TAMPEREEN TEKNILLINEN YLIOPISTO
TAMPERE UNIVERSITY OF TECHNOLOGY

BEGUM GOKCE SAGMANER

THE INTERFACIAL CHARACTERISTICS OF MULTILAYER SYSTEMS

Master of Science thesis

Examiner: Prof. Jurkka Kuusipalo
Examiner and topic approved by the
Faculty Council of the Faculty of
Materials Science
on 9 April 2016

ABSTRACT

BEGUM GOKCE SAGMANER: The Interfacial Characteristics of Multilayer Systems
Tampere University of Technology
Master of Science thesis, 68 pages, 27 Appendix pages
June 2016
Master's Degree Programme in Materials Science
Major: Polymers and Biomaterials
Examiner: Prof. Jurkka Kuusipalo

Keywords: multi layer systems, polymer coating, extrusion coating, polyester

The target of the study is to explore the opportunity of enhancing the adhesion of polyethylene terephthalate to the cardboard when the coating thickness of polyethylene terephthalate onto the cardboard is diminished. The reference coating weight is 40 g/m^2 and the target is to achieve same great adhesion with between 25-30 g/ m^2 coating weight.

According to the purpose of the thesis, literature study includes principles of extrusion coating, differences in extrusion coating of polyesters and polyolefins, adhesion theories and practical methods for adhesion improvement in extrusion coating. Literature study is followed by an experimental part, where adhesion improvement by changing processing parameters, modifying process equipments and trying different grades of polyethylene terephthalate was studied once the coating weight is diminished.

In the experimental part of the study, 5 adhesion trials are performed. In adhesion trial 1 and 2, PET 1 grade is utilized and PET 2 grade is utilized in adhesion trial 3,4,5. Firstly, both grades of PET are screened by performing with only corona as a pretreatment method by following same processing parameters. In screening trials, process variable is air gap. In the following adhesion trials, the effect of flame treatment by performing with various air gap levels and the effect of process equipment modifications by performing with various line speeds are studied for both of them by following same processing parameters. After trials have done, samples are analyzed. Analysis methods can be examined in two different groups: main and additional analysis methods. Main analysis methods are adhesion test by hand, grammage test and neck-in test. Additional analysis methods are heat sealing test, microscopy test and pinhole test. For the application of addditional test methods, it is decided that they will be performed if the adhesion level of polyethylene terephthalate on the cardboard reaches the level of 5 over 5 at the target coating weight.

The best attained adhesion of PET 1 is the level of 4 over 5 by performing only with corona during the screening trial. Normally, the best adhesion results is expected from modifications of process equipments results. The most probable reason of the situation is high melt temperature that caused thermal degradation.

In the case of adhesion of PET 2 grade, the best adhesion level is 4 over of 5 while performing with modified process equipments. In addition, coating weight results of PET 2 obtained from common test points are generally out of the target coating weight range. This shows that it is possible to reach almost same adhesion level at lower coating weight levels by utilizing PET 2 grade.

All in all, it is proven that it is possible to run the process at 30g$/m^2$ coating with almost perfect adhesion for both polyethylene terephthalate grades. Better adhesion can be achieved by performing with high line speeds, high screw speeds and minimum air gap values.

PREFACE

This thesis work has been conducted at Paper Converting and Packaging Laboratory, Departments of Materials Science of Tampere University of Technology.

Firstly, I would like to thank to my supervisors Prof. Jurkka Kuusipalo for his support and advice throughout the thesis. Then, I would like to thank Sanna Auvinen for providing me this thesis subject. Likewise, I would like to thank to Esa Suokas for sharing his experiences and his recommendations in critical points during all thesis trials. I would like to thank Marikki Laamanen, Petri Kuusipalo and Hilkka Koivuniemi-Mäkinen for helping and guiding me through all laboratory work and showing their patience to my endless questions.

Furthermore,I would like to thank to my one and only friends who are Anil Sirri Baslamisli and Ismailcan Ersahin. We grew up together, learned together and succeded together. The most especially, I am indebted to them for creating a huge smile on my face in every trouble I have faced and giving me the warmest hug once I needed.

The one who has been there for me for ages, the whom I feel I am the luckiest woman in the world, Cumhur Kusdemir. It is inexpressible to thank you for being near by me through all ups and downs, supporting me in all conditions with your unconditional love and first and foremost being my better half. May you be my future and my home forever.

Finally, for their unrequited love, patience and support, I could not thank to my mother, father and sister enough for the things they have done for me. Whatever I have achieved or will achieve anything worthy in my life, it is always thanks to you. Thus, I would like to dedicate this thesis for my beloved ones who are the gold pieces in my heart.

Tampere, June 2016

Begum Gokce Sagmaner

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LIST OF ABBREVIATIONS AND SYMBOLS

LDPE	Low Density Polyethylene, page 8
PE	Polyethylene, page 8
PP	Polypropylene, page 8
E/VAC	Ethylene vinyl acetate, page 8
E/BA	Ethylene butyl acrylate, page 8
E/MA	Ethylene methyl acrylate, page 8
E/EA	Ethylene ethyl acrylate, page 8
MPO	Modified polyolefins, page 8
PET	Polyethylene terephthalate, page 9
PMP	Polymethylpentane, page 9
PLA	Polylactide , page 9
PGA	Polyglycolide, page 9
HDPE	High Density Polyethylene, page 10
LLDPE	Linear Low Density Polyethylene, page 10
Mw	Molecular weight, page 11
MWD	Molecular Weight Distribution, page 11
TIAG	Time in Air Gap, page 22
TS	Test Series, page 33
TP	Test Point, page 33
Tmelt	Melt Temperature, page 49
CW	Coating weight, page 82
S-(number)	Sample-(number), page 82

W_A	Work of adhesion, page 16
W_C	Cohesive energy, page 16
γ_{LV}	Lifshitz-van der Waals, the capital of all long range interactions, page 16
SL	the term for surface in contact with the liquid, page 16
LV	he term for liquid in contact with its vapour, page 16
θ_{SL}	the contact angle formed between the liquid and solid, page 16
γ_S	the surface energy of solid that is measured precisely only in vacuum, page 16

1. INTRODUCTION

In today's world, packaging has an inevitable role in the quality of food products. Since it prevents breakage of the product , increases the shelf-life of foods and prevents the loss of foods that is occurred due to spoilage by protecting from detrimental effects of environment, chemicals and physical factors. To protect the product, packaging materials have great barrier properties against to oxygen, carbondioxide, moisture and other gases like flavors and aromas. Furthermore, it is possible to keep nutrients and colors in the product just like a first day without deterioration in a given time by blocking light. Thus, a wide variety of foods of year-round can be obtained and consumed without deterioration through suitable packaging methods. Beside these passive protection properties of packaging materials, they involve in the active roles to maintain the desired quality of product in a proper atmosphere. By based on this, there are three main functions of packaging properties: providing protection, utility and communication in physical, atmospheric and human environments. It is crucial that package should be optimized by considering the efficiency of all three functions in all three environments [23]

From past to today, the food packaging methods and materials have evolved by depending on the life style of the society, increase in the population and the demand for the quality. These factors are driving force to create new food categories and make an innovation in packaging [23]. For instance, busy lifestyle increases the demand of microwave products like pizza, pasta and soup type of foods and the demand leads to develop new packaging technology that comprises of polyethylene terephthalate coated cardboard.

To respond the market change and needs, polyethylene terephthalate has become a recommended material for extrusion coating, especially for special cases. To enable use in high temperatures, the board is coated with black or white polyethylene terephthalate which provides a high degree of protection from humidity, oxygen and grease, as well as good heat resistance. The food side of the packaging, coated with white polyethylene terephthalate , will remain pure white even when exposed to high temperatures. However, the utilization of polyethylene terephthalate in extrusion coating is quite challenging due to its processability and weak adhesion. Since, extrusion coating technology was primarily designed to coat polyethylene layer on paperboard. Processing problems may be solved by using some well-known methods: examining the effects of processing parameters, modifying process equipments and trying different grades of main polymer.

Adhesion states that the tendency of dissimilar bodies are being together by providing initimate interfacial contact, thus transfer of mechanical force is happened across the interface. As a precondition for great adhesion, wetting is a key factor since it provides to close contact between substrate and adherent. Therefore, it is important to control wetting and adhesion during processing in terms of that materials are to be held together or stand apart [25]. For porous substrates, such as cardboard, it is generally thought that adhesion is created by molten polymer penetration into the pores and around the fibers. This sets up conditions for good physical or mechanical bonds, although chemical bonds may also play a role. However, in the case of polyethylene terephthalate adhesion to cardboard, chemical adhesion is not expected. Since, polyethylene terephthalate is not eager to oxidize due to heteroatoms in the backbone and slightly low cross-linking level.

There are quite limited source in the literature about polyethylene terephthalate adhesion to the cardboard in extrusion coating probably due to the challenges in processing of polyethylene terephthalate, minor market share in coating business. However, it becomes priority to make innovation in chilled and frozen food packaging by considering the changes in demand. While increasing the quality of packaging, it is vital to reduce unfavorable environment impact and use the energy sources as minimum as possible. If all the factors are considered, certain benefits can be seen in aiming for enhancing polyethylene terephthalate to the cardboard in thinner coating thickness.

The overarching target of the study is to explore the opportunity of enhancing the adhesion of polyethylene terephthalate to the cardboard when the coating thickness of polyethylene terephthalate onto the cardboard is diminished. The reference coating weight is 40 g/ m^2 and the target was to achieve same great adhesion with between 25-30 g/ m^2 coating weight. The scope of the study is to determine the primary mechanisms behind the reduction in adhesion with decreasing coating thickness, develop models for quantitatively predicting adhesion performance and apply the improved understanding to practical advantage. According to purpose and scope of the thesis; the motivation for the current work began with the literature study of adhesion theories and practical methods for adhesion improvement in extrusion coating. Then, it is maintained with observations from an earlier study polyethylene terephthalate to enhance its adhesion to cardboard. In the experimental of the study, it is concentrated on the effects of processing parameters, modifications of process equipments and properties of polyethylene terephthalate .

2. THEORETICAL BACKGROUND

This chapter provides the basic knowledge for extrusion coating, the differences in processing of polyesters and polyolefins in extrusion coating, the theory of adhesion and practical methods for adhesion improvements that will be used throughout the thesis.

2.1 Extrusion Coating

With the effect of upward trend, more sophisticated packaging and industrial materials, laminated and multi layer flexible structures become extensively utilized for many applications. New developed multilayer laminate structures correspond the requirement of special packaging and industrial applications by having the unique barrier, sealability and structural properties in each layer and extrusion coating is one of the technology to manufacture these type of structures [21].The first developed continuous process to merge an extruded thermoplastic polymer on a substrate was extrusion coating. In the late 1940s, the very first manufacturing plant was established in the U.S. Over the years, the technology has been developed and turned into the most used polymer converting process in the world. Originally, the process was invented to provide moisture proofing by implementing thin polyethylene layer onto paperboard. In several years, the process has grown to fulfill various demands in the market, such as flexible packaging, photographic reproduction, health care, construction, and industrial applications [8]. Correspondingly, the utilized type of substrate and polymer have changed and varied by depending on the demand of the packaging industry.

In the following, a short outlook on extrusion coating process technology, materials for extrusion coating and common applications are presented.

2.1.1 Extrusion Coating Process Technology

The working principle of extrusion coating process is that a substrate is laminated with hot molten polymer as it is run through a cooled nip roll assembly. After laminate is cooled down by the chill roll, the web undergoes second operations like slitting, surface treatment, printing or decorating. Then, it is taken up by wind-up equipment [8] [26]. It combines two low-cost materials having different properties to create better performance packaging product. The aim of extrusion coating is to manufacture products having unique properties that an individual material can not have on their own by combining the best properties of each component. It is expected to have valuable, additional properties of these products like enhanced heat sealability, tear or crease resistance, appearance, better barrier properties to prevent the permeability of water or oxygen and other gases, additional chemical resistance and improved printing ability [7].

In the following Figure 2.1, an extrusion coating line assembly is illustrated.The main components in the extrusion coating process are the extruder, pressure rolls and chill rolls .

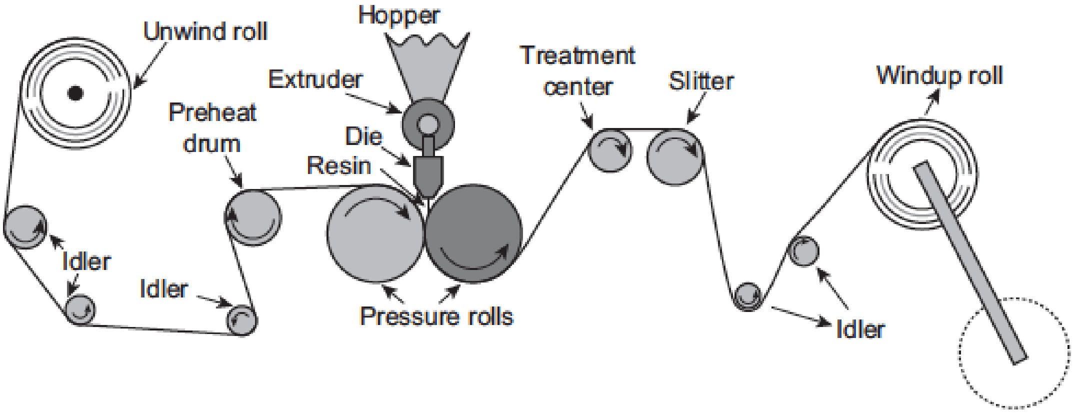

Figure 2.1 *The illustration of extrusion coating line assembly [7]*

The main functions of extruder in the coating process is to melt the polymer pellets completely and homogeneously by the help of shear forces and heat and then, deliver the highest material output to the die at an acceptable melt temperature. Material output of extruder is based on the screw geometry, the back pressure of the extruder and the rheology of the polymer.There are some requirements that the coating extruder fulfills its main functions as it should be: minimum variations in output, stable melt temperature, the least purging time during polymer changes, minimum residence time and minimum time for a step change from drool to the targeted process output. By considering these requirements, a single screw is mostly preferred with having shallower, longer feeds for the coating extruder. [21] [8] [5] [12].

As polymer left from extruder, it flows from the die through to the nip between a chill roll and a pressure roll placed directly below the die as seen in Figure 2.1. Polymer melt is pressed onto the substrate by the help of the pressure between the roll and pressure rolls. The desired thickness of the film on the substrate is achieved by higher moving speed of substrate than the extruded film. The distance between the die lip and the nip to chill roll contact point is called as air gap or draw distance and the time is spent in the air gap which is called as time in air gap. When the polymer film is flowing in the air gap, it contacts with the air in certain time so then the distance and the time spent in air gap play key role for the performance of polymer coating. The degree of adhesion strength is changed by depending on the length of the air gap and time in the air gap [21] [14].

As mentioned earlier, heated chill roll is also determinant component in terms of the polymer's coating performance. The function of chill roll is to solidify the molten film and press the melt onto the substrate. The optimum temperature of chill roll is around 15-30°C and to maintain the temperature uniformly, water is circulated inside the roll as a cooling medium. It is crucial to provide sufficent water to control and distribute temperature effectively at the surface of the roll. To provide maximum cooling, it would be better if the web revolve around chill roll as long as possible before passing the next roll. Also, it is known that heated chill roll has positive effect on adhesion. However, this method is not applicable for all polymers. If temperature of the chill roll is reached to really high values, it leads to some problems that polymer melt may be sticked to the surface of chill roll or partial delamination may occur between the substrate and the coated web. In addition, surface of the chill roll is another key element to improve the quality of the coating. If rough rolls are used, matte finish is obtained. On the other hand, glossy and smooth finish is obtained if smooth rolls are utilized. The surface of the chill roll can be metallic. [21] [10].

Even the purpose of the extrusion coating is to manufacture the product having uniform coating thickness with great adhesion, some defects can occur due to troubleshots during processing, structure of substrate and/or characteristics of polymer. Thick or thin coating in the machine direction, voids, pinholes, orange peel and contamination due to gels or foreign material can be given as an example of these defects.Voids and pinholes may be formed because of extremely high line speed that causes to be drawn melt too much. This situation is resulted with poor adhesion between polymer coating and substrate. Other defects are occurred by reason of poor processing conditions and/or less amount of feed. Process has to be run with the proper die length, uniform and constant melt temperature and pressure. High melt temperature may lead to thermal degradation of the polymer [8] [7].

2.1.2 Coextrusion and Extrusion Lamination

Coextrusion coating and extrusion laminating are additional technologies to extrusion coating for manufacturing multilayer structures.

Coextrusion performs as an integrated to extrusion coating process that makes it powerful method because of combining in single structure. It is a single-step process that begins with more than two different polymers. They are simultaneously conveyed in the coating extruder and combined in a single die to create a multilayer structured, thin sheet or film as presented on the left picture of Figure 2.2. Coextrusion is more attractive method compared to conventional multistep lamination and coating processes in terms of cost and complexity [8] [4].

In extrusion laminating, two different substrates are fed from the both side of the extruded film as distinct from extrusion coating and the illustration of the process is exemplified on the right picture of Figure 2.2. It seems like a three-ply process, that comprises of two substrates and molten film. However, it can be turned multilayer process, if coextrusion is utilized for the production of molten film. This process can be called as combined co-extrusion and lamination line [7].

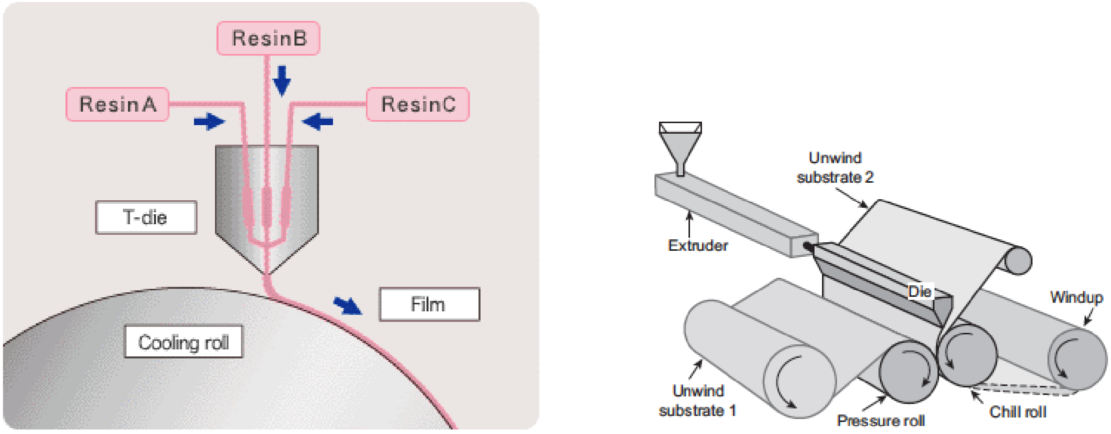

***Figure* 2.2** *Illustration of co-extrusion(on the left side,[7]) and extrusion lamination (on the right side, [16]) processes*

2.1.3 Materials for Extrusion Coating

An in depth understanding of the synergy between the polymer, the substrate and the process capability is essential if the extrusion coater is to manufacture products at optimum cost and performance balance. In this section, key substrates and polymers used for extrusion coating were discussed.

The handle and extensibility of a packaging film will depend on stiffness and thickness. Ideally, a film or laminate should have high stiffness and minimum extensibility. By depending on the purpose of the packaging and desired functions, suitable materials are selected.

Low density polyethylene(LDPE) was the first plastic for extrusion coating process approximately 60 years ago. Although LDPE still remains the largest single category of coating plastic, there are a wide range of polymers that are applicable for extrusion coating nowadays. They are listed in the following [8]:

Polyolefins Polyethylene (PE) and Polypropylene (PP)

Copolymers Ethylene vinyl acetate (E/VAC), Ethylene butyl acrylate (E/BA), Ethylene methyl acrylate (E/MA) and Ethylene ethyl acrylate (E/EA)

Adhesives Acid copolymers and Modified polyolefins (MPO)

Other polymers Polyesters (eg. Polyethylene terephthalate (PET)) and Polymethylpentane (PMP)

Biopolymers Starch, Polylactide (PLA) and Polyglycolide (PGA)

The most common substrates used in extrusion coating are paper, paperboard, plastic film and aluminium foil.

2.1.4 Common application

The major application area of extrusion coated and laminated products is food packaging and this is specified as given in the following:

- liquid packaging for milk, wine or other liquids
- frozen food packaging for ready-to-serve foods
- heavy duty packaging for sugar, salt or grains
- light packaging for moisture-absorbing foods, spices, seasoned goods, processed meat or medical products
- gas film packaging and vacuum packaging for heat sterilization

Additionally, it is applicable for other packaging purposes such as photo film packaging, photo sensitive paper, agricultural ans industrial chemicals, packaging of machine parts, detergent cartoons and cup, plate boards for oven [21] [22].

2.2 The Differences in Extrusion Coating of Polyesters and Polyolefins

A number of studies have been conducted to explain the effects of processing parameters, rheology and morphology of polymers on extrusion coating process. The significance of this part is to understand what the challenges are in extrusion coating of polyesters and how they can be solved and improved by comparing extrusion coating of polyolefins. Extrusion coating process is generally preferred and designed to laminate paper by using polyolefins, especially LDPE. The reasons why the polyolefins are commonly used in extrusion coating are that they are low-cost materials, easy to process and applicable for many applications. By depending of changing life style and demands, polyester, PET, has been commonly used for several years. Especially nowadays, it is quite attractive for production of dual-ovenable trays [8] [6].

Molecular weight, molecular weight distribution and long or short chain branching degree have inevitable effects on processing of polymers. Therefore, the basic chemistry of PE and PET will be reviewed in the following .

The PE comprises of two carbon atoms and four hydrogen atoms with zigzag backbone, that is presented in the first scheme of Figure 2.3, and it is formed by undergoing the radical polymerization. The most common polyethylenes are LDPE, HDPE and LLDPE as seen in the 4th, 5th and 6th scheme of Figure 2.3. LDPE is unique one with regards to its high degree of long branching. Crystallinity of PE is determined with its degree of branching. Side chains prevent the crystallization of linear main chain and this situation leads to decrease in the density, crystallinity [13].

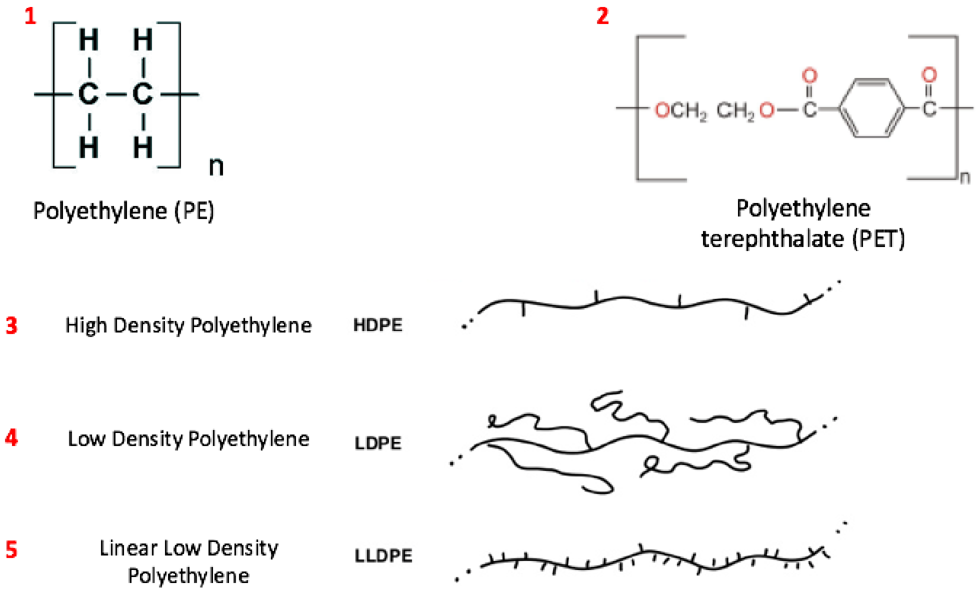

Figure 2.3 The chemical structure of polyethylene (1), polyetyhlene terephthalate (2) and different polyethylenes (3,4,5) [24]

For the production of PET, monomers are manufactured first by esterification reaction and it is schematically illustrated in Figure 2.4. Then, PET is manufactured by polymerization of polymers in one or more than one polycondensation reaction. The number of polycondensation reaction can be changed according to desired average Mw of PET. The chemical structure of PET is presented in the second scheme of Figure 2.3. As a by-product of this reversible reaction, water is formed. It means that esters and polyesters can exposure hydrolytic degradation in the presence of moisture at average temperature. Hence, it is vital to dry with desiccated air pellets before processing, modify the screw design slightly and utilize the air pins with a minimum air gap [6].

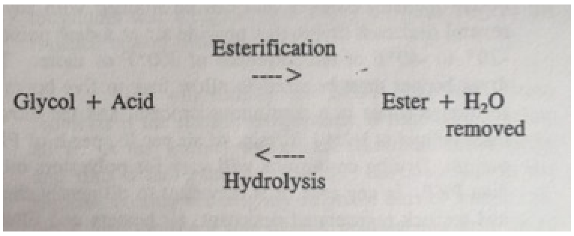

Figure* *2.4 *The production of the monomer of polyethylene terephthalate by esterification reaction [6]*

In typical processing conditions, PET has poor melt strength and more sensitive shear compared to LDPE. The reason of this situation is that PET has relatively low molecular weight and narrow molecular weight distribution than LDPE and LDPE has high melt strength through its high degree of long chain branched structure [15]. In addition, PET tends to neck-in more than LDPE due to its narrow MWD. Neck-in is one of the key factor in extrusion coating and indicated how much coating draws in across the web. If the neck-in increases, thickness of the material increases and this means high material cost. Since LDPE has quite broad MWD, it can be accepted as an excellent polymer in terms of neck-in. Even it seems challenging for PET compared to LDPE, neck-in can be improved for PET by decreasing air gap [2].

As seen in Figure 2.3, PET includes oxygen molecules and they provide some polarity to PET. The surface tension of PET, 42 mJ/m^2 is higher than surface tension of LDPE, 32 mJ/m^2. The natural high surface tension of PET is the result of its polarity. Although it has polarity, it does not have still any tendency to oxidize in air gap in the same way with LDPE [20].

The another reasons why PET is challenging for extrusion coating are due to its high melting temperature, intrinsic viscosity and density. PET has quite high melting temperature, 235°C, compared to LDPE, 120-136°C, therefore the melt temperature of PET in extruder is relatively high. Although high melt temperature is good to obtain great adhesion of polyester to the substrate, it leads to additional neck-in and low viscosity. The both of them together make complicated to control the output. To prevent this situation, lower line speed values are suggested [6] [20].

As mentioned before, extrusion coating process is mainly designed for LDPE. Since properties of PET differ from LDPE, screw design of extruder is another major concern for extrusion coating of PET. When it is considered that frequent screw changes are challenging, costly and loss of time, the use of hybrid, all-purpose extruders is required. The screw having a shallower, longer feed and a shorter transition section with metering is able to handle several materials including polyethylene and polyester. The one critical thing related screw design is barrier flight clearance. It is tighter for LDPE that brings on generating excessive frictional heat with PET [6].

2.3 Theory of Adhesion

In this part, the theory of adhesion will be discussed and by depending of the characteristics of PET, it will be explained which adhesion theory is suitable for the adhesion of PET to cardboard or not.

Adhesion represents that the tendency of dissimilar bodies are being together by providing initimate interfacial contact. As a precondition for great adhesion, wetting is a key factor since it provides to close contact between substrate and adherent. Wetting and penetration can be considered as same in the sense of the phenomena of adhesion [25]. Actually, they differ from each other. Wetting can happen in both porous and non-porous substrate; in other respects, the adhesion phenomena of penetration is more physical for porous subtrates, that will be discussed in following sections (see in 2.3.1 Mechanical Interlock Theory) [22].

Surface energy and polarity of both adherent and substrate are critical elements in terms of wetting. To obtain great adhesion, surface tension must be enough to wet out the surface. Furthermore, the polarity of both participants should be compatible with each other to provide optimum wettability and this is happened as an independently from temperature. In the figure, great and poor wettings of liquid adherent over a solid substrate are exemplified [22].

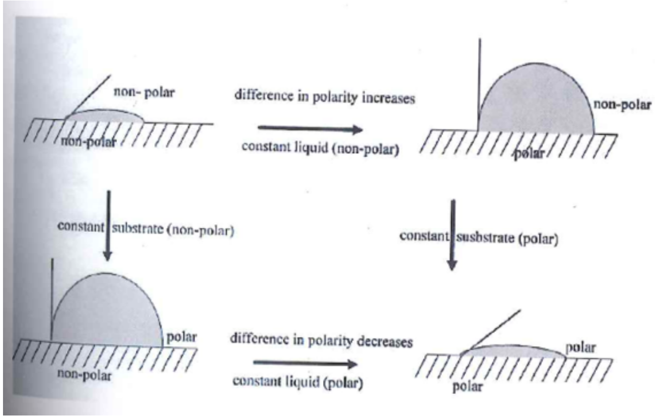

Figure 2.5 *Polarity miss-match effect on contact[22]*

It is well known fact that wetting of a surface by a liquid is affected by the roughness of the surface. The analysis of three-dimensional situations related to uneven or heterogeneous solid surface is one of the major theoretical problem. In practice, it states that the physical properties of the surface (eg. surface roughness, shape and particle size) affect wetting behavior as much as the chemical properties of the surface (eg. heterogeneity). The contact angles are key characteristics of wet processing of solid substrates such as froth flotation since they provide a simple and yet effective evaluation of the hydrophobicity of a solid surface [18].

There are five main adhesion theories: physical bonding, chemical bonding, diffusion and interdiffusion theory, mechanical interlock theory and weak boundary layer theory. In the case of polyethylene terephthalate adhesion, the expected adhesion theory is mechanical interlock theory, especially penetration by considering characteristics of PET and its adhesion onto paperboard substrate.

2.3.1 Mechanical Interlock Theory

The basic idea behind the mechnical interlocking theory is that when adherent materials or adhesive may enter the irregularities (pores) onto the substrate prior to hardening, it contributes adhesive bonds with porous materials such as wood, textiles paper. Therefore, the mechanical coupling or interlocking is one of the key adhesion mechanism for adhesion to the surface of the porous substrates [1][11].

In Figure 2.6, the mechanism of mechanical interlocking is illustrated. The theory states that surface roughness and irregularities provide to increase the contact area between substrate and adherent. Thus, adherent can penetrate easily to depths of hundreds of micrometers once the pore openings are getting larger. The depth of penetration is based on the porosity of wood, the viscosity of the molten adherent material, the pressure and duration of bonding [1][11].

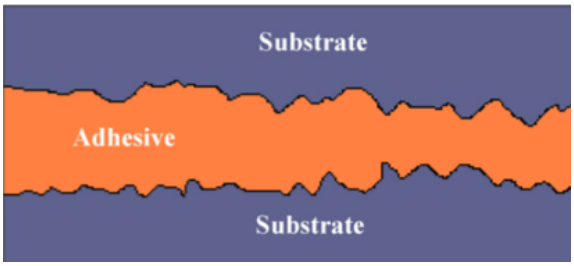

***Figure* 2.6** *Illustration of mechanical coupling between two substrates [1]*

The adhesion is created by penetration of the molten polymer into the pores and around fibers, solidification of melt and thus forming interlocks in a macroscopic level. Penetration is important for the mechanical interlocking theory since the requirement for an intimate contact between polymer and substrate. The penetration of the molten polymer to the substrate can be enhanced by preheating the paper before it reaches the nip and/or increasing the chill roll temperature. The heated substrate and warmer chill roll allow the molten polymer to flow on the substrate for a longer time before solidification and in addition to permit relaxation of the polymer chains. This makes mechanical interlocking easier and also relieves stresses created by crystallization that would reduce the adhesion [22] [1] .

2.3.2 Surface Tension and Energy

A direct measurement of intermolecular forces is called as surface tension. As a result of attraction of the bulk material for the surface layer, the tension is created in surface layers and the attraction has a tendency to decrease he number of molecules in the surface region. By depending on the tendency, the distance between the molecules is getting larger and to fulfill this, it requires work to be done and returns work to the system upon a return to a standard configuration. This situation answers the question why tension exists and why there is a surface free energy. The advantage of this thermodynamical mechanism compared to other adhesion mechanism, it is not necessary to provide a molecular interaction to obtain great adhesion. It is enough to keep the process equilibrium at the interface [22] [25] [1].

In neutral environments such as air, the thermodynamics of the polymer system will attempt to reduce the surface free energy as minimum as possible by orientating the surface into the non-polar region of the polymer. Once the surface of the polymer is in contact with a polar substance like water, great adhesion needs that the interfacial tension should be minimized. There are three zones for liquids interacting with polymers and that the surface tension of the solid is a function of the surface tension of the liquid. In Zone 1, which is called as the unperturbed zone, the surface tension of a polymer seperate from the surface tension of the wetting liquid. There is a linear dependence between the surface tension of the polymer and the surface tension of the liquid in the other two zones, called as the depolarisation zone (polymer surface tension is lower than the unperturbed zone), and the additional polarisation zone (polymer surface tension in the unperturbed zone is lower than in the additional polarisation zone) [25].

The surface free energy of liquids is identical with their surface tension. With solids, the phenomenon is getting more complex because of stretching and stresses. The most basic methods for surface energy estimation depend on upon contact angle measurements. There a drop of a known liquid is dropped on to the surface. It will be exhibit a sphere with a specific contact angle, from which the surface energy can be found. When a droplet of a liquid is in equilibrium on a surface, as in Figure 2.7., it adopts a form to minimize the potential energy [25] .

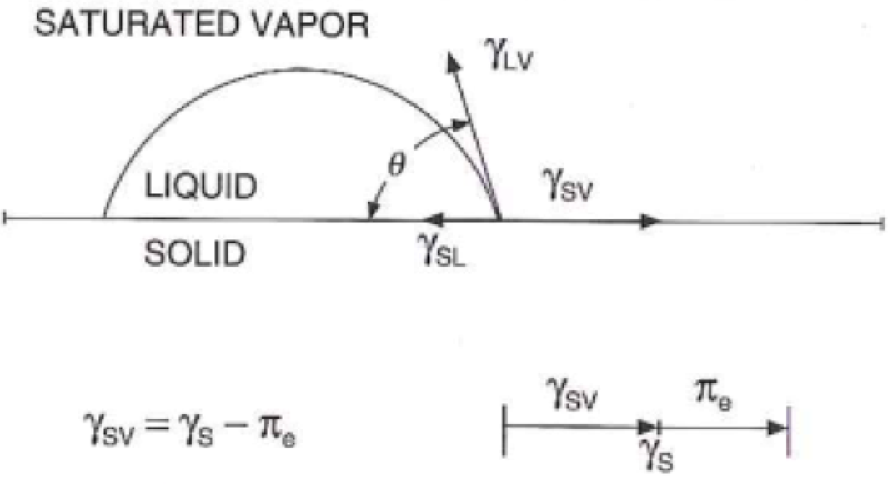

***Figure* 2.7** *Contact angle equilibrium [25]*

When a droplet of a liquid is in equilibrium on a surface, as in Figure 2.7, it adopts a form to minimize the potential energy. Considering the adsorption of vapour and thus the reduction in solid surface energy in the form of spreading pressure or equilibrium film pressure, $\pi_S(\gamma_{SV} = \gamma_S - \pi_S)$, is given in [25] [22] Equation 2.1:

$$\gamma_S = \gamma_{SL} + \gamma_{LV} cos\theta_{SL} + \pi_S \tag{2.1}$$

where SL is the term for surface in contact with the liquid , and LV is the term for liquid in contact with its vapour, θ_{SL} designates the contact angle formed between the liquid and solid, γ_S refers to the surface energy of solid that is measured precisely only in vacuum. By following this approach, the reversible work of adhesion between a liquid droplet and a solid surface at rest can be calculated once neglecting the spreading pressure and the related equation is presented in the following[25] [22] :

$$W_A = \gamma_S + \gamma_{LV} + \gamma_{SL} = \gamma_{LW}(1 + cos\theta_{SL}) \tag{2.2}$$

In the Equation 2.2, the term, W_A represents the work of adhesion and the term, γ_{LV} represents Lifshitz-van der Waals, the capital of all long range interactions. Unit for surface tension is generally $dynes/cm$ or mN/m and for the surface energy mJ/m^2 [25] [22].

The surface tension is based on molecular parameters such as molecular weight and chemical compositions of the polymer. Decrease in surface tension is of critical importance in the terms of adhesion to improve the wetting of the substrate material, which allows the chemical bonding to be more effective. The surface tension of polymer can be decreased by blending and functionalizing polymers and that contributes the penetration and spreading. Spreading states that solid surface is replaced by a solid with liquid interface and a liquid surface after the contact. Spreading happens once the contact angle is equal to zero, and it also indicates the movement of liquid with respect to the contact angle. A precondition for total wettability and spontaneous spreading is that the work of adhesion is higher than the cohesive energy, ($W_C = 2\gamma_L$), of the liquid [25]:

$$W_A \geq W_C \tag{2.3}$$

For instance, the surface tension of polyethylene terephthalate, 43 dyne/cm is higher than surface tension of polyethylene, 31 dyne/cm. Therefore, total wettability and the work of adhesion of polyethylene is slightly greater than polyethylene terephtha-

Table 2.1 *Goals of process varying and most common variables affectingto each goal*

Goal	Variable
Oxidation	Melt temperature, nip distance, time in the air gap, ozone, crystallinity
Penetration	Melt Temperature, viscosity, nip pressure, coating weight, rough and warm substrate
Wettability	Polarity, functional groups, primers, surface energy modifications

late. To reach good enough adhesion in challenging conditions where the polyethylene has, the one of the crucial thing should be considered reduction of surface tension and energy of polyethylene terephthalate [22] [25] [1].

2.4 Practical Methods For Adhesion Improvement

This chapter focuses on practical methods used for adhesion improvement in extrusion coating. Information revealed in the literature concerning PET is limited, therefore the section can not introduced comprehensively.

Process parameters such as melt temperature, air gap, line speed and chill roll temperature on adhesion have huge impact on adhesion, therefore they are the most studied topics in extrusion coating. By depending on this, they are discussed in this section. For example references [17] [9] [3] [8] [12] [6] [21] give a detailed description of process conditions variables and their input the adhesion. Loosely, it can be said that adhesion improvement in extrusion coating depends on the rate of oxidation, penetration and wettability. In Table 2.1, the most important and well-known variables are listed. Due to characteristics of PET, penetration was studied mainly as an adhesion improvement methods in the thesis.

Besides, processing parameters, type, degree of the resin and blending adhesion promoting additive or polymer have inevitable effect to enhance adhesion. In thesis trials, two different grade of PET were used. Therefore, it does not mention effect of blending adhesion promoting additive or polymer in this part of the thesis.

2.4.1 Resin

Resin viscosity and type play key roles in adhesion. To select the most appropriate grade for a particular extrusion coating application, there are so many variables and contradictory requirements by based on desired laminate properties and processing performance. Heat sealing, curling, flexibility, impact strength, adhesion. oil resistance and barrier properties can be given as an example.

Since adhesion is the main focus area in the thesis study, two different PET grade were analyzed. It is expected that a lower viscosity (high melt index) PET grade will penetrate to porous substrate and achieve higher adhesion level according to literature[21].Low viscosity resin is able to penetrate more porous substrate so then it can form a strong mechanical bonding upon cooling [3] .

2.4.2 Substrate Treatment

Paper and paperboard are commonly used substrate in extrusion coating, however their surface properties are not enough to obtain great adhesion due their low surface energy, incompatibility and chemical inertness, the presence of contaminants and weak boundary layers. In that case, surface treatments provides to form special functional groups on the surface of the substrate. By arranging crystallinity and roughness of the surface and/or removing contaminants and weak boundary layers, surface energy can be increased, special interactions with other functional groups can be created and surface cross-linking can be improved [12].

There are four main substrate treatments method to enhance the adhesion of the coating to the substrate: flame, corona, ozone and plasma treatments. Flame, corona and plasma treatment changes physical or chemical properties of a thin surface layer without altering bulk properties.In this part of thesis, only corona and flame treatments are examined by based on scope of the thesis. Ozone treatment is not in question for this case, since it is commonly utilized to improve the rate of oxidation of polymer film in extrusion coating. In addition, there is a traditional method which is chemical primer that is used where the other methods are low-efficient [12].

Flame Treatment

In flame treatment, the thermic energy of a gas flame is used to create oxygen radicals and they improve the polarity and surface energy of substrate by attacking the substrate surface. The main reasons why flame treatment is utilized paper and paperboard substrate are to [9][3];

- clean the contamination on paperboard like dust and foreign particles
- remove naps and excess proud surface fibres
- improve the bonding between substrate and adherent by cross-linking of oxygen and releasing N_2 molecules onto the substrate from flame.

In flame treatment, combustion reaction is occurred that is rapid exothermic reaction between fuel gas and atmospheric oxygen. As a fuel gas, propane (C_3H_8), natural gas or methane(CH_4) and butane(C_4 H_{10}) are commonly utilized in flame treatment. The gases are combusted with atmospheric oxygen and at the end of the reaction, water and carbondioxide are generated. [9][12].

In this study, propane gas was used trough adhesion trials that was performed to understand effect of flame treatment on adhesion.

The structure of single flame presented in Figure 2.8 can be examined into three conical zones

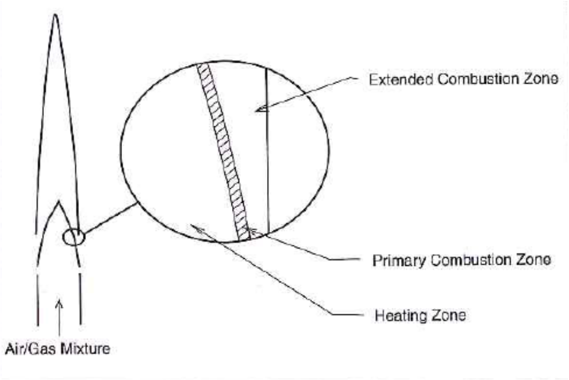

Figure 2.8 *Flame structure [12]*

Heating zone (1st zone) : The mixture of air and gas is ignited with 500°C.

Primary combustion zone(2nd zone) : Actual combustion starts. Flame has a deep blue and shining cone. Just above the primary combustion zone, the flame reaches its highest temperature, 1750°C, in which ion activity and concentration are at maximum levels. The active area of the flame is utilized to treat surface.

Secondary combustion zone(3rd zone) : Temperature decreases along with treatment efficieny.

Furthermore, flame treatment properties are as crucial as flame treatment equipment. To obtain great flame treatment results on substrate, there are some key parameters [9]:

- the rate of gas
- the air to gas ratio
- the distance between burner and substrate
- the line speed

Corona Treatment

Corona treatment is one of the surface treatment methods by utilizing low temperature corona discharge to notify changes on the surface properties. To generate the corona plasma, high voltage is applied to an electrode that has a sharp tip. By implementation of high voltage, the air is ionized in the gap of the treater and a gaseous conductor is formed. Ozone, atomic oxygen, oxygen free radicals and the free electrons comprise of the corona [3][12] .

The free electrons includes 10 ev energy by the time and they use the energy to reach the substrate surface. Typical bond energies found in treated substrates are given in Table 2.2.

The corona treatment improves surface energy by creating polar groups on the surface of the substrate. This improves wetting and adhesion properties of the surface. Other adhesion mechanisms include crosslinking of surface regions and increasing

Table 2.2 *Typical bond energies found in treated substrates [3]*

Bond Types	Bond Energy (ev)
H-H	4.5
C-H	4.3
C-C	3.6
C-O	3.6
O-H	4.8
O-O	1.4
C-C	7.4
O_2	5.1

the cohesive strength of the substrate by elimination of low molecular weight fractions from the surface, altering of the surface morhpohology and improving surface micro-roughness and formation of electret on the surface [12].

Compared to flame treatment, corona treatment does not last longer and leads to the treatment of the reverse side. In the sense of adhesion, some studies claim that flame treatment improves adhesion properties better than corona treatment. However, some of them completely conflicts the studies or state that they have equal contribution. It is difficult to make a decision one way or the other for this issue, since it is based on interaction of treated substrate with utilized polymer as well. Therefore, it is widely recommended to use both of them together to attain the best possible adhesion.

2.4.3 Melt Temperature

There are two key points related to melt temperature in extrusion coating: its uniformity and the degree of melt temperature.

Non-uniform temperature may be cause bands of variable adhesion across the web. In the case of polyester adhesion,the degree of feed zone temperature in one of the key parameter to provide melt temperature uniformity in the extruder [3] [21] .

Since, PET has quite high melting temperature, 235°C, the melt temperature of PET in extruder is higher than 235°C by based on the degree of PET. It is favorable to set the melt temperature high to keep the melt hotter through the air gap so the polymer can stay slightly longer in molten state at the nip point. Additionally, it improves flow-ability of the polymer in an around tiny fibers or imperfections or on the substrate surface by reducing viscosity. Besides, it is critical to determine optimum melt temperature. Excessive melt temperature may lead to form unwanted smoke, discoloration, odour and thermal degradation of polymer [3] [21] [2].

2.4.4 Air Gap

The distance between the die lip and the nip to chill roll contact point is called as air gap shown in Figure 2.9 or draw distance and the time is spent in the air gap which is called as time in air gap [8] . When the polymer film is flowing in the air gap, it contacts with the air in certain time so then the distance and the time in air gap (TIAG) play key role for the performance of polymer coating and penetration to porous substrates like paper and paperboard [17] [8] .

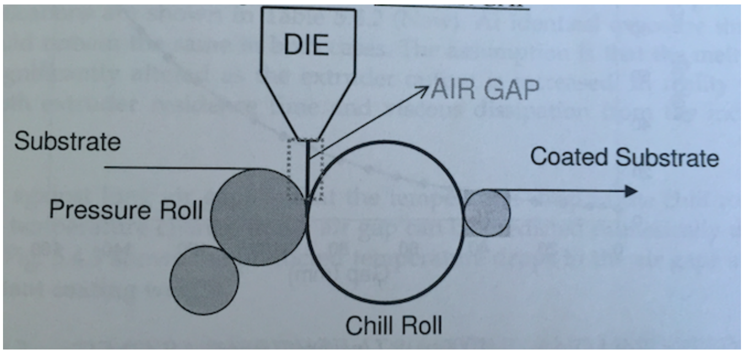

Figure* *2.9 *Schematically representation of position of air gap in extrusion coating [8]*

Adhesion increases once the length of the air gap increases so then polymer coating thickness and neck-in increase. High neck-in values is not desired in extrusion coating because it makes ineligible coated paperboard due to narrow coating width to manufacture final product [21].

The optimizing of length of air gap is crucial in the sense of the degree of oxidation and cooling in the air gap. It is known that adhesion increases with increasing TIAG despite an increase in cooling in the air gap. Since, chemical bonding increases with increase in TIAG. In other words it improves oxidation[17]. However, in the case of adhesion of PET to the paperboard, chemical adhesion is not expected by depending on characteristics of PET. Therefore, it suggests to keep air gap as minimum as possible if great adhesion is desired in thinner coatings [6].

2.4.5 Line Speed

When line speed is reduced, the time, the polymer is in the air gap, increases and by depending on this, the amount of heat loss increases. Decreasing time spent in the nip, by increasing the line speed, would be expected to get polyester coating closer to the center of the nip impression before solidified, and therefore also improve adhesion [6] [8].

Furthermore, it is suggested to perform the process with higher line speeds if thinner coating is desired [21].

2.4.6 Chill Roll Temperature

The responsibility of chill roll in extrusion coating is to cool down the molten polymer.Additionally, chill roll surface determines the appearance of coated material and these can influence the adhesion in extrusion coating. A high gloss finish allows greater polymer contact with the surface which promotes sticking to the chill roll [3].

When chill roll is heated, it contributes to obtain better adhesion. However, the temperature of the chill roll influence the sticking of the polymer to roll. [3]. PET does not have sticking problem when heated chill roll is used. However, it is minor contributor to improved adhesion. [6].

3. EXPERIMENTAL PART

In this chapter, the details of the implementation steps for the thesis work are described. Throughout this chapter, the following steps will be covered respectively: target of experimental part, materials and machine used in the experimental part, thesis trials and analysis methods.

3.1 Target of Experimental Part

The overarching aim of the experimental part was to enhance the adhesion of PET coating on cardboard when the coating weight of PET was diminished on cardboard. For this purpose, 5 adhesion trials were performed to screen the effect of different grades PET on adhesion and to investigate the effect of key processing parameters and equipment modifications on adhesion. As extrusion coating in big scale line needs several kilos of polymer per hour, it was reasonable to look for an alternative method for screening, modifying processing parameters and equipment. Therefore, pilot line in packaging laboratory of Tampere University of Technology was utilized to perform adhesion trials.

To analyze the samples, three main and three additional testing methods were used. Main analysis methods were adhesion test by hand, grammage test and neck-in test. Additional analysis methods were heat sealing test, microscopy test and pinhole test. Additional test methods were applied if the adhesion level of PET on the cardboard reaches the level of 5 over 5 at the target coating weight. The reason of the approach is that sample preparations of these methods were time consuming and only yes-no based answers can be obtained.

In the following sections, utilized materials, process line, adhesion trials and analysis methods were explained comprehensively. Results obtained from adhesion trials were presented and discussed associated to each other.

3.2 Materials and Machine Used in Experimental Part

In the experimental part of the thesis, different grades of PET, LDPE and different grades of commercial cardboard were utilized as a main coating polymer, an encapsulation polymer and a substrate to manufacture thesis samples, respectively. Grades and properties of materials are presented detailed in the following sections.

The pilot line in packaging laboratory of Tampere University of Technology was utilized to perform adhesion trials and is described in the section of pilot line.

3.2.1 Substrate

Commercial cardboard was used through all thesis trials. It comprises of three layers. For the adhesion trials, two different grades of commercial cardboard were used.

3.2.2 Encapsulation Polymer

In all thesis trials, LDPE was used as an encapsulation polymer and also same grade LDPE was used for all trials. Utilized LDPE grade was especially designed for extrusion coating of high volume production of boards and papers applications. Besides that, the main reason of utilization of this grade of LDPE in all thesis trials is its high melt flow rate.

3.2.3 Coating Materials

The main coating polymer of adhesion trials was PET and two different grade of PET were used for manufacturing of the thesis samples; PET 1, PET 2. In this study, PET 1 was utilized for adhesion trial 1 and and PET 2 was utilized for adhesion trial 3, 4 and 5.

3.2.4 Pilot Line

As mentioned in the target of experimental part (see section 7), the pilot line of Tampere University of Technology in PAK laboratory presented in Figure 3.1 was used to screen effect of different grades PET and processing parameters on adhesion. In this pilot line, coextrusion coating, coextrusion laminating, cast film coextrusion, surface treatment, dispersion coating and edge trimming service can be performed.

Figure 3.1 *The pilot line line of Tampere University of Technology in PAK laboratory*

Sections of plot line were presented schematically in Figure 3.3. In extrusion coating process, a thin film of very hot molten PET was pressed onto a cardboard (substrate) passing through a cooled nip roll assembly. The substrate was fed from unwinder part to the process. Before combining the polymer melt with the substrate, corona treatment was applied to the substrate. According to run plan, flame treatment was applied as an additional pretreatment method for some trials. The PET melt at uniform temperature was drawn down into the nip formed by the chill roll and the rubber sleeved pressure roll. The substrate was simultaneously fed into the same nip where it combines with the molten polymer under pressure. Then, the cooled web was separated from chill roll and it continued by passing through stripper rolls and post heat treatment. Finally, it was pulled by haul-off with constant tension and winding equipment. During processing, chill roll has vital role to determine both product quality and productivity, therefore improvements on chill roll was performed to investigate effect on adhesion [8].

Main unwind, second unwind for the lamination, rewind, press roll, chill roll, edge trimming and plastic systems dehumidifiers and containers comprise of main units of the pilot line [19]. They were performed with various configuration combinations and processing parameters for each adhesion trial and these were presented as a table under the related section of thesis for each adhesion trial.

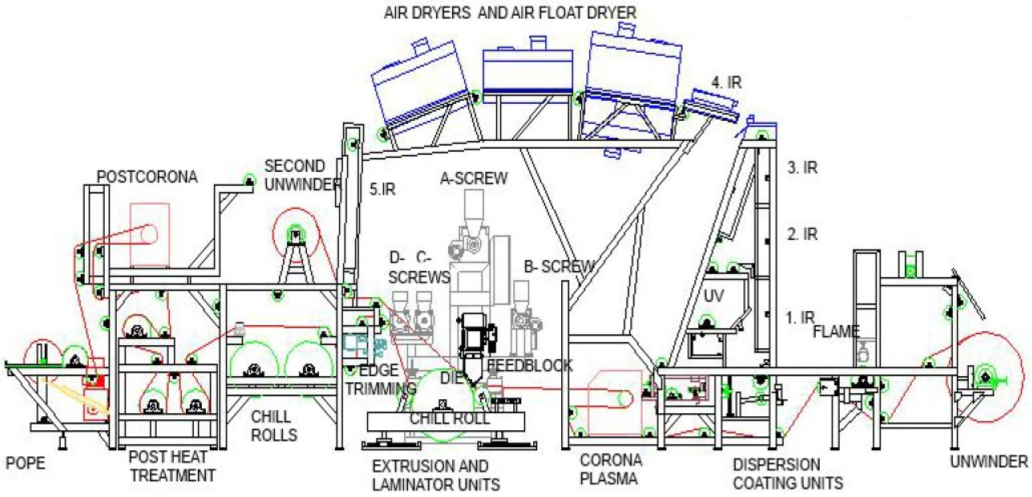

***Figure* 3.2** *Schematically representation of section of pilot line [19]*

***Table* 3.1** *Machine specifications of Extruder A and D [19]*

	Extruder A	Extruder D
Diamater (mm)	60 mm	30 mm
L/D Ratio	30	25
Maximum Output (kg/h)	90 kg/h LDPE (changes depending on the polymer)	20 kg/h LDPE (changes depending on the polymer)
Transducers	Polymer melt temperature and pressure measurement transducers	Polymer melt temperature and pressure measurement transducers
Loader	Hopper loader	Hopper loader

There were four different extruders for coextrusion coating in the pilot line: extruder A, B, C and D. For thesis trials, two of them were utilized. Extruder A for main coating polymer and Extruder D for encapsulation polymer were used. Extruder properties of both type A and D were presented in Table 3.1. In the trials, the selector plugs design of the pilot line was web A-A-A-A-A D/D in order of web, multilayer structure and encapsulation. These two extruders were combined at the beginning of die system. As a die system, internally deckled extrusion T-type die was used and it has lip heaters and edge encapsulation system [19].

In the pilot line, corona, atmospheric plasma, flame, UV ozone were applicable surface treatment methods. Corona and flame treatment were applied as a surface treatment to subtrate for thesis trials. Vetaphone Corona-Plus was utilized for corona treatment with ceramic electrodes by performing maximum 4 kW power. On the other hand, Hill Gmbh, type EF 75-1 (Burner CE62-500) was used for flame treatment. It was performed by propane gas supply and maximum output heat was 50 kW. Also, flame treatment unit was moveable and it has adjustable air gap [19].

3.3 Thesis Trials

In the following, the target and method of the thesis trials are explained detailed. For the study, 5 adhesion trials were performed in extrusion coating line of Tampere University of Technology. Extrusion coating line was modified according to purpose of the adhesion trial. Firstly, processing parameters and process improvement methods were determined and they were repeated for different grades of PET. This approach was chosen to obtain senseful and comparable data about interfacial characteristics various grades of PET when adhered to cardboard in changeable processing conditions. Furthermore, screening trial was done to understand behavior of the polymers in determined processing conditions for utilized each grade of PET.

During each thesis trial, extrusion coating line was run individually for each test series. After each test series was completed, the process line stopped and the coated cardboard roll was unwind. With this approach, fluctuation problems because of rapidly changes in processing were minimized. In each test series, approximately 20 m coated cardboard was manufactured for each test points.

3.3.1 Adhesion Trial 1

In adhesion trial 1, the effect of flame pretreatment and air gap on adhesion was investigated by using PET 1 as main coating polymer and thick commercial cardboard as a substrate. For this purpose, the trial was divided into two stages: screening part and actual part. The detailed processing parameters of both screening and actual parts of adhesion trial 1 and the configuration of used equipment were listed in Table 3.2.

By following the target of the thesis, flame was utilized as a pretreatment method in actual part additionally to investigate the effect of flame pretreatment on adhesion.

Table 3.2 *The list of processing parameters and equipment of adhesion trial 1*

Processing Parameters and Equipments	Screening Part	Actual Part
Equipments		
Extruder	A	A
Plug	AAAAA-DD	AAAAA-DD
Screenpack	Standard	Standard
Nip roll	Medium grade	Medium grade
Chill roll	Configuration 1	Configuration 1
Pretreatment Methods	Corona	Corona and Flame
Parameters		
Deckles (mm)	550	550
Die gap (μm)	600	600
Nip pressure (bar)	5.50	5.50
Melt temperature (°C)	high	high

Flame treatment has not been already installed to extrusion coating line. Therefore, process line was modified and flame treatment was placed after corona treatment. This configuration was shown in Figure 3.3. In Figure 3.3, the picture on left side flame treatment can be seen and the position of the equipment can be understood by compared to the picture on right side. There is a roll in the front of orange cable at the right picture and the flame treatment was installed above that roll. Corona treatment was just behind of this configuration which was placed next to yellow big pipes as given in the picture on right side.

In the screening part, only corona was used as a pretreatment method and in the actual part, both corona and flame were used. The reason of why corona pretreatment is mandatory for both cases is that the adhesion level of polymers on the board can not be higher than 2 without using corona by based on reference data related this project. Furthermore, 5 test series were designated by based on combination of different screw speed and line speed values and each test series was run by 7 different air gap levels from level 1 (minimum) to level 7 (maximum). Thus, the effect of air gap on adhesion was examined as well. Both screening and actual part were performed by following same test series and process variables to obtain comparable results. In Table 3.3, the logic of test series and process variables of the adhesion trial 1 was illustrated.

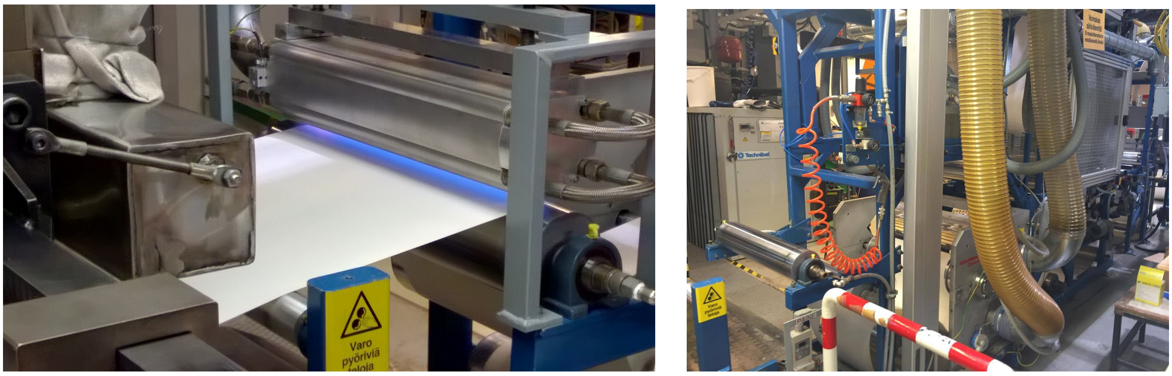

***Figure* 3.3** *The position of flame treatment in extrusion coating line during actual part of adhesion trial 1*

***Table* 3.3** *Test series and process variables data of the adhesion trial 1*

Test Series			Process variable: Air gap (mm)						
	Screw Speed (rpm)	Line Speed (m/min)	TP-1	TP-2	TP-3	TP-4	TP-5	TP-6	TP-7
TS-1	level 1	level 3	level 1	level 2	level 3	level 4	level 5	level 6	level 7
TS-2	level 2	level 3	level 1	level 2	level 3	level 4	level 5	level 6	level 7
TS-3	level 3	level 4	level 1	level 2	level 3	level 4	level 5	level 6	level 7
TS-4	level 4	level 6	level 1	level 2	level 3	level 4	level 5	level 6	level 7
TS-5	level 5	level 6	level 1	level 2	level 3	level 4	level 5	level 6	level 7

3.3.2 Adhesion Trial 2

The aim of the adhesion trial 2 was to understand the effect of process equipment modifications and line speed on adhesion. In the trial, PET 1 as main coating polymer and thick commercial cardboard as a substrate were utilized like in adhesion trial 1. To understand effect of process equipment modifications, second configuration of chill roll was used.

Air gap was kept at the minimum level (level 1) by based on neck-in results of adhesion trial 1. On the other hand, the rest processing parameters were like in adhesion trial 1 and the details of them were presented in the Table 3.4.

Table 3.4 *The list of processing parameters and equipment of adhesion trial 2*

Processing Parameters and Equipments	
Equipments	
Extruder	A
Plug	AAAAA-DD
Screenpack	standard
Nip roll	Medium grade
Chill roll	configuration 2
Pretreatment Methods	Corona
Parameters	
Deckles (mm)	550
Die gap (μm)	600
PET 1 Nip pressure (bar)	5.50
Melt temperature (°C)	high
Air gap (mm)	level 1 (minimum)

Table 3.5 *Test series and process variables data of the adhesion trial 1*

Test Series		Process variable: Line speed (m/min)				
	Screw Speed (rpm)	TP-1	TP-2	TP-3	TP-4	TP-5
TS-1	level 1	level 1	level 2	level 3	level 4	
TS-2	level 2	level 1	level 2	level 3	level 4	level 6
TS-3	level 3	level 2	level 3	level 4	level 6	level 9
TS-4	level 4	level 2	level 3	level 4	level 6	level 9
TS-5	level 5	level 3	level 4	level 6	level 9	level 10

Since the effect of line speed was observed, the machine was performed with various level of line speed at determined screw speeds. During trial, 5 test series were designated according to screw speed values changing from level 1 to level 5 and each test series were performed by determined line speeds values changing from level 1 (minimum) to level 10 (maximum). However, each test series was not able to perform exactly same line speed because of runnability problems. In Table 3.5, the logic of test series and process variables of the adhesion trial 2 were exemplified.

Table 3.6 *The list of processing parameters and equipment of adhesion trial 3*

Processing Parameters and Equipments	
Equipments	
Extruder	A
Plug	AAAAA-DD
Screenpack	Standard
Nip roll	Medium grade
Chill roll	Configuration 1
Pretreatment Methods	Corona
Parameters	
Deckles (mm)	510
Die gap (μm)	600
Nip pressure (bar)	5.50
Melt temperature (°C)	Low

3.3.3 Adhesion Trial 3

Adhesion trial 3 was a screening trial since different grade of PET was used that is PET 2. In this trial, screening part of adhesion trial 1 was followed by changing processing parameters slightly to adapt new grade of PET. For instance, melt temperature for adhesion trial 3 was set as lower values by based on typical moulding temperature and melting temperature of PET 2 and deckles distance was reduced to 510 mm. Detailed processing parameters and the configuration of used equipment of adhesion trial 3 were listed in Table 3.6.

Since it is a screening trial, the logic of screening part of adhesion trial 1 was followed For instance, test series were designated by based on the combination of screw speed and line speed and process variable was air gap values like in previous one. However, some line speed values were slightly changed for some test series. The reason of behind this situation is that the relation between screw speed and line speed has an effect on the polymer coating weight and according to that, these values should slightly modify to obtain polymer coating weight around target when the grade of PET is changed. These changes in test series and process variables data of the adhesion trial 3 were shown clearly in Table 3.6.

***Table 3.7** Test series and process variables data of the adhesion trial 3*

Test Series			Process variable: Air gap (mm)						
	Screw Speed (rpm)	Line Speed (m/min)	TP-1	TP-2	TP-3	TP-4	TP-5	TP-6	TP-7
TS-1	level 1	level 3	level 1	level 2	level 3	level 4	level 5	level 6	level 7
TS-2	level 2	level 4	level 1	level 2	level 3	level 4	level 5	level 6	level 7
TS-3	level 3	level 5	level 1	level 2	level 3	level 4	level 5	level 6	level 7
TS-4	level 4	level 7	level 1	level 2	level 3	level 4	level 5	level 6	level 7
TS-5	level 5	level 8	level 1	level 2	level 3	level 4	level 5	level 6	level 7

3.3.4 Adhesion Trial 4

The purpose of the adhesion trial 4 was to understand effect of process equipment modifications and line speed on adhesion like in adhesion trial 2, however distinctly from adhesion trial 2, PET 2 and thin commercial cardboard were used as a main coating polymer and substrate for the trial, respectively. The exactly same configuration of chill roll in adhesion trial 2 was utilized for adhesion trial 4. Furthermore, adhesion trial 2 was followed by changing processing parameters, which are deckles and melt temperature, slightly to adapt new grade of PET. The detailed processing parameters of adhesion trial 4 and the configuration of used equipment during the trial are listed in Table 3.8.

Although the logic of the adhesion trial 2 was followed for run data, process variable values were slightly changed to adapt new grade of PET like in processing parameters of this trial. During trial, 5 test series were designated according to different screw speed values changing from level 1 to level 5 same as previous one, however each test series were performed by 4 different line speeds values changing from level 1 to level 9 in this trial. For example, the highest line speed was level 9. Since, it was investigated that standard deviation of polymer coating weight was extremely higher than acceptable limits. In Table 3.9, the logic of test series and process variables of the adhesion trial 4 was illustrated.

Table 3.8 *The list of processing parameters and equipment of adhesion trial 4*

Processing Parameters and Equipments	
Equipments	
Extruder	A
Plug	AAAAA-DD
Screenpack	standard
Nip roll	Medium grade
Chill roll	configuration 2
Pretreatment Methods	Corona
Parameters	
Deckles (mm)	510
Die gap (μm)	600
Nip pressure (bar)	5.50
Melt temperature (°C)	low
Air gap (mm)	level 1 (minimum)

Table 3.9 *Test series and process variables data of the adhesion trial 1*

Test Series		Process variable: Line speed (m/min)			
	Screw Speed (rpm)	TP-1	TP-2	TP-3	TP-4
TS-1	level 1	level 1	level 2	level 3	level 4
TS-2	level 2	level 1	level 2	level 3	level 4
TS-3	level 3	level 2	level 3	level 4	level 6
TS-4	level 4	level 3	level 4	level 6	level 9
TS-5	level 5	level 3	level 4	level 6	level 9

3.3.5 Adhesion Trial 5

The adhesion trial 5 was performed to understand the effect of flame treatment and air gap like in the actual part of adhesion trial 1 by using PET 2 as a main coating polymer and thick commercial cardboard as a substrate. Process parameters and the configuration of used equipment were similar to adhesion trial 3 (See Table 3.6). There was only one difference between adhesion trial 3 and 5: pretreatment method. In adhesion trial 5, both corona and flame were utilized as pretreatment method.

Process line was modified again and flame treatment was placed before corona treatment and the configuration was shown in Figure 3.4. In Figure 3.4, corona treatment was placed next to yellow big pipes as given in the picture on right side and as it can be seen in same picture, there is a screen cabinet behind the corona treatment. Flame treatment was installed behind that cabinet in adhesion trial 5 and the close-up photograph of the configuration was given on the left side in figure 9. This configuration of the flame treatment was exact opposite from the actual part of adhesion trial 1 by based on results of the trial. In that trial, first corona was turned on then flame and as a result of this, substrate burnt. Therefore, the flame treatment was started first then corona in adhesion trial 5.

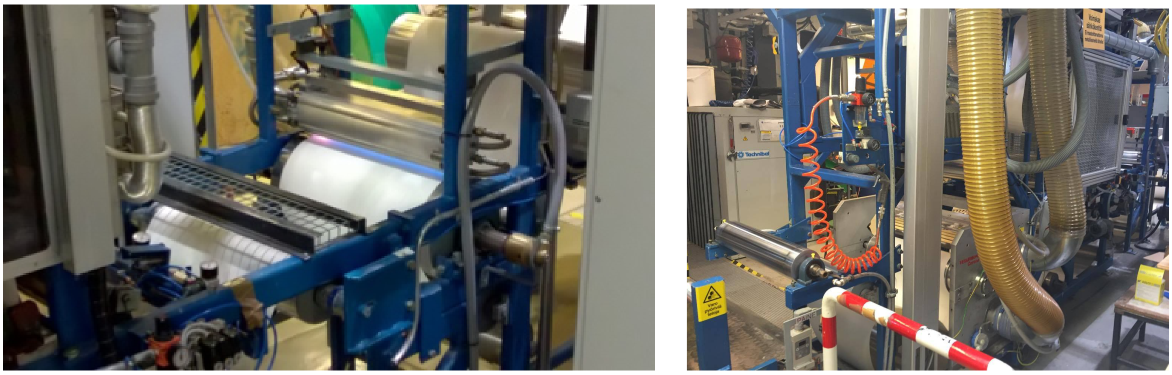

Figure 3.4 *The position of flame treatment in extrusion coating line during actual part of adhesion trial 1*

The logic of the adhesion trial 3 was exactly followed for run data of adhesion trial 5 (See Table 3.7). In this trial, one more test series was performed additionally. The parameters of the test series were that the screw speed was at level 1 and the line speed was at level 2.

3.4 Analysis Methods

In the following, the used testing methods are presented for analyzing the samples. The major part of testing was purposed and planned to improve the adhesion of PET to the cardboard in extrusion coating while diminishing coating thickness. By considering the target of the thesis, there were 3 main analysis methods which are grammage test, adhesion test and neck-in test and some additional test which were heat sealing test, pinhole test and microscopy test.

All samples were tested by same way to avoid experimental errors. By considering this, 5 parallel samples were taken in each test point for the analysis by starting from the beginning of the line and leaving approximately 30 cm interval between each parallel sample. Approximately 20 m coated cardboard line was manufactured for each test points in each test series.

3.4.1 Grammage Test

The aim of grammage test was to measure average polymer coating weight on cardboard. There are two types of grammage test method: conventional method and coating removal method. The reason of why two different grammage test method were used is that thick substrate material has high moisture holding capacity and it leads to obtain unacceptable error margin and standard deviation values in results. Because of that, we found some solutions to avoid from this situation:

- If the adhesion level was maximum 2, only the coated top layer were weighed, since it was easy to peel polymer coating from cardboard without fiber and with really less amount of fiber.
- If it was not easy to peel the coated layer and the adhesion level is 3 or higher than 3, the conventional method was applied first. If the standard deviation was found higher 2, it was given up from conventional method and continued with coating removal method.

For all grammage test methods, 5 different samples were prepared by cutting from middle of the cardboard in circle shaped for each test point and the illustration of the way of taking grammage test sample were shown schematically in Figure 3.5.

The size of each samples was 0.01 m^2 and to cut them, circle shape cutting equipment was used. The grammage test sample and circle shape cutting equipment were exemplified in Figure 3.6.

Precisa 500 M-200 C model analytical scale was used in all weight measurements that was shown in Figure 3.7.

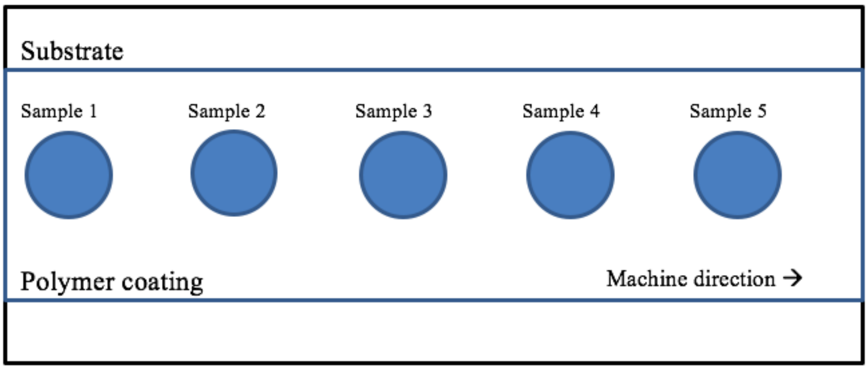

Figure 3.5 *Schematically illustration of the way of taking samples*

Figure 3.6 *The grammage test sample example and circle shape cutting equipment*

Conventional Method

In the conventional method, samples were weighed with substrate. Firstly, the average coating weight of substrate was found exactly. The substrate samples were prepared by following same procedure explained before. Then, each samples was measured and the average of them was taken.

After that, the weight of samples started to be measured. The difference between the weight of sample and substrate was calculated for each them. Finally, the average coating weight and standard deviation were calculated by using results of 5 different samples taken from each test points for all test series.

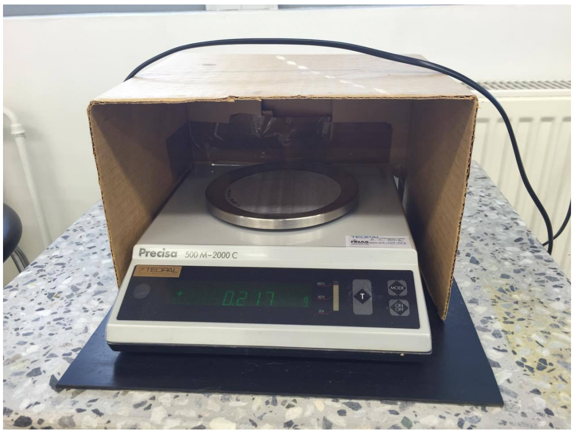

Figure 3.7 *Analytical scale, used in weight measurements: Precisa 500M-200C*

Coating Removal

This is a in-house method. The approach in coating removal is to remove fibers from plastic coating layer by using solution. If there is less amount of fiber, it does not need to leave in solution and in this case, the means of less amount fiber is that the adhesion level is 2 or less than 2. If there are huge amount of fibers, sample leaves in solution until all the fibers are removed from the surface of plastic film coating.

After that , coating weight is calculated by following same procedure explained in conventional method part.

3.4.2 Adhesion Test

Adhesion test is used to evaluate adhesion between polyolefins/ polyesters and fiber-based substrates. In this method, the polymer coating was separated from the fiber substrate. To separate, the symbol of X was drawn on the sample by using sharp curved knife like shown in the first picture of Figure 3.8.The crucial point was that only the film layer should be cutted not cardboard and the knife should be as sharp as razor. After that, film layer was peeled off from cross point through machine direction like shown in the third picure of Figure 3.8. This testing method is schematically shown in the following figure.

The evaluation of adhesion test was done by determining the extent of fiber tear like shown in the fourth picture of Figure 3.8. The evaluation was based on visual

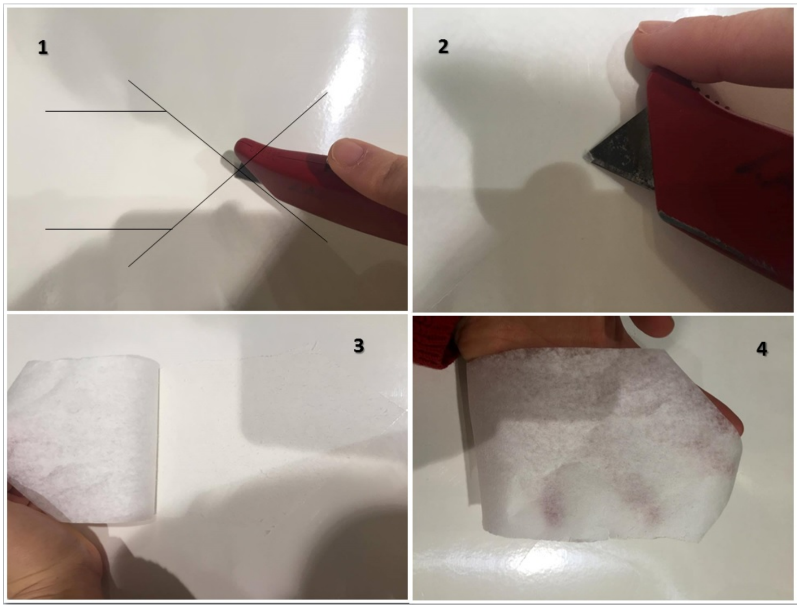

Figure 3.8 *The schematically representation of adhesion test by hand method*

observation. The criteria of evaluation was given in the following:

0 : Layers do not adhere

1 : Layers peel off each other

2 : Layers peel off each other, some fibers are removed

3 : Fiber tear <50% of surface area

4 : Fiber tear >50% of surface area

5 : 100% fibre tear (total fiber tear)

If cohesive failure occurred in one of the samples, the interface was not the weakest point, and good adhesion was not achieved.

3.4.3 Neck-in Test

Neck-in test was applied to find reduction in width. In this test method, the width of coated cardboard was measured from both end sides of the coated cardboard where the coating was finished. This test was applied to 5 different place in same coated cardboard sample for each test point. Then, the difference between width and deckles was taken for each measurement. Finally, the average and standard deviation of results taken 5 different place in same coated cardboard sample were calculated.

3.4.4 Additional Tests

Pinhole test, heat sealing test and microscopy test were comprises of additional test methods. These test methods were not applied all test points in each test series. Since, these test methods did not provide progressional results to improve the adhesion level of PET on the cardboard. For instance, it can be obtained results from pinhole test only by based yes or no. On the other hand, the sample preparation of microscopy and heat sealing test took too much time and the information related to the main target of the thesis did not obtained from them. Therefore, it was decided that additional test method will be applied if the adhesion level of PET on the cardboard reaches 5 at the target coating weight.

4. RESULTS AND DISCUSSION

In this chapter, the results from the trials are presented and discussed. Results were evaluated and presented mainly by based on adhesion level and process variable. Most of quantitative data was collected to utilize in qualitative purposes, therefore real statistics reading is not executed for the data. In the following sections, the graphical representation of results is presented for all trials. The numerical representation of results can be found in the Appendix A, B and C.

4.1 Results of Adhesion Trial 1

In the part, results from adhesion trial 1 are introduced. As mentioned in the thesis trials part, adhesion trial 1 comprised of two stages which are the screening part and actual part.

4.1.1 Screening Part

In this part, results of screening part of trial 1 were examined. As mentioned before, only corona was utilized as pretreatment method and the effect of air gap on adhesion was studied in the screening part of adhesion trials for PET 1 grade. Both main and additional test methods were applied to analyze the samples.

In the study, since the target was to reach great enough adhesion when the coating was diminished, the relation of coating weight and adhesion results was introduced firstly in Figure 4.1. It is obvious that adhesion level increases if coating weight increases. Our coating weight target is between 25 and 30 g/m^2. Once the range of coating weight was analyzed, the best adhesion level, 4.5 over 5, was obtained from test series performing at level 5 screw speed, level 6 line speed and level 4 air gap. As an overview, higher adhesion level results were obtained at higher screw speeds like level 4 and level 5, higher line speed, level 6, and over 28 g/m2 coating weight.

The main point was that the adhesion level increases if the melt temperature increases. Since high melt temperature reduces the viscosity of resin and slightly increases the solidification time of PET[5]. Therefore, it was one of the expected results to improve the adhesion. By based on the information, high adhesion level was obtained at high screw speed values because melt temperature increases by increasing of screw speed in extrusion coating.

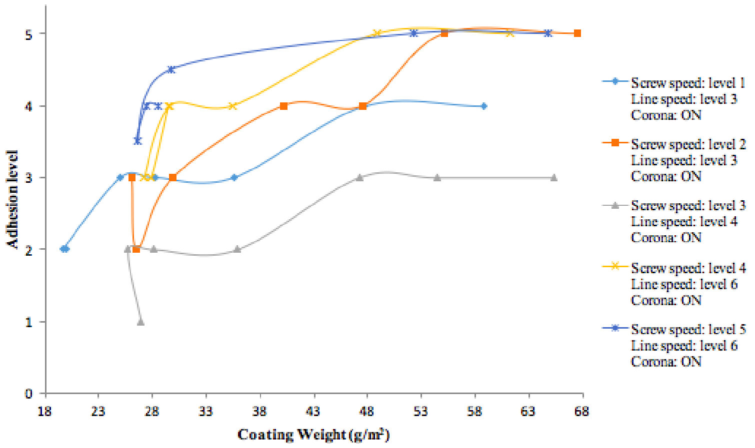

Figure 4.1 *Adhesion level versus coating weight(g/m^2) graph of screening part of trial 1*

In Figure 4.2 and Figure 4.3, the effect of air gap on adhesion and coating weight was represented. Normally, air gap should be kept as minimum as possible to reach great adhesion of PET on cardboard since large air gaps may lead to premature cooling of the melt and increase in viscosity. Inversely, higher adhesion levels were reached while performing with high air gap values according to In Figure 4.2. The reason behind the behavior is that coating weight was reached over 40 g/ m^2 at those levels as seen in Figure 4.3.

The target coating weight is determined by the line speed and the extruder output rate. In our case, increase in coating weight was based on changes in air gap, since line speed and extruder output rate was kept constant in each test series. According to literature [21], large air gap gives better drawdown but worse neck-in. Therefore, both neck-in and coating weight inreased by increasing air gap as seen in Figure 4.3 and Figure 4.4. Once neck-in values were getting higher, the width of the coating was getting narrower and thickness of the coating was getting higher. Getting thicker coating provided good adhesion cardboard. On the other hand, getting narrower coating weight caused to obtain ineligible and unusable width of laminate.

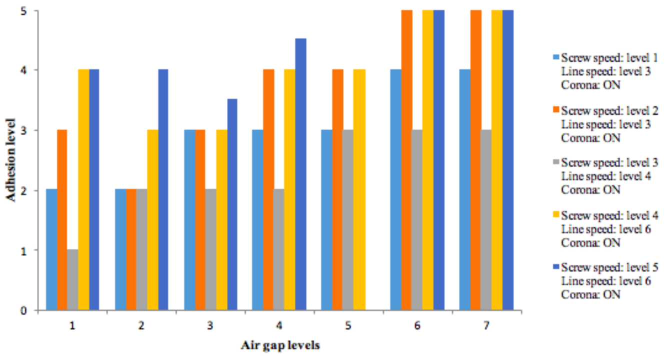

Figure *4.2 Adhesion level versus air gap levels graph of screening part of trial 1*

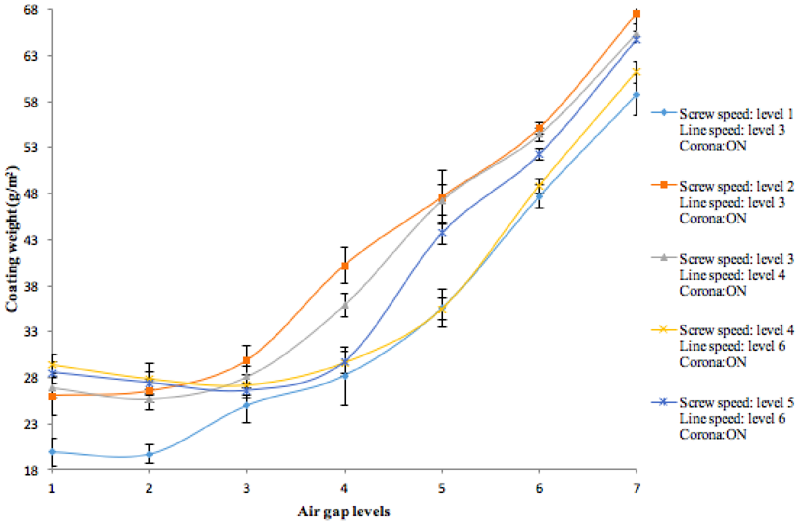

Figure *4.3 Coating weight(g/m^2)versus air gap levels graph of screening part of adhesion trial 1*

To understand effect of air gap on adhesion comprehensively, its effect on drawdown was examined. It was vital that PET 1 grade had good drawdown, particularly if low coating weights were to be obtained without causing tear-offs and voids. For this purpose, pinhole tests were applied to all test points in each test series and no pinhole observed for all of them.

Furthermore, edge trimming was another limiting factor by optimizing processing parameters of extrusion coating. To avoid from this situation, neck-in should be kept minimum. In our case, neck-in values were good enough even performing at low level air gaps without edge bead.

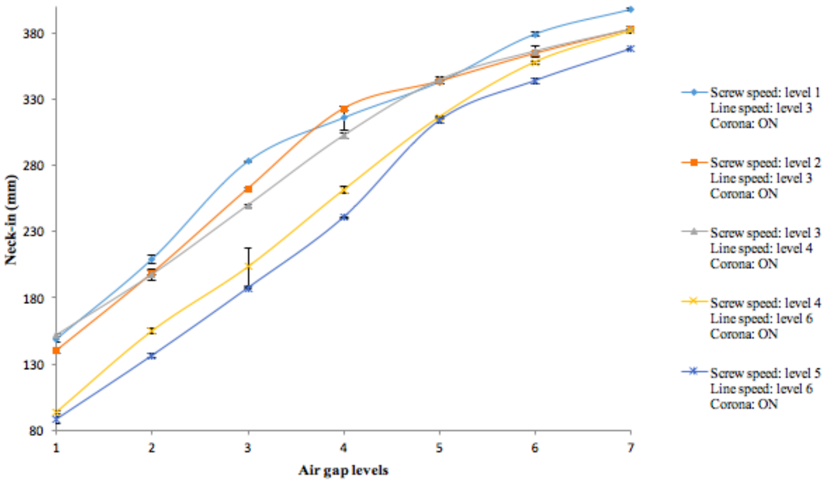

Figure 4.4 *Neck-in(mm) versus air gap levels graph of screening part of trial 1*

In conclusion, it was understood and decided according to results of screening part of adhesion trial 1 that:

- The most considerable result close to target was 4 over 5 adhesion level at 28.54 g/m^2 coating and it was obtained while processing at level 5 (maximum) screw speed, level 6 line speed and level 1 (miminum) air gap.
- Corona pretreatment was the key requirement in extrusion coating when it comes to adhesion; however it was not enough on his own to reach great adhesion with target coating weight for PET 1 grade.
- The air gap level must be kept as low as possible for PET 1 grade to obtain the maximum useable width of the laminate without voids and tear-offs.

4.1.2 Actual Part

After screening processing behavior of PET 1 grade, it was investigated in actual part whether both corona and flame pretreatment improve the adhesion of PET on cardboard or not. For this purpose, process was run by following same materials and processing parameters and samples were analyzed same as screening part. The only thing is that additional test methods were not used for actual part samples because of the reason indicated in results of screening part of adhesion trial 1.

In the actual part, adhesion level was quite low even coating weight results slightly higher than screening parts results by based on comparison of Figure 4.1 and Figure

4.5. If we looked the overall picture, the maximum adhesion level was 3 over 5 and adhesion values were slightly high at high screw speeds, line speeds and coating weights. However, test points obtained 3 over 5 adhesion values were out of target. Since the coating weight of samples was higher 30 g/ m^2 coating and neck-in were quite high at those points. On the other hand, the most acceptable adhesion level close to target was 2.5 over 5 in 26.56 g/ m^2 coating weight and it was obtained while operating at level 4 screw speed, level 6 line speed and level 1 (minimum) air gap.

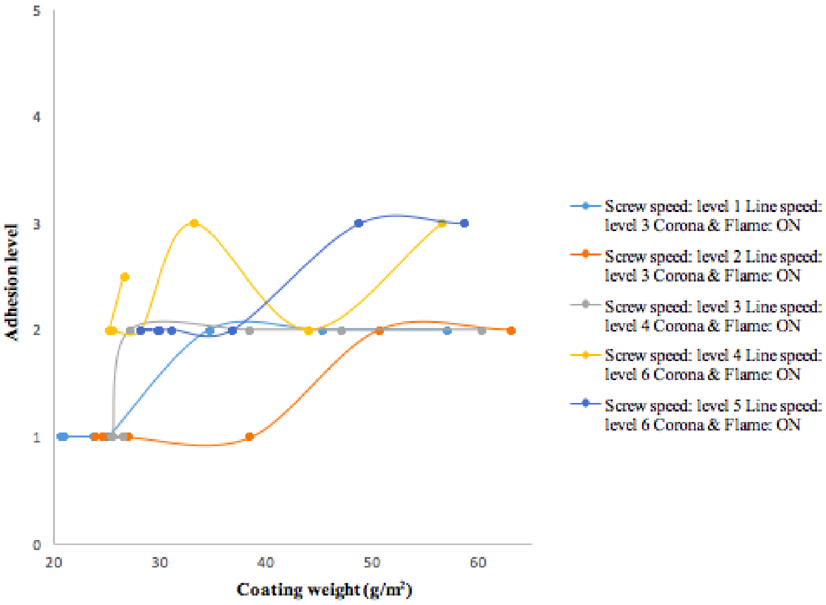

Figure 4.5 *Adhesion level versus. coating weight(g/m^2) graph of actual part of adhesion trial 1*

Even coating weight of samples were getting increasing with air gap, adhesion levels changed from 1 to 2 between the coating weight range of 25 and 30 g/m^2 according to Figure 4.6 and Figure 4.7. By based on the result, it cannot be certainly said that flame pretreatment did not work. It was hypothesized that the reason behind the situation was the configuration of corona and flame was wrong. In the actual part, substrate was exposed to first corona treatment and then flame treatment. During corona discharge, electrons were accelerated into the surface of the substrate leading to the long chains to rupture, generating a multiplicity of open ends and free valences were formed. This means that the surface became more receptive to the molten polymer by being dried and warmed. After corona treatment, flame treatment caused to burn all formed free radicals on the substrate. Because of that, the effect of flame treatment on adhesion was not examined.

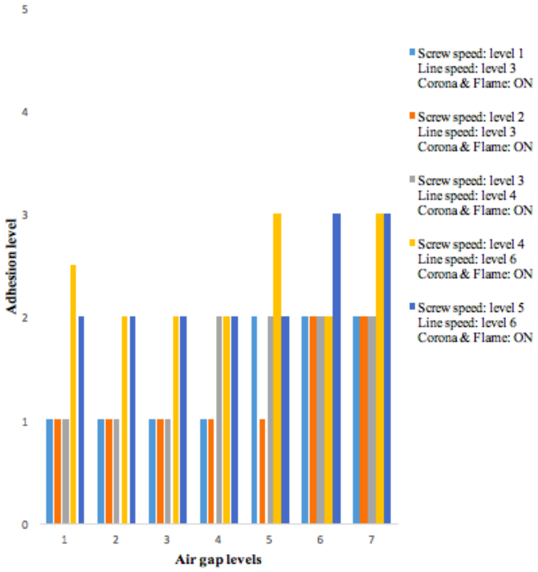

Figure 4.6 *Adhesion level versus air gap levels graph of actual part of adhesion trial 1*

However, it is not applicable and senseful, thus neck-in quite narrow. At high air gap level with low line speed and screw speed values, still high adhesion level was observed by the reason of increase in coating thickness where is higher than the target.

Although effect of air gap on neck-in was same as the screening part of trial 1 and this represents that high neck-in values were reached as air gap was increased as it was represented in Figure 4.8 , there were two remarkable differences between results. The first one was that the reduction in the width was not as low as in the screening part and the second was that width of the samples were big or slightly varied compared the screening part samples. This situation may be resulted from changes in viscosity.

All in all, the most vital result was deduced from actual part of adhesion trial 1 that the configuration of flame and corona should be changed as a vice-versa to study effect of flame treatment on adhesion in future trials.

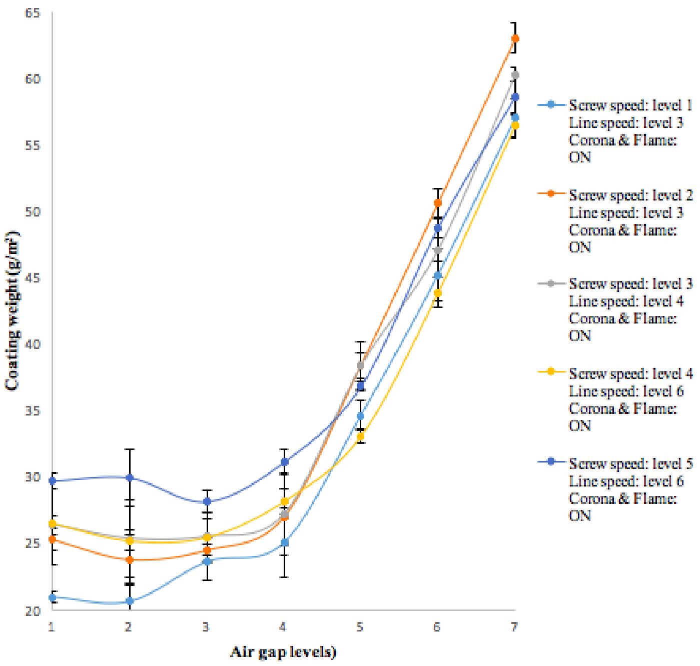

Figure 4.7 *Coating weight(g/m^2) versus air gap levels graph of actual part of adhesion trial 1*

4.2 Results of Adhesion Trial 2

In this trial, the aim was to examine effect of process equipment modifications and line speed on adhesion by using PET 1 grade. To analyze samples, only main testing methods were used by following same steps in previous trials.

It was expected some improvements in the adhesion results of adhesion trial 2 at the target coating weight compared to previous trials through process modification but unfortunately results were slightly problematic in terms of runnability and non-homogenous adhesion behavior on cardboard. On the other hand, no differences observed in processing behavior of PET 1 grade. Also, adhesion of PET on cardboard increased when coating weight increases again as presented in Figure 4.9. The attained highest adhesion level was 4 over 5 at 30.98 g/m^2 coating according to Figure 4.9 while performing at level 2 line speed, level 3 screw speed and level 1 (minimum) air gap. This test point did not have any runnability and non-homogenous adhesion problems.

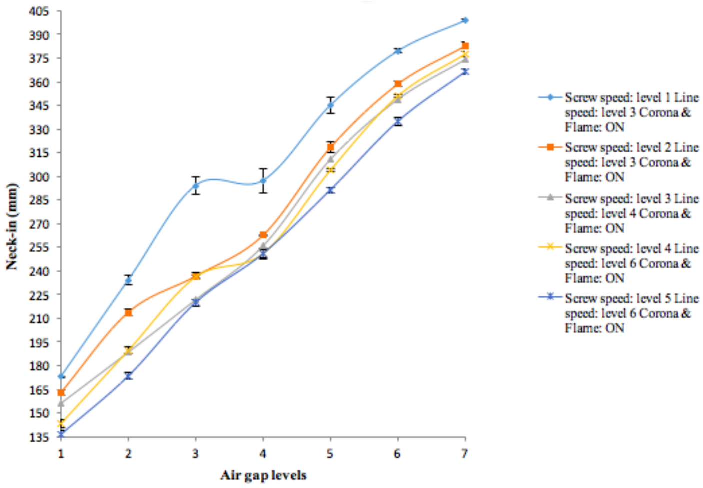

Figure 4.8 *Neck-in(mm) versus air gap levels graph of the actual part of adhesion trial 1*

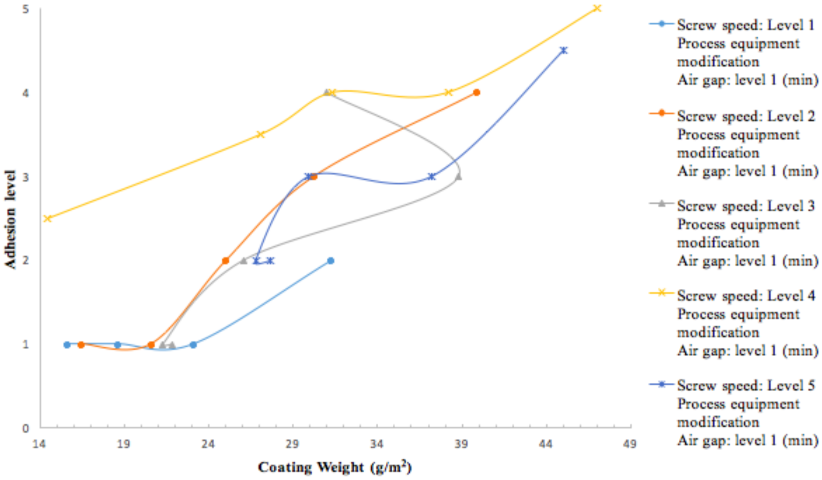

Figure 4.9 *Adhesion level versus coating weight(g/m^2) graph of adhesion trial 2*

The main problems in the most of the results of trial 2 were cohesive failure and varied adhesion. The results must be analyzed carefully and precisely in order to avoid incorrect interpreting, as heavily fiber tear indicated more cohesive failure in the layer of the cardboard than true peeling between the polymer and cardboard interface. This expression states that adhesion strength at that level was higher than cohesion strength. Normally, this behavior is expected to occur if there is great chemical adhesion. In our case, chemical adhesion was not a matter of discussion. Instead of chemical adhesion, there were great penetration and strong polar-polar interactions.

To minimize those problems, samples of the trial were left at least 2 days to mature and then they were started to be tested. Also, it was known that adhesion depends on the time to some extent mostly if there is chemical adhesion; it can show a rise as a function of time until some point or it can fail within the time due to environmental attacks. By considering this, adhesion was measured in two days time interval. No sharp differences were observed with respect to adhesion. For instance, the adhesion level of the sample was varied between the level of 2 and 3. After the time interval, it stabilized at adhesion level of 2 or 3 even stayed still same as varied. It was proved that there was no chemical adhesion and the time interval had no effect to improve the penetration of PET on cardboard.

Technically, these variations in adhesion could not be presented in Figure 4.9 but their adhesion behavior and notes taken during testing was explained explicitly in Appendix A, B and C. In Figure 4.10, the picture was given to exemplify dominant adhesion behavior of samples of the trial.

Figure 4.10 *Example for variation in adhesion from the level of 3 to 5*

According to graphs given in Figure 4.11 and Figure 4.12, adhesion values were decreasing to some extent and then stabilized with increase of line speed. Additionally, coating weight decreases with increase of line speed. Since the higher line speed helps to reduce coating weight and coating weight has more dominant effect on adhesion, they were expected results.

In addition, it was noticed in Figure 4.12 that there was a limitation in terms of line speed for PET 1 grade. When the line speed was getting higher than level 6 , STDEV reached extremely high and the process lost its runnability. The most interesting

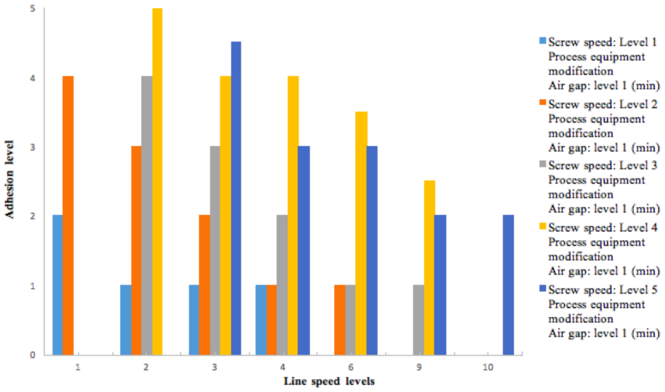

Figure 4.11 *Adhesion level versus line speed levels graph of adhesion trial 2*

result was that STDEV was found as higher than coating weight while performing at maximum (level 5) screw speed and maximum (level 10) line speed. The result showed that although it was possible to obtain really thin coatings without holes or voids, runnability of the process disappeared.

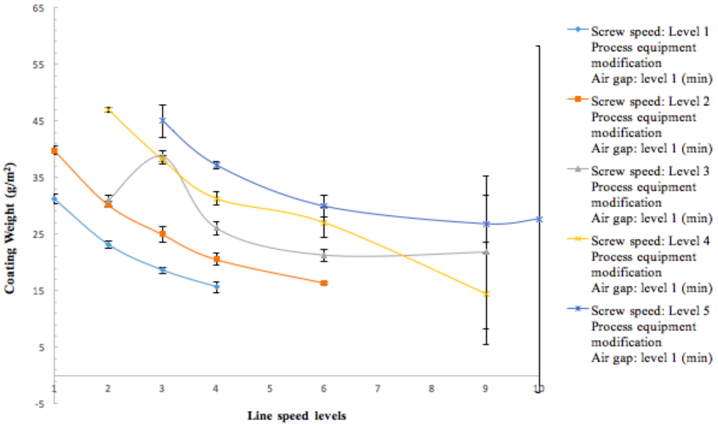

Figure 4.12 *Coating weight(g/m^2) versus line speed levels graph of adhesion trial 2*

Neck-in values were obtained almost linear with the increase of line speed by keeping air gap at the same level (minimum) during the trial runs as seen in Figure 4.13 . In addition, neck-in results for all test series and test points were senseful and useful. These results proved that air gap should be kept as minimum as possible to obtain useful and eligible polymer coating width. Since oxidation was not necessary for PET as discussed before, it would better to keep at that level. Another advantage of shortening air gap was to improve melt curtain stability.

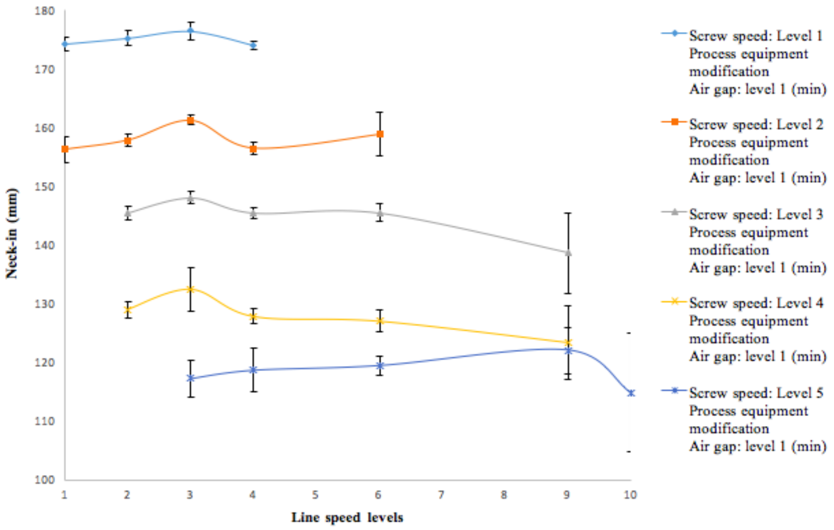

Figure 4.13 *Neck-in(mm) versus line speed levels graph of adhesion trial 2*

Another thing was that neck-in values were slightly decreasing with increase of screw speed. Normally, neck-in and drawdown are a function of melt flow index and melt temperature. Normally, neck-in increases and drawdown improves with increasing melt temperature. In this trial PET 1 grade behaved totally opposite. It was assumed that polymer swelling ratio has more dominant effect on adhesion for this case. Due to increase in screw speed and melt temperature, polymer swelling ratio increased so then neck-in decreased. This hypothesis can be supported drawdown ratio since neck-in decreases but drawdown is worse as the swelling ratio increases according to literature [21]. While operating with maximum screw speed, maximum line speed and really high melt temperature; the drawdown ratio was quite low because STDEV was extremely high. Still, there were no breakage on the coating but it was the limit.

In Figure 4.14 , the comparison of results of screening part of trial 1, actual parts of trial 1 and trial 2 were done in terms of adhesion, coating weight and neck-in while performing with same process variables. According to adhesion level versus test points graph in Figure 4.14, better adhesion was reached by performing high line speeds and high screw speeds. This information can be proved from the coating weight versus common test points graph of Figure 4.14 . The target coating weight range was mostly provided by performing with higher screw speed and line speed values. It was good thing since operating at higher line speeds was desired in the real manufacture line regarding to economic aspects. Additionally, obtaining lower neck-in results while performing with high screw speed, line speed and minimum air gap values strengthened the hypothesis.

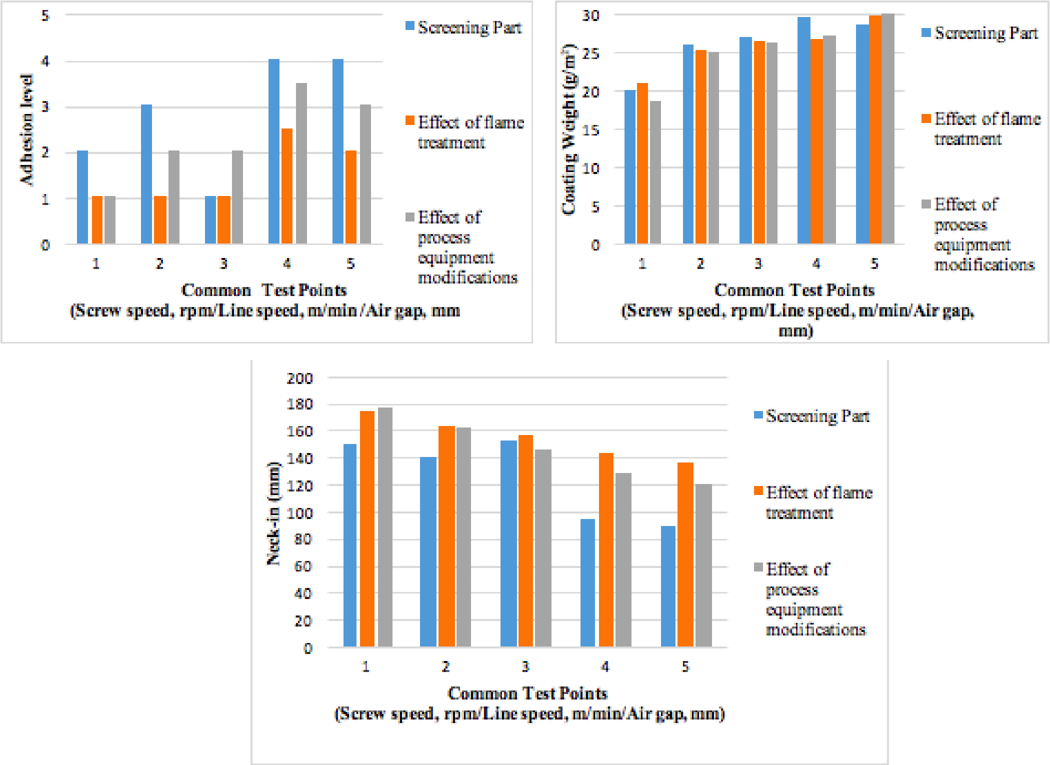

Figure 4.14 *Comparison of adhesion, coating weight and neck-in results of PET 1 by performing only with corona treatment, corona and flame treatments and modified process equipments at the common test points*

Another thing was that the best attained adhesion of PET 1 was the level of 4 over 5 by performing only with corona. For this case, actual part of trial 1 was not in question because effect of flame treatment was not observed in proper way. Normally, it was expected from process equipment modification results. One of the reasons of this situation may be due to chill roll configuration. Another reason may be due to variational moisture level of the substrate. Thick commercial cardboard is three-layer substrate and accordingly, its moisture level shows difference between winter time and spring time. During winter time, they were kept inside but during summer time, they were kept outside. Therefore, this situation caused changes in their quality. This trial was performed in winter time with dry substrates so then this affects adhesion results unfavorably. Beside all of them, the most probable reason was high melt temperature of PET 1 grade during adhesion trial 2, which was operated by modified process equipments. The higher melt temperature may lead to thermal degradation of polymer. When polyester undergoes the thermal degradation, water and acetaldehyde form as degradation product according to literature and they are not conducive to great adhesion [5]. The hypothesis was supported by checking mass pressure and the temperature of cooling pipe placed under the hopper.

4.3 Results of Adhesion Trial 3

Through adhesion trial 1 and trial 2, effect of corona treatment, flame treatment and process equipment modifications in different line speeds and air gap values was studied for PET 1 grade. In the rest of the trials, the effect of same matter was investigated by following same processing variables for PET 2. Thus, the effect of different grades of PET was examined as well.

The investigation of adhesion behavior of PET 2 was begun with adhesion trial 3 by screening part. In this trial, previous screening trial was followed as that the substrate was treated only with corona and process variable was air gap.

According to Figure 4.15, the highest adhesion level of the grade of PET was 2 over 5 by performing higher air gap values and at that level, coating weight result were the out of target. In the target coating weight and eligible coating width range, the adhesion level was dramatically poor, only 1 over 5. As compared to previous screening trial results, PET 1 grade showed better performance than PET 2 grade in terms of adhesion on cardboard. According to literature, the polymer having low viscosity is able to penetrate into the porous substrate more and establish stronger mechanical bonds upon cooling compared to the polymer having high viscosity. Extrusion coating is both dynamic and kinetic process and also it would be tricky to compare them by depending only melt temperature and viscosity. For instance, there was another crucial difference between them: heat capacity. By based on their heat capacities, drop in temperature of the PET differs from die to nip and this affects cooling in the air gap and the nip so then adhesion.

In this trial, the effect of air gap on adhesion was investigated by performing with only corona as pretreatment method and the results was presented in the Figure 4.16 and Figure 4.17 in the sense of adhesion and coating weight, respectively. According previous trial results, adhesion levels improved with an increase of coating weight; in

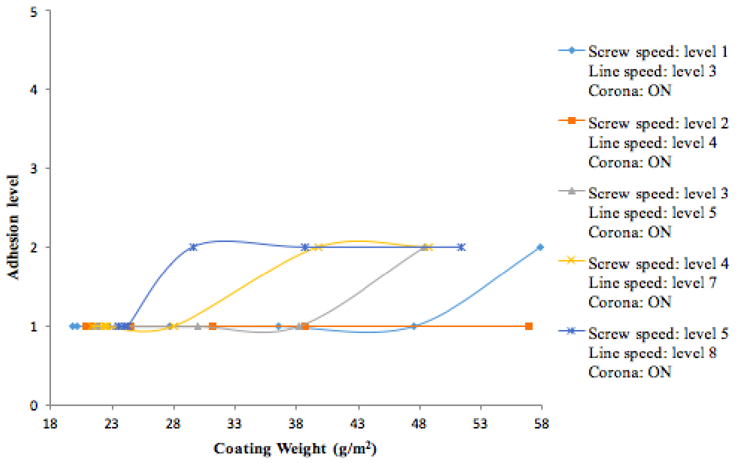

Figure 4.15 *Adhesion level versus coating weight(g/m^2) graph of adhesion trial 3*

other words, an increase of air gap. Although, coating weight was getting increase with air gap in this trial, samples had negligible improvement in their adhesion levels or even not. The result showed that coating weight was not as dominant as on adhesion of PET 2 grade compared to PET 1 grade.

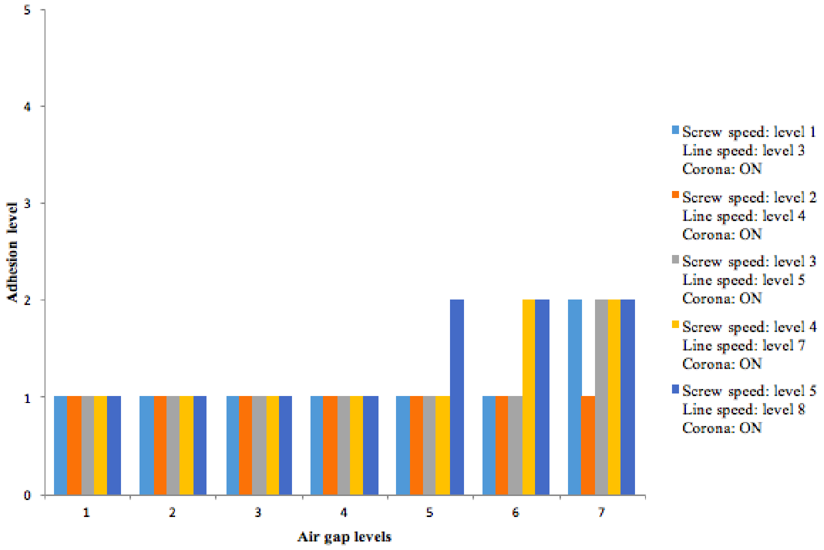

Figure 4.16 *The adhesion level versus air gap levels of adhesion level of trial 3*

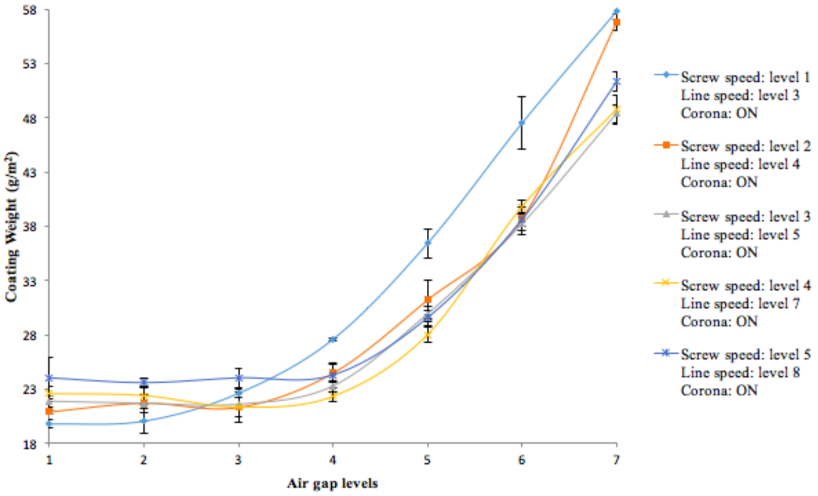

Figure 4.17 *The coating weight (g/m^2) versus air gap levels graph of adhesion trial 3*

Furthermore, this can be proved with neck-in results given in Figure 4.18. When air gap increased, coating weight increased so then drawdown ratio decreased. These factors led to increase neck-in as an expected. In addition, there were not sharp and non-negligible differences between the neck-in results of PET 1 and PET 2 by performing with similar processing parameters and same coating weight.

All in all, it was not reached desired adhesion level at target coating weight by operating with PET 2 in screening part and its performance during processing was quite poor compared to PET 1 in terms of adhesion.

4.4 Results of Adhesion Trial 4

The aim of adhesion trial 4 was to study the effect of process equipment modifications and line speed on adhesion of PET 2 grade by following process variables of adhesion trial 2 that was performed with same purpose by utilizing PET 1 grade. The main difference between adhesion trial 2 and 4 was the substrate. In adhesion trial 4, thin commercial cardboard was used as distinct from adhesion trial 2.

This trial was performed with five different test series by depending on screw speeds same as before. However, the results of the test series performing with maximum screw speed did not indicate in Figure 4.20, Figure 4.21 and Figure 4.23. Since, the test series completely failed in all line speeds and voids were formed on the coating of cardboard as exemplified in Figure 4.19.

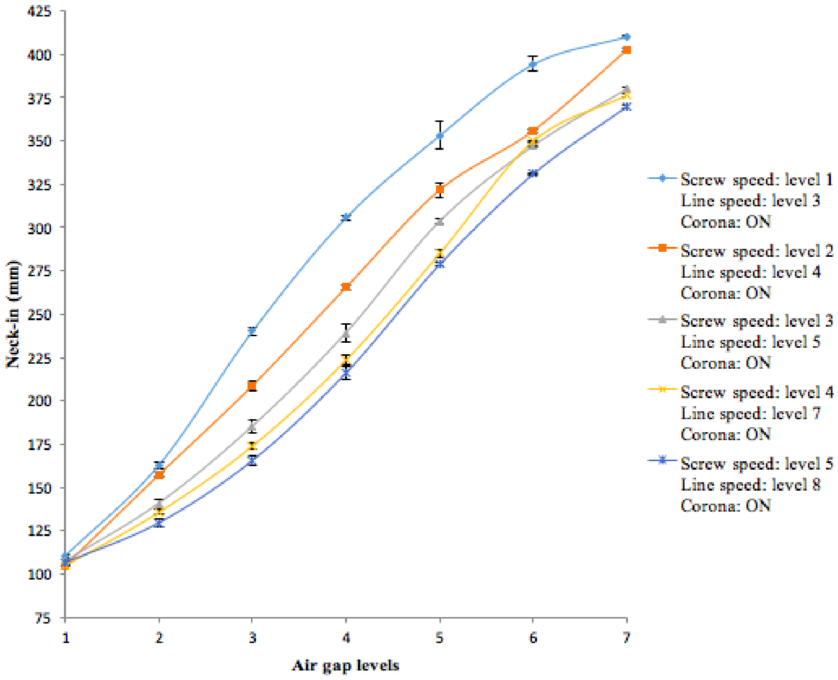

Figure 4.18 *Neck-in(mm) versus air gap levels graph of adhesion trial 3*

Figure 4.19 *An example for damaged samples of the test series performing with maximum screw speed level during adhesion trial 4*

The main reasons of the failure were high melt temperature and dependently low melt strength. It is the well-known fact for the use of polyesters in extrusion coating that increases in melt temperature with the increase of screw speed improves the adhesion of polyesters to the substrate. However, if the melt temperature is getting higher above and beyond, it will be additional neck-in and reduced viscosity lower than expected [6]. Hence, this situation concluded with damaged samples and uncontrollable process.

In Figure 4.20, the relation of adhesion and coating weight was represented. According to graph, adhesion of PET 2 improved by increasing screw speed and in parallel with melt temperature. The most senseful adhesion level was 4 over 5 at 27 g/m^2 coating weight when the process run with level 4 screw speed, level 6 line speed and minimum air gap. In that test series, the melt temperature was 281°C that was the best possible melt temperature for PET 2 while operating with modified process equipments. This was proven with the failure of the test series performing maximum screw speed.

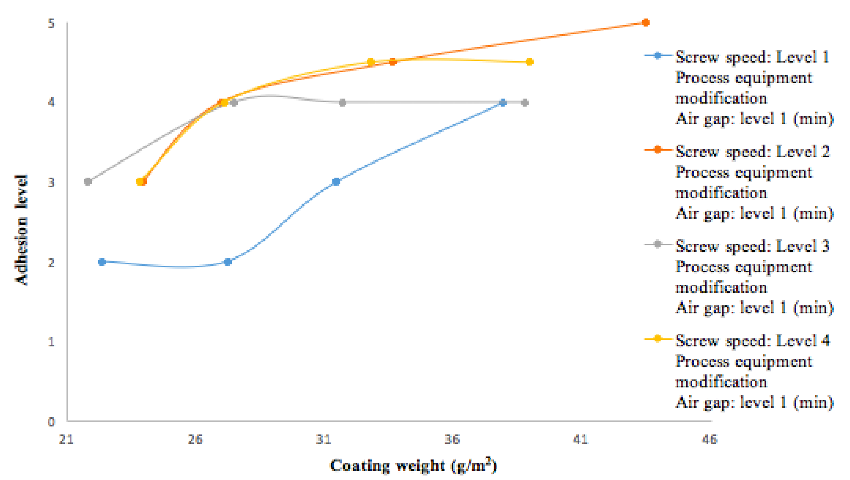

Figure 4.20 *Adhesion level versus coating weight(g/m^2) graph of adhesion trial 4*

By based on Figure 4.21, as expected, adhesion level decreased with the increase of line speed because increase in line speed led to reduce coating thickness. From that perspective, PET 2 grade showed same kind of behavior as PET 1 in terms of adhesion. However, it had better adhesion quality than PET 1 while performing with modified process equipments. Substrate may be the most probable reason of the result. Since, thin substrate was used in the trials. The moisture level of thin substrate was slightly lower than thicker one, therefore it provides better adhesion and less varied neck-in results.

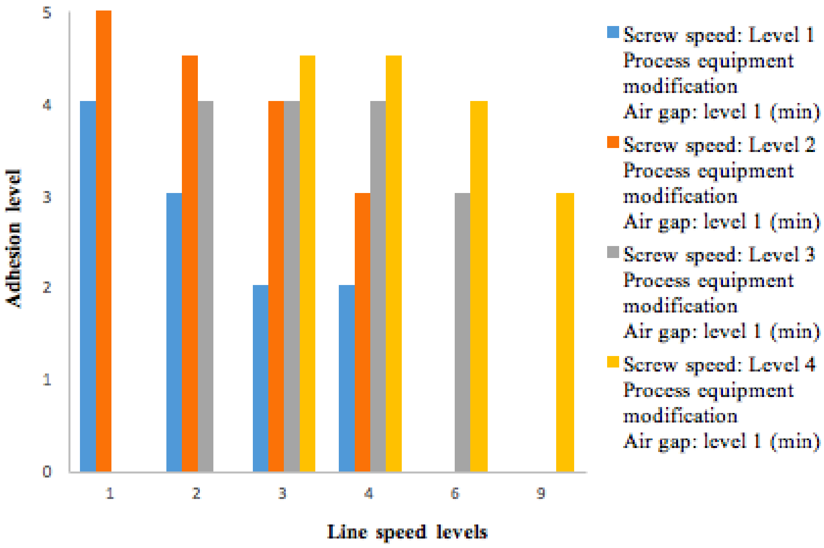

Figure* *4.21* *Adhesion level versus line speed levels graph of adhesion trial 4

Although it was reached the higher adhesion levels with expected coating weight by utilizing PET 2 , its processing was extremely challenging in extrusion coating beside PET 1 . The first reason of this situation was melt temperature as mentioned before and the second thing is line speed. When the line speed and screw speed were increased comparatively together, it was resulted with ultrahigh melt temperature and unsteady low coating weight.

Neck-in results of the trials were great regarding Figure 4.23. The coating did not have any serious stability problem from the edges and also, it was proven with STDEV that was lower than 1, mostly. In addition, neck-in results of the trial was almost half of neck-in results of previous trial, which was performed for PET 1 grade to understand the effect of process equipment modifications on adhesion. It was good thing to keep the neck-in results as minimum as possible. In this case,

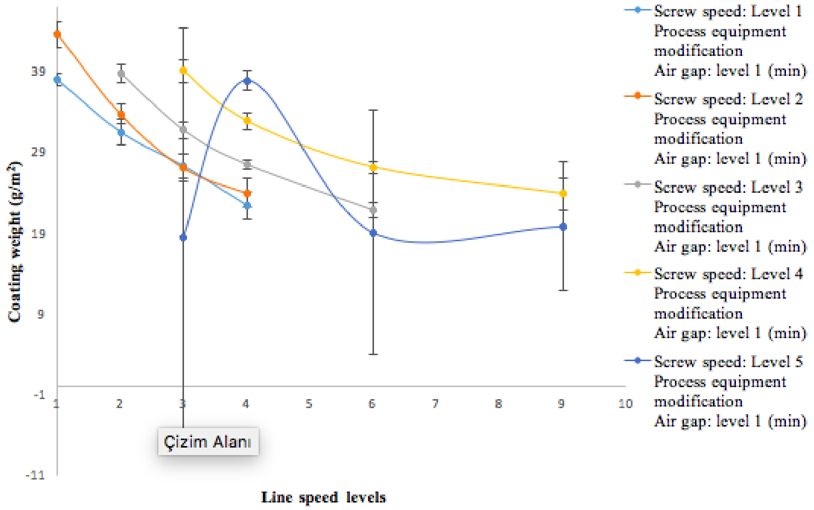

***Figure** 4.22 Coating weight(g/m^2) versus line speed levels graph of adhesion trial 4*

the attracted point was how the neck-in results were that much different, although draw down ratio of both PET 1 grade and PET 2 grade was so close each other at same coating thickness. It indicated that the elasticity of PET 2 grade was higher than the elasticity of PET 1 grade.

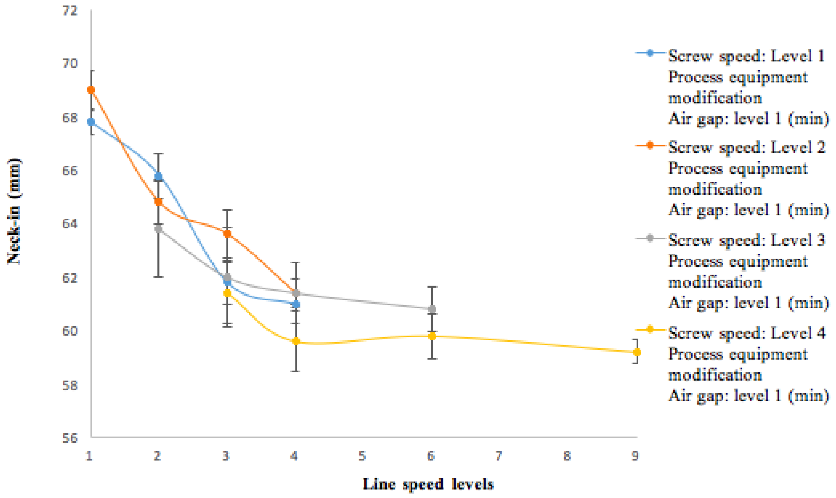

***Figure** 4.23 Neck-in(mm) versus line speed levels graph of adhesion trial 4*

4.5 Results of Adhesion Trial 5

As understood from adhesion trial 4, the effect of heat had an inevitable effect to improve the adhesion of PET 2 to the cardboard and by considering this; it was decided to repeat the trial, which was performed to study the effect of flame treatment on adhesion, for PET 2 grade by rearranging the configuration of flame treatment. For this purpose, processing parameters of actual part of adhesion trial 1 was followed; however, the configuration of the flame and corona treatment was installed as a vice-versa.

According to Figure 4.24, it is discernable that the configuration was as it should be, since adhesion level improved by increasing coating weight and air gap. On the other hand, intended adhesion level was not obtained at target coating weight range. At that level, the most considerable adhesion results were the level of 3 over 5 at 25,94 g/m^2 coating weight and the level of 4 over 5 at 30,12 g/m^2 coating weight while performing with level 4 screw speed and level 7 line speed. It can be indicated that the effect of heat had positive contribution to the adhesion of PET 2 grade as against to PET 1 grade. Nevertheless, it was not still enough to reach expected adhesion level and product specifications only by changing processing parameters and making equipment modification and applying substrate treatment methods.

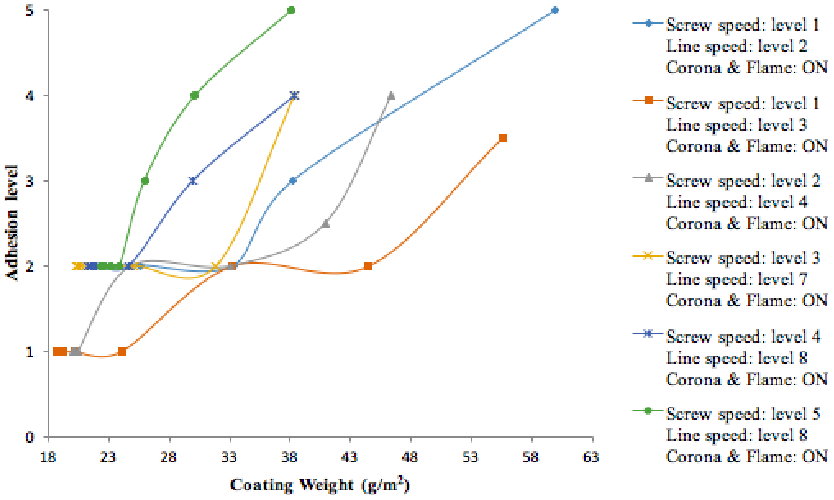

Figure 4.24 Adhesion level versus coating weight(g/m^2) graph of adhesion trial 5

The overall graphical representation of effect of air gap on coating weight, adhesion and neck-in was given in Figure 4.25. The reason of why each graph was not discussed individually was that PET 2 grade did not affected from increase in air gap unexpectedly. Same as happened in previous trials increase in air gap caused to increase coating weight and neck-in, therefore adhesion was improved.

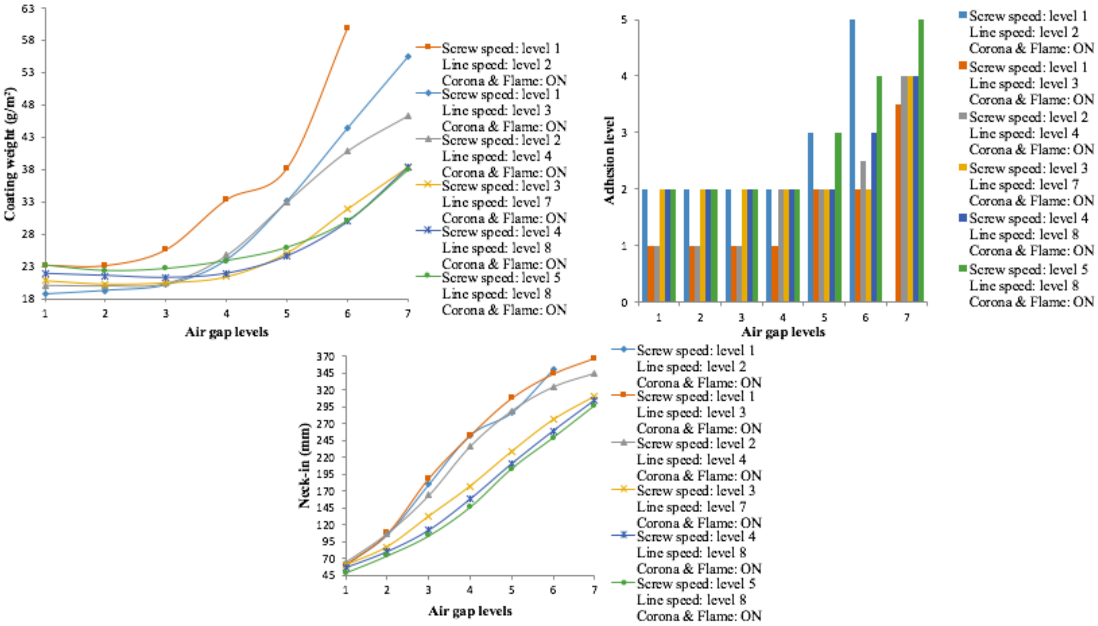

Figure 4.25 *The graphical representation of the effect of air gap on coating weight(1st graph),adhesion(2nd graph) and neck-in(3rd graph) for adhesion trial 5*

As an remarkable result, only the test point performed with level 3 air gap was failed while performing with maximum screw speed and level 8 line speed. The failure was not continuous as presented in Figure 4.26. For example, if 20 m coated cardboard sample was obtained from the test point, it covers only 1 m; however still it should be countable as a processing failure. In this case, melt temperature was not responsible from this fault, since it stayed stable if screw speed was not changed.

In Figure 4.27, the comparison of results of adhesion trial 3, 4 and 5 were done in terms of adhesion, coating weight and neck-in while performing with same process variables. According to adhesion level versus test points graph in Figure 4.27, adhesion of PET 2 grade improved better while performing with modified process equipments. As distinct from previous comparison results of PET 1 grade, neck-in and coating weight results of PET 2 grade were not that much close while operating with almost same processing parameters. As a reason of the situation, it can be indicated that PET 2 presented different rheological behavior by comparison of its previous runnability results.

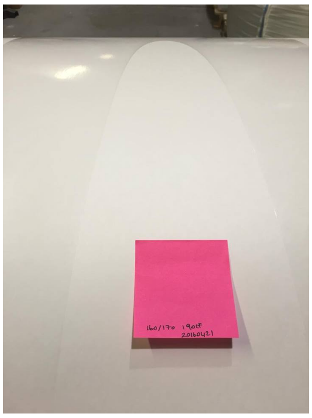

Figure 4.26 *Damaged sample while performing with maximum screw speed and 8th level line speed during trial 5*

In addition, coating weight results of PET 2 obtained from common test points were generally out of the target coating weight range. This showed that it is possible to reach almost same adhesion level at lower coating weight levels by utilizing PET 2 grade.

According to neck-in results graph in Figure 4.27, neck-in results in screening part were almost two times higher than the rest of the trial results of PET 2 . It was mostly related to mass pressure and temperature of cooling pipe under the hopper. They were quite high during screening part of PET 2 grade. Normal, these parameters have indirect effect on adhesion and in the case of adhesion of polyester to cardboard, it turns the key parameter. Once these values were quite high, it caused low shear and low elasticity and this resulted with high neck-in values.

All in all, the fulfilled highest adhesion level for both PET grades was same which is 4 over 5 and it was reached approximately at same coating weight which was around 27-28 g/m^2 coating weight. On the other hand, they presented different adhesion behavior in the sense of the effect of heat because of the reasons depending on substrate, characteristics of the PET and the position of the corona and flame treatment. Additionally, machine stability problem can be countable in a roundabout way. In each test series, we were waiting only 3-5 minutes while passing one test point to another one to reach stabile enough. The time showed difference between each test point and test series so it could affect the results slightly. Nevertheless, it was proven that it is possible to run the process at 30g/m^2 coating with almost perfect adhesion for both PET grades.

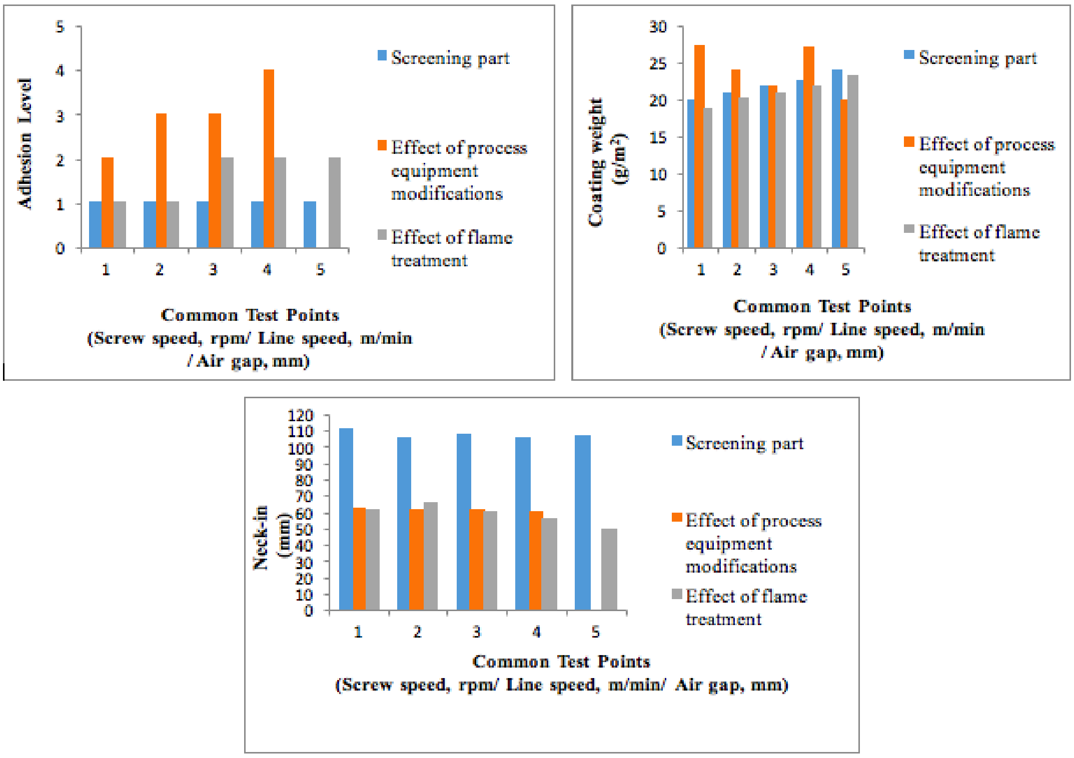

Figure 4.27 *Comparison of adhesion, coating weight and neck-in results of PET 2 grade by performing only with corona, corona and flame and modified process equipments at the common test points*

5. CONCLUSIONS

The busy lifestyle increases the demand of microwave foods and the demand leads to develop new packaging technology that comprises of polyethylene terephthalate coated cardboard. Beside this, packaging waste and their negative effects on environment are getting increasing too. Hence, increase efficiency of food packaging and handling the waste become major priority. Correspondingly, in this thesis work, it was focused on adhesion phenomenon of polyethylene terephthalate in extrusion coating once coating thickness was diminished. It is well-known that polyethylene terephthalate is not as good adherent material as polyethylene to the most of the substrate and in the case of adhesion of polyesters, the coating weight has huge impact. Therefore, it has been tempting to give a try to find out ways to improve polyethylene terephthalate's adhesion onto the cardboard once coating weight is diminished. The target in paractical part of this study was to explore the opportunity of enhancing the adhesion of polyethylene terephthalate to the cardboard by changing processing parameters, modifying process equipments and using different grades of the polymer when the coating thickness of polyethylene terephthalate onto the cardboard was between 25-30 g/m^2. The reference coating weight was 40 g/ m^2 and it is desired to great adhesion level same as reference at 25-30 g/m^2 coating. The scope of the study was to determine the primary mechanisms behind the reduction in adhesion with decreasing coating thickness, develop models for quantitatively predicting adhesion performance and apply the improved understanding to practical advantage.

According to purpose and scope of the thesis; the motivation for the current work began with the literature study of principles of extrusion coating, differences in extrusion coating of polyesters and polyolefins, adhesion theories and practical methods for adhesion improvement in extrusion coating. The expected adhesion theory was penetration by based on characteristics of polyethylene terephthalate. To improve the penetration, the effects of resin, substrate treatment (corona and flame treatment), melt temperature, air gap, line speed and process equipment modifications were investigated.

In the experimental of the study, it was concentrated on the effects of processing parameters, modifications of process equipments and properties of polyethylene terephthalate. For this purpose, 5 adhesion trials were performed. In adhesion trial 1 and 2, PET 1 grade was utilized and PET 2 grade was utilized in adhesion trial 3,4,5. Adhesion trial 1 comprised of two parts: screening part and actual part. In the screening part, only corona was used as a pretreatment method and process variable was air gap. In actual part, process variable and parameters was exactly same with screening part. As distinct from screening part, both corona and flame were utilized as a pretreatment method in actual part to study effect of flame treatment in extrusion coating. The target of the adhesion trial 2 was to understand effect of process equipment modifications and line speed on adhesion of polyethylene terephthalate. For this purpose, corona was used as a pretreatment method, process variable was line speed. These trials were followed for PET 2 grade as well to obtain senseful and comparable data about the effect interfacial characteristics various grades of polyethylene terephthalate when adhered to cardboard in changeable processing conditions. Screening of PET 2 grade was done in adhesion trial 3 by following previous processing parameters. The effect of process equipment modifications and flame treatment was studied in adhesion trial 4 and 5 for PET 2, respectively.

To analyze the samples, there were three main and three additional testing methods. Main analysis methods were adhesion test by hand, grammage test and neck-in test. Additional analysis methods were heat sealing test, microscopy test and pinhole test. For the application of addditional test methods, it is decided that they will be performed if the adhesion level of polyethylene terephthalate on the cardboard reaches the level of 5 over 5 at the target coating weight. The target was not fulfilled, therefore additional test methods were not applied.

In the case of adhesion of PET 1 grade, better adhesion was reached by performing high line speeds and high screw speeds. This hypothesis was supported with that the target coating weight range was mostly provided by performing with higher screw speed and line speed values. It was good thing since operating at higher line speeds was desired in the real manufacture line regarding to economic aspects. Additionally, obtaining lower neck-in results while performing with high screw speed, line speed and minimum air gap values strengthened the hypothesis. Another thing was that the best attained adhesion of PET 1 was the level of 4 over 5 by performing only with corona. For this case, actual part of trial 1 was not in question because effect of flame treatment was not observed in proper way due to wrong configuration of corona and flame treatment equipments in the process line. Normally, the best adhesion results was expected from the trial including modified process equipments. The most probable reason of the situation was high melt temperature of PET 1 grade during the trial. The higher melt temperature may lead to thermal degradation of polymer.

In the case of adhesion of PET 2 grade, the best adhesion level were obtained as 4 over of 5 while performing with modified process equipments. In addition, coating weight results of PET 2 obtained from common test points were generally out of the target coating weight range. This showed that it is possible to reach almost same adhesion level at lower coating weight levels by utilizing PET 2 grade.

Neck-in results in screening part were almost two times higher than the rest of the trial results of PET 2 grade. It was mostly related to mass pressure and temperature of cooling pipe under the hopper. They were quite high during screening part of PET 2 grade. Normal, these parameters have indirect effect on adhesion and in the case of adhesion of polyester to cardboard, it turned the key parameter. Once these values were quite high, it caused low shear and low elasticity and this resulted with high neck-in values.

All in all, the fulfilled highest adhesion level for both polyethylene terephthalate grades was same which is 4 over 5 and it was reached approximately at same coating weight which was around 27-28 g/m^2 coating weight. Also, it was proven that it is possible to run the process at 30g/m^2 coating with almost perfect adhesion for both polyethylene terephthalate grades. In the case of adhesion of PET onto the cardboard, better adhesion results can be achieved by performing with high line speeds, high screw speeds and minimum air gap values and coating thickness have stronger effect on the adhesion compared to the effect of processing parameters and modifications.

5.1 Proposals for further actions

In this study, the general overview was obtained about adhesion of polyethylene terephthalate onto cardboard under changeable process conditions once coating weight was diminished. To achieve desired adhesion properties at target coating weight, the scope of the thesis was not wide enough.

There are some issues that can be considered in further steps of the project. Machine stability problem can be countable in a roundabout way. In each test series, we were waiting only 3-5 minutes while passing one test point to another one to reach stabile enough. The time showed difference between each test point and test series so it could affect the results slightly. The another issue is the characteristics of utilized polyethylene terephthalate. In some trials, used polyethylene terephthalate grade behaved different from expected through process. Therefore, it would be useful to take a sample after and before processing. Thus, comprehensive information about changes in properties of the polymer can be obtained. Lastly, the characteristics of pure polyethylene terephthalate was not enough to achieve great adhesion in thinner coatings, therefore polymer blends can be tried.

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APPENDIX A. COATING WEIGHT RESULTS

In Appendix A, numerical representation of coating weight results of all adhesion trials are given. Description of test series and test points can be found thesis trials section.

Table 1 *Coating weight results of screening part of adhesion trial 1*

Air Gap, mm	CW of S-1, g/m^2	CW of S-2, g/m^2	CW of S-3, g/m^2	CW of S-4, g/m^2	CW of S-5, g/m^2	Average Coating Weight, g/m^2	STDEV
Test Series 1							
TP-1	20.10	22.30	19.50	19.00	18.60	19.90	1.45
TP-2	18.70	20.10	21.20	19.30	19.10	19.68	0.99
TP-3	25.00	24.40	22.40	27.70	25.40	24.98	1.91
TP-4	27.30	25.00	25.70	31.40	31.70	28.22	3.15
TP-5	34.50	33.70	37.50	34.00	38.00	35.54	2.05
TP-6	47.40	47.90	46.00	49.60	47.50	47.68	1.29
TP-7	61.60	60.50	58.30	56.30	57.10	58.76	2.24
Test Series 2							
TP-1	25.90	23.30	24.90	28.70	27.30	26.02	2.09
TP-2	26.40	24.90	25.90	28.20	27.20	26.52	1.26
TP-3	28.10	30.50	30.90	31.60	28.20	29.86	1.61
TP-4	39.60	38.50	40.40	39.00	43.40	40.18	1.93
TP-5	52.40	44.90	45.60	47.70	47.30	47.58	2.93
TP-6	54.70	55.20	56.10	54.80	54.70	55.10	0.60
TP-7	66.50	66.00	67.30	67.10	70.80	67.54	1.89
Test Series 3							
TP-1	26.70	27.80	28.10	26.40	25.50	26.90	1.06
TP-2	25.80	26.00	24.60	24.70	27.30	25.68	1.10
TP-3	27.50	28.40	29.60	28.50	26.40	28.08	1.20
TP-4	35.00	35.80	38.10	35.10	35.40	35.88	1.28
TP-5	48.40	45.60	47.00	45.80	49.40	47.24	1.65
TP-6	55.10	54.70	54.80	54.30	53.20	54.42	0.74
TP-7	64.90	65.50	66.70	65.80	64.10	65.40	0.97

***Table* 2** *Coating weight results of screening part of adhesion trial 1 (cont.)*

Air Gap, mm	CW of S-1, g/m^2	CW of S-2, g/m^2	CW of S-3, g/m^2	CW of S-4, g/m^2	CW of S-5, g/m^2	Average Coating Weight, g/m^2	STDEV
Test Series 4							
TP-1	29.20	29.30	31.10	29.20	28.30	29.42	1.02
TP-2	27.80	30.20	28.80	26.40	26.10	27.86	1.70
TP-3	28.40	28.30	26.30	26.30	26.70	27.20	1.06
TP-4	29.40	29.60	29.50	31.40	28.30	29.64	1.11
TP-5	35.80	35.30	34.50	37.30	34.50	35.48	1.16
TP-6	48.00	50.10	49.10	48.30	48.70	48.84	0.82
TP-7	60.80	61.30	60.80	60.00	63.20	61.22	1.20
Test Series 5							
TP-1	29.50	27.50	27.50	28.20	30.00	28.54	1.15
TP-2	26.10	26.90	29.10	28.10	26.90	27.42	1.18
TP-3	26.20	25.50	26.90	26.70	27.60	26.58	0.79
TP-4	32.30	29.90	29.40	28.30	28.60	29.70	1.59
TP-5	42.40	42.80	44.50	43.40	45.30	43.68	1.20
TP-6	51.90	51.30	52.40	52.90	52.90	52.28	0.69
TP-7	65.20	64.70	64.60	64.40	65.00	64.78	0.32

Table *3 Coating weight results of actual part of adhesion trial 1*

Air Gap, mm	CW of S-1, g/m^2	CW of S-2, g/m^2	CW of S-3, g/m^2	CW of S-4, g/m^2	CW of S-5, g/m^2	Average Coating Weight, g/m^2	STDEV
Test Series 1							
TP-1	21.30	21.40	21.00	20.40	20.80	20.98	0.40
TP-2	20.90	19.70	19.80	22.60	20.20	20.64	1.19
TP-3	21.30	23.70	24.80	24.10	24.20	23.62	1.36
TP-4	29.40	24.80	24.70	22.30	24.10	25.06	2.63
TP-5	34.80	35.30	34.80	32.70	35.60	34.64	1.14
TP-6	44.00	46.00	47.50	46.00	42.60	45.22	1.92
TP-7	59.10	57.10	56.20	55.20	57.50	57.02	1.46
Test Series 2							
TP-1	25.20	25.40	24.60	24.70	26.60	25.30	0.80
TP-2	23.10	25.90	23.60	25.20	21.20	23.80	1.85
TP-3	23.40	24.40	26.00	24.10	24.60	24.50	0.95
TP-4	25.70	30.10	28.30	25.20	25.60	26.98	2.13
TP-5	40.00	40.10	37.50	38.60	35.90	38.42	1.77
TP-6	50.50	50.10	52.40	49.60	50.40	50.60	1.07
TP-7	61.80	64.30	63.60	63.50	61.80	63.00	1.14
Test Series 3							
TP-1	27.30	26.10	27.00	26.10	25.60	26.42	0.70
TP-2	22.10	26.20	29.90	25.10	23.60	25.38	2.96
TP-3	28.70	24.60	25.40	24.20	24.70	25.52	1.83
TP-4	25.20	25.70	25.70	26.70	32.60	27.18	3.08
TP-5	39.00	39.50	38.60	37.30	37.40	38.36	0.98
TP-6	46.80	47.50	48.50	46.30	46.40	47.10	0.91
TP-7	60.70	60.60	59.30	60.30	60.60	60.30	0.58

Table 4 *Coating weight results of actual part of adhesion trial 1 (cont.)*

Air Gap, mm	CW of S-1, g/m^2	CW of S-2, g/m^2	CW of S-3, g/m^2	CW of S-4, g/m^2	CW of S-5, g/m^2	Average Coating Weight, g/m^2	STDEV
Test Series 4							
TP-1	24.60	27.50	26.20	31.30	23.20	26.56	3.11
TP-2	25.10	25.60	26.10	25.30	24.10	25.24	0.74
TP-3	24.90	27.50	25.20	24.00	25.90	25.50	1.31
TP-4	27.10	28.60	29.30	28.70	27.20	28.18	0.98
TP-5	33.80	33.00	32.50	33.50	32.70	33.10	0.54
TP-6	44.00	42.50	43.50	45.60	43.80	43.88	1.12
TP-7	55.60	57.50	56.30	55.70	57.10	56.44	0.84
Test Series 5							
TP-1	29.50	30.00	30.30	28.90	29.90	29.72	0.54
TP-2	33.60	30.10	29.30	28.20	28.60	29.96	2.16
TP-3	27.00	28.50	28.50	27.70	29.10	28.16	0.82
TP-4	32.60	30.00	31.10	30.60	31.30	31.12	0.97
TP-5	36.30	37.10	36.70	37.10	36.90	36.82	0.33
TP-6	50.00	48.70	48.30	48.50	48.20	48.74	0.73
TP-7	58.60	59.20	60.30	57.50	57.40	58.60	1.21

Table 5 *Coating weight results of adhesion trial 2*

Line speed, m/min	CW of S-1, g/m^2	CW of S-2, g/m^2	CW of S-3, g/m^2	CW of S-4, g/m^2	CW of S-5, g/m^2	Average Coating Weight, g/m^2	STDEV
Test Series 1							
TP-1	31.8	30.8	31.3	32.3	29.9	31.22	0.93
TP-2	23.3	23.9	23.6	22.3	22.5	23.12	0.69
TP-3	18.9	19.3	18.7	17.9	18.1	18.58	0.58
TP-4	14	16.2	16.1	16.2	15.6	15.62	0.94
Test Series 2							
TP-1	40.7	40.1	39.1	40.2	39.1	39.84	0.71
TP-2	31	29.7	30.4	29.8	30.2	30.22	0.52
TP-3	26.9	24.1	23.4	25.4	25.3	25.02	1.34
TP-4	20	20.1	21.7	21.7	19.4	20.58	1,06
TP-5	16.6	16.4	16.1	16.3	16.7	16.42	0.24
Test Series 3							
TP-1	29.60	31.90	30.80	31.80	30.80	30.98	0.93
TP-2	37.80	38.20	40.00	38.60	39.30	38.78	0.88
TP-3	26.50	25.10	24.50	26.60	27.50	26.04	1.22
TP-4	19.80	21.60	22.90	20.80	21.20	21.26	1.13
TP-5	9.60	8.00	24.50	41.00	25.90	21.80	13.53
Test Series 4							
TP-1	46.8	46.7	47.4	46.7	47.6	47.04	0.43
TP-2	38.5	37	37.8	38.5	39.1	38.18	0.80
TP-3	30.7	32.6	31	29.9	32.4	31.32	1.15
TP-4	30.1	24.6	28.1	24.1	28.4	27.06	2.59
TP-5	6.4	8.9	18.3	28.5	10.1	14.44	9.04
Test Series 5							
TP-1	43.1	45.3	44	42.8	49.9	45.02	2.90
TP-2	36.1	37.6	31	37.7	37.4	37.18	0.65
TP-3	28.6	33.4	29.6	29.2	28.8	29.92	1.98
TP-4	30.3	32.5	23.3	20	27.8	26.78	5.10
TP-5	6.1	11.5	77.5	37	6.2	27.66	30.65

Table 6 *Coating weight results of adhesion trial 3*

Air Gap, mm	CW of S-1, g/m^2	CW of S-2, g/m^2	CW of S-3, g/m^2	CW of S-4, g/m^2	CW of S-5, g/m^2	Average Coating Weight, g/m^2	STDEV
Test Series 1							
TP-1	19.5	20.2	20	19.3	20.1	20	0.40
TP-2	21	19.5	18.3	20.7	20.9	20	1.16
TP-3	22.3	22.4	22.7	22.5	23.3	23	0.40
TP-4	27.4	27.6	27.7	27.8	27.6	28	0.15
TP-5	37.2	37	37.5	36.4	34.2	36	1.33
TP-6	45.9	44.7	47.5	49	50.7	48	2.39
TP-7	57.9	57.5	56.8	59.1	58.1	58	0.84
Test Series 2							
TP-1	21	21	20.8	21.1	20.7	20.92	0.16
TP-2	21.3	20.8	22	23	21.5	21.72	0.83
TP-3	20	22.8	22.9	20.7	20.2	21.32	1.42
TP-4	24.5	23.7	24.3	25.8	24.1	24.48	0.79
TP-5	31.9	33.1	30.7	32	28.4	31.22	1.79
TP-6	38.2	39.5	40.2	38.2	37.6	38.74	1.07
TP-7	57	57	56.3	56	57.9	56.84	0.74
Test Series 3							
TP-1	21.3	21.3	22.3	22.4	21.9	21.84	0.53
TP-2	19.2	23.1	22.6	22.2	21.2	21.66	1.54
TP-3	20	22.7	23.6	20.1	21.4	21.56	1.59
TP-4	23.2	23.7	22.7	22.8	23.8	23.24	0.50
TP-5	30.9	29.6	29.2	29.5	30.3	29.90	0.69
TP-6	37.1	39.6	38.4	37.5	38.4	38.20	0.97
TP-7	49.2	48.6	49.1	47.6	47.5	48.40	0.81

Table 7 *Coating weight results of adhesion trial 3 (cont.)*

Air Gap, mm	CW of S-1, g/m^2	CW of S-2, g/m^2	CW of S-3, g/m^2	CW of S-4, g/m^2	CW of S-5, g/m^2	Average Coating Weight, g/m^2	STDEV
Test Series 4							
TP-1	21.5	22.4	23.3	23	22.8	22.60	0.70
TP-2	21.8	22.5	22.4	21.7	23.7	22.42	0.80
TP-3	22.5	20.8	20.5	21	22	21.36	0.85
TP-4	23	22.1	22.6	22	22	22.34	0.44
TP-5	27.3	29.1	28.3	27.7	27.7	28.02	0.70
TP-6	39.6	40.1	39.7	40.6	39	39.80	0.60
TP-7	47.4	48.1	50.7	49.3	48.3	48.76	1.28
Test Series 5							
TP-1	23.1	22.6	25.6	26.6	22.2	24	1.96
TP-2	24.2	23.2	23.4	24	23.1	24	0.49
TP-3	23	25.3	24.2	23.7	23.9	24	0.84
TP-4	22.8	24.4	23.6	24.7	25.9	24	1.17
TP-5	28.5	29.5	29.7	30.2	29.9	30	0.65
TP-6	38.3	38.3	39.5	39.1	38	39	0.63
TP-7	52.2	52.3	50.1	50.8	51.4	51	0.93

Table 8 *Coating weight results of adhesion trial 5*

Air Gap, mm	CW of S-1, g/m^2	CW of S-2, g/m^2	CW of S-3, g/m^2	CW of S-4, g/m^2	CW of S-5, g/m^2	Average Coating Weight, g/m^2	STDEV
Test Series 1							
TP-1	18.9	18.7	18.4	19	18.4	18.68	0.28
TP-2	20.1	19.2	19.9	17.8	19.2	19.24	0.90
TP-3	18.9	21	20.6	20.4	19.7	20.12	0.83
TP-4	22.9	22.8	25.1	25	24.3	24.02	1.11
TP-5	32.6	32.1	34	34.9	32.2	33.16	1.23
TP-6	43.5	43	44.1	44.4	47.3	44.46	1.68
TP-7	54.6	55.6	55.4	56.7	55.5	55.56	0.75
Test Series 2							
TP-1	20.5	20.1	19.5	18.9	21.5	20.1	0.99
TP-2	18.4	21	23	18.7	19.1	20.04	1.94
TP-3	21.7	21	20.4	18.2	20.8	20.42	1.33
TP-4	25.7	24.3	27.1	23.5	23.2	24.76	1.63
TP-5	31.8	34.2	34.5	33.1	31.6	33.04	1.33
TP-6	39.8	40.3	41.2	41.6	41.5	40.88	0.79
TP-7	45.7	46.9	46.5	46.6	46.1	46.36	0.47
Test Series 3							
TP-1	19.2	20.9	22.4	20.9	20.3	20.74	1.16
TP-2	19	19.6	22.9	19.7	20	20.24	1.53
TP-3	21.9	19.9	21.2	19.2	20	20.44	1.09
TP-4	20.1	19.7	21.5	22.9	22.7	21.38	1.46
TP-5	25.2	26.5	24.5	24.4	24.8	25.08	0.85
TP-6	33.5	31.6	33.2	29.8	31.3	31.88	1.51
TP-7	38	38.1	37.8	39.1	39	38.4	0.60

Table 9 *Coating weight results of adhesion trial 5 (cont.)*

Air Gap, mm	CW of S-1, g/m^2	CW of S-2, g/m^2	CW of S-3, g/m^2	CW of S-4, g/m^2	CW of S-5, g/m^2	Average Coating Weight, g/m^2	STDEV
Test Series 4							
TP-1	21.2	22.9	21.2	21.4	22.7	21.88	0.85
TP-2	21.4	23	22.2	19.9	21.3	21.56	1.15
TP-3	20.9	21.9	22	20.3	21.2	21.26	0.71
TP-4	21.8	21.7	22.8	22.3	21	21.92	0.68
TP-5	24.7	26	23.8	23.5	25.1	24.62	1.01
TP-6	31.5	30.2	29.7	29.4	29	29.96	0.97
TP-7	38.6	38.4	37.2	38.5	39.3	38.4	0.76
Test Series 5							
TP-1	24.1	22.8	23.3	22.8	22.6	23.12	0.61
TP-2	22.2	21.8	21.5	22.5	23.5	22.3	0.77
TP-3	23	20.6	21.7	23.9	23.9	22.62	1.44
TP-4	23.9	21.9	25.2	25.5	22.8	23.86	1.54
TP-5	26	25.4	26.1	26.7	25.5	25.94	0.52
TP-6	29.9	29.2	30.4	30.7	30.4	30.12	0.59
TP-7	36.9	38.8	38.8	37.8	37.8	38.02	0.80

Table 10 *Coating weight results of adhesion trial 4*

Line speed, m/min	CW of S-1, g	CW of S-2, g	CW of S-3, g	CW of S-4, g	CW of S-5, g	Average Coating Weight, g/m^2	STDEV
Test Series 1							
Board (start)	1,814	1,812	1,817	1,81	1,801	181.08	0.61
Board (end)	1,846	1,853	1,853	1,875	1,871	185.96	1.26
Board (average)						183.52	3.45
TP-1	2,207	2,215	2,224	2,221	2,205	37.92	0,84
TP-2	2,149	2,154	2,155	2,124	2,165	31.42	1.53
TP-3	2,103	2,094	2,115	2,129	2,094	27.18	1,50
TP-4	2,034	2,065	2,075	2,069	2,048	22.30	1.68
Test Series 2							
Board (start)	1,872	1,874	1,864	1,859	1,867	186.72	0.61
Board (end)	1,866	1,872	1,879	1,896	1,884	188.34	0.89
Board (average)						187.53	1.15
TP-1	2,288	2,304	2,315	2,329	2,313	43.45	1.51
TP-2	2,203	2,203	2,228	2,221	2,202	33.61	1.22
TP-3	2,125	2,161	2,161	2,129	2,149	26.97	1.72
TP-4	2,088	2,109	2,112	2,133	2,131	23.93	1.84
Test Series 3							
Board (start)	1,886	1,896	1,907	1,892	1,88	189.22	1.03
Board (end)	1,869	1,885	1,864	1,872	1,878	187.36	0.81
Board (average)						188.29	1.32
TP-1	2,272	2,268	2,261	2,289	2,261	38.73	1.15
TP-2	2,207	2,197	2,212	2,197	2,185	31.67	1.04
TP-3	2,152	2,153	2,167	2,158	2,156	27.43	0.60
TP-4	2,096	2,102	2,106	2,111	2,088	21.77	0.89

Table 11 *Coating weight results of adhesion trial 4*

Line speed, m/min	CW of S-1, g/m^2	CW of S-2, g/m^2	CW of S-3, g/m^2	CW of S-4, g/m^2	CW of S-5, g/m^2	Average Coating Weight, g/m^2	STDEV
Test Series 4							
Board (start)	1,858	1,893	1,854	1,866	1,888	187.58	1.83
Board (end)	1,876	1,876	1,890	1,881	1,872	187.9	0.69
Board (average)						187.74	0.23
TP-2	2,288	2,269	2,251	2,269	2,258	38.96	1.40
TP-3	2,199	2,196	2,198	2,216	2,217	32.78	1.04
TP-4	2,140	2,151	2,159	2,141	2,150	27.08	0.79
TP-5	2,114	2,118	2,098	2,147	2,100	23.8	1.97
Test Series 5							
Board (start)	1,872	1,853	1,871	1,881	1,858	186.7	1.13
Board (end)	1,874	1,897	1,882	1,87	1,879	188.04	1.04
Board (average)						187.37	0.95
TP-1						18.47	25.87
TP-2						37.81	1.18
TP-3						19.02	15.07
TP-4						19.78	8.02

APPENDIX B. NECK-IN RESULTS

In Appendix B, numerical representation of neck-in results of all adhesion trials are given. Description of test series and test points can be found thesis trials section.

***Table 12** Neck-in results of screening part of adhesion trial 1*

Air gap, mm	Neck-in of S-1, mm	Neck-in of S-2, mm	Neck-in of S-3, mm	Neck-in of S-4, mm	Neck-in of S-5, mm	Average Neck-in, mm	STDEV
Test Series 1							
TP-1	145	151	150	149	149	149	2.28
TP-2	204	213	211	209	209	209	3.35
TP-3	283	284	282	282	283	283	0.84
TP-4	309	307	314	320	329	316	8.93
TP-5	343	341	344	343	343	343	1.10
TP-6	379	377	378	379	382	379	1.87
TP-7	399	398	396	397	398	398	1.14
Test Series 2							
TP-1	138	140	142	143	138	140	2.28
TP-2	197	197	200	200	199	199	1.52
TP-3	263	261	261	263	263	262	1.10
TP-4	325	324	321	322	321	323	1.82
TP-5	345	344	343	342	342	343	1.30
TP-6	363	365	365	365	363	364	1.10
TP-7	383	382	383	382	382	382	0.55
Test Series 3							
TP-1	151	150	153	151	153	152	1.34
TP-2	199	199	202	196	191	197	4.16
TP-3	251	249	249	250	249	250	0.89
TP-4	304	303	303	301	300	302	1.64
TP-5	343	345	341	345	347	344	2.28
TP-6	369	369	369	361	361	366	4.38
TP-7	384	384	385	380	379	382	2.70

Table 13 *Neck-in results of screening part of adhesion trial 1 (cont.)*

Air gap, mm	Neck-in of S-1, mm	Neck-in of S-2, mm	Neck-in of S-3, mm	Neck-in of S-4, mm	Neck-in of S-5, mm	Average Neck-in, mm	STDEV
Test Series 4							
TP-1	93	92	94	95	95	94	1.30
TP-2	157	156	157	152	153	155	2.35
TP-3	220	213	205	190	188	203	14.02
TP-4	263	264	262	259	258	261	2.59
TP-5	315	316	316	317	317	316	0.84
TP-6	358	357	358	359	356	358	1.14
TP-7	383	381	380	381	381	381	1.10
Test Series 5							
TP-1	85	89	92	87	87	88	2.65
TP-2	138	137	135	136	135	136	1.30
TP-3	186	187	188	187	188	187	0.84
TP-4	240	241	241	240	241	241	0.55
TP-5	312	314	315	316	312	314	1.79
TP-6	343	342	347	344	343	344	1.95
TP-7	368	368	368	367	368	368	0.45

Table 14 *Neck-in results of actual part of adhesion trial 1*

Air gap, mm	Neck-in of S-1, mm	Neck-in of S-2, mm	Neck-in of S-3, mm	Neck-in of S-4, mm	Neck-in of S-5, mm	Average Neck-in, mm	STDEV
Test Series 1							
TP-1	173	172	173	173	173	173	0.45
TP-2	234	231	232	236	238	234	2.86
TP-3	303	290	289	292	297	294	5.81
TP-4	306	301	299	295	286	297	7.50
TP-5	351	348	343	347	337	345	5.40
TP-6	380	381	380	380	377	380	1.52
TP-7	399	399	399	400	398	399	0.71
Test Series 2							
TP-1	162	165	161	164	162	163	1.64
TP-2	214	214	215	216	210	214	2.28
TP-3	234	238	236	239	237	237	1.92
TP-4	263	263	263	263	262	263	0.45
TP-5	323	321	318	316	315	319	3.36
TP-6	361	356	358	359	360	359	1.92
TP-7	385	384	384	382	380	383	2.00
Test Series 3							
TP-1	158	156	157	156	154	156	1.48
TP-2	172	171	192	203	205	189	16.38
TP-3	224	222	223	220	220	222	1.79
TP-4	251	249	249	258	273	256	10.20
TP-5	309	310	311	311	313	311	1.48
TP-6	349	350	349	349	347	349	1.10
TP-7	375	373	373	375	374	374	1.00

Table 15 *Neck-in results of actual part of adhesion trial 1 (cont.)*

Air gap, mm	Neck-in of S-1, mm	Neck-in of S-2, mm	Neck-in of S-3, mm	Neck-in of S-4, mm	Neck-in of S-5, mm	Average Neck-in, mm	STDEV
Test Series 4							
TP-1	139	144	145	146	142	143	2.77
TP-2	187	189	193	190	189	190	2.19
TP-3	235	236	236	237	237	236	0.84
TP-4	252	255	252	249	247	251	3.08
TP-5	305	303	305	304	303	304	1.00
TP-6	351	351	351	352	350	351	0.71
TP-7	379	377	376	379	377	378	1.34
Test Series 5							
TP-1	137	139	135	138	133	136	2.41
TP-2	172	172	172	175	176	173	1.95
TP-3	219	221	221	217	222	220	2.00
TP-4	253	252	247	250	253	251	2.55
TP-5	289	291	292	292	293	291	1.52
TP-6	339	335	333	333	334	335	2.49
TP-7	365	366	366	365	369	366	1.64

Table 16 *Neck-in results of adhesion trial 2*

Line Speed, m/min	Neck-in of S-1, mm	Neck-in of S-2, mm	Neck-in of S-3, mm	Neck-in of S-4, mm	Neck-in of S-5, mm	Average Neck-in, mm	STDEV
Test Series 1							
TP-1	176	174	174	173	174	174	1.10
TP-2	174	174	175	176	177	175	1.30
TP-3	174	176	177	178	177	176	1.52
TP-4	173	175	174	174	174	174	0.71
Test Series 2							
TP-1	159	157	156	156	153	156	2.17
TP-2	157	159	159	157	157	158	1.10
TP-3	160	161	162	161	162	161	0.84
TP-4	155	156	156	157	158	156	1.14
TP-5	163	161	159	158	153	159	3.77
Test Series 3							
TP-1	147	146	144	145	145	145	1.14
TP-2	149	148	147	147	149	148	1.00
TP-3	145	146	144	146	146	145	0.89
TP-4	146	147	145	146	143	145	1.52
TP-5	132	131	145	145	140	139	6.80
Test Series 4							
TP-1	127	130	130	130	128	129	1.41
TP-2	130	131	130	132	139	132	3.78
TP-3	130	127	127	127	128	128	1.30
TP-4	129	128	127	127	124	127	1.87
TP-5	117	122	122	134	122	123	6.31
Test Series 5							
TP-1	112	120	118	117	119	117	3.11
TP-2	120	120	121	112	120	119	3.71
TP-3	117	121	119	119	121	119	1.67
TP-4	119	122	123	118	128	122	3.94
TP-5	105	107	124	127	111	115	10.06

Table 17 *Neck-in results of adhesion trial 3*

Air gap, mm	Neck-in of S-1, mm	Neck-in of S-2, mm	Neck-in of S-3, mm	Neck-in of S-4, mm	Neck-in of S-5, mm	Average Neck-in, mm	STDEV
Test Series 1							
TP-1	111	109	111	111	111	111	0.89
TP-2	160	162	164	165	164	163	2.00
TP-3	238	238	239	243	243	240	2.59
TP-4	305	306	304	306	308	306	1.48
TP-5	360	359	355	352	340	353	8.04
TP-6	389	392	395	397	399	394	3.97
TP-7	410	410	410	411	411	410	0.55
Test Series 2							
TP-1	105	105	105	105	105	105	0.00
TP-2	158	159	155	157	158	157	1.52
TP-3	210	207	205	211	211	209	2.68
TP-4	267	264	265	267	264	265	1.52
TP-5	325	322	326	320	315	322	4.39
TP-6	357	356	356	354	356	356	1.10
TP-7	401	403	402	403	403	402	0.89
Test Series 3							
TP-1	108	107	107	105	108	107	1.22
TP-2	138	143	138	140	143	140	2.51
TP-3	183	180	185	187	190	185	3.81
TP-4	239	239	240	231	246	239	5.34
TP-5	304	304	304	304	301	303	1.34
TP-6	346	347	347	347	348	347	0.71
TP-7	379	381	381	381	378	380	1.41

Table 18 *Neck-in results of adhesion trial 3 (cont.)*

Air gap, mm	Neck-in of S-1, mm	Neck-in of S-2, mm	Neck-in of S-3, mm	Neck-in of S-4, mm	Neck-in of S-5, mm	Average Neck-in, mm	STDEV
Test Series 4							
TP-1	106	105	104	105	105	105	0.71
TP-2	136	134	136	138	135	136	1.48
TP-3	172	176	174	172	176	174	2.00
TP-4	225	222	227	221	223	224	2.41
TP-5	281	284	287	286	287	285	2.55
TP-6	350	350	349	350	350	350	0.45
TP-7	375	377	377	376	377	376	0.89
Test Series 5							
TP-1	106	106	108	107	105	106	1.14
TP-2	127	128	132	130	130	129	1.95
TP-3	169	165	162	163	168	165	3.05
TP-4	213	214	214	216	223	216	4.06
TP-5	278	278	279	278	280	279	0.89
TP-6	331	331	331	330	332	331	0.71
TP-7	370	370	369	370	371	370	0.71

Table 19 *Neck-in results of adhesion trial 5*

Air gap, mm	Neck-in of S-1, mm	Neck-in of S-2, mm	Neck-in of S-3, mm	Neck-in of S-4, mm	Neck-in of S-5, mm	Average Neck-in, mm	STDEV
Test Series 1							
TP-1	63	61	61	61	61	61	0.89
TP-2	105	108	108	110	107	108	1.82
TP-3	188	191	190	189	188	189	1.30
TP-4	252	252	254	253	252	253	0.89
TP-5	306	307	308	308	309	308	1.14
TP-6	340	343	345	344	346	344	2.30
TP-7	369	368	369	368	361	367	3.39
Test Series 2							
TP-1	64	64	64	64	69	65	2.24
TP-2	108	106	107	109	107	107	1.14
TP-3	167	168	162	158	163	164	4.04
TP-4	234	238	235	236	237	236	1.58
TP-5	284	289	290	290	289	288	2.51
TP-6	323	322	325	322	327	324	2.17
TP-7	342	342	346	345	346	344	2.05
Test Series 3							
TP-1	59	60	60	60	59	60	0.55
TP-2	85	86	86	88	91	87	2.39
TP-3	136	134	129	132	131	132	2.70
TP-4	176	177	176	178	179	177	1.30
TP-5	229	232	228	227	226	228	2.30
TP-6	275	277	278	275	273	276	1.95
TP-7	310	311	312	312	308	311	1.67

Table 20 *Neck-in results of adhesion trial 5 (cont.)*

Air gap. mm	Neck-in of S-1, mm	Neck-in of S-2, mm	Neck-in of S-3, mm	Neck-in of S-4, mm	Neck-in of S-5, mm	Average Neck-in, mm	STDEV
Test Series 4							
TP-1	58	57	56	55	56	56	1.14
TP-2	79	81	81	80	80	80	0,84
TP-3	110	110	111	112	116	112	2,49
TP-4	157	158	158	158	158	158	0.45
TP-5	210	209	208	210	212	210	1.48
TP-6	257	259	259	259	257	258	1.10
TP-7	301	301	300	309	307	304	4.10
Test Series 5							
TP-1	49	49	47	50	50	49	1.22
TP-2	74	76	73	75	73	74	1.30
TP-3	104	103	103	104	104	104	0.55
TP-4	144	145	145	147	148	146	1.64
TP-5	203	203	202	203	202	203	0.55
TP-6	248	249	248	247	249	248	0.84
TP-7	299	297	296	295	291	296	2.97

Table 21 *Neck-in results of adhesion trial 4*

Line Speed, m/min	Neck-in of S-1, mm	Neck-in of S-2, mm	Neck-in of S-3, mm	Neck-in of S-4, mm	Neck-in of S-5, mm	Average Neck-in, mm	STDEV
Test Series 1							
TP-1	67	68	68	68	68	67.80	0.45
TP-2	66	65	67	66	65	65.80	0.84
TP-3	62	61	61	63	62	61.80	0.84
TP-4	61	61	61	61	61	61	0
Test Series 2							
TP-1	70	68	69	69	69	69	0.71
TP-2	66	64	65	65	64	64.80	0.84
TP-3	63	65	64	63	63	63.60	0.89
TP-4	62	61	62	61	61	61.40	0.55
Test Series 3							
TP-1	62	64	66	65	62	63.80	1.79
TP-2	64	60	63	60	63	62.00	1.87
TP-3	60	61	63	62	61	61.40	1.14
TP-4	61	60	60	61	62	60.80	0.84
Test Series 4							
TP-2	60	61	63	61	62	61.40	1.14
TP-3	59	60	61	60	58	59.60	1.14
TP-4	59	60	61	59	60	59.80	0.84
TP-5	59	59	60	59	59	59.20	0.45

APPENDIX C. ADHESION RESULTS

In Appendix C, numerical representation of adhesion results of all adhesion trials are given. Description of test series and test points can be found thesis trials section.

Table 22 *Adhesion results of trial 1 and 3 and 5*

Air Gap, mm	Adhesion Levels			
	Screening Part of Adhesion Trial 1	Actual Part of Adhesion Trial 1	Adhesion Trial 3	Adhesion Trial 5
Test Series 1				
TP-1	2	1	1	1
TP-2	2	1	1	1
TP-3	3	1	1	1
TP-4	3	1	1	1
TP-5	3	2	1	2
TP-6	4	2	2	2
TP-7	4	2	2	3.5
Test Series 2				
TP-1	3	1	1	1
TP-2	2	1	1	1
TP-3	3	1	1	1
TP-4	4	1	1	2
TP-5	4	1	1	2
TP-6	5	2	1	2,5
TP-7	5	2	1	4
Test Series 3				
TP-1	1	1	1	2
TP-2	2	1	1	2
TP-3	2	1	1	2
TP-4	2	2	1	2
TP-5	3	2	1	2
TP-6	3	2	1	2
TP-7	3	2	2	4

Table 23 *Adhesion results of trial 1 and 3 and 5 (cont.)*

Air Gap, mm	Adhesion Levels			
	Screening Part of Adhesion Trial 1	Actual Part of Adhesion Trial 1	Adhesion Trial 3	Adhesion Trial 5
Test Series 4				
TP-1	4	2.5	1	2
TP-2	3	2	1	2
TP-3	3	2	1	2
TP-4	4	2	1	2
TP-5	4	3	1	2
TP-6	5	2	2	3
TP-7	5	3	2	4
Test Series 5				
TP-1	4	2	1	2
TP-2	4	2	1	2
TP-3	3.5	2	1	2
TP-4	4.5	2	1	2
TP-5	2	2	2	3
TP-6	5	3	2	4
TP-7	5	3	2	5

Table 24 *Adhesion results of trial 2*

Line Speed, m/min	Adhesion level
Test Series 1	
TP-1	2
TP-2	1
TP-3	1
TP-4	1
Test Series 2	
TP-1	4
TP-2	3 - Cohesive adhesion was occurred
TP-3	2
TP-4	1
TP-5	1
Test Series 3	
TP-1	4
TP-2	varied from 2-4
TP-3	2
TP-4	1
TP-5	mostly 1 but little region is 3
Test Series 4	
TP-1	5
TP-2	4
TP-3	varied between 3-5
TP-4	varied between 3-4
TP-5	varied between 1-4
Test Series 5	
TP-1	varied between 4-5
TP-2	varied between 2-4
TP-3	varied between 2-4
TP-4	varied between 1-3
TP-4	varied between 1-3

Table 25 *Adhesion results of trial 4*

Line Speed, m/min	Adhesion level
Test Series 1	
TP-1	4
TP-2	3
TP-3	2
TP-4	2
Test Series 2	
TP-1	5
TP-2	4.5
TP-3	4
TP-4	3
Test Series 3	
TP-1	4
TP-2	4
TP-3	4
TP-4	3
Test Series 4	
TP-2	4.5
TP-3	4.5
TP-4	4
TP-5	3