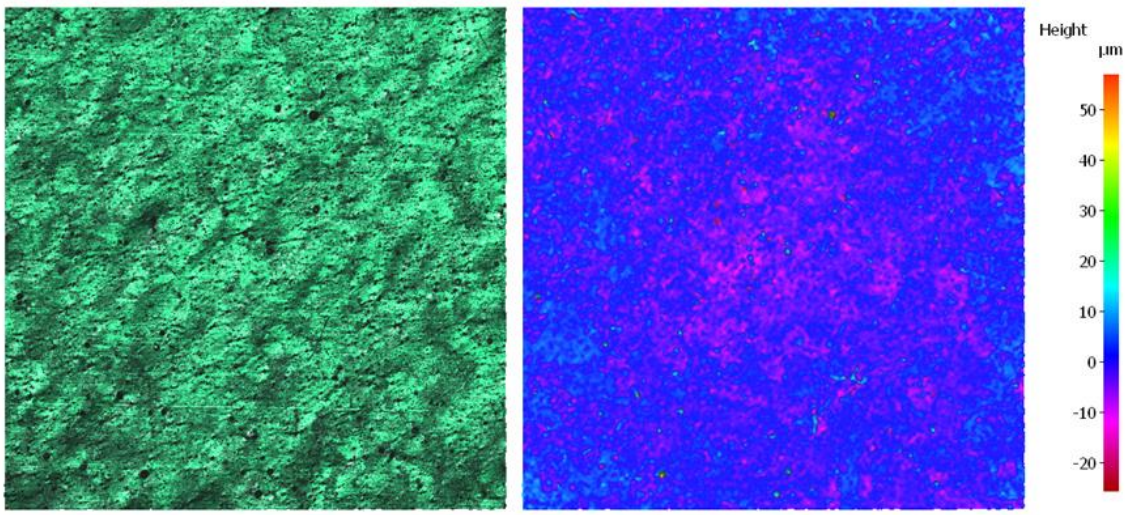

Figure 44. *Profilometer images from polymer film that has not been heat sealed. The area of the image is 2.81 mm x 2.81 mm. A)Live view image b) topography image.*

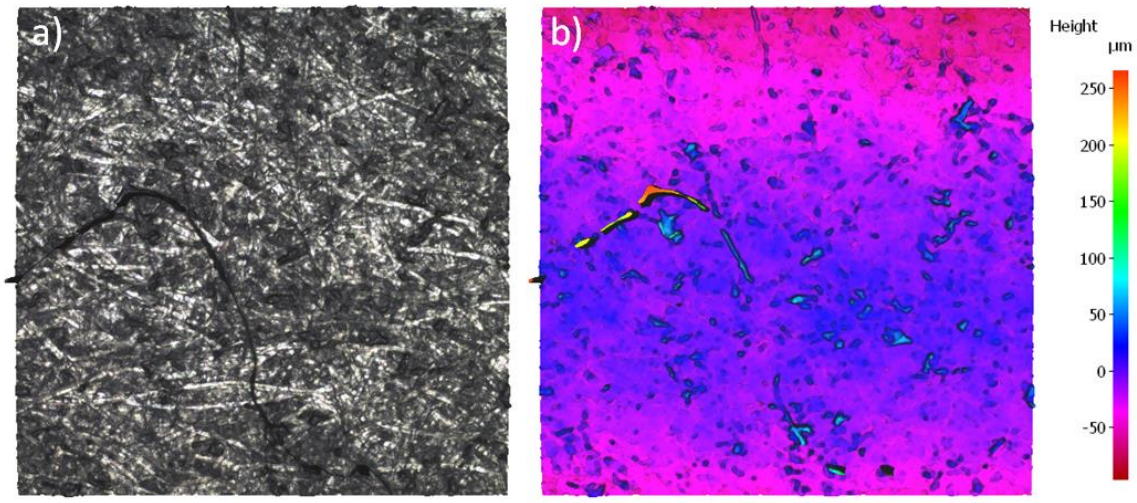

Figure 45. *Profilometer images of an area from sample 3 from polymer film that has been heat sealed with paper but has been torn apart with the paper. The area of the image is 2.81 mm x 2.81 mm. A) Live view image b) topography image.*

What affects to the *Sa* value difference before and after heat sealing is partly due to molten polymer being pressed against paper during the heat sealing process so the polymer film's surface reshapes because of this. The change of the surface can be seen from the profilometer images. After the sealing the polymer film surface looks to have rougher peaks. Samples 1, 2 and 3 have clearly higher average *Sa* than samples 4, 5 and 6. Samples did have different pressures and sealing times so this could affect how deep and from how many places molten polymer has penetrated into the paper which then could affect the surface roughness. Even if samples 4, 5 and 6 have higher sealing temperature and longer sealing time maybe the higher sealing pressure somehow smoothens the surface of the paper so that the molten polymer doesn't flow into the dimples of the paper so easily and these samples' *Sa* values are lower because of that. It is likely that there are more fibres

or parts of them attached to the polymer film surface with samples 1, 2 and 3. This is logical if the lower sealing pressure lets the molten polymer surround fibres better so then when the sample is peeled open more fibres gets torn due to mechanical interlocking. Because used sealing parameters and seal opening conditions are different than used in matrix-study these results cannot be compared with fibre amount index results which might have been interesting.

In Table 11 are the results for the paper samples. The values for the *Sa* before heat sealing are close to each other. The result of sample 1 catches the attention by giving the highest *Sa* value and also it has highest standard deviation. When considering the results after sealing and tearing the seals open this sample 1 is the only one that has lower *Sa* value after sealing. This result does not seem logical at all since the expectation is that when the seal is torn open it creates rougher and more uneven surface. But since the *Sa* value before the sealing had such a high standard deviation it could be that there has happened something when measuring the sample that creates error.

When comparing the results of *Sa* values before and after sealing on Table 11 it can be noted that the differences are small. There is no clear difference between the different sealing parameters. If sample 1 would be ignored from the results it would seem that samples 2 and 3 have slightly higher differences between before and after sealing results. But this conclusion can’t be made reliably because the differences are so small and the standard deviations for *Sa* are in most of the cases bigger than the calculated differences. In the earlier measurements with samples having same sealing conditions and peel angle but slower peel rate the samples sealed in harsher conditions had the higher *Sa* value. Also the earlier measurements suggested that a higher peel rate might create smoother paper surface.

Table 11. *Results from profilometer measurements done for paper.*

Sample	Average Sa [µm], before sealing	Standard Deviation	Average Sz [µm], before sealing	Average Sa [µm], after sealing	Standard Deviation	Average Sz [µm], after sealing	Difference in Sa (before sealing –after sealing)
1	6.3	2.7	61.2	6.2	0.4	134.0	-0.1
2	5.2	0.5	52.5	6.7	1.4	181.7	1.5
3	5.8	1.2	55.9	6.7	1.6	249.1	0.9
4	5.6	0.8	64.2	6.0	0.8	140.6	0.4
5	5.3	0.6	62.8	5.5	0.6	185.5	0.1
6	5.9	1.2	66.4	6.3	0.8	166.3	0.4

In Figure 46 and Figure 47 is shown the same paper sample before and after it has been heat sealed and the seal peeled open. The images have been taken from quite same spot of the sample because some of the same fibres can be seen on both of them. What can be noted from these images is that after tearing the polymer off from the paper some fibres are partly sticking out of the paper. The bar scales in Figure 46 b) and Figure 47 b) show that the height scale of the samples have big difference. Also in Table 11 are given *Sz*, maximum height of selected area, values for the samples. There is a notable difference between those values before and after sealing. These maximum height values support the idea that some of the fibres are pointing out of tea paper surface rather than just lying against the paper.

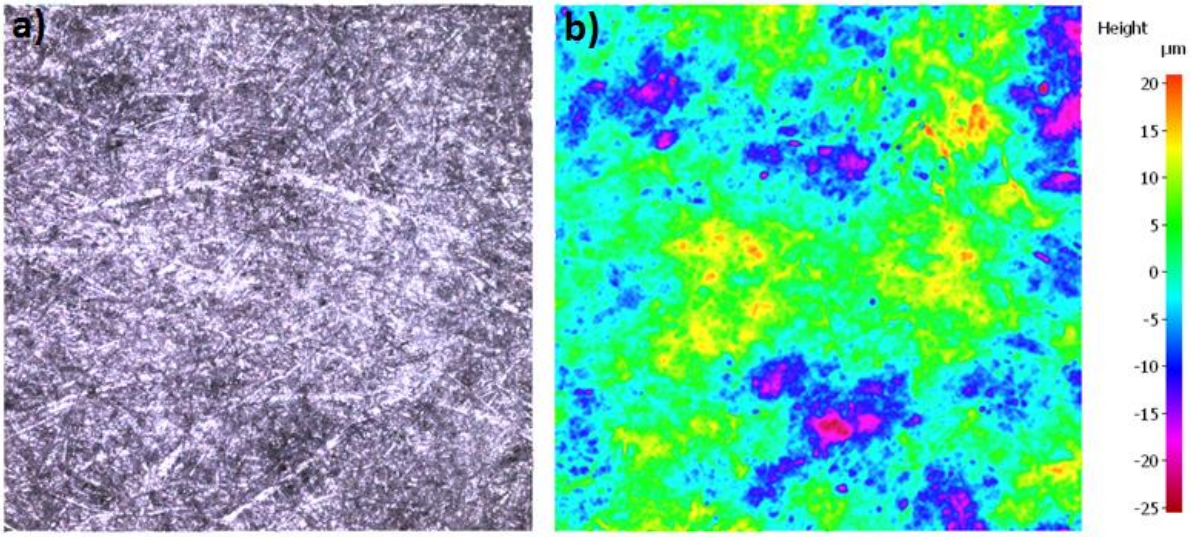

Figure 46. *Profilometer images from sample 1 from paper that has not been heat sealed. The area of the image is 2.81 mm x 2.81 mm. A Live view image, b) topography image.*

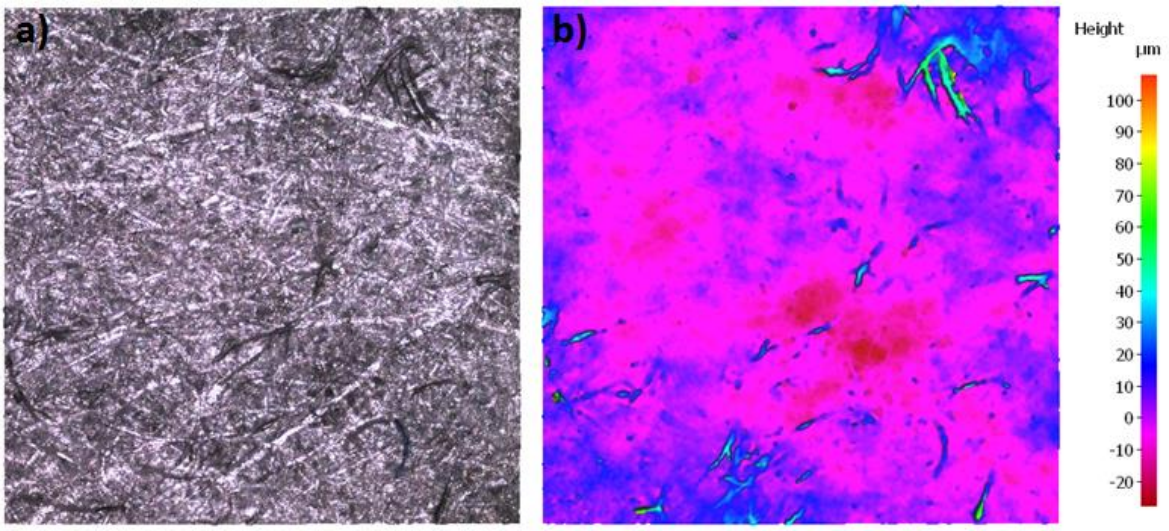

Figure 47. *Profilometer images from sample 1 from paper that has been heat sealed with polymer film which has been torn off. The area of the image is 2.81 mm x 2.81 mm. A) Live view image, b) topography image.*

When considering images and how evident it is that the paper surface changes so that there are fibres pointing out of it and the maximum height of selected area, it is peculiar that it does not transform to the results of profilometer measurements. This could suggest that there are perhaps some differences how the samples have been attached to the sample table or something else is causing variety and the measuring method should still be further developed.

6.10 The Angle Method measurements

The Angle Method created by Hishinuma was tested to find out if it would give any interesting information regarding heat seal or heat sealing conditions. What should be remembered here is that it was originally done to different materials (polymer film against polymer film) than what were investigated in this study.

Two sealing conditions were chosen for initial testing of the method and these were the following:

- 160 °C, 0.5 MPa, 1.5 s
- 180 °C, 0.7 MPa, 3.0 s.

Kraft paper A and Film A were used in these measurements. The samples for seal strength measurements were 10 mm wide strips instead of the 25 mm which were used in other tests. This was because 10 mm wide sealing bar was in use with the heat sealing equipment. The samples were prepared in the way that the seal edge was in 45° as was shown in Figure 8. The machine direction of the paper was the same as in other peeling tests. The 10 mm wide seal wouldn't have provided area on the test piece where the sealed area would have reached from side to side if 25 mm wide samples would have been used. This area where the seal reaches from edge to edge is an important part of the study since on the recorded load-time curve the seal strength measurement should indicate plateau if the failure mode is peel seal. Also seal strength at the plateau area should be same as if 10 mm wide samples were tested in the normal way according to ASTM standard F 88-00. The grip separation rate in these tests was 300 mm/min which means 150 mm/min peel rate.

Samples were cut from left, middle and right from the sealed area of the samples. Four of the middle position curves for different heat sealing conditions are presented in Figure 48 and Figure 49. From these images the possible and expected plateau area is not possible to see because there is only about 3 mm in length of the sample that has the seal reaching from side to side which means only slightly over one second in time.

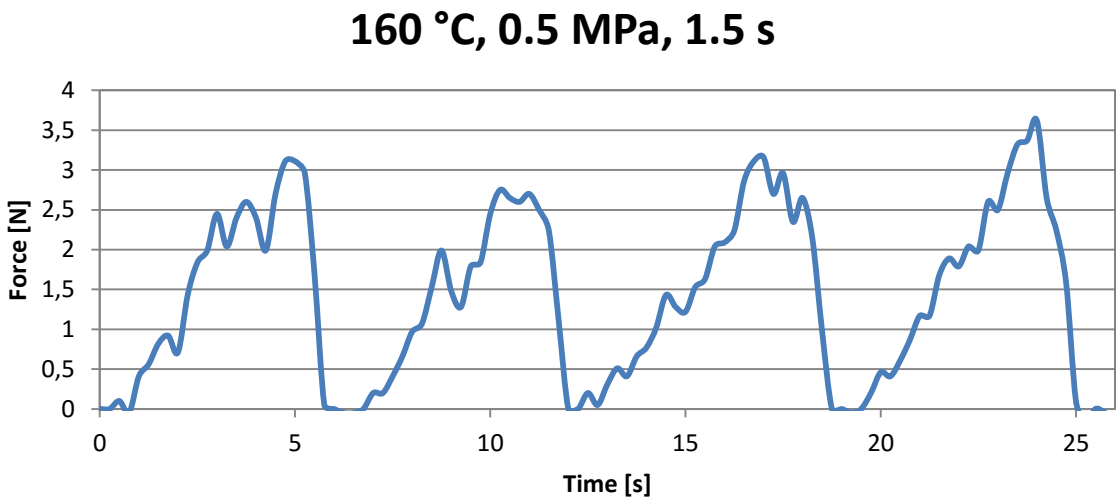

Figure 48. *The angle method curves for four 10 mm wide samples that were heat sealed at 160 °C with 0.5 MPa pressure for 1.5 s.*

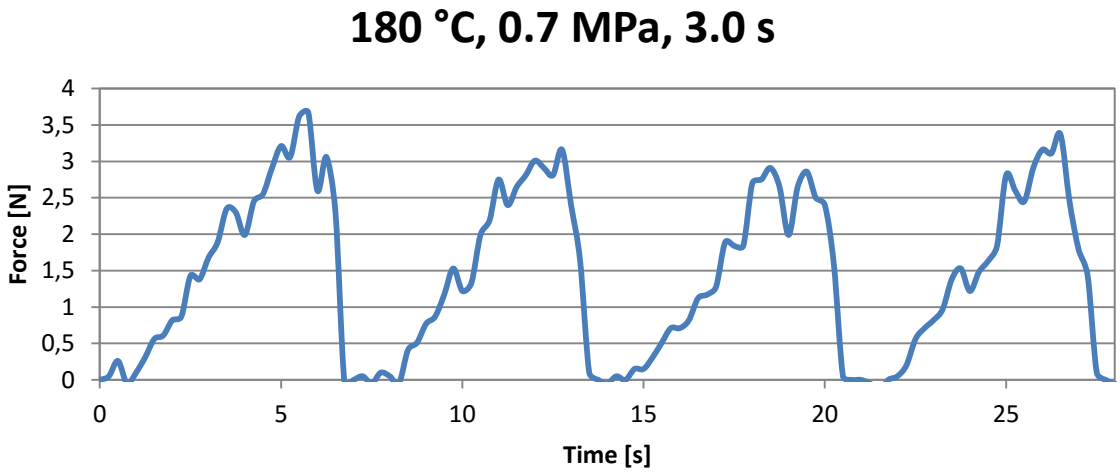

Figure 49. *The angle method curves for four 10 mm wide samples that were heat sealed at 180 °C with 0.7 MPa pressure for 3.0 s.*

When considering the area where the width of the seal grows gradually when peeled for example the second curve in Figure 48 has a bigger peak on it. But also the peel curves done according to ASTM standard F 88-00 have peaks and lower adhesion points on them so based on this information and the figures it is difficult to make any conclusions if there is or is not polyballs or pinholes on the seal edge. The two different heat sealing conditions do not give very different type of curves either. Of course they have difference in the maximum load level they reach but the peaks and low adhesion points on the curves seem quite similar.

Because of the similarity of the curves a new test setting was planned. It was thought that if gentle and harsh sealing conditions are used there would be difference on the load-time

curves. Also it was thought that by using wider samples the results would tell more. Two centimetres wide sealing bar with Teflon coating was used. The lower sealing bar was prepared from brass bar and on top of it a silicon plate was plastered. Two different series of samples were prepared which were:

- 130 °C, 0.5 MPa, 1.5 s
- 200 °C, 0.5 MPa, 3.0 s.

The expectation was that the samples sealed at 130 °C would give straighter force-time curve from the beginning where the peeled seal width increased incrementally and the other samples would have more peaks or low adhesion points on it. 15 mm wide samples were used. This setting made it possible to have longer distance where the seal on the sample was the whole width of the sample. This also shows little on the curves in Figure 50 and 51 so that the end of some of the curves are more level than in the earlier angle method tests.

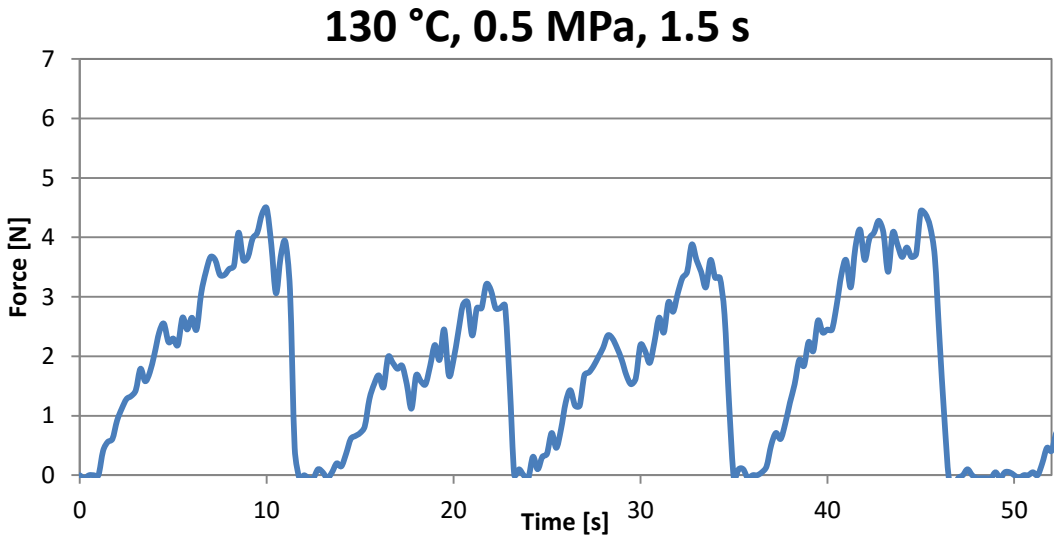

Figure 50. *The angle method curve for four 15 mm wide sample that were heat sealed at 130 °C, with 0.5 MPa pressure for 1.5 s*

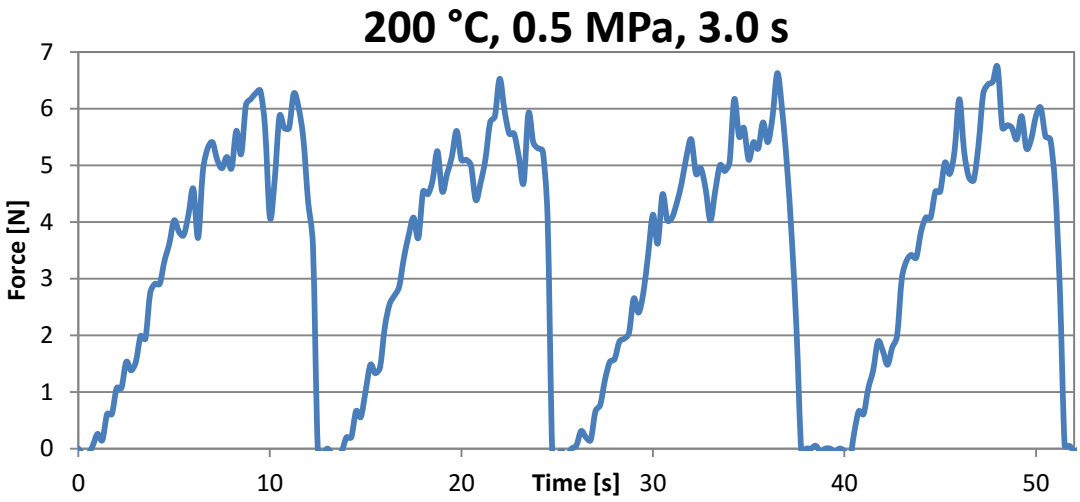

Figure 51. *The angle method curve for four 15 mm wide samples that were heat sealed at 200 °C with 0.5 MPa pressure for 3.0 s.*

Because the silicon anvil was prepared at TUT by pasting the silicon sheet on a sealing bar it is not as even from the thickness as the one used with 10 mm wide sealing bar. The paste was difficult to spread completely evenly on the sealing bar and after attaching the silicon sheet on the sealing bar the air bubbles form between them had to be pressed off. As a result of this it can be stated that the seals are not completely even from their whole area and probably affect the results.

If the curves are compared with each other regardless of the unevenness in them it looks as if the curves for gentler conditions (130 °C, 0.5 MPa, 1.5 s) have more variety on them. Especially the second and third curves seem to have bigger and wider peaks on them. But then the other curves are quite similar to the harsher condition's curves. The same goes for the other curves that are not presented here that it was difficult to find any clear differences between them.

As a conclusion for this method it can be stated that these experiments were not too successful in acquiring reliable information. Since Hishinuma had developed this method for different materials than what were used in this study it can change how the curves should be interpret. Because the curves done according to ASTM standard F 88-00 also have variety in the load-time curves it is very possible that those peaks and low adhesion points show in the angle method curve where the seal area width grows when peeling and makes it difficult to know if it is due to some defects on the seal edge.

7. CONCLUSION

Temperature, time and pressure are important parameters in heat sealing process. In this study about heat sealing paper with multi-layer polymer film it was noted that temperature has great effect on seal strength. Also sealing time is important parameter since it affects seal strength in the same manner as temperature: the longer sealing time the more heat is conducting to the polymer film and melting it. Based on the results in this study it can be suggested that after the sealing time is long enough for the sealing layer of the polymer film to melt and reach the set sealing temperature the sealing time does not increase seal strength anymore. Another observation was that after the sealing pressure is high enough it does not increase seal strength strongly anymore but if it is too low also seal strength stays low. These conclusions agreed with earlier studies about the subject even if those concentrated in heat sealing polymer against polymer.

It was pointed out in the theory part that pressure affects the heat sealing process in a different way than temperature and time. Its purpose is to bring the surfaces close to each other. It was noted that fibre amount index results did not correlate completely with seal strength results. For example with sealing time of 3.0 s and pressure of 0.7 MPa there was a clear drop of fibre amount index at temperatures 140°C to 170 °C even though seal strength with same parameters was high. When fibre amount index – temperature curves of samples with sealing time of 1.5 s but having different sealing pressures (0.2 MPa, 0.5 MPa and 0.7 MPa) were compared with each other it was noted that below about 180 °C with the lowest pressure the fibre amount index was also lowest. Whereas with temperatures above that the samples sealed with 0.2 MPa had the highest fibre amount index. These results suggest that high pressure might make the surface structure of the paper more compact so the molten polymer film does not penetrate into the structure of paper surface as freely and so decreasing mechanical interlocking. Why fibre amount index then is low for 0.2 MPa at low temperatures could be because the materials are not in as close contact as with higher pressure and thus the heat doesn't conduct to the polymer film as well. Then only when the temperature is fairly higher than the sealing temperature the polymer film is enough molten to flow into the paper surface shapes around the fibres.

Based on the different studies it would seem that fibre amount index is more often higher for the right side than for the left side of a sample. But in the case of studies that only include four or few more different sample series definite conclusions cannot be made since they don't have that much statistical value. That is why the matrix-study is more reliable for this and there 57.9 % of the samples had higher fibre amount index on right side which is not that far from 50 %. Also when studying the standard deviations of fibre amount index on matrix-study it was found that for 52.6 % the standard deviation of fibre

amount index was bigger on the right side. Based on these results the left and right side do not really differ so the manual peeling method cannot be said to cause differences here.

Standard deviations for fibre amount index were compared between samples that were peeled open manually and with an instrument. It was found that the samples peeled manually gave in general lower fibre amount index results. If the results of same sealing parameters were compared then the results suggested that with low fibre amount index values the standard deviations were small for manual method and with high fibre amount index values standard deviations were small for samples peeled with Instron. But if the samples peeled with Instron are compared also with matrix-study's results so that the samples having same fibre amount index are compared with each other using Instron doesn't give lower standard deviations for fibre amount index than the manual method.

Profilometer is a good way to study the changes that happen on the surface of paper and polymer samples in heat sealing process. The images showed clearly fibres that were pointing out of paper surface and also the fibres that were attached to the polymer film after peeling the seals open. Images also revealed that the polymer films surface topography changes during the heat sealing process. The fact that some of the fibres that were attached to the polymer film surface looked like they were partly covered by polymer supports the mechanical interlocking theory when considering the adhesion phenomena between paper and polymer film in heat sealing.

Measured results from different profilometer test support the observations made from the images. Especially for the polymer film there was clear difference in the average surface height values for different sealing conditions. In the case of paper there were differences but the changes weren't always so clear. Effect of peel angles and peel rates were studied from the paper samples and they suggested that with 90° peel angle the average surface height is smaller than with 180°. Higher peel rate gave smaller average surface height. But conclusions can't be made for sure how peel rate or angle affects to the surface roughness since these tests weren't very extensive and their standard deviations were quite big. More measurements should be done to see if they give same results. Also the measuring practise would need some developing still so that possible errors can be eliminated.

Seal strength measurements were done with two different peel angles: 90° and 180°. In theory it was suggested that peel angle should affect bonding strength of the materials so that 90° peel test gives twice the seal strength of 180° test. Two different kinds of results were obtained: with Kraft paper A and Film A 180° gave higher seal strength but with Kraft paper B and Film B the situation was the opposite. Neither of these material combinations did not follow the given equation exactly. It was noted that Film B was stiffer and tougher than the other film. Possibly the properties of the films caused this difference but it cannot be said for sure at this point since it would require more research and characterization of the films.

The tests done with the angle method turned out not to provide useful information. The interpretation of the curve was difficult and with different sealing conditions the curves did not show clear differences. It could be that this method is not valid when the heat sealed materials are paper and polymer instead of polymer-polymer paring. Since the load-time curves for the samples tested according to ASTM standard F88-00 already have some noise or low adhesion points on them rather than being straight, the curves obtained from measurements done according to the angle method possibly have this noise on their curves too which makes them harder to interpret. In addition the test setting that was used was somewhat flawed. Use of a wider sealing bar and wider samples would have been better. Also slower peel rate would have given more points to the curve and possibly give more informative curves.

8. SUGGESTIONS FOR FURTHER STUDIES

Since in this study it was not possible to explain why fibre amount index decreased and then increased back as a function of temperature with sealing pressure of 0.7 MPa and sealing time 3.0 s it would require some more research.

It was discovered in this study that the used material pairings behaved in the opposite way regarding seal strength for peel test conducted in 90 degree and 180 degree peel angles. The properties of the films are different for example it was noted that the Film B is stiffer and tougher than Film A. To find out why different material combinations give opposite results would need some more research and comparison of the properties of the films.

If it is wanted to study closer does the sealing bar heat unevenly a more comprehensive study should be made. This would mean more parallel samples than was done in this study. Also the sealing conditions could be chosen to be better for this purpose. There should be conditions chosen from the part of the seal strength-curve where seal strength is increasing rapidly since there the differences would show the most. Some reference point form plateau area would of course be advisable as well.

Since this study was not so extensive based on the results from profilometer measurement it is not possible to say how peel angle, peel rate or sealing conditions affect the surface roughness for sure. This study should be repeated with new samples to see if same kinds of results are obtained. Also more careful consideration should be paid to attaching the samples to the sample table so that they would be as straight as possible. This is because if the samples for example are more curved after heat sealing it might be that the program calculating the surface roughness cannot correct it completely and thus it affects the results.

For further studies using profilometer the polarizer should be tested when scanning transparent polymer film with bigger sample series to see if it gives less variable results than measurements done without it. Then comparison of surface roughness of unsealed film and film that has been heat sealed with paper and peeled apart from it could probably be done more reliably. In addition, it would be interesting if the results could be compared with fibre amount index results, which means that the heat sealing conditions and peeling conditions should be the same in both studies. This could give better idea to how much tearing conditions of the fibres affect the surface topography and what is due to other factors.

In the case of the angle method it would be advisable to try notably lower peel rates for the test. Then there would be more recorded points for the time-load curve. This might reveal more about the shapes of the curves and offer more information. The test with the

wider (20 mm) sealing bar failed in this study but if reliable wider sealing bars could be used it would be preferred over the 10 mm wide bar because also then there would be more recorded points on the load-time curve.

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APPENDIX 1: FT-IR SPECTRUMS

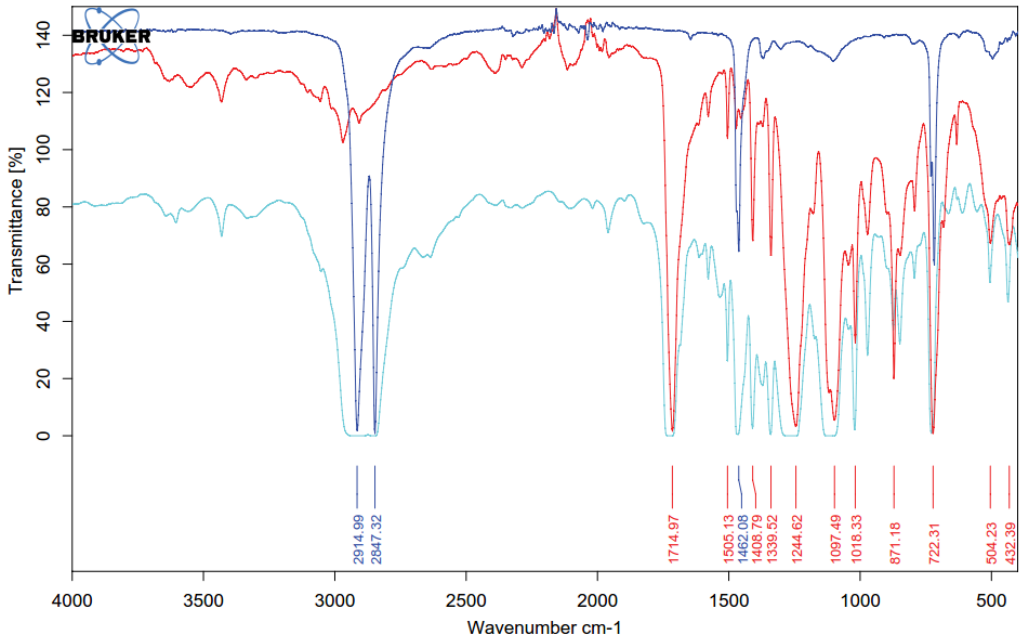

Figure 1. *FT-IR spectrum of Film A. Blue line is the spectrum of the PE surface, red is PET's and light blue is the spectrum run through the whole film.*

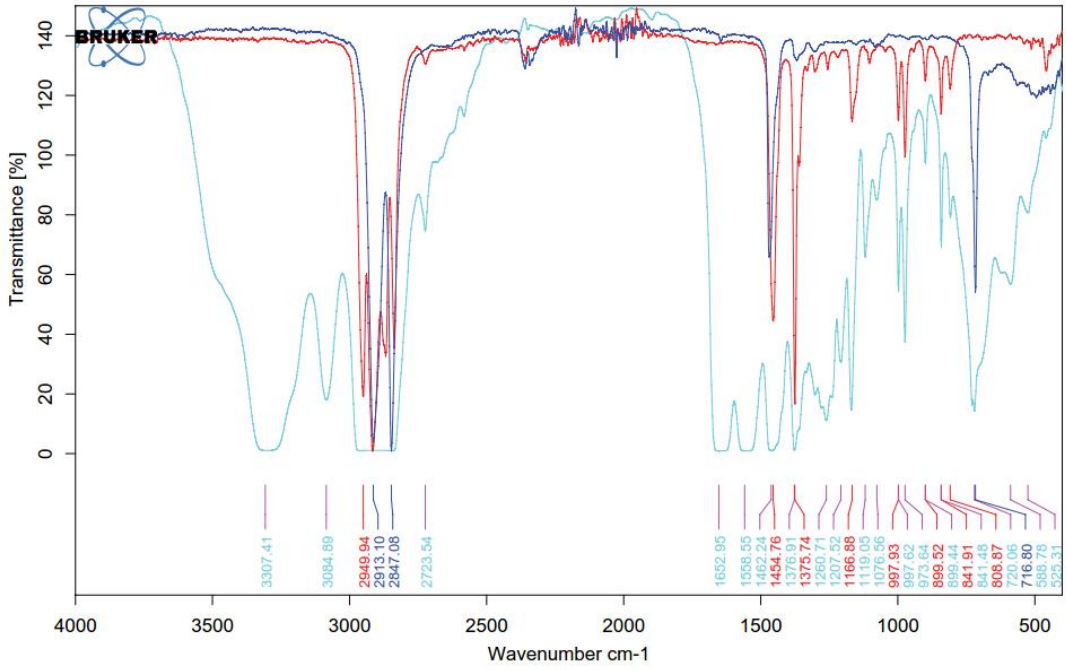

Figure 2. *FT-IR spectrum of Film B. The blue line is the spectrum of PE surface, the red one is PP's and the light blue is the spectrum run through the whole film.*

APPENDIX 2: STANDARD DEVIATIONS OF FIBRE AMOUNT INDEX FOR MATRIX-STUDY

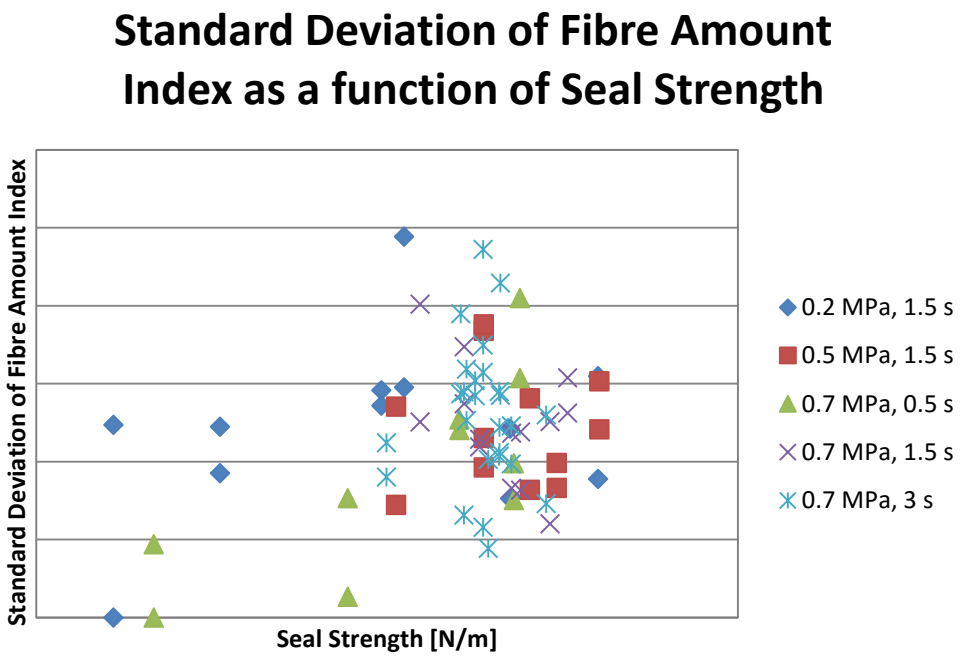

Figure 1. *Standard deviations of fibre amount index as a function of seal strength.*

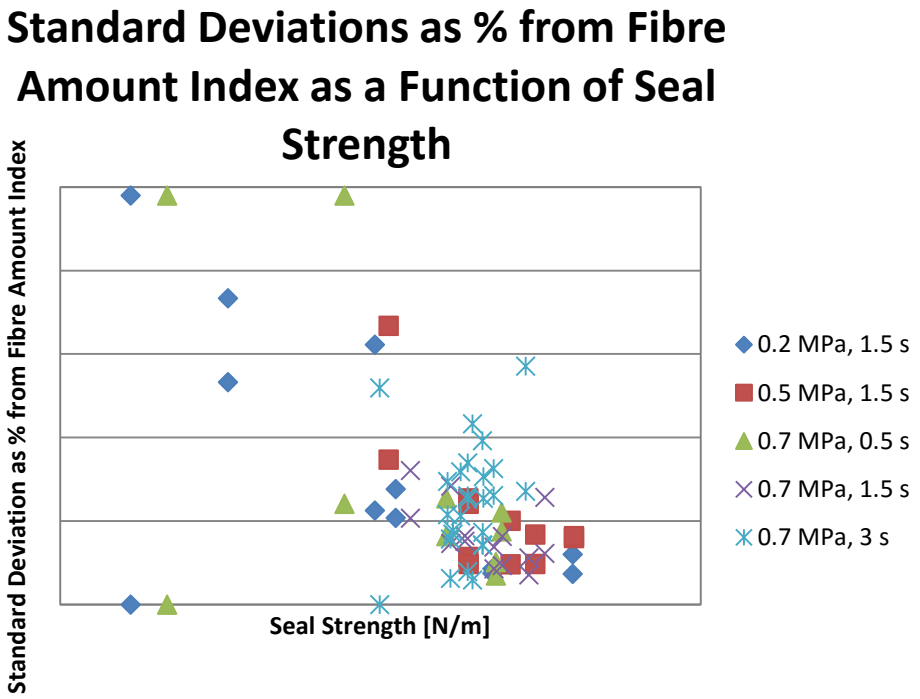

Figure 2. *Standard deviations of fibre amount index as % from fibre amount index as a function of seal strength.*

APPENDIX 3: STANDARD DEVIATIONS AS A FUNCTION OF FIBRE AMOUNT INDEX

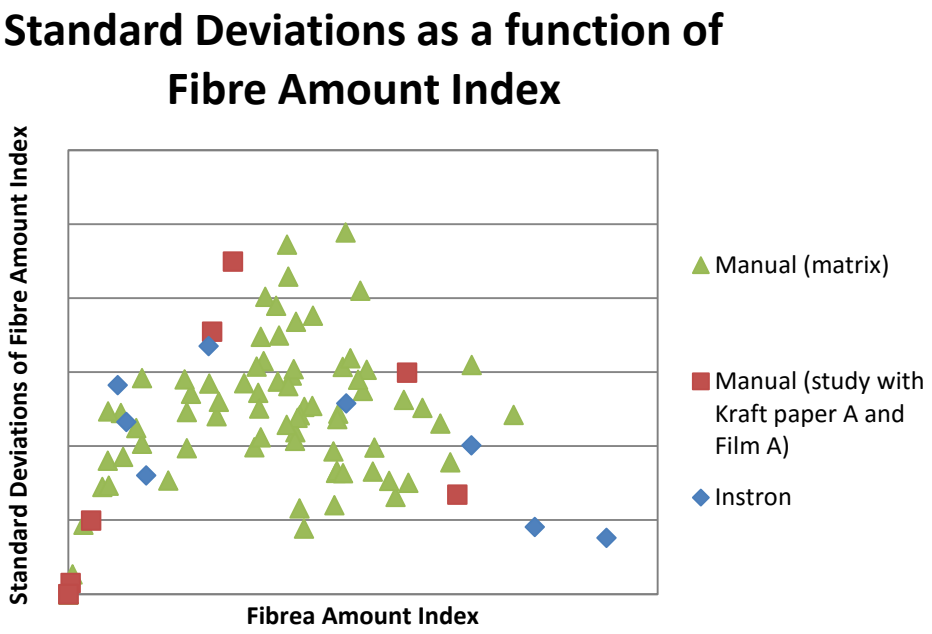

Figure 1. *Comparison of standard deviation of fibre amount index between peeling samples manually and with an equipment.*