Paper-based flexible laminates tendency to curl

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Investigating the effect of humidity and temperature

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Abstract

The main purpose of this study is to assess the impact of environmental conditions on paper-based flexible laminates tendency to curl. Any deviation in flatness may indeed affect materials processability. Two different paper-based flexible laminates are considered: a widely processed heat-sealable yogurt lid (paper / metallized polyethylene terephthalate / heat-sealing lacquer) and a prototype laminate (aluminium / paper / polyethylene). Packaging materials sheets (54*36 cm) are namely sampled from packaging materials rolls. Their tendency to curl is assessed by implementing the cross-cut method described in the German DIN 55403 standard (Deutsches Institut für Normung (DIN), 2014). Fifteen climates are tested i.e. five different relative humidity values (30%, 40%, 50%, 60% and 70%) and three different temperature values (20°C, 25°C and 30°C). The effect of a polyethylene wrapping is also evaluated as a potential solution to prevent or reduce curl. For the yogurt lid which is shown to be sensitive to climate conditions, deformation occurs in both machine and cross-machine directions: average curl values respectively reach 13 millimetres and 37 millimetres. An equation to predict curl parameters as a function of climate and room conditions is furthermore suggested (r^2 = 72%). For the prototype laminate which appears not to be sensitive to climate conditions, deformation happens to be much lower than for the first material and only occurs in the machine direction. Average curl values reach 3 millimetres in the machine direction and zero millimetres in the cross-machine direction. Finally, considering the material-specific results obtained for the two laminates, no general conclusion can be drawn as regard to the polyethylene wrapping effect.

Keywords: flexible laminates, paper, curl, relative humidity, temperature.

Executive summary

Introduction

Flexible packages have started replacing traditional containers. Between 2012 and 2017, the volume of F&B flexible packages actually increased by 11% in the world as opposed to 5% for the global packaging industry growth; by 5% in Western Europe as opposed to 1% for the Western Europe packaging industry growth (Euromonitor International, 2018). Flexible packaging is all the more interesting that it enables creative eye-catching designs which generate high market appeal, lower shipping and storage costs (smaller and lighter packages) as well as a lower environmental impact (fewer resources and less energy for production, lower transport-related CO_2 emissions and convenient features which may help reduce food waste) (Lingle, 2012; Hrinya, 2017).

Within this category, flexible multi-layer constructions (also called laminates) can be designed to meet specific performance requirements. Each layer indeed provides the multi-ply packaging material with a particular function or particular functions such as gas barrier, moisture barrier, light barrier, chemical resistance, puncture resistance, strength and heat sealing ability (Hrinya, 2017). Paper materials can namely be laminated: they are often selected for their stiffness and dead-fold properties (i.e. once folded, the material retains its shape and does not unfold - Riley A., 2012) as well as their environmentally-friendly appearance.

However, paper-based packaging materials are well-known for causing important issues during converting and filling processes due to paper hygroinstability (i.e. dimensional change due to fluctuations of the surrounding atmosphere moisture - Lindner, 2018). Curl phenomenon is a common problem which may happen in non-climatized manufacturing facilities. It can be defined as “an undesirable condition caused by uneven rates of absorption or evaporation of moisture, uneven rates of contraction or expansion, or internal stresses in the material” (Catty Corporation, 2017).

The purpose of this master thesis was to describe and assess the effect of climate conditions on paper-based flexible laminates tendency to curl so as to understand to what extent environmental conditions can affect their processability. It was suggested by and conducted for the German multinational food company Unternehmensgruppe Theo Müller - the sixteenth biggest milk processor in the world in 2016 (Cornall, 2016).

Objectives

The following main objectives were defined:

1. Identify the main factors leading to paper-based flexible laminates curl phenomenon (namely environmental conditions);
2. Study the behaviour of two different paper-based flexible laminates under varying relative humidity (RH) and temperature conditions;
3. Assess climate-related risk for both materials;
4. Test the effect of low-density polyethylene wrapping (abbreviated to PE wrapping) as a solution to prevent or reduce packaging materials tendency to curl.

Materials and method

Two different laminates were considered:

- a widely processed 60-µm thick heat-sealable yogurt lid made up of paper and metalized polyethylene terephthalate, coated with a heat seal lacquer (Pap/mPET/HSL);
- a 67-µm thick prototype laminate made up of aluminium foil, paper and polyethylene film (Alu/Pap/PE).

Considering the very few packaging materials rolls which were made available as well as their dimensions (a single 106-mm diameter test roll for the Alu/Pap/PE prototype laminate), it was decided to sample materials sheets from rolls.

The cross-cut method was selected as test method for the determination of the tendency to curl (Deutsches Institut für Normung (DIN), 2014). On the whole, it consists in cutting crosses in the packaging material web using a given cutting pattern and measuring specific distances, in the machine direction (MD) (= a parameter in mm) and the cross-machine direction (CD) (= b parameter in mm) (see Figure 0.1). Regardless of the direction, two different ways of measuring the tendency to curl are distinguished. When the distance or gap between the material edges is shorter than 5 mm, the tendency to curl should be measured as the shortest height between the table and the material edges. On the other hand, when the distance between the material edges is longer than 5 mm, the tendency to curl should be measured as the shortest distance between the material edges.

Laminates tendency to curl was assessed under fifteen climates i.e. under five RH values (30%, 40%, 50%, 60% and 70%) and three temperature values (20°C, 25°C and 30°C), which were defined based on RH and temperature data recorded within a factory and documented by the quality team.

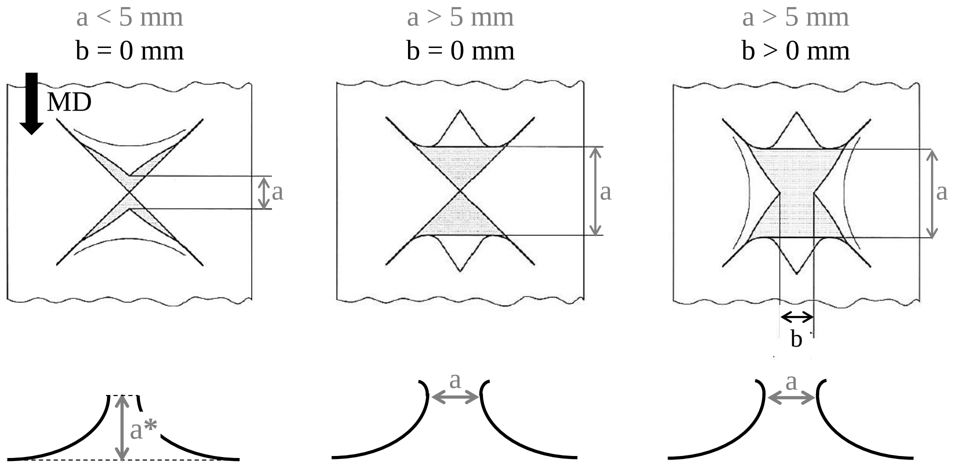


Figure 0.1 The cross-cut method (adapted from Deutsches Institut für Normung (DIN), 2014).

Results and discussion

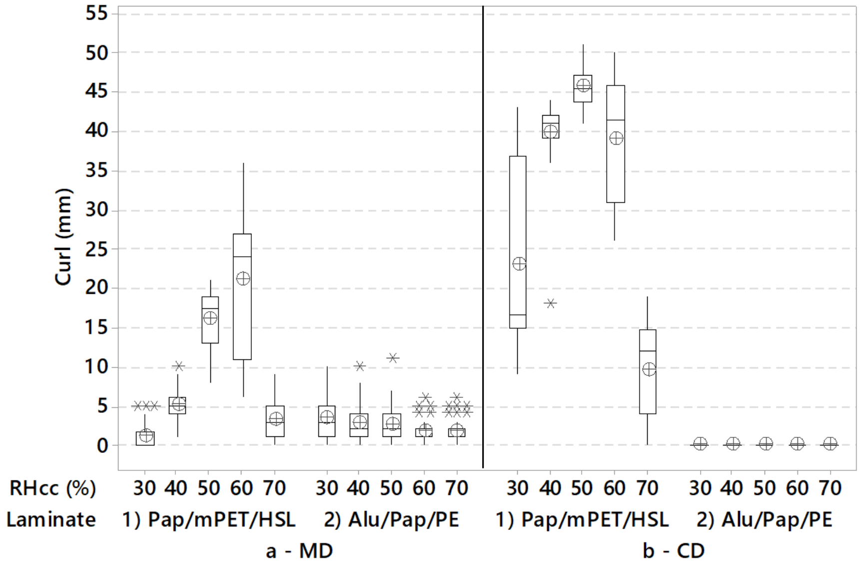
Main factors leading to paper-based flexible laminates curl

Numerous factors can lead to paper-based flexible laminates curl, at different levels (cellulose, paper, laminate, roll) and different stages (converting, warehousing, filling). RH and temperature happened to be the most well-documented factors. However, this does not mean that they are the most significant ones since no hierarchy has been described yet. There is currently a lack of information as regard to paper-based flexible laminates curl phenomenon.

Impact of relative humidity and temperature on the two studied laminates behaviour (Figure 0.2 and Figure 0.3)

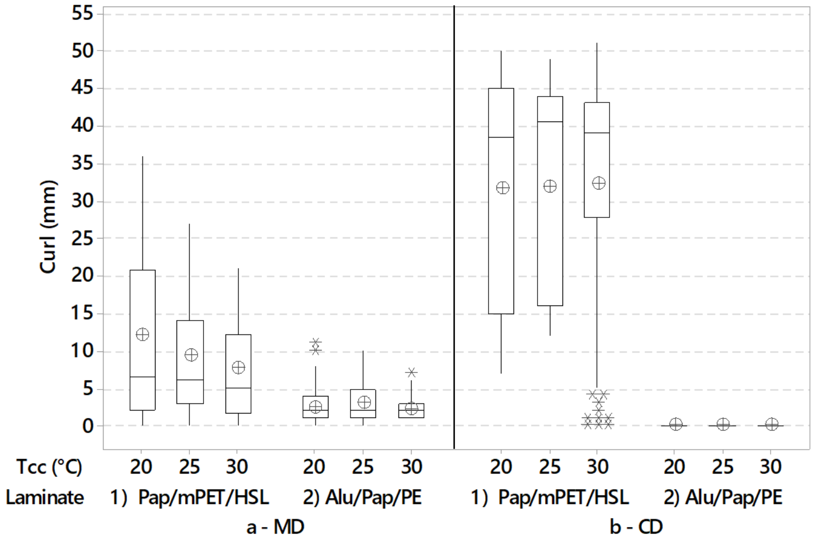
Concerning the Pap/mPET/HSL laminate, deformation occurred both in the MD and the CD: average curl values respectively met 13 mm and 37 mm. RH and temperature were both shown to have a significant effect on its tendency to curl; RH more significantly impacted its tendency to curl than temperature. Highest curl average values were reached for 50% and 60% RH whereas lowest ones were reached for 30% and 70% RH. The material was demonstrated to be sensitive to climate conditions.

Concerning the Alu/Pap/PE prototype, deformation only occurred in the MD: the average curl value met 3 mm. Whatever the environmental conditions, the CD-oriented curl parameter was always equal to 0 mm. On whole, the laminate tendency to curl was shown to be low and not sensitive to climate conditions.



Note: Data for 20°C, 25°C and 30°C were aggregated.

Figure 0.2 Boxplot[1] comparing the RH effect on the studied laminates tendency to curl.



Note: Data for 30%, 40%, 50%, 60% and 70% were aggregated.

Figure 0.3 Boxplot comparing the temperature effect on the studied laminates tendency to curl.

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[1] Box plots are a standardized way of representing data distributions based on five statistical measures: minimum, first quartile, median, third quartile and maximum. The data set average value is marked using this symbol: ⊕.

Materials climate-related risk assessment

The Pap/mPET/HSL laminate climate-related risk was characterized as high. It was namely possible to observe and describe paper fibres hygroexpansion in the MD and the CD under various RH. Considering the given laminate structure, paper may directly exchange water with the environment (no barrier).

The Alu/Pap/PE prototype laminate climate-related risk was characterized as low. Considering the given laminate structure, paper is sandwiched between aluminium and PE which probably protect it i.e. prevent it from absorbing water.

Solution to reduce paper-based laminates tendency to curl (Figure 0.4)

In the case of the Pap/mPET/HSL laminate, PE wrapping led to a stronger tendency to curl (while limiting absolute variations in curl) and was therefore not considered as protective.

In the case of the Alu/Pap/PE laminate, PE wrapping led to lower curl averages in the MD (while enabling relatively high absolute variations in curl) and was thus considered as protective.

These opposite observations made the PE wrapping effect complex to analyse. Considering the given material-specific results, it was not possible to draw a single general conclusion for all the paper-based flexible laminates.

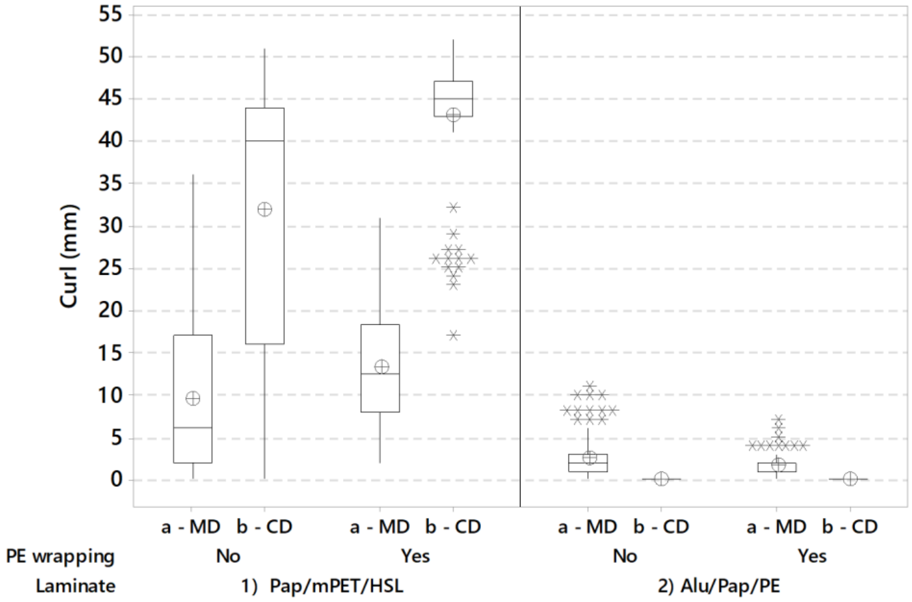

Figure 0.4 Boxplot comparing the effect of PE wrapping on the two studied laminates tendency to curl.

Conclusions

Recommendations usually made to avoid paper-based packaging materials curl phenomenon were shown not to be applicable to paper-based laminates. Indeed, RH and temperature ranges (45-60% RH and 20-25°C) recommended by paper suppliers did not appear particularly meaningful. Considering the given material-specific results, laminates behaviour may depend on the different materials which are bound together and should not be boiled down to the behaviour of one single material. Consequently, this kind of study should be conducted to assess RH and temperature effects on the tendency to curl of any single laminate (no possible extrapolation from one material to another).

Further research recommendations

This thesis investigated the effect of humidity and temperature as regard to paper-based flexible laminates tendency to curl. Two main research areas were finally suggested. Firstly, conducting a similar study at the roll scale (as opposed to the sheet scale) which would probably be more meaningful from the industry perspective. Differences in curl might indeed be expected depending on the location of the samples towards the roll core. Secondly, undertaking a study comparing the effect of the lamination process nature to the one of the environmental conditions on paper-based flexible laminates tendency to curl. Changing the lamination process could indeed reduce the tendency to curl much more than trying to protect packaging materials from "harmful" environmental conditions. The idea behind would be to hierarchize the different factors leading to curl so as to define effective solutions to this problem.

Acknowledgments

This master thesis unfortunately brings the amazing two-year international Food Innovation and Product Design (FIPDes) experience to a close. I feel very fortunate to have been part of the sixth cohort. I will never forget what happened during these semesters in France, Ireland, Sweden and Germany. Especially all the people I met, could they be colleagues, professors, students or else flatmates.

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Aretsried, May 2018

Clémentine Muller

List of acronyms and abbreviations

a	Machine direction-oriented curl parameter measured as the shortest distance between the material edges
a*	Machine direction-oriented curl parameter measured as the shortest height between the table and the material edges
Alu/Pap/PE	Aluminium/Paper/Polyethylene (laminate)
b	Cross-machine direction-oriented curl parameter measured as the shortest distance between the material edges
CD	Cross-machine direction
F&B	Food and beverage(s)
LDPE	Low-density polyethylene
MD	Machine direction
Pap/mPET/HSL	Paper/Metallized polyethylene terephthalate/Heat-sealing lacquer (laminate)
PE wrapping	Low-density polyethylene wrapping
RH	Relative humidity
RH_{cc}	Climate chamber relative humidity
RH_r	Room relative humidity
T_{cc}	Climate chamber temperature
T_r	Room temperature

1 Introduction

Packaging can be defined as "a coordinated system made up of any materials of any nature, to be used for preparing goods for containment, protection, transport, handling, distribution, delivery and presentation" (Hellström & Olsson, 2016). Packaging materials may be selected as regard to physicochemical and functional properties, operational constraints and marketing considerations. The aim of the thesis is to assess the impact of climate conditions on paper-based flexible laminates tendency to curl. Curl, as opposed to flatness, may indeed affect packaging materials processability. The given study was suggested by and conducted for the German multinational food company Unternehmensgruppe Theo Müller, the sixteenth biggest milk processor in the world in 2016 (Cornall, 2016). This first chapter justifies its relevance, defines the research problem and sets the study boundaries.

1.1 Background

According to BillerudKorsnäs (2016), urbanisation, sustainability, mobility revolution, digitalization and connectivity, value chains transformation and customerisation are the six main megatrends that are currently shaping the world. In the field of the packaging industry, packaging sustainability has thus been described as a strong strategic tool for brands: 72% of the consumers would actually be ready to pay 10 to 20% more for products that are packed in packages showing sustainable benefits (BillerudKorsnäs, 2016; Riley S., 2018; Sabo, 2018). Packaging sustainability can be addressed in different ways: by reducing packaging materials environmental impact, increasing logistical efficiency and/or minimizing food waste (Molina-Besch & Pålsson, 2016). However, because of waste regulations and taxes, most of the improvements happened to be packaging materials-related: companies often focus on minimising the use of resources (Riley S., 2018).

In this context, flexible laminates are particularly attractive to industrials since they enable to decrease landfilled packaging materials weight. Containers or bottles are said to require at least twice the amount of resources used for films for instance (Packaging & Converting Intelligence, 2016; Ecolean, 2017). More generally, multi-layer packaging appears as an optimal solution as regard to meeting consumer needs (convenience and personalisation), fulfilling product requirements (customized protection and extended shelf-life without adding preservatives) and complying with industrial constraints (cost mainly) (Packaging & Converting Intelligence, 2016; Waste360, 2018; Sabo, 2018).

However, plastic- and aluminium-based laminates may not always look natural enough for the growing number of “environmentally-conscious consumers”. How to make these packages look greener then? Well, by creating a “sustainable design”. In other words, by using earthy colours, brown packaging or paper-like materials; in the best case, by laminating paper (Packaging & Converting Intelligence, 2015). In fact, paper and board are usually considered as the most sustainable packaging materials (BillerudKorsnäs, 2017). Reasons include their renewable origin (wood fibres), biodegradability and high recycling rates (Fortin, 2012; Packaging & Converting Intelligence, 2015).

1.2 Research problem

Laminating paper certainly provides packaging with stiffness and dead-fold properties (i.e. once folded, the material retains its shape and does not unfold - Riley A., 2012) and/or a greener appearance. However, this packaging material is well-known for causing important issues during converting and filling processes due to its hygroinstability (i.e. the dimensional change due to fluctuations of the surrounding atmosphere moisture - Lindner, 2018). Besides, multi-layer structures which combine different materials showing various physicochemical properties may also be responsible for some problems among which curl i.e. “an undesirable condition caused by uneven rates of absorption or evaporation of moisture, uneven rates of contraction or expansion, or internal stresses in the material” (Catty Corporation, 2017). In laminated structures, an individual lamina is actually restrained by the other laminae and is not free to expand which generates stresses (Free University of Brussels (ULB), n.d.). The current lack of information as regard to paper-based flexible laminates curl phenomenon motivates this study.

1.3 Purpose and aims

The purpose of this thesis is to describe and assess the effect of climate conditions on paper-based flexible laminates tendency to curl so as to understand to what extent environmental conditions can affect their processability. It aims at:

- Identifying the main factors leading to paper-based flexible laminates curl phenomenon (namely environmental conditions);
- Studying the behaviour of two different paper-based flexible laminates under varying relative humidity (RH) and temperature conditions;
- Assessing climate-related risk for both materials;
- Testing the effect of low-density polyethylene wrapping (abbreviated to PE wrapping) as a solution to prevent or reduce packaging materials tendency to curl.

1.4 Focus and demarcation

This study focuses on climate-related curl which should be distinguished from reel-related curl i.e. curl which may happen to packaging materials stored for a long time on reels, tightly wound around a narrow diameter core. Reel-related curl is therefore characterized as a deviation from flatness in the machine direction (Iggesund Paperboard, 2010).

The test method for the determination of the tendency to curl was chosen from the three methods which are described in the German standard DIN 55403 (Deutsches Institut für Normung (DIN), 2014).

Materials were limited to two different paper-based flexible laminates. The first one is made up of paper and metalized polyethylene terephthalate; it is coated with a heat seal lacquer. No curl-related problem has been encountered yet. The idea is to document the tendency to curl for a widely processed packaging material. The second one consists of aluminium foil, paper and polyethylene film. Its tendency to curl appears as a critical point for the development of a new packaging solution. The given materials are respectively abbreviated to Pap/mPET/HSL and Alu/Pap/PE.

2 Literature review

This second chapter is a narrative literature review of paper-based flexible laminates properties, manufacturing processes and curl phenomenon. Insights into the food and beverage (F&B) flexible packaging industry are also introduced. As very few detailed pieces of information have been published concerning paper-based flexible laminates, assumption was made that technical data for paper and paper-based packaging materials are applicable to paper-based laminates. Scientific articles were supplemented with technical reports from the paper packaging industry. Packaging suppliers actually directly face curl issues and need to provide their customers with solutions to prevent or at least reduce curl.

2.1 The “flexible” packaging industry

In 2017, F&B packages represented 73% of the global packaging industry in volume i.e. about 3 223 883 million retail/trade-off units; 81% of Western Europe packaging industry in volume i.e. about 599 561 million retail/trade-off units. Among the different packaging types, flexible packaging and rigid plastics clearly appear as the two most important categories (see Figure 2.1). They respectively account for 40% and 27% of the total packaging volumes in the world; 49% and 20% of the total packaging volumes in Western Europe; 35% and 31% of the total packaging volumes in Germany (Euromonitor, 2018)[2]. (Euromonitor International, 2018).

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[2] Figures from Euromonitor International (2018) are expressed in volume and namely in retail/off-trade units which correspond to the number of packaging units sold to the consumer through all retail channels. A 1-litre plastic bottle would account for one retail/off-trade unit volume for instance.

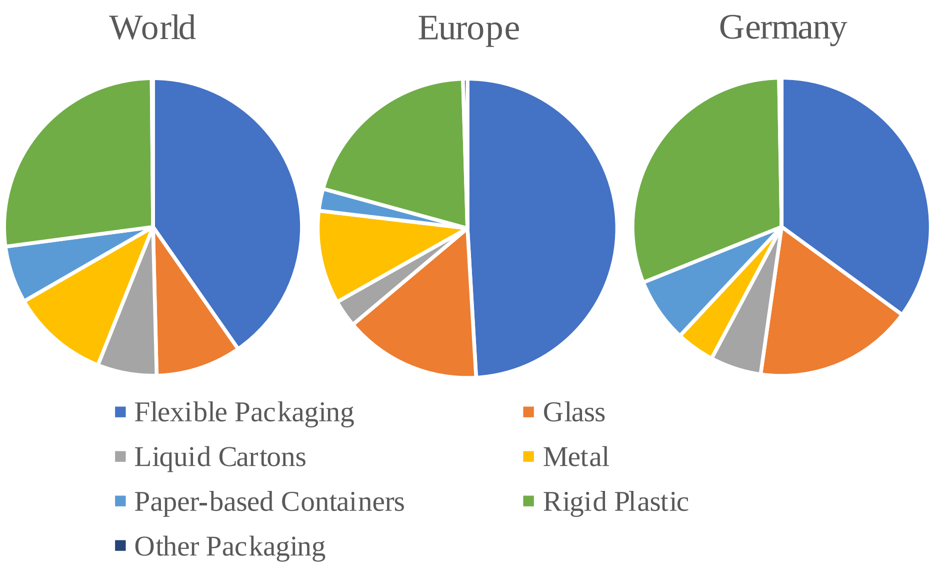


Figure 2.1 Volume share of the different packaging types for F&B in 2017 (Euromonitor International, 2018).

2.1.1 **A growing demand for flexible plastic packaging solutions**

Flexible packaging may be defined as “a package that can change shape when filled with its contents” (Hrinya, 2017). It namely includes bags, pouches, wraparound labels, shrink or stretch sleeves, and lidding. In 2017, flexible packages for F&B represented 29% of the global and the Western Europe packaging industries in volume. Between 2012 and 2017, the volume of F&B flexible packages increased by 11% in the world as opposed to 5% for the global packaging industry growth; by 5% in Western Europe as opposed to 1% for the Western Europe packaging industry growth (Euromonitor International, 2018). Actually, flexible packages have started replacing traditional containers. From the packaging industry perspective, they enable creative eye-catching designs which generate high market appeal, lower shipping and storage costs (smaller and lighter packages) as well as a lower environmental impact (fewer resources and less energy for production, lower transport-related CO_2 emissions and convenient features which may help reduce food waste). From the consumer perspective, they particularly fit millennials on-the-go lifestyles by providing them with more convenient (easy-to-open and resealable features for example) and “sustainable” packaging solutions (Lingle, 2012; Hrinya, 2017).

Among the different F&B flexible packaging types, flexible plastic appears as the predominant category (see Figure 2.3). It accounts for 77% of the total flexible packaging volumes in the world; 74% of the total flexible packaging volumes in

Western Europe and in Germany (Euromonitor International, 2018). Flexible plastic packaging covers hermetically sealed packs, flow wrap, over wrap and any other flexible plastic wrapping material (metallized plastic included) that is not a stand-up pouch. It mainly corresponds to the primary packages (i.e. packages in contact with the product) of confectionery, bakery products, frozen food, snacks or else pasta/rice (Euromonitor International, 2013).

Figure 2.2 Examples of flexible plastic packages.

Besides, it can be noticed that if paper-based multilayer flexible packaging (Flexible Aluminium/Paper and Flexible Paper/Plastic) only reaches 4% of the total flexible packaging volumes in the world and 5% of the total flexible packaging volumes in Western Europe, it actually represents 9% of the total flexible packaging volumes in Germany i.e. 4 309.3 million retail/trade-off units (Euromonitor International, 2018).

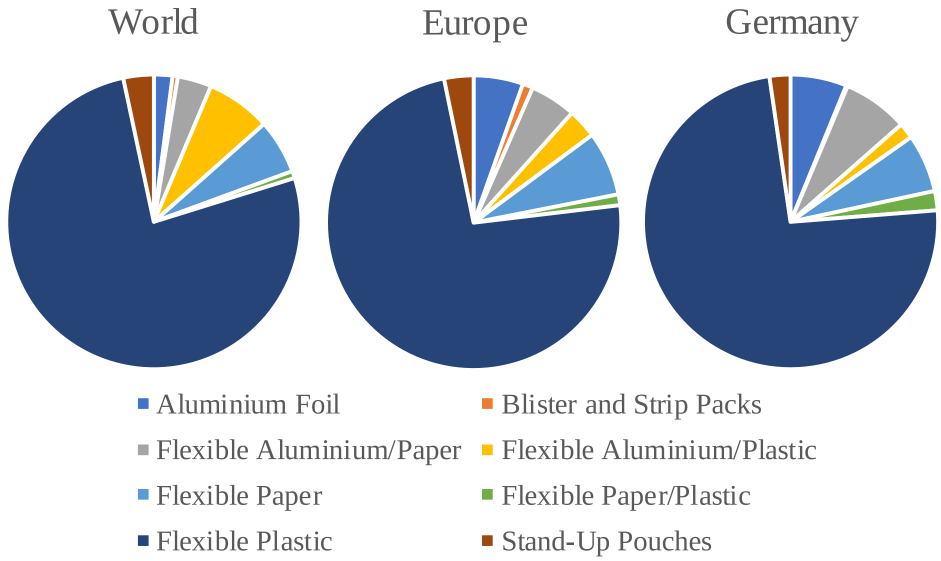


Figure 2.3 Volume share of the different flexible packaging types for F&B in 2017 (Euromonitor International, 2018).

2.1.2 The nature of flexible laminates

Flexible laminates or composites may be defined as flexible multi-layer constructions. Each layer provides the multi-ply packaging material with a particular function/particular functions such as gas barrier, moisture barrier, light barrier, chemical resistance, puncture resistance, strength and heat sealing ability (see Figure 2.4). As a result, packaging materials can be designed to meet specific performance requirements (Hrinya, 2017). Common laminates are described in Figure 2.5.

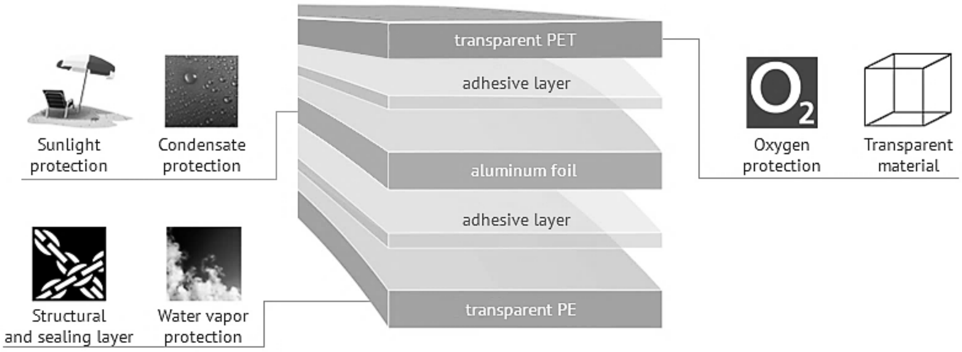
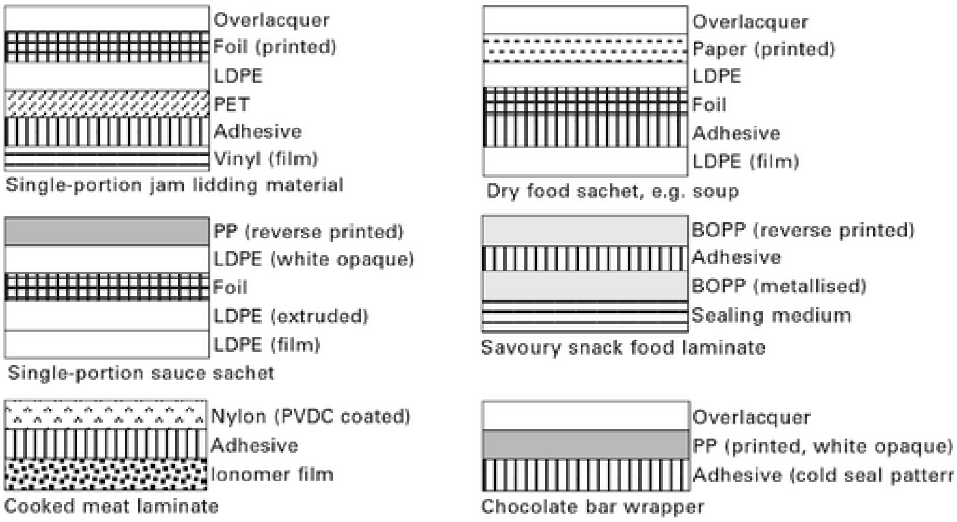


Figure 2.4 Description of one flexible multilayer packaging material for ketchup (Uniflex, n.d.).



Note: LDPE = Low-density polyethylene, PET = Polyethylene terephthalate, PP = Polypropylene, BOPP = Biaxially-oriented polypropylene, PVDC = Polyvinylidene chloride

Figure 2.5 Examples of flexible laminates for different food applications (Riley A. , 2012).

2.2 Paper-based laminates materials properties

No single film can satisfy all packaging requirements, hence the advantage of combining different materials physicochemical and functional properties into one single multilayer structure that is greater than the sum of its parts. Flexible packaging requires substrates (mainly paper, aluminium and polymers) but also adhesives, primers or else coatings. In the case of F&B, when selecting the different packaging materials components, one strong constraint concerns the compliance with food contact materials regulations i.e. respecting the specific migration limits (Lingle, 2012; Hrinya, 2017). This section focuses on the different components which are part of the two studied laminates, Pap/mPET/HSL and Alu/Pap/PE.

2.2.1 **Paper**

Paper lamination provides packages with stiffness and dead-fold properties (Riley A. , 2012).

Paper is made up of cellulose fibres which are hygroscopic. Therefore, fibres exchange moisture with their environment until equilibrium with that environment is reached: they absorb moisture from a humid atmosphere while releasing moisture in a dry atmosphere (Glatfelter, 2005). There is almost no way to prevent paper from reaching an equilibrium with the surrounding air (Iggesund Paperboard, 2010).

Table 2.1 introduces common physical quantities which are usually referred to when discussing paper hygroexpansion i.e. “the dimensional change due to fluctuations of the RH of the surrounding atmosphere which affects the moisture content of the paper” (Lindner, 2018).

Normal paper moisture content usually reaches 2 to 10% depending on the type of paper, its past moisture history and the environmental conditions to which it is exposed to. When exposed to extreme conditions, paper moisture content may reach 0.5 to 13% (Glatfelter, 2005).

Paper hygroinstability can cause important issues during both converting and filling processes. Indeed, moisture significantly affects most of the paper properties: dimensions, flatness, conductivity, strength and fold. An increase in the environmental humidity namely leads to an increase in cellulose fibre dimensions while a decrease in the environmental humidity has the opposite effect. Considering that fibres are usually oriented in the machine direction (MD) (see Figure 2.6) and that individual fibres diameter changes two to five times more than their length, paper deformation mainly occurs in the cross-machine direction (CD) (see Figure 2.7) and its thickness (Iggesund Paperboard, n.d.; Glatfelter, 2005; Lindner, 2018).

Table 2.1 Overview of some physical quantities related to paper hygroexpansion.

Physical quantity	*Explanation*	*Formula*	*Unit*
Relative humidity (RH)	The “measure of the amount of water in the air, at a specific air temperature, expressed as a percentage of the maximum amount of water the air can hold at that temperature [saturation]” (Glatfelter, 2005). Ratio between the partial pressure of water vapor at the considered temperature $p_{H2O}(T)$ and the saturation pressure of water vapor at the same temperature $p^*_{H2O}(T)$ (Vaisala Oyj, 2013).	$RH = \frac{p_{H_2O}(T)}{p^*_{H_2O}(T)} * 100$	%
Paper moisture content (MC_{paper})	The percentage of the total paper weight which is water. Ratio between the mass of water (the mass of the paper m minus its mass after having been oven-dried m_{od}) and the mass of paper (Reeb & Milota, 1999).	$MC_{paper} = \frac{m - m_{od}}{m} * 100$	%
Coefficient of Moisture or Hygroscopic Expansion (β)	Ratio between the material relative dimensional change ΔL (%) and the corresponding variation in its moisture content ΔMC (%) (Wu & Suchsland, 1996; TA Instruments, 2007; Iggesund Paperboard, 2010).	$\beta = \frac{\Delta L}{\Delta MC}$	%/%

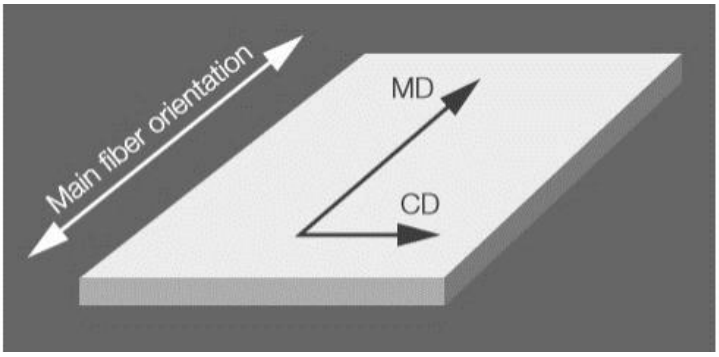
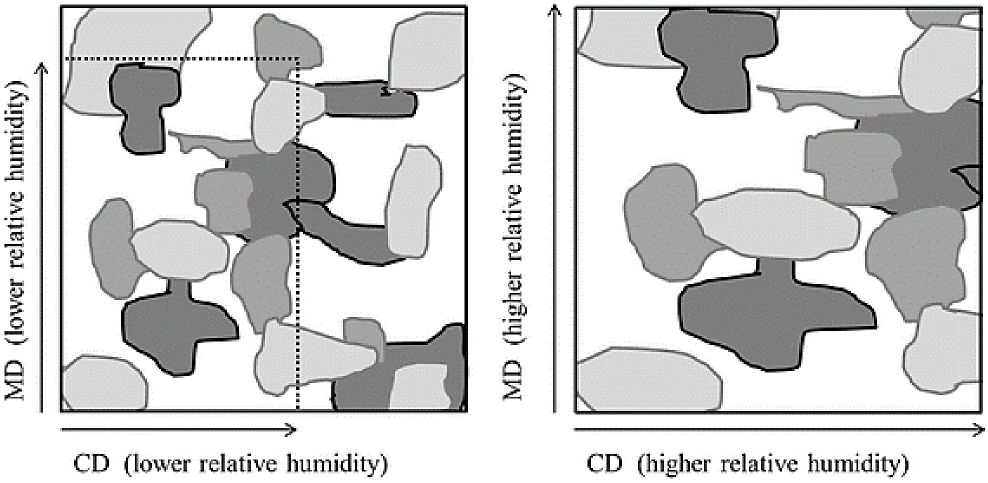


Figure 2.6 Orientation of cellulose fibres (Iggesund Paperboard, 2010).



Note: This figure is a schematic representation of a speckle pattern which was obtained using a digital correlation technique. Ink was spread on the paper surface and pictures were taken for different RH values.

Figure 2.7 Paper deformation due to moisture (Lindner, 2018).

However, curl will disappear when paper moisture content reaches the equilibrium RH i.e. the RH of the atmosphere at a particular temperature at which paper neither absorbs nor releases moisture (Glatfelter, 2005; Dignesh, Krishnan, Rajasekaran, & Kumar, 2013).

2.2.2 **Aluminium foil and metallized films**

Aluminium lamination provides packages with barrier properties to light, moisture, gases and grease (Iggesund Paperboard, 2010). It also shows dead-fold properties (Marsh & Bugusu, 2007). Aluminium is dimensionally stable (United States Patent No. U.S. 3,098,780, 1963).

Aluminium foil is obtained by rolling pure aluminium metal into thin sheets which thickness may range from 6.3 to 12 µm (Dixon, 2011).

On the other hand, aluminium-metallized films correspond to polymer films coated with a very thin layer of aluminium which thickness may reach 30 nm. The most common metallized polymers are oriented polypropylene (OPP), biaxially oriented polypropylene (BOPP), polyethylene terephthalate (PET) and biaxially oriented nylon (BON). The involved process is called vacuum coating or vacuum metallizing: aluminium evaporates inside a vacuum chamber and condenses to form a solid coating. Aluminium-metallized films represent cheaper alternatives to aluminium foils (Marsh & Bugusu, 2007).

2.2.3 **Heat sealing agents**

Lacquers or coatings can be used to provide packaging materials with heat sealing ability which plays a role in packaging integrity. Three different types can be distinguished: hot melts, water-based and wax-based. Heat sealing lacquer/coating selection is made as regard to its adhesion for the given material to be applied to as well as its physicochemical and functional properties such as viscosity, coefficient of friction, minimum seal temperature, strength or peel resistance, scuff resistance, chemical resistance or else temperature resistance (Paramelt, 2011).

Heat sealing ability can also be provided by coating packaging materials with heat sealable polymers like polyethylene (PE). The corresponding process is called extrusion coating (see Figure 2.8). Plastic granules are converted to the molten state under heat and pressure in the extruder. The resulting molten polymer is extruded through a slit die as a thin web which is rapidly cooled down by a chill roll (Brown, 1992; Iggesund Paperboard, 2010; Qenos Pty Ltd, 2015).

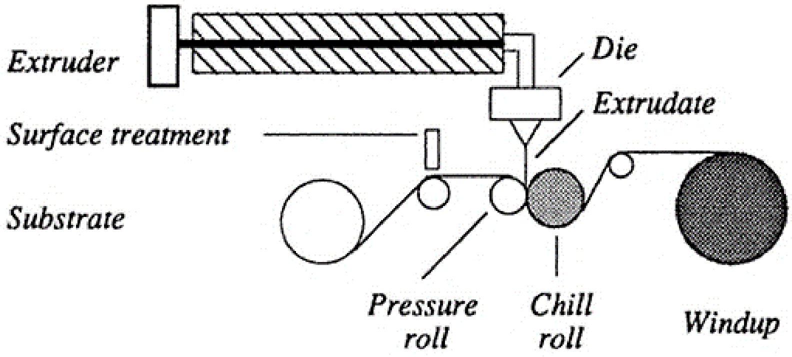


Figure 2.8 Extrusion coating of substrates with thermoplastics (Brown, 1992).

2.2.4 **Adhesives**

An adhesive can be defined as “a substance capable of holding at least two surfaces together in a strong and permanent manner” (Ebnesajjad & Landrock, 2014). In the case of laminated flexible packaging, three different types of adhesive systems can be used: solvent-less adhesives, water-based and solvent-based. The solvent-less technology is currently the dominant technology (Multifilm Packaging Corporation, n.d.; Hrinya, 2017; Henkel, 2018). It shows the advantage to cut production time (faster line speeds) and energy costs (no drying step). It also enables to reduce chemicals emissions (volatile organic compounds as regard to solvent-based adhesives) which leads to an improved operational safety and a lower environmental impact (Brown, 1992; Lingle, 2012; Blumsack & Caimmi, 2017; Henkel, 2018).

Solvent-less adhesives correspond to reactive chemical systems which are 100% solid and thus do not require any drying process. Two-component systems based on reactive polyurethane chemistry are by far the most important adhesives for food packaging (Dixon, 2011; Schumacher & Schindler, 2013; Hrinya, 2017). Two-component adhesives actually consist of a resin (pre-polymer resulting from the reaction of a di-isocyanate and polyols) and a hardener (diol or polyol) which are separated in two different containers and should be mixed in specific proportions to initiate cross-linking reactions (see Figure 2.9) (Henkel, 2015; Dixon, 2011). One component systems can also be used. In this respect, the adhesive isocyanate groups react with the moisture of the atmosphere and the moisture of the web (paper moisture for example) which leads to cross-linking reactions (Petrie, n.d.; Brown, 1992; Henkel, 2015).

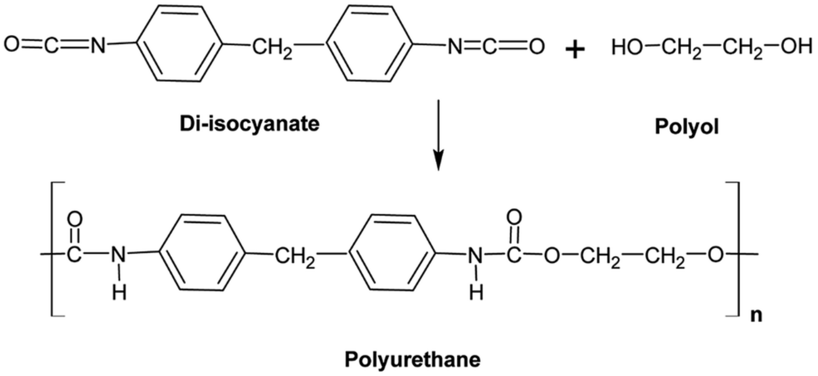


Figure 2.9 Reaction between an isocyanate and a hydroxyl group to form a urethane linkage - Case of a two-component polyurethane-based adhesive system (Akindoyo, et al., 2016).

Water-based or waterborne adhesives correspond to “adhesives which are made from materials that can be dispersed or dissolved only in water” (Ebnesajjad & Landrock, 2014). There are two general types of water-based adhesives:

- Solutions i.e. “stable dispersions of two or more immiscible liquids held in suspension by small percentage of substances called emulsifiers” (Ebnesajjad & Landrock, 2014);
- Latexes i.e. “stable dispersions of a polymeric material in an essentially aqueous medium” (Ebnesajjad & Landrock, 2014). Latex can be natural, synthetic or else artificial. Natural latex refers to the material primarily obtained from the rubber tree.

In the case of laminated flexible packaging materials, synthetic emulsion polymers are mainly used (Petrie, n.d.). Examples of polymers exclusively used for water-based adhesives include vinyl acetate, acrylic, polyurethane or else polyvinyl alcohol (Ebnesajjad & Landrock, 2014). Water-based adhesives are gaining importance in the flexible packaging industry because of improved toxicological safety (risk of primary aromatic amines migration into food in the case of not fully reacted solvent-less and solvent-based systems for instance), time saving (no food safety-related storage step between laminating and slitting, and between slitting and filling) and cost reduction (cheaper than solvent-based adhesives for example) (Schumacher & Schindler, 2013; Sergio, 2016; Shaw, 2017).

Solvent-based adhesives could be defined as adhesives which are made from materials that can be dispersed or dissolved only in organic solvents. The main polymers used for solvent-based adhesives include nitrocellulose, cellulose acetate butyrate, cyclized rubber, polyisobutylene and polyurethane (Ebnesajjad & Landrock, 2014).

Finally, Table 2.2 summarizes and compares the main characteristics of the solvent-less, water-based and solvent-based adhesives.

Table 2.2 Comparison of the different adhesives types for flexible packaging (adapted from Adhesives.org, n.d.; Leib & Jopko, 2008; Schumacher & Schindler, 2013; Ghosh, 2015; The Dow Chemical Company & ChemPoint, 2017).

Main characteristics	***Solvent-less***	***Water-based***	***Solvent-based***
Use of existing laminator	No	Yes	Yes
Relative machine cost	Low	Medium-High	Medium-High
Tension control	Superior	Average-Good	Average-Good
Typical line speed (m/s)	3.5-7.5	2.5-6	1.5-5
Drying	No	Yes	Yes
Adhesive solid content (%)	100	40-50	30-60
Adhesive layer thickness (µm)	1-2	> 3	3-5
Elasticity of the adhesive layer	Low	Medium	High
Adhesive waste	Low	Medium-High	High
Factory footprint	Low-Medium	Medium-High	Medium-High

2.2.5 **Primers**

Primers have been defined as “chemicals which when applied as a discrete layer to the substrate surface provide a chemical affinity between the extruded web and the substrate and hence lead to better adhesion” (Qenos Pty Ltd, 2015). They can be solvent- or water-based; reactive or non-reactive systems. Primer selection is specific to a particular application and the final use of the laminated material. The most common priming systems include polyurethanes, polyethylene imine, polyethylene chloride and ethylene-acrylic acid copolymer and ionomer. Paper-based materials, aluminium foil and polyesters are part of the substrates which often require the use of primers (Qenos Pty Ltd, 2015).

2.2.6 Other miscellaneous coatings

Coatings or lacquers can generally be classified into two categories: decorative or protective. Protective functions may cover scuff/scratch resistance, corrosion resistance, improved barrier properties or else improved material release (easy to unwind from the reel). On the other hand, decorative functions deal with more “marketing aspects” such as gloss/matt effect, print protection and colour stability (Riley A., 2012; Mieth, Hoekstra, & Simoneau, 2016).

2.3 Paper-based laminates manufacturing processes

Lamination consists in bonding two or more independent webs using adhesives, pressure and/or temperature (Dixon, 2011; Riley A., 2012; Ebnesajjad & Landrock, 2014). Bonding requires cohesion between molecules of the same layer and adhesion between the different layers (Ghosh, 2015). Three different processes may be implemented to bond paper-based laminates layers: adhesive lamination, extrusion lamination and hot melt lamination (Dixon, 2011). Process selection often depends on packaging converters process familiarity, currently installed equipment and/or cost constraints ("Extrusion or adhesive lamination?", 2017).

2.3.1 Adhesive lamination

Three different kind of adhesive lamination processes can be distinguished: solvent-less lamination, dry bond lamination and wet bond lamination (Dixon, 2011). As previously said, the solvent-less technology appears as the dominant technology (Multifilm Packaging Corporation, n.d.; Hrinya, 2017; Henkel, 2018). Adhesive lamination process selection is generally based on the nature of substrates to be laminated as well as the adhesive type, its conversion method and/or its application method (Ashter, 2014).

Solvent-less lamination consists in bonding two webs by using adhesives which are cured i.e. their physical properties are modified by chemical reaction which may be condensation, polymerization, vulcanization or cross-linking. This is usually accomplished by the action of heat and catalyst, alone or in combination, with or without pressure (Ebnesajjad & Landrock, 2014). The adhesive is generally heated (decrease in its viscosity) to apply it on the first web which is nipped to the second one as described in Figure 2.10. The resulting two-ply material is then cooled down and wound up. Metering rolls help control the adhesive thickness prior to application: they namely enable to apply much thinner adhesive layers compared to solvent-based systems. No drying step is required.

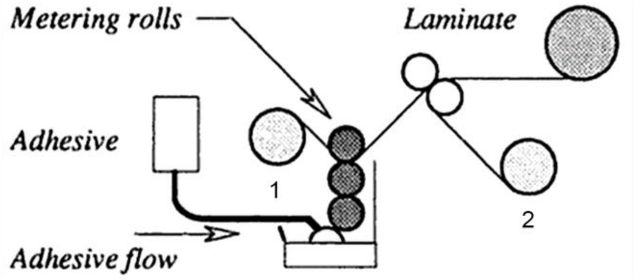


Figure 2.10 Solvent-less lamination (Brown, 1992).

Dry bond lamination consists in bonding two webs by using adhesives which are dry when passing the resulting two-ply material through a nip roller as shown in Figure 2.11. The cooling step leads to an increase in the adhesive viscosity which is responsible for bonding. Dry bond lamination is used when two non-porous materials need to be laminated. Water- or organic solvent-based adhesives can be applied. In the case of aluminium foil-based laminates, adhesives are usually applied to the aluminium foil. The solvent is then driven off in a drying oven. In order to ensure good adhesion, it is very important that the solvent completely evaporates before combining the two webs (Brown, 1992; Riley A., 2012).

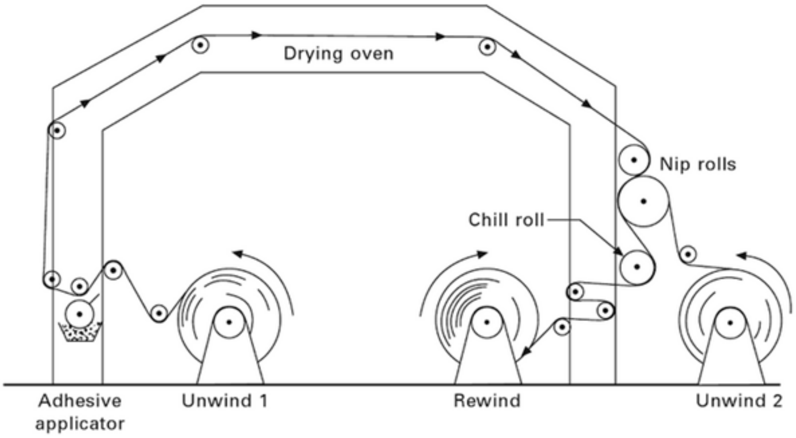


Figure 2.11 Dry bond lamination (Riley A. , 2012).

Wet bond lamination consists in bonding two webs by using adhesives which are still wet when passing the resulting two-ply material through a nip roller as described in Figure 2.12. In the case of paper-aluminium laminates, water- or solvent-based adhesives are applied to the aluminium foil: materials laminated to the aluminium foil need to be porous so that the liquid medium of the adhesive evaporates and the adhesive properly dries. A low paper moisture content is important to ensure good adhesion. High drying temperatures improve bonding and water resistance. However, care needs to be taken during drying in order not to over-dry paper and thus damage it (Brown, 1992; Riley A., 2012).

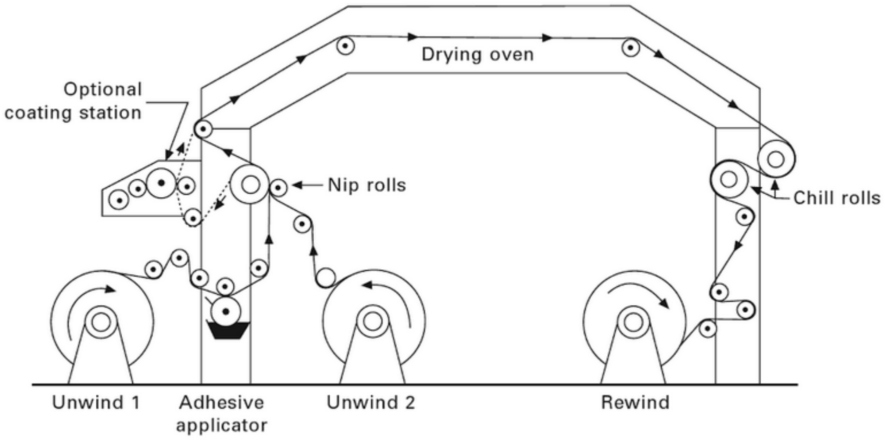


Figure 2.12 Wet bond lamination (Riley A. , 2012).

Table 2.3 summarizes and compares the main characteristics of the solvent-less, dry and wet bond processes.

Table 2.3 Comparison of the different adhesive lamination processes (adapted from Petrie, n.d.; Brown, 1992; Rolando, 2000).

Main characteristics	*Solvent-less bond*	*Dry bond*	*Wet bond*
Adhesive technology	Solvent-less	Water-based Solvent-based	Water-based Solvent-based
Adhesive application weight (g/m²)	1-3	2-3	2-8
Drying oven	No	Yes	Yes
Substrates particularity	Any combination of substrates	Any combination of substrates	Requires one porous substrate
Applications	Long runs	Short customised runs	Short customised runs

2.3.2 **Extrusion lamination**

Extrusion lamination consists in bonding two webs by extruding a thin layer of molten plastic, applying it to one web surface and combining the two webs as described in Figure 2.13. The resulting two-ply material passes through chilled nip rolls: it cools down the plastic layer which solidifies and thus performs as an adhesive. No drying step is required.

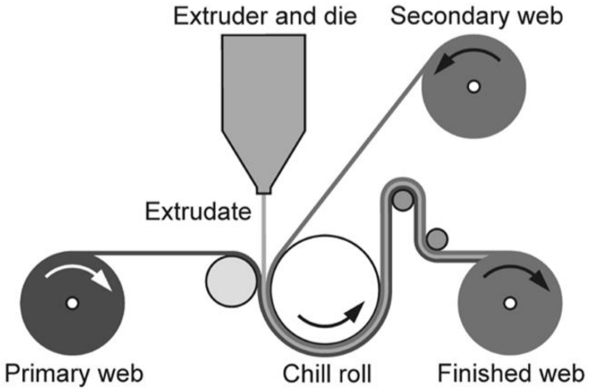


Figure 2.13 Extrusion lamination (Dixon, 2011).

2.3.3 Hot-melt lamination

Hot-melt lamination consists in bonding two webs by applying melted polymer resins, waxes or resin-wax combinations to one web surface and combining the two webs as shown in Figure 2.14. The resulting two-ply material passes through nip rolls before being cooled down. No drying step is required. Hot-melt lamination is quite similar to extrusion lamination.

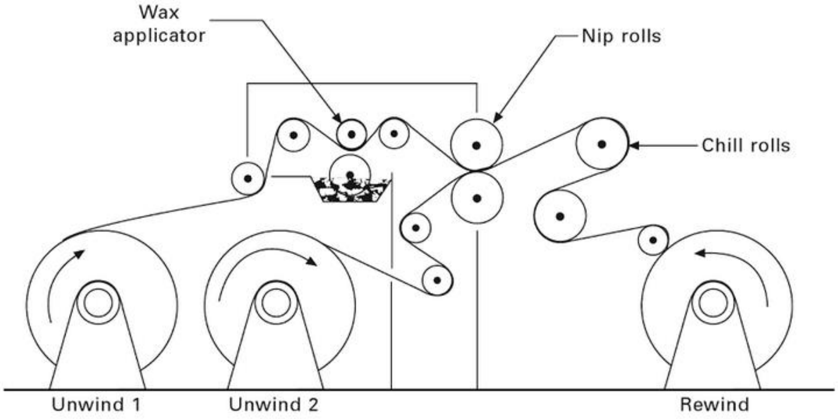
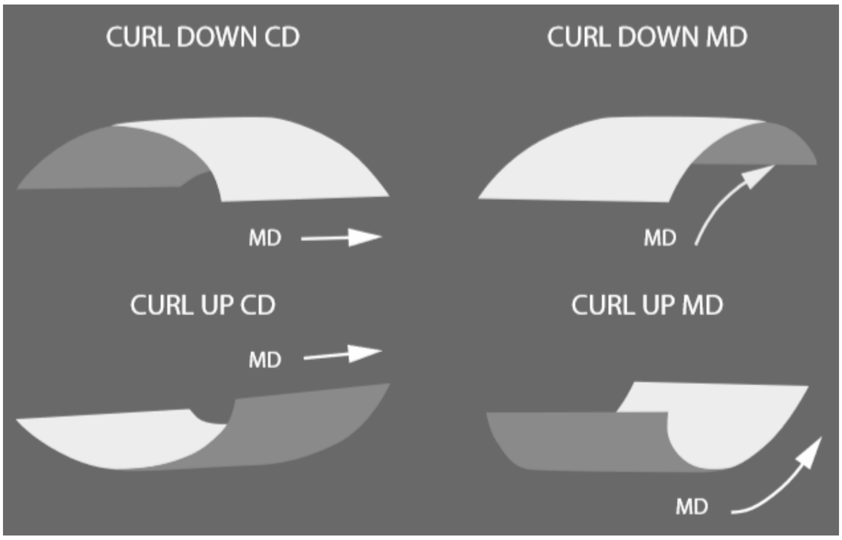


Figure 2.14 Wax bond lamination (Riley A. , 2012).

2.4 Paper-based laminates curl phenomenon

Curl is usually defined as a deviation from flatness or out-of-plane dimensional instability which may be observed under varying conditions of humidity and temperature (Carlsson, 1981; Lu & Carlsson, 2001; Uesaka, 2002; Iggesund Paperboard, 2010; Decker et al., 2010; Dignesh, Krishnan, Rajasekaran, & Kumar, 2013); “an undesirable condition caused by uneven rates of absorption or evaporation of moisture, uneven rates of contraction or expansion, or internal stresses in the material” (Catty Corporation, 2017). Curl measurement has three major components: its magnitude, the angle of the curl axis in relation to the MD and the side towards which the sheet curls (International Organization for Standardization (ISO), 2005). Different types of curl can be described as illustrated in Figure 2.15. The type of curl may vary according to the shape and the size of the material sample which tends to reach the most stable form by minimizing the total strain energy required for curl (Uesaka, 2002). Laminates made up of materials which show different physicochemical properties are particularly prone to curl, especially when the web structure is asymmetric (Iggesund Paperboard, 2010; Catty Corporation, 2017). Indeed, an individual lamina is restrained by the other laminae and is not free to expand which generates stresses (Free University of Brussels (ULB), n.d.). The difference in materials moisture-induced dimensional instability may namely cause deformation and thus dramatically affect laminates mechanical viability (TA Instruments, 2007; Decker et al., 2010). Under varying moisture conditions, paper often appears to be responsible for curl (Uesaka, 2002).



Note: Climate-related curl is said to be oriented in the CD (Iggesund Paperboard, 2010).

Figure 2.15 Different types of curl (Iggesund Paperboard, n.d.).

2.4.1 Framework of factors leading to the curl phenomenon

The main factors which may cause paper-based packaging materials and paper-based flexible laminates curl are summarized in Table 2.4.

Table 2.4 Framework of factors leading to paper-based laminates curl.

Levels	***Factors***	***References***
Filling process	RH of the atmosphere Temperature Packaging pattern shape and size Web tension	United States Patent No. U.S. 3,098,780, 1963 Heiss, 1980 Fellows & Axtell, 1993 Hertlein, 1998 Uesaka, 2002 Glatfelter, 2005 Husson, 2007 Iggesund Paperboard, 2010 Dignesh, Krishnan, Rajasekaran, & Kumar, 2013 Walker, 2017 Nautiyal, 2016
Warehousing	RH of the atmosphere Temperature Duration	United States Patent No. U.S. 3,098,780, 1963 Heiss, 1980 Fellows & Axtell, 1993 Hertlein, 1998 Uesaka, 2002 Glatfelter, 2005 Husson, 2007 Iggesund Paperboard, 2010 Dignesh, Krishnan, Rajasekaran, & Kumar, 2013 Nautiyal, 2016
Roll	Width Internal diameter External diameter Weight Protection	Nentwig, 2006
Laminate	Structure/construction Laminated materials (nature, moisture expansion coefficients, thermal expansion coefficients) Adhesives/binders Coatings	Free University of Brussels (ULB), n.d. TA Instruments, 2007 Decker et al., 2010 Iggesund Paperboard, 2010 Dixon, 2011

Table 2.4 Framework of factors leading to paper-based laminates curl (continued).

Levels	*Factors*	*References*
Converting processes	RH of the atmosphere Temperature Lamination process Drying (time, temperature) Individual web tension	Brown, 1992 Glatfelter, 2005 Iggesund Paperboard, 2010 Dignesh, Krishnan, Rajasekaran, & Kumar, 2013 Smith, 2015 Hrinya, 2017
Paper	Paper nature Fibres (type, purity, quantity) Fillers (type and quantity) Adhesives/binders (type and quantity) Manufacturing treatment (degree of fibre treatment) Moisture content Past moisture history	United States Patent No. U.S. 3,098,780, 1963 Glatfelter, 2005 Iggesund Paperboard, 2010 Dixon, 2011 Dignesh, Krishnan, Rajasekaran, & Kumar, 2013
Cellulose	Fibre morphology (length, diameter, curliness) Fibre orientation Single fibre sorption Inter-fibre contacts Microfibril angle	Glatfelter, 2005 Iggesund Paperboard, 2010 Lindner, 2018

Considering the given numerous factors and the fact that this study aims at describing and assessing the effect of climate conditions on two paper-based flexible laminates, RH, past moisture history and temperature will therefore mainly be discussed. These three factors what is more appeared as the best-documented ones.

2.4.2 Relative humidity

During winter or in the case of cold weather, paper RH may be higher than the environmental RH leading paper to release moisture and shrink. In the case of paper-aluminium laminates, the packaging material is expected to curl towards the paper side (United States Patent No. U.S. 3,098,780, 1963; Glatfelter, 2005). An environmental RH lower than 35% (Glatfelter, 2005) or even 45% (Iggesund Paperboard, n.d.) may be responsible for paper-based packaging materials curl.

In the case of rainy weather or humid environments, paper RH may be lower than the environmental RH leading paper to absorb moisture and swell. In the case of paper-aluminium laminates, the packaging material is expected to curl towards the aluminium foil side (United States Patent No. U.S. 3,098,780, 1963; Glatfelter, 2005). An environmental RH higher than 60% (Glatfelter, 2005; Iggesund Paperboard, n.d.) may be responsible for paper-based packaging materials curl.

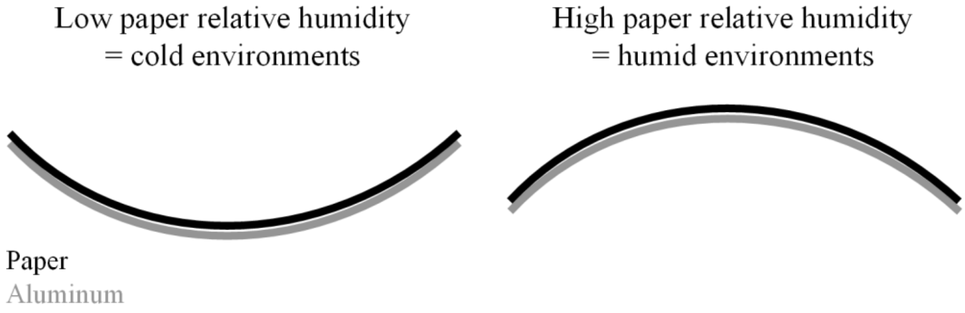


Figure 2.16 Effect of RH on paper-aluminium laminate curl.

More generally, a difference in the environmental and paper RH equal to or higher than 10% may be responsible for paper-based packaging materials curl (Heiss, 1980). Indeed, paper moisture expansion coefficient ranges from 0.020 to 0.060%/% in the MD and from 0.030 to 0.180%/% in the CD: a 10%-change in moisture content on the whole induces 0.2 to 1.8% hygroexpansion (Uesaka, 2002).

2.4.3 **Past moisture history**

The moisture content of any hygroscopic packaging material (namely paper) is impacted by its past moisture history (Uesaka, 2002; Iggesund Paperboard, 2010). Figure 2.17 shows an example of pulp moisture sorption isotherms where A is a desorption isotherm and B is an adsorption isotherm. The moisture content is different at any given RH depending on whether the material was brought into equilibrium from a higher RH or brought into equilibrium from a lower RH. (Dwan, 1987; Parker, Bronlund & Mawson, 2006). This is known as the hysteresis effect i.e. “a lagging effect in which a memory of the previous state is retained” (Iggesund Paperboard, 2010). Consequently, the moisture content of any hygroscopic packaging material varies depending on earlier climatic conditions and can hardly be estimated.

As many paper properties display complex hysteresis phenomena when plotted against RH, it is preferable to monitor paper properties as a function of moisture content rather than RH (Uesaka, 2002).

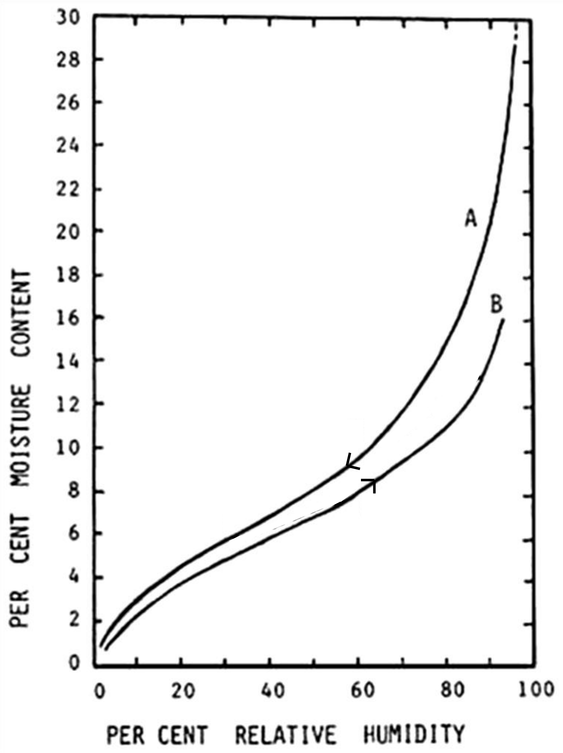


Figure 2.17 The hysteresis effect – Example of pulp moisture sorption isotherms (fixed temperature) (adapted from Dwan, 1987).

2.4.4 **Temperature**

During winter or in the case of cold weather, paper temperature may be lower than the environmental temperature leading paper to cool locally down the air and increase the environmental RH. Therefore, cold paper is expected to absorb moisture (Glatfelter, 2005). In the case of paper-aluminium laminates, the packaging material is expected to curl towards the aluminium foil side (United States Patent No. U.S. 3,098,780, 1963). A temperature lower than 10-15°C is said to be "cold" (Glatfelter, 2005).

During summer or in the case of warm weather, paper temperature might be higher than the environmental temperature leading paper to warm locally up the air and decrease the environmental RH. However, paper generally reaches the temperature equilibrium before curl occurs. Therefore, warm environments would not significantly affect paper-based packaging materials. A temperature higher than 25-30°C is said to be "warm" (Glatfelter, 2005).

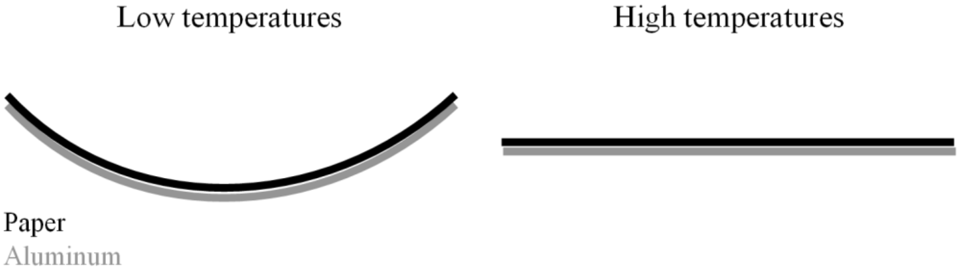


Figure 2.18 Effect of temperature on paper-aluminium laminate curl.

Paper thermal coefficients range from 2.0 to 7.5*10^{-6} K^{-1} in the MD and from 7.9 to 16.2*10^{-6} K^{-1}in the CD: a 100°C-change in temperature causes 0.02 to 0.16% thermal expansion. These figures namely demonstrate that paper thermal expansion is less significant than its hygroexpansion (Uesaka, 2002).

2.4.5 Practices to avoid paper curl

Table 2.5 summarizes all the practices that have been suggested in the literature in order to avoid paper-based packaging material curl (Fellows & Axtell, 1993; Glatfelter, 2005; Iggesund Paperboard, 2010).

In order to prevent or limit curl, the paper original moisture content should remain constant across the different processes steps (Glatfelter, 2005; Iggesund Paperboard, 2010; Riley A., 2012). Paper-based packaging materials are usually packed into water-proof wrapping which should be removed as late as possible. The reason is around 65% of the total moisture which will be absorbed in one hour is actually gained within the first 30 seconds of exposure to the new environment. When properly packed and stored, paper may retain a stable moisture content for more than two years (Glatfelter, 2005).

Packaging materials should be acclimatized to the manufacturing environment before being processed. The suggested warming-up periods depend on the pallet or roll weight and the difference in temperature between the packaging materials and the environment as described in Table 2.6.

Low RH environments (RH below 35%) can be corrected quite simply by using inexpensive humidification systems to spray water and/or steam into the air (Glatfelter, 2005; Husson, 2007). High RH environments however require substantial investments to decrease RH and reach values lower than 55-60% (Glatfelter, 2005).

According to the literature, paper-related curl problems are very rarely encountered when taking the previous precautions (Glatfelter, 2005).

Table 2.5 Framework of practices to prevent paper-based packaging materials curl.

Process steps	*Practices*	*References*
Filling process	Do not process packaging materials just after delivery At constant temperature: 20-25°C At constant RH: 45-60% (automatic online monitoring)	Fellows & Axtell, 1993 Glatfelter, 2005 Decker et al., 2010 Iggesund Paperboard, 2010
Unwrapping	Acclimatization to temperature conditions in the production area (see Table 2.6) Unwrap packaging materials just before processing them (“last-minute”)	Fellows & Axtell, 1993 Glatfelter, 2005 Iggesund Paperboard, 2010
Warehousing	On shelves or pallets (not on the floor) Neither outside nor under a roof Far from any heating/cooling or humidifying system Within intact moisture-proof wrapping Uptight rolls At constant temperature: 20-25°C At constant RH: 45-60% (automatic online monitoring)	Fellows & Axtell, 1993 Glatfelter, 2005 Decker et al., 2010 Iggesund Paperboard, 2010
Laminate converting	Material re-moistening	Uesaka, 2002 Decker et al., 2010
Paper converting	Reduce paper ability to take up moisture by using stabilizers, sizers or humectants	United States Patent No. U.S. 3,098,780, 1963 Dignesh, Krishnan, Rajasekaran, & Kumar, 2013

Table 2.6 Recommended warming-up times before removing wrapping (Iggesund Paperboard, n.d.).

Pallet or roll weight	*Initial temperature difference between board and room (°C)*		
(kg)	*10*	*20*	*30*
400	2 days	2 days	3 days
800	2 days	3 days	4 days
1,200	2 days	4 days	5 days

3 Materials and method

The aim of the study is to describe and assess the effect of climate conditions on two different paper-based flexible laminates tendency to curl so as to understand to what extent environmental conditions can affect their processability. This third chapter introduces the materials to be studied, the suggested test method to assess their tendency to curl, the selected environmental conditions (RH and temperature) and the designed experimental plan. It also justifies the experimental design which happened to mainly be constrained by the use of a climate chamber (samples dimensions and number for example) and the characteristics of the packaging materials which were made available (quantity of rolls and required exposure duration for instance).

3.1 Packaging materials

The first material is a Pap/mPET/HSL laminate which is used as heat-sealable yogurt lid. No curl-related problem has been encountered yet. The idea is to document the tendency to curl for a widely processed laminate. Paper and metallized polyethylene terephthalate are bounded through dry lamination. Further details are given in Table 3.1.

Table 3.1 Description of the Pap/mPET/HSL laminate.

Layers	*Thickness (μm)*	*Grammage (g/m²)*
Heat-sealable lacquer (polyolefin-, acrylate-, styrene-based)	-	2.9
Metalized polyethylene terephthalate	12	-
Polyurethane-based adhesive	-	2.5
Sulphate-based and single-side coated paper	-	43.0
Nitrocellulose-based inks	-	-
Nitrocellulose-based varnish	-	0.8
Laminate	60	69.0

The second material is an Alu/Pap/PE laminate. The tendency to curl happens to be critical for its processability. Paper and aluminium are bounded through dry bond lamination; paper and PE are bounded through solvent-less lamination. Further details are given in Table 3.2.

Table 3.2 Description of the Alu/Pap/PE laminate.

Layers	*Thickness (µm)*	*Grammage (g/m²)*
Polyethylene	30	27.7
Polyurethane-based adhesive lacquer	-	2.5
Uncoated smooth paper	30	30
Acrylate-based adhesive	-	2.5
Aluminium foil	7	18.9
Polyurethane-based primer	-	1
Nitrocellulose-based inks	-	-
Nitrocellulose -based coating	-	0.75
Laminate	67	83.4

Besides, packaging material rolls are often delivered wrapped in transparent low-density polyethylene (LDPE) bags. Considering the recommendations made to avoid paper curl (see Table 2.5 page 37), it seemed relevant to assess the "protective" effect of this low-density polyethylene wrapping (PE wrapping) then. The LDPE film used to wrap packaging materials samples was cut from 0.050 mm-thick LDPE side gusseted bags which are described in Figure 3.1 (Polyden Folienfabrik GmbH, article code: 55963).

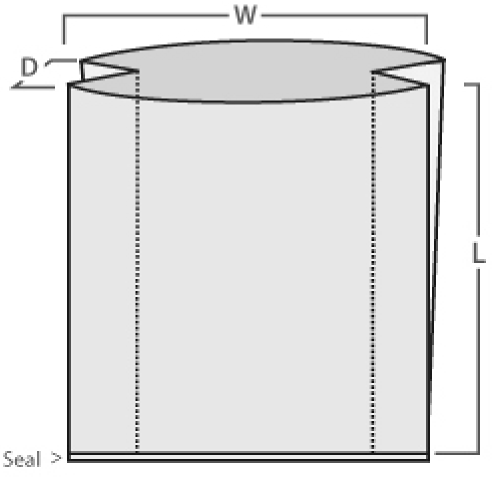


Figure 3.1 Description of the provided LDPE side gusseted bags (International Plastics, n.d. - L = 2000 mm, W = 1250 mm, D = 850 mm).

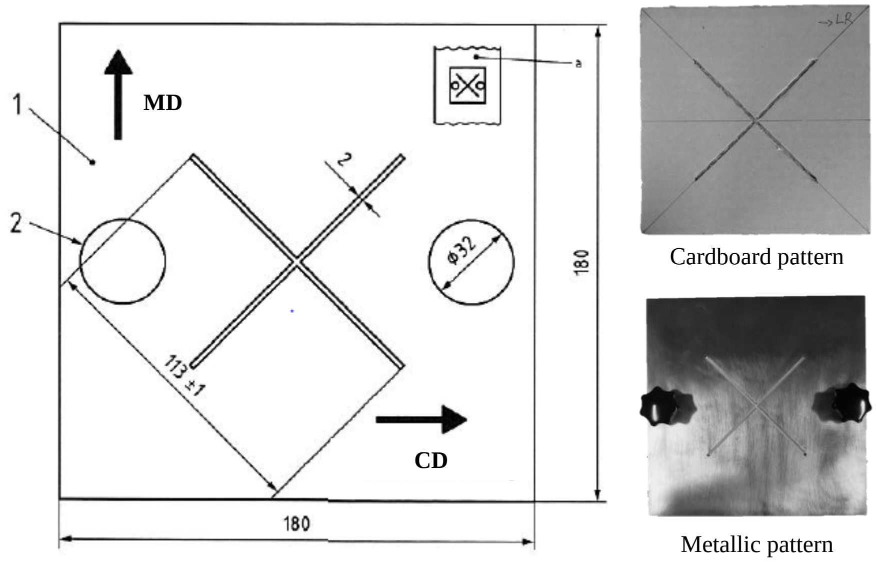
3.2 Test method to assess the tendency to curl

The German standard DIN 55403 (Deutsches Institut für Normung (DIN), 2014) describes three different methods to assess packaging films tendency to curl:

- The round sample method (“Rundprobenverfahren” in German);
- The determination of the flatness temperature (“Planlagetemperature”);
- The cross-cut method (“Kreuzschnittverfahren”).

Pre-tests were conducted to compare these three methods and decide on the one to be implemented (see A.1 Test method selection page 95). The cross-cut method was finally selected. It shows the advantage to be quite simple, quick and affordable to carry out meaning that other stakeholders would actually be able to implement it (quality control operators and packaging suppliers for instance). Required materials namely include a weather station, a cutting mat, a specific cutting pattern, a cutter, a ruler and tape. Both low and high tendency to curl can moreover be characterized.

It on the whole consists in cutting crosses in the packaging material web using a pattern (see Figure 3.2 and Figure 3.3) and measuring the distance between the edges, in the MD (a parameter in millimetres) and the CD (b parameter in millimetres) (see Figure 3.4). By assessing the tendency to curl in the MD and the CD, this method also appears to be very informative. As previously explained (see Figure 2.7 page 23), climate-related paper deformation mainly occurs in the CD meaning that the b parameter may particularly be relevant to study the effect of moisture and temperature on packaging materials behaviour.



Cardboard pattern

Metallic pattern

Note: Distances are expressed in millimetres (mm).

Figure 3.2 The cutting pattern (Deutsches Institut für Normung (DIN), 2014).

Figure 3.3 Cutting crosses using the cutting pattern.

The standard distinguishes two different ways of measuring the tendency to curl, regardless of the direction (see Figure 3.4 to Figure 3.7). When the distance or gap between the material edges is shorter than 5 millimetres, the tendency to curl should be measured as the shortest height between the table and the material edges (a* parameter). On the other hand, when the distance between the material edges is longer than 5 millimetres, the tendency to curl should be measured as the shortest distance between the material edges (a parameter).

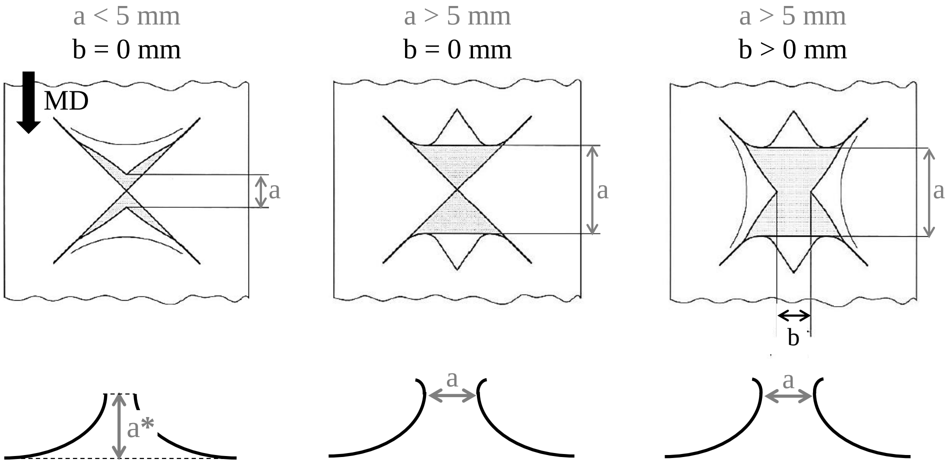


Figure 3.4 The cross-cut method (adapted from Deutsches Institut für Normung (DIN), 2014).

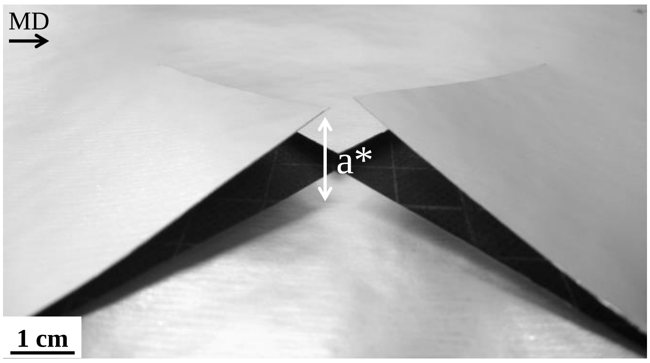


Figure 3.5 Illustration of the case "a < 5 mm".

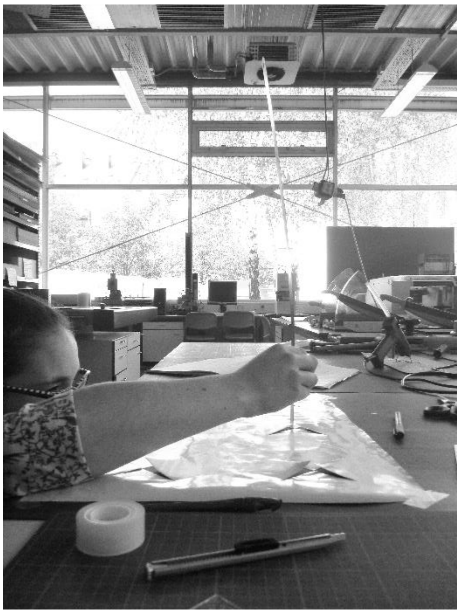


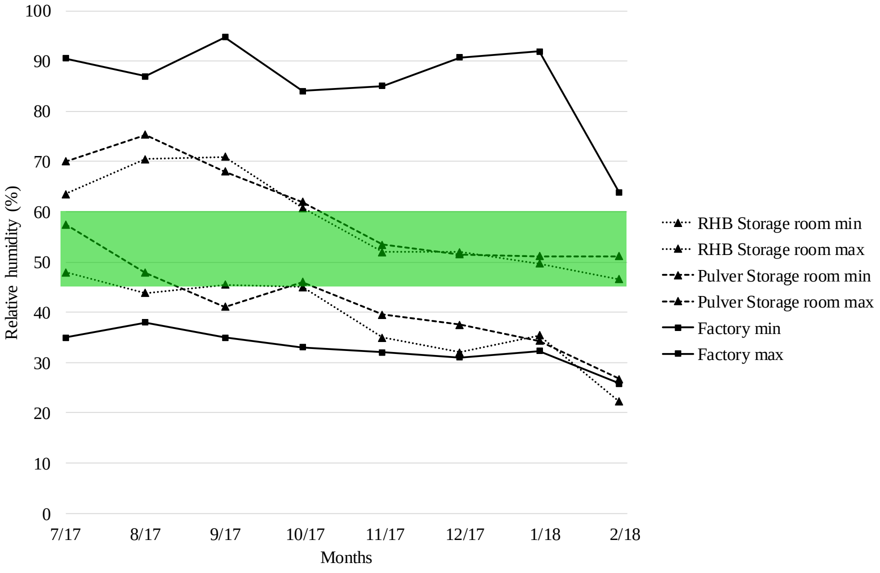
Figure 3.6 Measuring the a* parameter - Case of "low" tendency to curl.

Figure 3.7 Measuring the a parameter - Case of "high" tendency to curl.

3.3 Environmental conditions to be studied

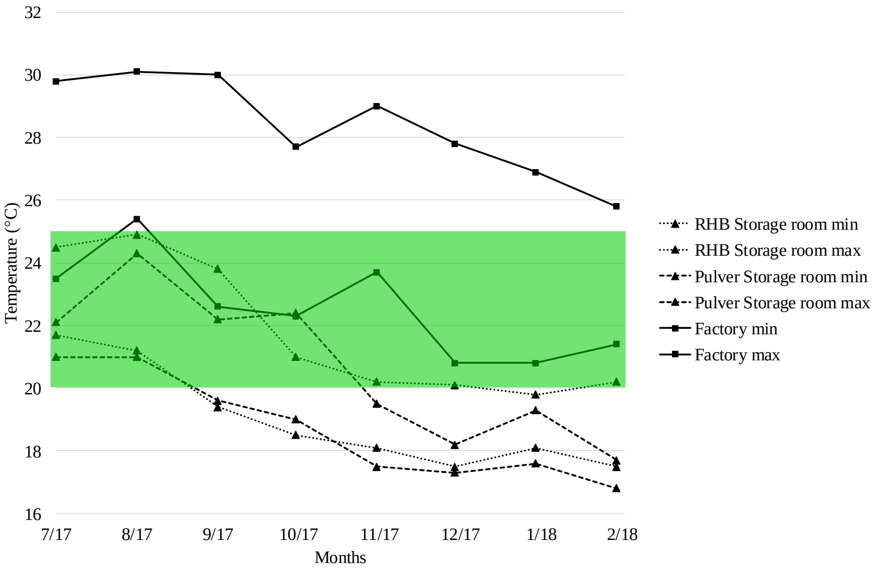
As previously highlighted (see Paper-based laminates curl phenomenon), RH and temperature may cause paper-based packaging materials curl. Therefore, environmental conditions within non-climatized storing and manufacturing areas have been analysed. In relation to materials processability, it was decided to mainly focus on the factory climate.

RH and temperature have been documented by the quality team since July 2017. However, all the data recorded before January 2018 has been saved in the form of graphs which do not enable any detailed analysis (different scales for various time ranges). Consequently, graphs representing the minimum and maximum range of RH and temperature were created so as to perform a first assessment (see Figure 3.8 and Figure 3.9). It appears that the range of RH is much larger within the factory, from 30 to 90%. RH values are furthermore far from the recommendations made for paper packaging materials (i.e. from 45 to 60%). The same analysis can be made for temperature. Temperature range happens to be much larger within the factory, from 18 to 30°C, and far from the recommendations for paper packaging materials (i.e. from 20 to 25°C). As a result, curl problems may be encountered when processing paper-based laminates in such environment.



Note: The green range shows the recommendations made by paper suppliers (Glatfelter, 2005; Iggesund Paperboard, 2010). The fact that RH was measured every 30 minutes explains the lower maximum value for February 2018: RH within the factory probably reached 90% but was not recorded (discontinuous measurement).

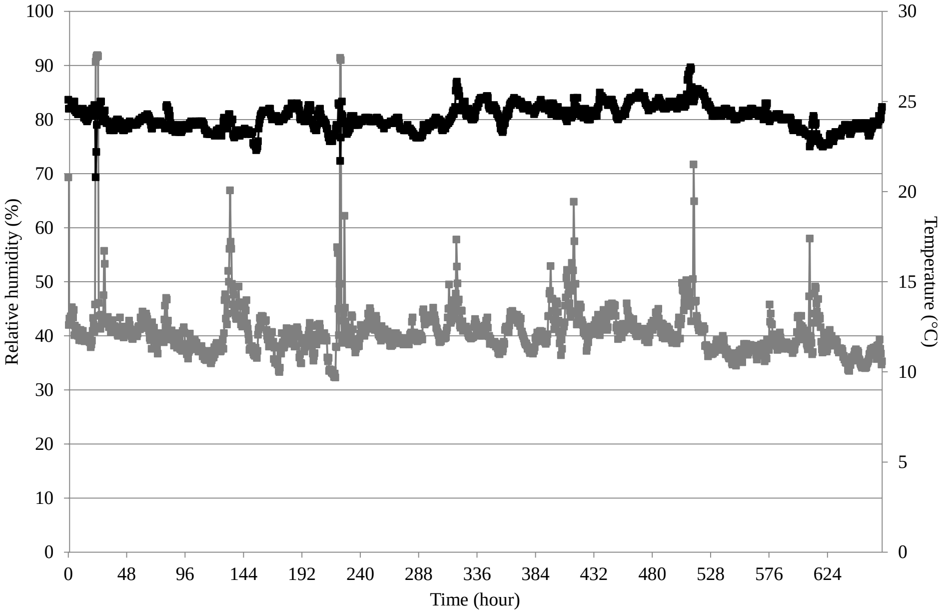
Figure 3.8 RH variation in the storage rooms and the factory from July 2017 to February 2018 (minimum and maximum values).



Note: The green range shows the recommendations made by paper suppliers (Glatfelter, 2005; Iggesund Paperboard, 2010). The fact that temperature was measured every 30 minutes might affect the results (discontinuous measurement).

Figure 3.9 Temperature variation in the storage rooms and the factory from July 2017 to February 2018 (minimum and maximum values).

Numerical data was made available for January 2018 and analysed so as to refine this first assessment. A graph plotting RH and temperature within the factory as a function of time was drawn (see Figure 3.10). According to the quality team, 12-hour cleaning processes are conducted every 90 hours on the different production lines. Indeed, cleaning processes can be identified through RH peaks. This observation demonstrates that the first representation using minimum and maximum curves does not really represent reality. Outside cleaning phases, RH seems to be lower than 50% (at least for January 2018). Thus, curl risk can be assessed as low if packaging materials are removed from the production area during cleaning; high if they are stored within the factory while cleaning the equipment. Even if not enough data could be collected so as to describe a seasonal effect, it seems that cleaning processes can cause much “harmful” variations in RH than seasonality.

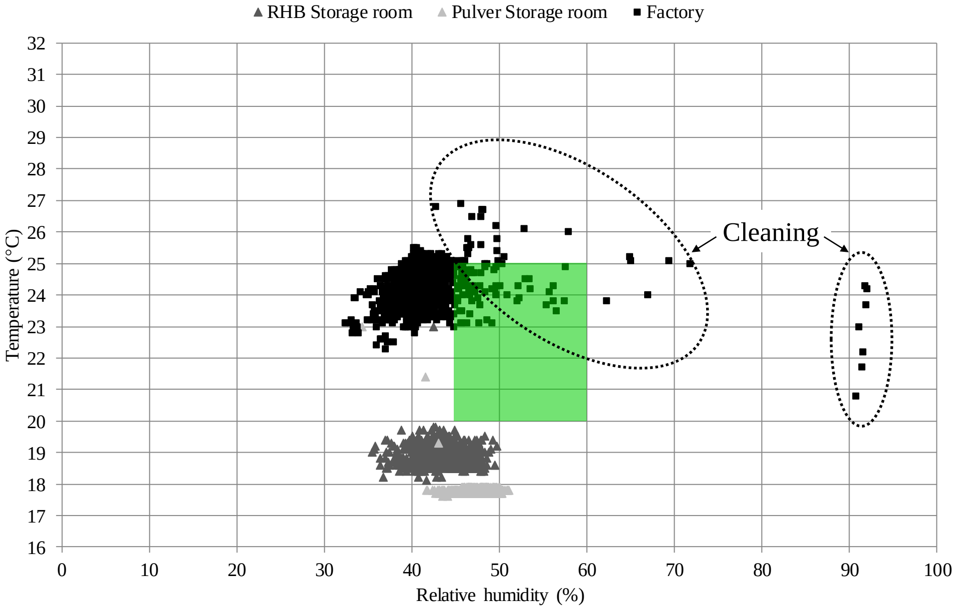


Note: Black dots represent temperature measurements; grey dots represent RH measurements. Data was recorded every 30 minutes.

Figure 3.10 RH and temperature variation within the factory in January 2018.

Another graph plotting RH as a function of temperature was drawn (see Figure 3.11). It namely shows the advantage to delimit specific areas and isolate “outsiders”. This representation leads to the same conclusion as previously: environmental conditions both within storage rooms and factory do not fit the guidelines for paper packaging materials.

As data for January may not be sufficient to describe annual changes in climate conditions, data from Figure 3.8, Figure 3.9 and Figure 3.11 all needs to be combined to define the experimental plan. For instance, temperature is said to easily reach 30°C in summer which is shown in Figure 3.9 but cannot be observed in Figure 3.11. The environmental conditions to be studied are presented in Figure 3.12. Considering the greater impact of RH on packaging materials tendency to curl and the large RH range that may be reached within the factory, five RH values were chosen: 30%, 40%, 50%, 60% and 70%. Extremely high RH values were not considered since they should not be reached if good manufacturing practices were respected. In order to describe the effect of temperature while limiting the number of experiments to be implemented, three temperatures were selected: 20°C, 25°C and 30°C. Finally, packaging materials will be studied under fifteen different climates using a climate testing chamber (Weiss-Voetsch Environmental Testing Instruments (Taicang) CO. Ltd., model: C 340, -40).



Note: The green area shows the recommendations made by paper suppliers (Glatfelter, 2005; Iggesund Paperboard, 2010). The dotted ellipses isolate measurements corresponding to cleaning phases.

Figure 3.11 Environmental conditions within the storage rooms and the factory in January 2018.

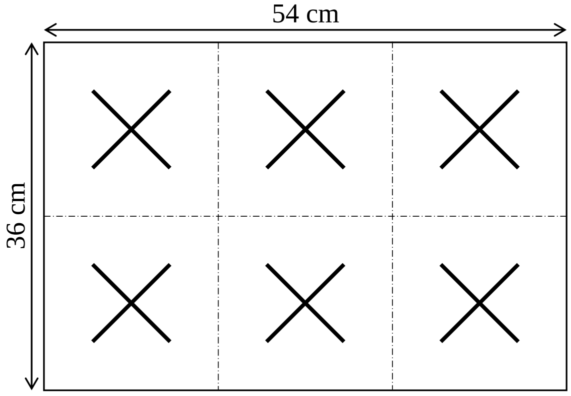
Note: The 15 dots correspond to the 15 climates to be studied.

Figure 3.12 Environmental conditions to be studied (experimental plan).

3.4 Experimental design

3.4.1 **Sampling technique**

Considering the very few packaging materials rolls which were made available as well as their dimensions (a single 106-mm diameter test roll for the Alu/Pap/PE prototype laminate), it was decided to study packaging materials sheets. Samples dimensions were defined by taken both the climate testing chamber and the cutting pattern dimensions into consideration. According to the German standard DIN EN 20 187:1993, materials should be arranged so that the air flow can “freely access” their surfaces i.e. they cannot be stacked on the top of each other. The climate testing chamber was namely equipped with two grills (70*53 cm each). Besides, pre-tests showed that a bare minimum of six repetitions should be implemented in order to collect “relevant” data. As a result, four identical samples enabling six measurements each could be fitted into the climate testing chamber (see Figure 3.13).



Note: A sample corresponds to six cutting patterns (see Figure 3.2).

Figure 3.13 A sample.

The sampling method is explained in Figure 3.14. In the case of the Pap/mPET/HSL laminate, the provided material roll width was 45 cm which highly constrained the samples location on the web (a single option). In the case of the Alu/Pap/PE laminate, the provided roll width was 106 cm. As regard to both roll width and weight, it was necessary to unwind this material using an unwinder system. By comparison with the first laminate, the idea was to collect materials from the middle part of the web and to position the sample pattern (54*36 cm) in the MD. However, because of time constraints, the sample pattern was oriented in the CD (less packaging to be unwound).

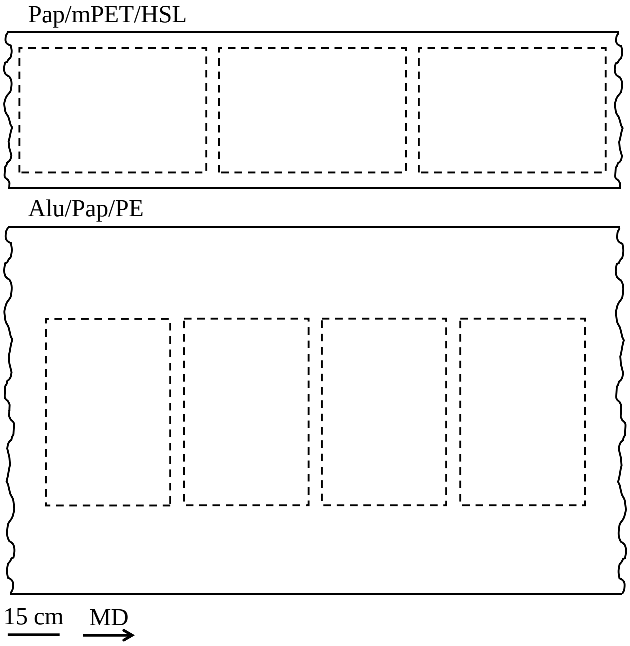


Figure 3.14 Sampling technique from rolls.

3.4.2 **Samples pre-conditioning**

As previously highlighted (see 2.4 Paper-based laminates curl phenomenon), paper-based materials past moisture history may affect their tendency to curl. Consequently, samples of the same material were all prepared on the same day and stored together in a single sleeve till carrying out the experiments. Since all the experiments could not take place on the same day, it also appeared significant to establish a “common starting point” (calibration). It was suggested to pre-condition all the samples under a standard climate i.e. 23°C and 50% (Deutsches Institut für Normung (DIN), 1993; Iggesund Paperboard, 2010) before setting the RH and temperature values of interest.

3.4.3 **Samples exposure duration**

Packaging materials exposure duration corresponds to the time it takes them to reach the equilibrium moisture content under given climate conditions. At the equilibrium, packaging materials neither absorb nor release moisture i.e. their mass is constant. As the time required to reach the equilibrium moisture content may vary for different climate conditions, packaging materials samples mass should always be monitored using an analytical lab scale (Sartorius, model: BP221S).

The methodology is described in the German standard DIN EN 20 187 (Deutsches Institut für Normung (DIN), 1993): 100 cm²-samples are cut from the material of interest using a circular sample cutter (Karl Schröder KG, model: Probenschneider PS 100), stored in the climate chamber with the four sheets to be studied, and weighted at different time intervals. The assumption is that 100-cm² samples behave as the sheets do i.e. small and big samples reach the moisture content equilibrium at the same time. The equilibrium is said to be reached when the variation in samples weight has been lower than 0.25% of their total mass for a one-hour interval (at least). Pre-tests showed that packaging materials exposure duration may approximately last 1h30 for the Pap/mPET/HSL laminate (see Table 3.3) and 1h for the Alu/Pap/PE laminate (see Table 3.4). If the equilibrium is not reached after the pre-defined time interval, samples are simply stored for a longer period of time in the climate chamber.

Table 3.3 Monitoring the 100-cm² samples mass - Case of the Pap/mPET/HSL laminate, at 23°C and 50% RH.

Sample	$m_{t(0h)}$ (g)	$m_{t(0,5h)}$ (g)	$m_{t(1,5h)}$ (g)	$\Delta m_{t(0-0,5h)}$ (%)	$\Delta m_{t(0,5-1,5)}$ (%)
1	0,6695	0,6730	0,6738	0,52	0,12
2	0,6666	0,6702	0,6708	0,54	0,09
3	0,6745	0,6778	0,6782	0,49	0,06

Note: $m_{t(0,5h)}$ corresponds to the 100-cm² sample mass in grams at 0,5 hours.

$\Delta m_{t(0-0,5h)}$ corresponds to the 100-cm² sample mass variation in percent within the first 0,5 hours.

Table 3.4 Monitoring the 100-cm² samples mass - Case of the Alu/Pap/PE laminate, at 23°C and 50% RH.

Sample	$m_{t(0h)}$ (g)	$m_{t(0,5h)}$ (g)	$m_{t(1,0h)}$ (g)	$\Delta m_{t(0-0,5h)}$ (%)	$\Delta m_{t(0-1,0)}$ (%)
1	0,8774	0,8771	0,8777	0,03	0,07
2	0,8811	0,8804	0,8809	0,08	0,06
3	0,8757	0,8753	0,8759	0,05	0,07

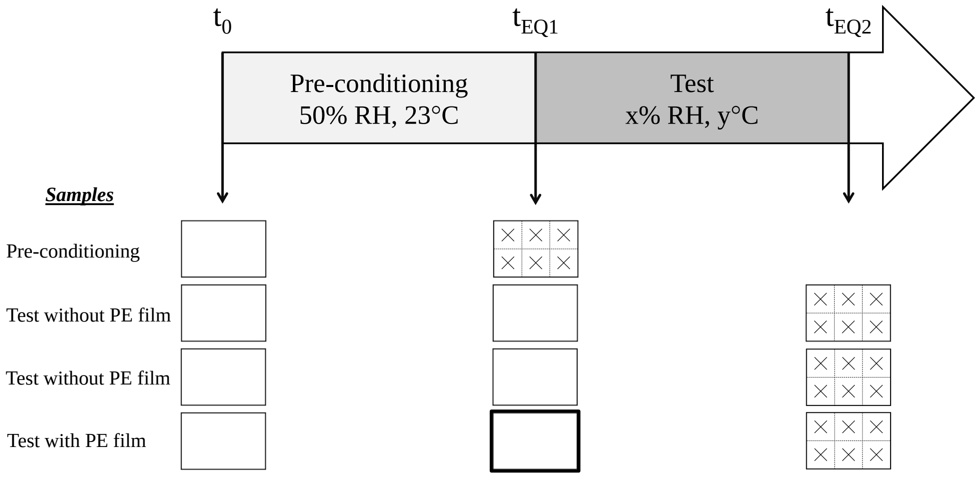
3.4.4 **Experimental procedure**

The overall procedure is illustrated in Figure 3.15.

At the beginning, 100-cm² circular samples (at least 2) are cut and weighted. The four packaging material sheets of a single material are fixed to the climate chamber grills using tape and put in the equipment at the same time as the circular samples (see Figure 3.16). RH and temperature are respectively set at 50% and 23°C for the pre-conditioning step. After having left the materials for 30 minutes inside the climate chamber under the specified conditions, circular samples are taken out of the equipment, weighted and put back in the equipment. If the samples weight has not significantly changed within the first 30 minutes (case of the Alu/Pap/PE laminate), a second measurement should be done 30 minutes later i.e. 1h after the experiment began. At that time, the RH equilibrium is reached. On the contrary, if the samples weight has significantly changed within the first 30 minutes (case of the Pap/mPET/HSL laminate), a second measurement should be done 1h later i.e. 1h30 after the experiment began. At that time, the RH equilibrium is reached. Afterwards, one sheet is taken out of the climate chamber, wrapped in PE film (see Figure 3.17 and Figure 3.18) and put back in the equipment; another sheet is taken out so as to assess the tendency to curl at the end of pre-conditioning phase. The climate chamber RH and temperature should be set to the values of interest (x% and y°C) before doing the measurements in order to save time. The sheet is fixed to the table using tape and six crosses are cut using the cutting pattern (see Figure 3.19). After having waited for two minutes, tendency to curl parameters are measured (Deutsches Institut für Normung (DIN), 1993). Room RH and temperature have to be recorded: a huge difference between the room and climate chamber conditions may namely affect the results.

The "test" phase procedure is similar to the pre-conditioning phase one. After having left the materials for 30 minutes inside the climate chamber under the specified conditions, circular samples are taken out of the equipment, weighted and put back in the equipment. This manipulation is repeated either 30 minutes or 1h later depending on the laminate. When the variation in circular samples weight has been lower than 0.25% of their total mass for a one-hour time interval, measurements are done one sheet at a time for the three remaining sheets. It is important to leave the samples which have not been assessed yet inside the climate chamber to prevent them from acclimatizing to the room climate conditions. Here again, room RH and temperature have to be recorded.

As explained above (see 3.4.1 Sampling technique), four samples enabling six repetitions each can be stored in the climate chamber. Besides, measurements have to be done at the end of the pre-conditioning step (calibration) and for the tests with and without PE wrapping i.e. three tests in total. The fourth sample was used to strengthen the results obtained for the test without PE wrapping considering that this experiment is the most meaningful one when assessing the climate impact on materials processability.



Note: An "empty" rectangle corresponds to a packaging material sheet.
Samples showing six crosses indicate that measurements are done at this/these step(s).

Figure 3.15 Overall procedure.

Figure 3.16 Picture of the 4 packaging material sheets and 100-cm² circular samples inside the climate chamber at t_0.

Figure 3.17 A sample wrapped in PE film at t_{EQ1} (front view).

Figure 3.18 A sample wrapped in PE film at t_{EQ1} (rear face).

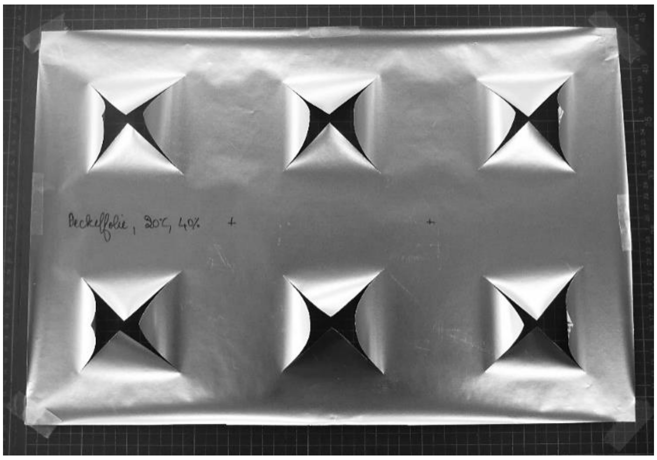


Figure 3.19 Example of a Pap/mPET/HSL laminate sheet after having cut six crosses at t_{EQ2}.

3.4.5 Experimental plan

The experimental plan is detailed in Table 3.5. Fifteen experiments were planned per laminate i.e. thirty experiments in total. In the case of the Pap/mPET/HSL laminate, 3h45 were necessary to carry out one experiment - sampling excluded, exposition and measurements included. All the data was collected within eight days (2 tests per day). In the case of the Alu/Pap/PE laminate, 2h45 were needed to conduct one experiment which would have enabled to get all the data within five days (3 tests per day). However, as the tendency to curl happened to be very low for this material i.e. lower or equal to 5 millimetres, it was not accurate to measure curl parameters as the shortest lengths between the material edges (see Figure 3.4 page 42). The shortest heights between the material edges and the table had to be considered instead. This "change in the test method" finally led to a seven-day experimental phase. Eventually, this study represented fifteen full days of lab work.

Table 3.5 Detailed experimental plan for one packaging material.

Experiment	*Temperature (°C)*	*RH (%)*	*PE wrapping*	*Number of measurements*
1	23	50	No	6
	20	30	No	12
	20	30	Yes	6
2	23	50	No	6
	25	30	No	12
	25	30	Yes	6
3	23	50	No	6
	30	30	No	12
	30	30	Yes	6
4	23	50	No	6
	20	40	No	12
	20	40	Yes	6
5	23	50	No	6
	25	40	No	12
	25	40	Yes	6
6	23	50	No	6
	30	40	No	12
	30	40	Yes	6
7	23	50	No	6
	20	50	No	12
	20	50	Yes	6

Table 3.5 Detailed experimental plan for one packaging material (continued).

Experiment	*Temperature (°C)*	*RH (%)*	*PE wrapping*	*Number of measurements*
8	23	50	No	6
	25	50	No	12
	25	50	Yes	6
9	23	50	No	6
	30	50	No	12
	30	50	Yes	6
10	23	50	No	6
	20	60	No	12
	20	60	Yes	6
11	23	50	No	6
	25	60	No	12
	25	60	Yes	6
12	23	50	No	6
	30	60	No	12
	30	60	Yes	6
13	23	50	No	6
	20	70	No	12
	20	70	Yes	6
14	23	50	No	6
	25	70	No	12
	25	70	Yes	6
15	23	50	No	6
	30	70	No	12
	30	70	Yes	6

4 Results and discussion

This fourth chapter aims at describing the influence of RH and temperature on the tendency to curl of the selected paper-based laminates, Pap/mPET/HSL and Alu/Pap/PE, as well as assessing the effect of PE wrapping as a potential solution to prevent or at least reduce the curl phenomenon. As the materials curl magnitude happened to be very different and required to measure curl parameters in two different ways as explained in the German standard DIN 55403 (Deutsches Institut für Normung (DIN), 2014), results are introduced and discussed one laminate at a time before comparing the two laminates. Most of the graphs were drawn using Minitab; statistical analysis was implemented using RStudio.

4.1 Case of the Pap/mPET/HSL laminate

The tendency to curl of this first paper-based laminate was generally much higher than 5 millimetres both in the MD and the CD. As a result, according to the DIN 55403 standard (2014) (see also Figure 3.4 page 42), all the measurements were done considering the shortest distance or gap between the material edges and this, even when the distance between the material edges was shorter than 5 millimetres. The reason is mixing lengths and heights may have been questionable from the data analysis perspective.

4.1.1 **Effect of the pre-conditioning phase**

The effect of the pre-conditioning step was assessed by plotting the two curl parameters values measured for the samples which were only exposed to the standard climate i.e. 50% RH and 23°C (see Figure 4.1). It can be stressed that the MD-oriented curl is almost equal to 50 millimetres for all the samples (with the exception of the test for 30% RH) meaning that pre-conditioning would actually homogenize the tendency to curl in this direction. However, the same observation cannot be made for the CD-oriented curl which range appears to be quite broad, from 5 to 35 millimetres approximately. Yet, this pre-conditioning step was designed to establish a “common starting point” for all the tests which could not take place on the same day, under the exact same conditions (calibration). Different curl values at the beginning of the test phase may in fact affect the final results. Explanations for this observation could be the variation in room climate conditions from one experiment to another as well as the fibres diameter higher moisture sensitivity (compared to fibres length - see Figure 2.7 page 23). In other words, curl in the CD might have been homogenized but the samples would have acclimatized to the room conditions before doing the measurements.

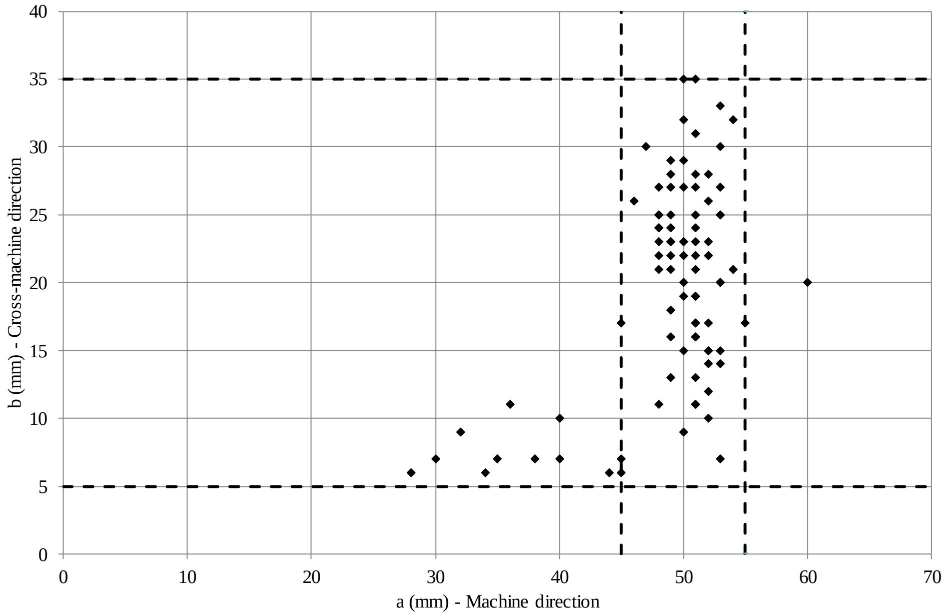


Figure 4.1 Effect of the pre-conditioning step (50% RH and 23°C) on the Pap/mPET/HSL laminate tendency to curl.

4.1.2 Effect of relative humidity and temperature on curl

Firstly, the effect of RH and temperature was qualitatively assessed by plotting the two curl parameters values for the studied climates (see Figure 4.2). Five different symbols were used for the five different RH; three different colours were chosen for the three different temperatures. This representation namely helps identify different groups which correspond to specific RH. Within these RH-related groups, it may what is more be possible to distinguish some temperature-related sub-groups (in the case of 70% RH for example). Thus, mapping both curl parameters seems to be particularly relevant or informative. It can be observed that the tendency to curl increases from 30 to 60% RH whereas it decreases from 60 to 70% RH. This result is quite surprising. Indeed, one would expect the tendency to curl to be higher for "extreme" RH conditions i.e. 30% and 70% RH which respectively correspond to dry and humid environments. One would also expect the tendency to curl to be lower for 50% RH which is defined as the optimal RH for paper-based packaging materials. This may suggest that recommendations for paper-based packaging may not be applicable to paper-based laminates; laminates behaviour may heavily depend on the different materials which are bound together and should not be boiled down to the behaviour of one single material. As regard to temperature, it may also be difficult to identify a logical trend. For 70% RH, the tendency to curl is lower at 30°C and higher at 25°C whereas for 50% RH, the tendency to curl is lower at 25°C and higher at 20°C. It can even be pointed out that the tendency to curl decreases as a function of temperature in the case of 60% RH. According to the literature, one would either expect no significant temperature effect or similar and better results for 20°C and 25°C which correspond to the optimal temperature range for paper-based packaging materials.

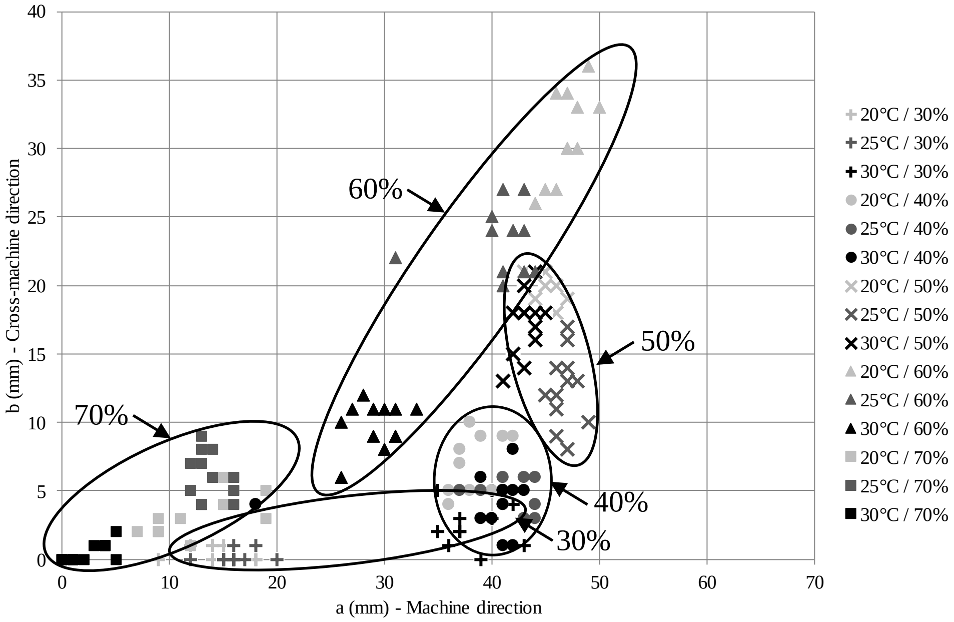
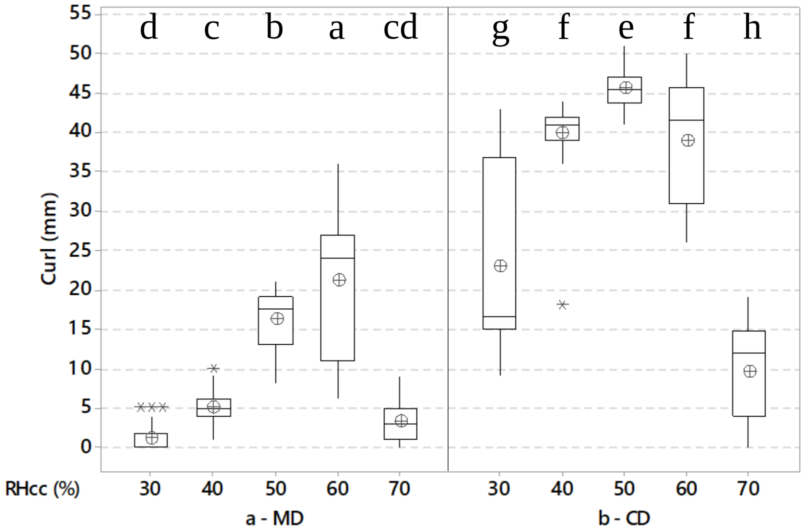


Figure 4.2 Effect of RH and temperature on the Pap/mPET/HSL laminate tendency to curl (test without PE wrapping).

Secondly, a descriptive analysis of the effect of RH and temperature was carried out by drawing box plots. The analysis was conducted one factor at a time. As regard to RH (see Figure 4.3), as already said, the material behaves in the complete opposite way to what could be expected: the tendency to curl happens to be lower for "extreme" RH conditions. In both directions, the same trend can be observed. The average curl value increases from 30 to 60% RH and decreases from 60 to 70% RH in the MD; it increases from 30 to 50% RH and decreases from 50 to 70% RH in the CD. Besides, it can be noticed that deformation does occur to a greater extent in the CD. At 50% RH, deformation in the CD is almost equal to threefold as much the deformation in the MD for instance. This could be related to paper hygroinstability: paper fibres diameter actually varies more than their length under varying moisture conditions. Lastly, the material appears to be particularly sensitive to moisture: CD-oriented curl doubles from 30% to 50% RH and is divided by four from 50% to 70% RH for example. Indeed, the laminate seems to absorb much water at the beginning of the exposition phases (see Table 3.3 page 50).



Note: RH_{cc} stands for climate chamber RH. Data for 20°C, 25°C and 30°C were aggregated. Series which do not "share" a common letter are significantly different (multiple comparisons with Bonferroni adjustment). Comparisons are made one direction at a time i.e. results for the MD and the CD are not compared.

Figure 4.3 Box plot[3] showing the effect of RH on the Pap/mPET/HSL laminate tendency to curl, in the MD and the CD.

Concerning temperature (see Figure 4.4), it affects the tendency to curl in the MD: the higher the temperature, the lower the tendency to curl. MD-oriented curl is namely halved from 20°C to 30°C. However, no significant effect can be described in the CD.

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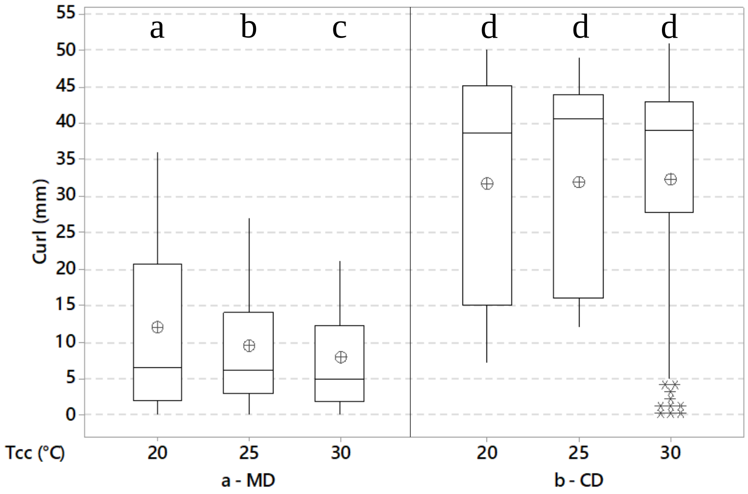
[3] Box plots are a standardized way of representing data distributions based on five statistical measures: minimum, first quartile, median, third quartile and maximum. The central rectangle spans the first quartile (25% of the data are below this value) to the third quartile (75% of the data are below this value). The line inside the rectangle shows the median (middle of the data set). Extremities of the vertical lines respectively indicate the minimum (bottom extremity) and maximum (top extremity), excluding outsiders which are identified using stars. Here, the data set average value has been added: it is marked with a square cross surrounded by a circle.



Note: T_{cc} stands for climate chamber temperature.
Data for 30%, 40%, 50%, 60% and 70% were aggregated. Series which do not “share” a common letter are significantly different (multiple comparisons with Bonferroni adjustment). Comparisons are made one direction at a time.

Figure 4.4 Box plot showing the effect of temperature on the Pap/mPET/HSL laminate tendency to curl, in the MD and the CD.

When studying one factor at a time, RH appears to more significantly affect the material tendency to curl compared to temperature: CD-oriented curl varies from 10 to 46 millimetres as regard to RH whereas it ranges from 31 to 32 millimetres as regard to temperature for instance (see Figure 4.3 and Figure 4.4). Nevertheless, the interaction between these two factors should not be neglected. Line graphs can be used to identify the interaction between two independent variables. In the case of no interaction, lines are expected to be parallel. According to Figure 4.5 and Figure 4.6, there is evidence of an interaction between RH and temperature.

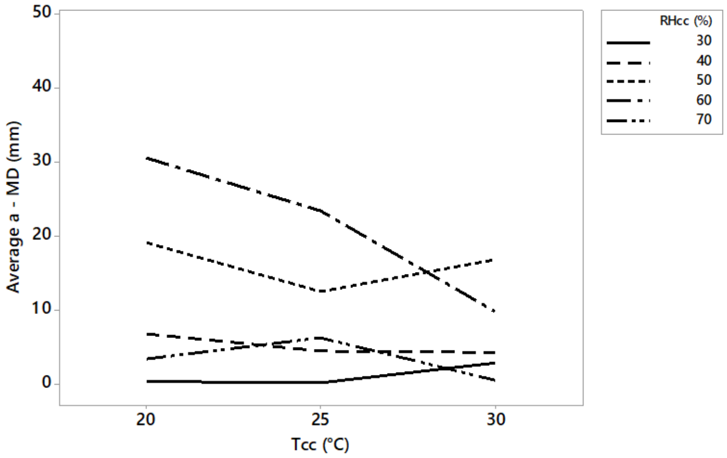


Figure 4.5 Line graphs demonstrating the interaction between RH and temperature effects on the Pap/mPET/HSL laminate average curl values in the MD.

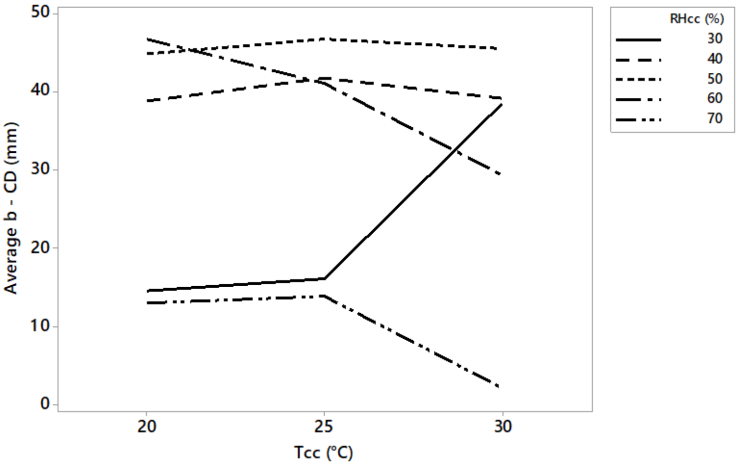


Figure 4.6 Line graphs demonstrating the interaction between RH and temperature effects on the Pap/mPET/HSL laminate average curl values in the CD.

Finally, a more quantitative analysis was performed so as to predict the tendency to curl as a function of RH and temperature. A first multiple linear regression based on the climate chamber temperature, the climate chamber RH and the interaction between these two terms was suggested. The following model was considered:

$$\boldsymbol{Y_i = b_0 + b_1 * T_{cci} + b_2 * RH_{cci} + b_3 * T_{cci} * RH_{cci} + E_i \ for\ i = 1{:}183} \tag{4.1}$$

where Y_i was defined as the ratio of B_i (curl in the CD for the i-th measurement) and A_i (curl in the MD for the i-th measurement), T_{cci} as the climate chamber temperature for the i-th measurement, RH_{cci} as the climate chamber RH for the i-th measurement, E_i as the residual error for the i-th measurement. Implementing regressions with the b/a curl ratio as response surface indeed leads to higher coefficients of determination r^2 i.e. better predictions than focusing on individual curl parameters. Outputs from the multiple linear regression are given in Table 4.1, Table 4.2, Figure 4.7 and Figure 4.8.

The regression shows that it exists a relationship between the ratio of curl parameters and the climate chamber conditions. However, only 44% of the variation in the b/a ratio is explained by the variation in the climate chamber RH and temperature: this first model is not precise enough to predict the laminate tendency to curl. Besides, according to the residual plot, residuals do not seem to be randomly distributed meaning that the assumption of homocedasticty (variance homogenity) is not satisfied for the given model. What is more, the quantile-quantile plot stresses a deviation from the normal distribution: all the points should lie on the reference line i.e. the first bisector. In a nutshell, this first model seems to be questionable.

Table 4.1 Results of the first multiple linear regression analysis - Analysis of variance table.

	Df	*Sum sq.*	*Mean sq.*	*F value*	*Pr(>F)*
Regression	3	3.3432	1.1144	46.6225	2.5794E-22
Residuals	179	4.2786	0.0239		
Total	182	7.6218			

Table 4.2 Results of the first multiple linear regression analysis - Regression Analysis.

The regression equation is:
Curl ratio = -0.5874519 + 0.0159698*T_{cc} + 0.0209945*RH_{cc} - 0.0004710*T_{cc}:RH_{cc}

	Coef. value	*Std. error*	*T*	*Pr(>|t|)*	*Signif (5%)*
(Intercept)	-0.5874519	0.2657381	-2.211	0.0283	*
T_{cc}	0.0159698	0.0105340	1.516	0.1313	
RH_{cc}	0.0209945	0.0051602	4.3069	7.09E-05	*
T_{cc}:RH_{cc}	-0.0004710	0.0002052	-2.295	0.0229	*

S = 0.1546 / R-sq = 43.86% / R-sq(adj) = 42.92%

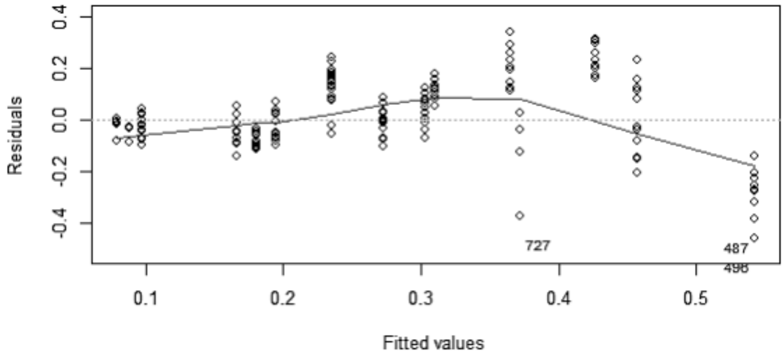


Figure 4.7 Residual plot for the first multiple linear regression (homoscedasticity assumption).

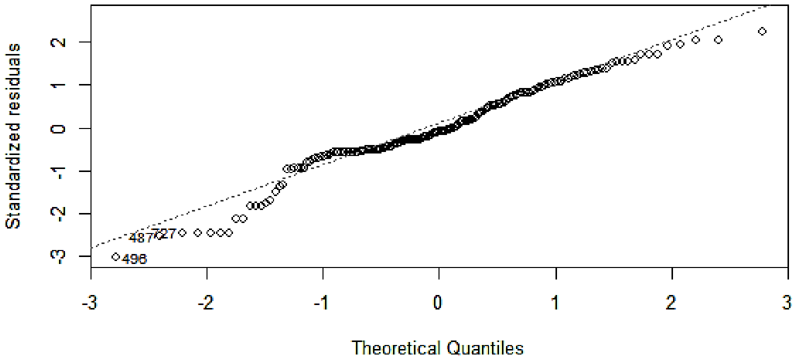


Figure 4.8 Quantile-quantile plot for the first multiple linear regression (normality assumption).

Considering that the room climate conditions where the measurements are done may affect the results (noise), a second multiple linear regression based on the climate chamber and room temperatures, the climate chamber and room RH as well as the interactions between these different terms was implemented. The following model was considered:

$$\begin{aligned}
&\boldsymbol{Y_i = b_{10} + b_{11} * T_{cci} + b_{12} * RH_{cci} + b_{13} * T_{ri} + b_{14} * RH_{ri} + b_{15} * T_{ri} * RH_{ri}} \\
&\boldsymbol{+b_{16} * T_{cci} * T_{ri} + b_{17} * RH_{cci} * RH_{ri} + b_{18} * RH_{cci} * T_{ri} + b_{19} * T_{cci} * RH_{ri}} \\
&\boldsymbol{+b_{20} * T_{cci} * RH_{cci} + b_{21} * T_{cci} * T_{ri} * RH_{cci} + b_{22} * T_{cci} * T_{ri} * RH_{ri}} \\
&\boldsymbol{+b_{23} * T_{ri} * RH_{cci} * RH_{ri} + b_{24} * T_{cci} * RH_{cci} * RH_{ri}} \\
&\boldsymbol{+b_{25} * T_{cci} * RH_{cci} * T_{ri} * RH_{ri} + E_i \;\; for\; i = 1{:}\,183}
\end{aligned} \tag{4.2}$$

where Y_i was defined as the ratio of B_i (tendency to curl in the CD for the i-th measurement) and A_i (tendency to curl in the MD for the i-th measurement), T_{cci} as the climate chamber temperature for the i-th measurement, RH_{cci} as the climate chamber RH for the i-th measurement, T_{ri} as the room temperature for the i-th measurement, RH_{ri} as the room RH for the i-th measurement, E_i as the residual error for the i-th measurement. Final outputs from this second multiple linear regression are given in Table 4.3, Table 4.4, Figure 4.9 and Figure 4.10. Some iterations enabled to simplify the initial model: non-significant terms were excluded.

This second regression shows that it exists a relationship between the ratio of curl parameters, the climate chamber RH and the room climate conditions. The climate chamber temperature term was eliminated because it was not significant. In this respect, one may think that the temperature equilibrium has already been reached when doing curl measurements which would explain that no effect can be detected. Besides, 72% of the variation in the b/a ratio is explained by the variation in the climate chamber RH and the room climate conditions. According to the residual plot, residuals seem to be quite randomly distributed: the assumption of homocedasticty is satisfied for the given model. The quantile-quantile plot gives evidence of a normal distribution: almost all the points lie on the first bisector (some outlyers can be identified). In a nutshell, this second model is more precise than the first one and would enable a relatively good prediction of the laminate tendency to curl.

Table 4.3 Results of the second multiple linear regression analysis - Analysis of variance table.

	Df	*Sum sq.*	*Mean sq.*	*F value*	*Pr(>F)*
Regression	6	5.4933	0.9155	75.7018	3.7351E-46
Residuals	176	2.1286	0.0121		
Total	182	7.6218			

Table 4.4 Results of the second multiple linear regression analysis - Regression Analysis.

The regression equation is:
Curl ratio = 26.7144821 - 1.1990761*T_r - 0.4997555*RH_r - 1.2390396* RH_{cc} + 0.0321654*RH_{cc}:RH_r + 0.0555821*RH_{cc}:T_r - 0.0014321*RH_{cc}:RH_r:T_r

	Coef. Value	***Std. error***	***T***	***Pr(>|t|)***	***Signif (5%)***
(Intercept)	26.7144821	10.5168907	2.540	0.011950	*
T_r	-1.1990761	0.4830126	-2.482	0.013989	*
RH_r	-0.4997555	0.3543190	-1.410	0.160177	
RH_{cc}	-1.2390396	0.2988832	-4.146	5.27E-5	*
RH_{cc}:RH_r	0.0321654	0.0091053	3.533	0.000526	*
RH_{cc}:T_r	0.0555821	0.0136987	4.057	7.46E-5	*
RH_{cc}:RH_r:T_r	-0.0014321	0.0004181	-3.425	0.000766	*

S = 0.1097 / R-sq = 72.37% / R-sq(adj) = 71.26%

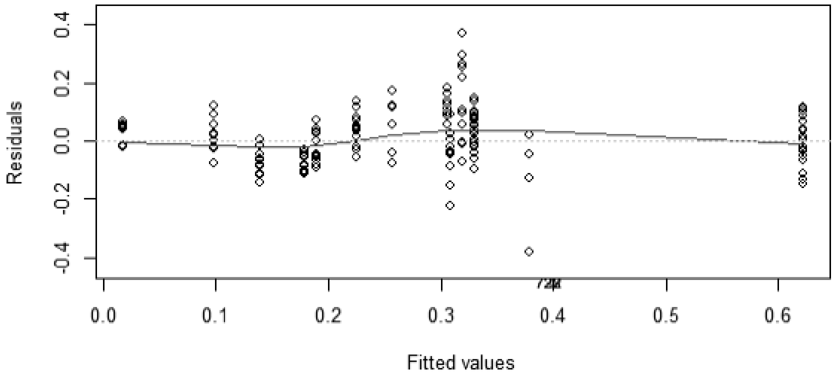


Figure 4.9 Residual plot for the second multiple linear regression (homoscedasticity assumption).

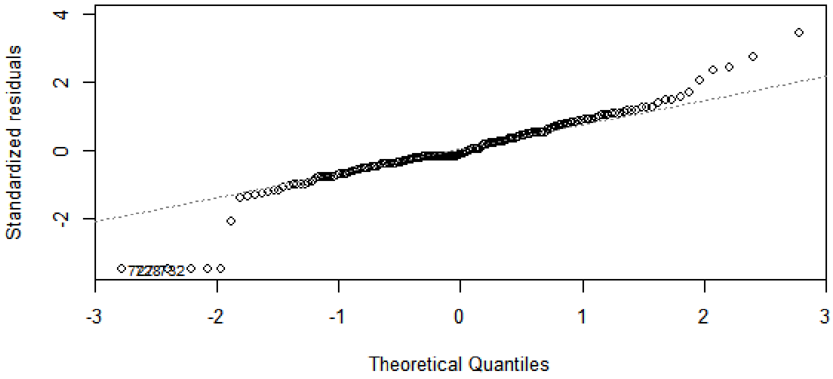
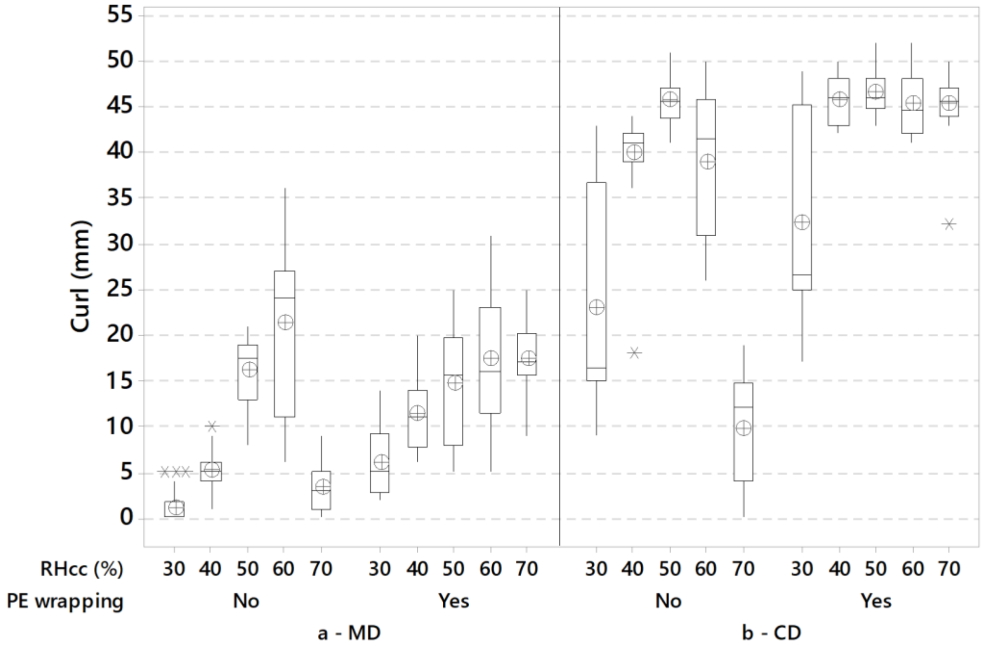


Figure 4.10 Quantile-quantile plot for the second multiple linear regression (normality assumption).

4.1.3 Effect of PE wrapping

In this study, the effect of PE wrapping will be considered as “protective” if it leads to a reduction in curl (compared to the test without PE wrapping).

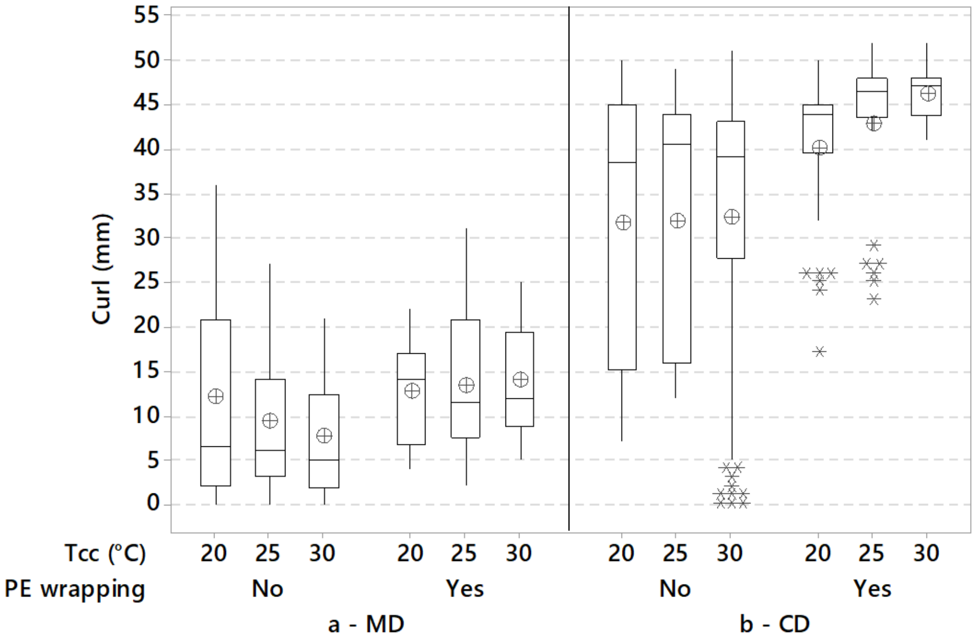
In order to describe the effect of PE wrapping, a first analysis was carried out by drawing box plots. Curl values are represented by focusing on the RH factor on the one hand and on the temperature factor on the other hand. According to Figure 4.11, the main difference between the tests with and without PE wrapping can be observed at 70% RH, both in the MD and the CD. The laminate average curl with PE wrapping is in fact equal to threefold as much its curl without PE wrapping.



Note: Data for 20°C, 25°C and 30°C were aggregated.

Figure 4.11 Box plot illustrating the effect of RH on the Pap/mPET/HSL laminate tendency to curl, in the MD and the CD, with and without PE wrapping.

According to Figure 4.12, it can be noticed that PE wrapping leads to higher curl averages as well as reduces measures dispersion (or variability) for the studied temperatures. On the whole, the effect of PE wrapping could be described as negative considering that it may lead to a stronger tendency to curl.



Note: Data for 30%, 40%, 50%, 60% and 70% were aggregated.

Figure 4.12 Box plot illustrating the effect of temperature on the Pap/mPET/HSL laminate tendency to curl, in the MD and the CD, with and without PE wrapping.

Secondly, the absolute variation in the average curl during the test phase was studied by creating contour graphs. Curl variation for the i-th climate was calculated with the following formula:

$$\Delta\overline{\boldsymbol{curl}}_{\boldsymbol{i}} = \frac{\overline{\boldsymbol{curl}}(\boldsymbol{t_{EQ2}})_{\boldsymbol{i}} - \overline{\boldsymbol{curl}}(\boldsymbol{t_{EQ1}})_{\boldsymbol{i}}}{\overline{\boldsymbol{curl}}(\boldsymbol{t_{EQ1}})_{\boldsymbol{i}}} * \mathbf{100}\ \ \boldsymbol{for}\ \boldsymbol{i} = \mathbf{1}{:}\,\mathbf{15} \tag{4.3}$$

where $\overline{curl}(t_{EQ1})_i$ is defined as the average curl at the beginning of the test phase for the i-th climate and $\overline{curl}(t_{EQ2})_i$ as the average curl at the end of the test phase for the i-th climate (see also Figure 3.15 page 52). According to Figure 4.13, when comparing the results with and without PE wrapping, in both directions, it can be stressed that black areas are larger for the test without PE wrapping. Light grey areas are furthermore larger for the test with PE wrapping. Thus, PE wrapping may “retain the initial conditions” which are responsible for a high tendency to curl at the end of the test phase. Table 4.5 further shows that, in the MD, the average absolute variation in curl reaches 36% without PE wrapping as opposed to 12% with PE wrapping; in the CD, it meets 60% without PE wrapping as opposed to 35% with PE wrapping. It can be concluded that the PE wrapping effect is quite complex to analyse: one may namely think that low curl averages would go with low absolute variations in curl averages which is not the case here.

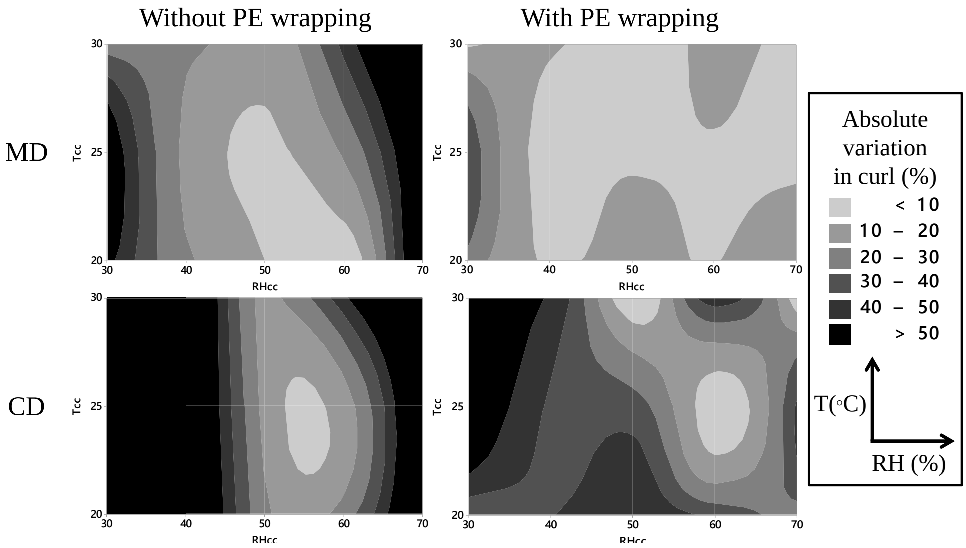


Figure 4.13 Contour chart representing the absolute variation in the Pap/mPET/HSL laminate curl as a function of RH and temperature, in the MD and the CD, with and without PE wrapping.

Table 4.5 Overview of the Pap/mPET/HSL laminate absolute variation in curl with and without PE wrapping.

Absolute variation in curl (%)	***MD***		***CD***	
	Without PE	***With PE***	***Without PE***	***With PE***
Average	36	12	60	35
Minimum	4	3	15	1
Maximum	95	38	98	67

4.1.4 **Conclusions from the analysis of the laminate tendency to curl**

The Pap/mPET/HSL laminate deformation occurs both in the MD and the CD: average curl values respectively meet 13 millimetres and 37 millimetres (see Figure A.5 page 113). Deformation does occur to a greater extent in the CD in relation to the fact that individual fibres diameter changes two to five times more than their length under varying moisture conditions (Iggesund Paperboard, n.d.; Glatfelter, 2005; Lindner, 2018). Considering the given laminate structure, paper may directly exchange water with the environment (no barrier).

The laminate is sensitive to climate conditions. RH appears to more significantly impact its tendency to curl than temperature. Highest curl average values are reached for 50% and 60% RH whereas lowest ones are reached for 30% and 70% RH. These figures differ from the paper packaging industry recommendations: technical data for paper and paper-based packaging materials would thus not be applicable to paper-based laminates.

Despite its climate sensitivity, no curl-related problem has been encountered for this material yet. This stresses the importance of the filling process nature according to which the critical curl limit has to be defined i.e. some processes require the material to be perfectly flat whereas some others do not. In the case of a demanding filling process, the material climate-related risk would probably be high.

PE wrapping limits the absolute variation in curl but eventually leads to a stronger tendency to curl. In the case of this Pap/mPET/HSL laminate, PE wrapping cannot be considered as protective.

4.2 Case of the Alu/Pap/PE laminate

The tendency to curl of this second paper-based laminate surprisingly happened to be very low: less than or equal to 10 millimetres in the MD and equal to zero millimetres in the CD. As a result, the tendency to curl which was initially measured as the shortest distance or gap between the material edges (a parameter) had to be measured as the shortest height between the table and the material edges (a* parameter) (Deutsches Institut für Normung (DIN), 2014 - see also Figure 3.4 page 42). It was eventually decided to measure the two MD-oriented curl parameters, a and a*, so as to get a deeper understanding of the DIN 55403 standard.

4.2.1 **Understanding the recommendations made in the DIN 55403 standard as regard to the cross-cut test method**

The change from a to a* which is suggested in the case of low tendency to curl could indicate a strong relationship between these two parameters. Furthermore, when doing measurements, it was clear that the higher the a value, the higher the a* value (positive correlation). Consequently, a linear regression was undertaken in order to identify the nature of the relationship between a and a*. The following model was considered:

$$A_i = b_0 * A^*{}_i + E_i \;\; for\; i = 1{:}180 \tag{4.4}$$

where A_i was defined as the curl length for the i-th measurement, A^*_i as the curl height for the i-th measurement and E_i as the residual error for the i-th measurement. In the case of flat samples, the distance between the material edges and the height are both equal to zero millimetres: the intercept was thus set to zero. Outputs from the linear regression are given in Table 4.6, Table 4.6, Figure 4.14, Figure 4.15 and Figure 4.16. The data set includes curl values for the test without PE wrapping (pre-conditioning step and test with PE wrapping excluded).

This regression shows that there is a very strong relationship between a and a*. The coefficient of determination r^2 almost reaches 90% i.e. 90% of the variation in a is explained by the variation in a*. However, according to the residual plot, residuals do not seem to be randomly distributed meaning that the assumption of homocedasticty (variance homogenity) would not be satisfied for the given model. Besides, the quantile-quantile plot stresses a deviation from the normal distribution: all the points should lie on the first bisector. In a nutshell, this model may be questionable. Part of the problem may come from the quite low precision when measuring the a parameter. Using a 0.5-millimetre graduated ruler and increasing the number of samples or repetitions would have a positive effect on the results.

Table 4.6 Results of the linear regression analysis - Analysis of variance table.

	Df	*Sum sq.*	*Mean sq.*	*F value*	*Pr(>F)*
Regression	1	1662.22	1662.22	1567.80	3.54E-90
Residuals	179	189.78	1.06		
Total	180	1852.00			

Table 4.7 Results of the linear regression analysis - Regression Analysis.

The regression equation is:
Curl length MD = 0.363009*Curl height MD

	Coef. value	*Std. error*	*t*	*Pr(>|t|)*	*Signif (5%)*
Curl height MD	0.363009	0.009168	39.59	1.77E-90	*

S = 1.03 / R-sq = 89.75% / R-sq(adj) = 89.19%

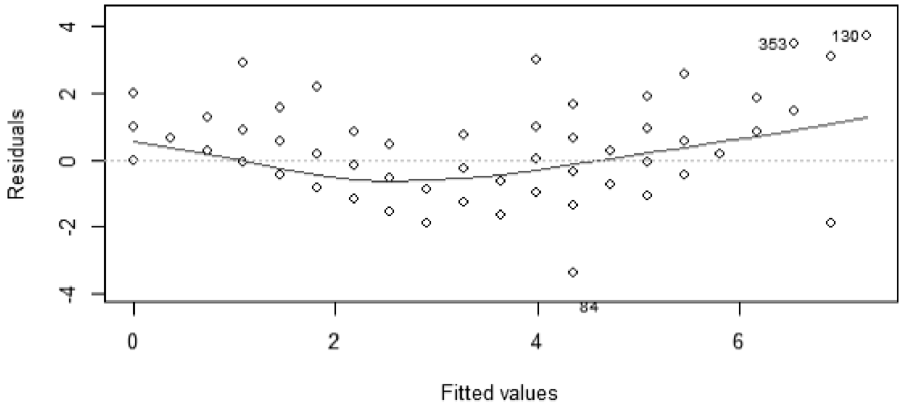


Figure 4.14 Residual plot for the linear regression (homoscedasticity assumption).

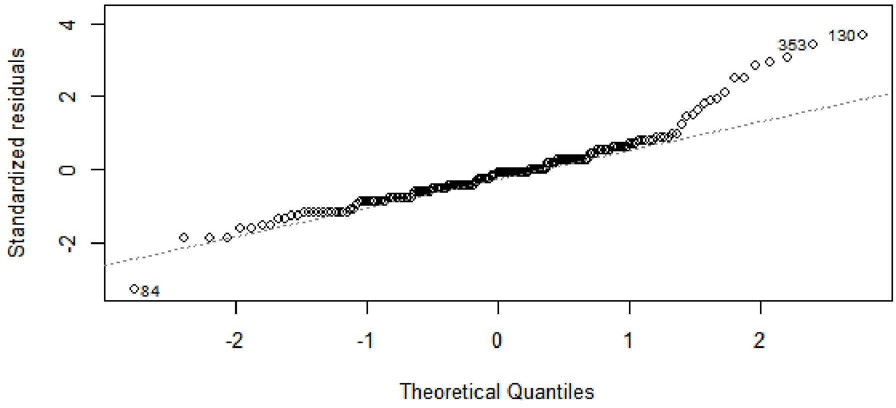


Figure 4.15 Quantile-quantile plot for the linear regression (normality assumption).

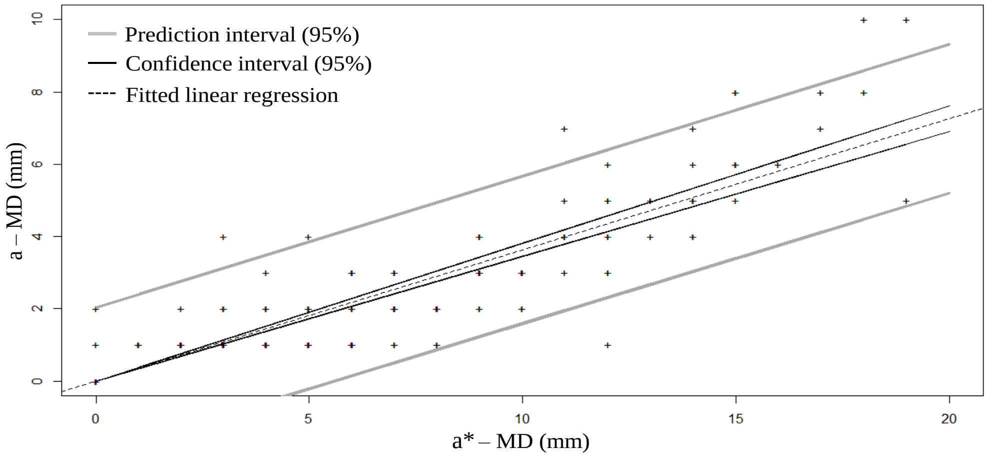


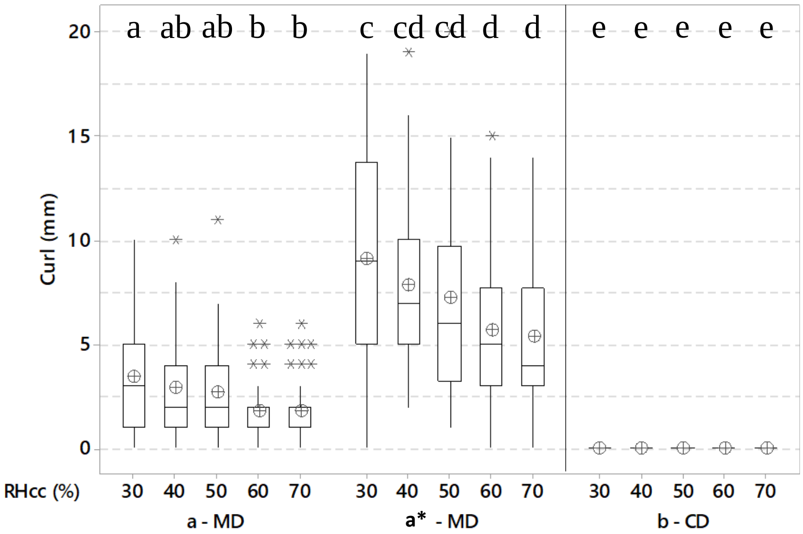
Figure 4.16 Linear relationship between a and a* parameters for the Alu/Pap/PE laminate.

4.2.2 **Effect of the pre-conditioning step**

In the case of this second laminate, the tendency to curl is equal to zero millimetres in the CD. Therefore, it is not possible to draw the same graph as previously (see Figure 4.1 page 58). Instead, the distribution of a and a* values can be analysed. At the end of the pre-conditioning step, the a value ranges from 0 to 12 millimetres; the a* value ranges from 0 to 19 millimetres. These figures may suggest that this calibration step did not homogenize the tendency to curl in the MD which may in fact affect the final results. Here again, the influence of room climate conditions may be mentioned.

4.2.3 **Effect of relative humidity and temperature**

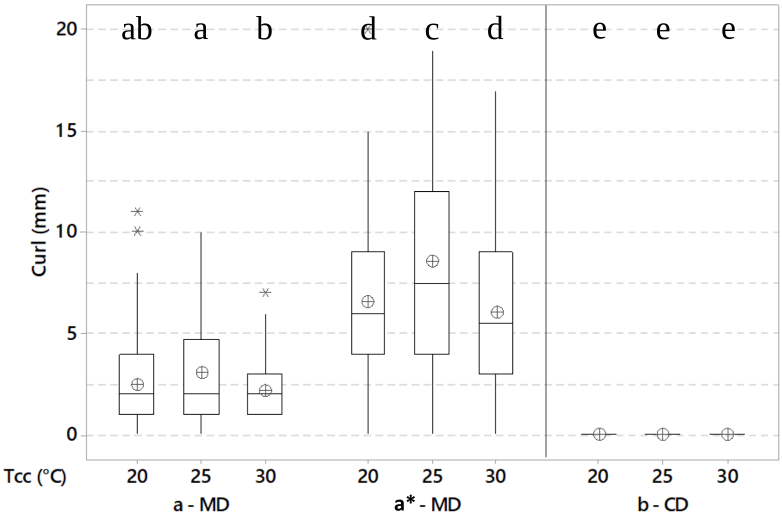
As for the first material, a descriptive analysis of RH and temperature effects was carried out by drawing box plots. The analysis was conducted one factor at a time for the MD-oriented curl. If it can be observed that an increase in RH leads to a reduction in the average laminate curl (see Figure 4.17), RH effect does not however appear to be very significant. This result can be related to the fact that the laminate does not absorb much water. Indeed, according to Table 3.4 (see page 50), the Alu/Pap/PE 100 cm²-samples mass remained almost constant during the exposition phases. Sandwiching paper between aluminium and PE may therefore help prevent or reduce curl.



Note: Data for 20°C, 25°C and 30°C were aggregated. Series which do not “share” a common letter are significantly different (multiple comparisons with Bonferroni adjustment). Comparisons are made one parameter at a time i.e. results for a, a* and b are not compared.

Figure 4.17 Box plot showing the effect of RH on the Alu/Pap/PE laminate tendency to curl, in the MD and the CD.

Concerning temperature (see Figure 4.18), a slight still significant increase in the a* parameter can be noticed at 25°C.



Note: Data for 30%, 40%, 50%, 60% and 70% were aggregated. Series which do not "share" a common letter are significantly different (multiple comparisons with Bonferroni adjustment). Comparisons are made one parameter at a time.

Figure 4.18 Box plot showing the effect of temperature on the Alu/Pap/PE laminate tendency to curl, in the MD and the CD.

When studying one factor at a time, RH and temperature do not show to strongly impact the laminate tendency to curl. Nonetheless, the interaction between these two factors should not be neglected. According to the line graphs given in Figure 4.19 and Figure 4.20, as all the different lines are not parallel, there is evidence of an interaction between RH and temperature.

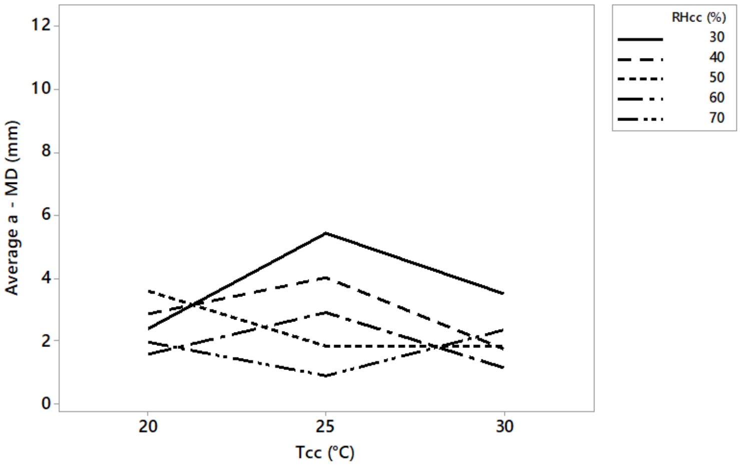


Figure 4.19 Line graphs demonstrating the interaction between RH and temperature effects on the Alu/Pap/PE laminate average curl values in the MD - Case of the "a" parameter.

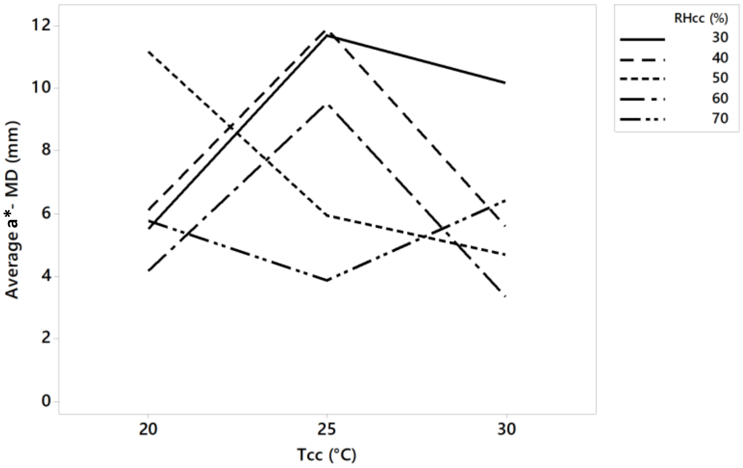


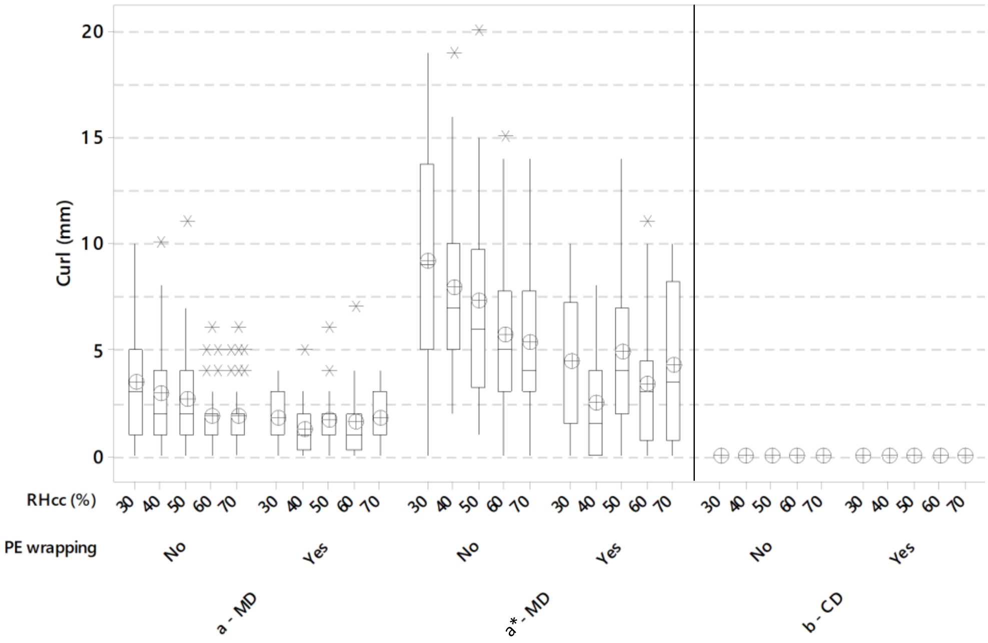
Figure 4.20 Line graphs demonstrating the interaction between RH and temperature effects on the Alu/Pap/PE laminate average curl values in the MD - Case of the a* parameter.

Unfortunately, it was not possible to implement statistics for the Alu/Pap/PE laminate so as to predict curl as a function of climate chamber and room conditions. Neither a nor a* enables such meaningful analyses (very low coefficients of determination - see A.3.3 Predict curl as a function of climate chamber and room conditions page 133).

4.2.4 **Effect of PE wrapping**

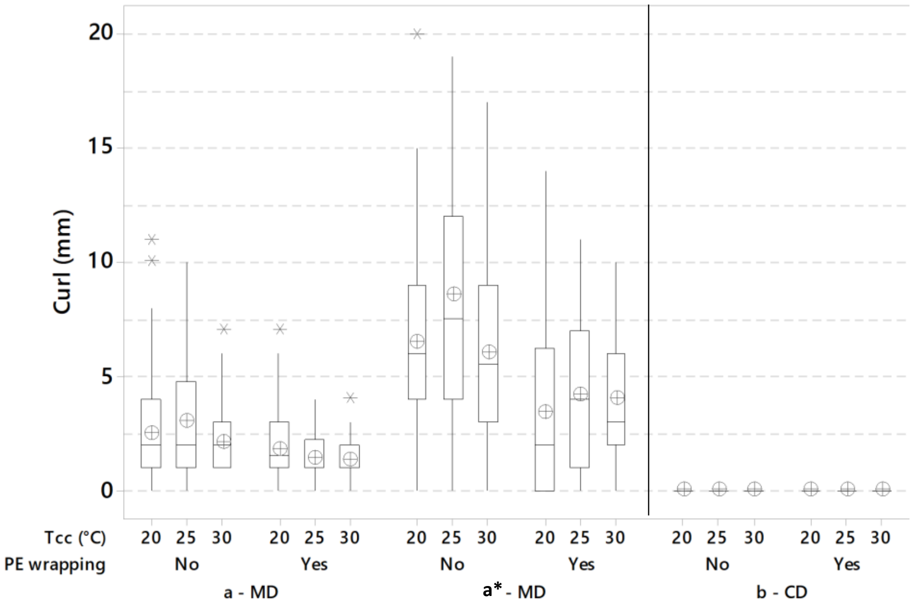
As defined earlier, the effect of PE wrapping will be considered as “protective” if it leads to a reduction in curl (compared to the test without PE wrapping).

A first analysis was carried out by drawing box plots. According to Figure 4.21, when focusing on the RH factor, it can be stressed that PE wrapping leads to lower curl averages in the MD. At 30% RH, the a* parameter average value for the test without PE wrapping is namely equal to twice as much its average value with PE wrapping for instance. According to Figure 4.22, when considering the temperature factor, it can be pointed out that PE wrapping does also lower curl. At 20°C, the a* parameter average value for the test without PE wrapping is equal to twice as much its average value with PE wrapping for example. On the whole, the effect of PE wrapping could be described as positive.



Note: Data for 20°C, 25°C and 30°C were aggregated.

Figure 4.21 Box plot illustrating the effect of RH on the Alu/Pap/PE laminate tendency to curl, in the MD, with and without PE wrapping.



Note: Data for 30%, 40%, 50%, 60% and 70% were aggregated.

Figure 4.22 Box plot illustrating the effect of temperature on the Alu/Pap/PE laminate tendency to curl, in the MD, with and without PE wrapping.

Secondly, the absolute variation in the average curl during the test phase was studied by creating contour graphs. According to Figure 4.23, when comparing the results with and without PE in the MD, black areas appear to be relatively large for the test without PE wrapping meaning that the average curl strongly varied during the test phase. Table 4.8 further shows that, in the MD, the average absolute variation in curl reaches 31% without PE wrapping as opposed to 51% with PE wrapping; in the CD, it meets 47% without PE wrapping as opposed to 58% with PE wrapping. In this case, high absolute variations in curl averages lead to lower curl averages.

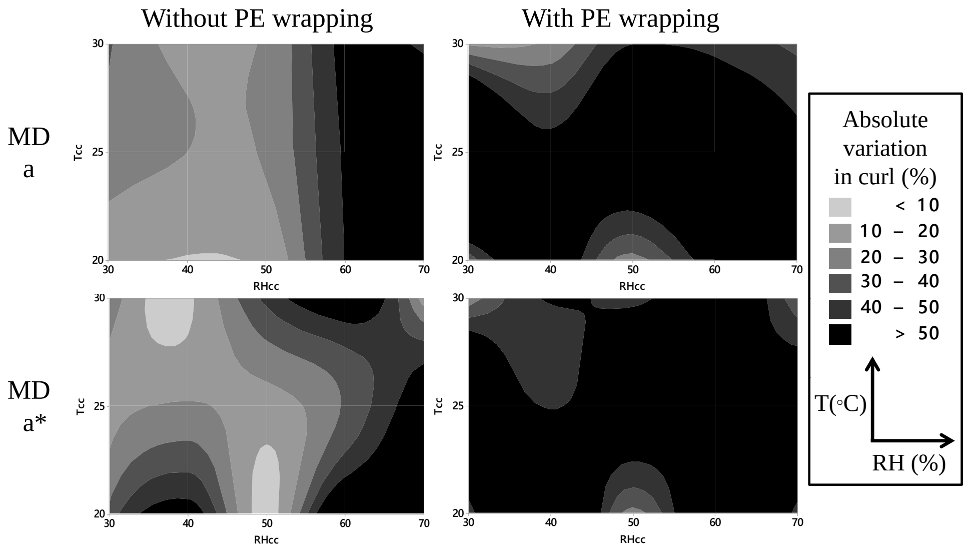


Figure 4.23 Contour chart representing the absolute variation in the Alu/Pap/PE MD-oriented curl as a function of RH and temperature, with and without PE wrapping.

Table 4.8 Overview of the Alu/Pap/PE laminate absolute variation in curl with and without PE wrapping.

Absolute variation in curl (%)	*MD*		*CD*	
	Without PE	*With PE*	*Without PE*	*With PE*
Average	31	51	47	58
Minimum	7	19	6	23
Maximum	80	86	72	100

4.2.5 Conclusions from the analysis of the laminate tendency to curl

The Alu/Pap/PE laminate deformation only occurs in the MD: the average curl value meets 3 millimetres (see Figure A.6 page 132). Whatever the environmental conditions, the CD-oriented curl parameter is always equal to zero millimetres.

The laminate does not appear to be sensitive to climate conditions. In fact, RH and temperature do not really impact the laminate tendency to curl. This may be explained by the fact that paper is sandwiched between aluminium and PE which may namely protect it i.e. prevent it from absorbing water.

While not being sensitive to climate, the material tendency to curl has been identified as a critical point for the development of a new packaging solution. This highlights the demanding nature of the filling process which requires the material to be (perfectly) flat. Leaving aside the filling process, the material climate-related risk could be described as low, namely compared to the Pap/mPET/HSL laminate.

PE wrapping leads to lower curl averages in the MD while enabling relatively high absolute variations in curl. In the case of this Alu/Pap/PE laminate, PE wrapping can be considered as protective.

5 Conclusions and recommendations

This fifth chapter corresponds to the conclusion of the study. It aims at summarizing the main outcomes obtained for the two selected paper-based laminates, emphasizing the limits of the study as well as providing the reader with meaningful recommendations for future researches dealing with similar or related topics. In this respect, researchers, packaging developers as well as packaging suppliers may benefit from the given recommendations, making the study all the more relevant.

5.1 Conclusions

5.1.1 **Main factors leading to paper-based flexible laminates curl**

Numerous factors can lead to paper-based flexible laminates curl, at different levels (cellulose, paper, laminate, roll) and different stages (converting, warehousing, filling). Therefore, a framework of factors based on a literature review of scientific articles and technical reports was suggested (see Table 2.4 page 32). RH and temperature happened to be the most well-documented factors. However, this does not mean that they are the most significant ones since no hierarchy has been described yet: there is currently a lack of information as regard to paper-based flexible laminates curl phenomenon.

5.1.2 **Impact of relative humidity and temperature on the selected Pap/mPET/HSL and Alu/Pap/PE laminates behaviour**

Two different packaging materials were considered: a widely processed heat-sealable yogurt lid (Pap/mPET/HSL) and a prototype laminate (Alu/Pap/PE). Their tendency to curl was assessed under fifteen climates i.e. under five RH values (30%, 40%, 50%, 60% and 70%) and three temperature values (20°C, 25°C and 30°C). The cross-cut method which is described in the German DIN 55403 standard was implemented as test method.

Concerning the Pap/mPET/HSL laminate for which no curl-related problem has been encountered yet, deformation occurs both in the MD and the CD: average curl values respectively meet 13 millimetres and 37 millimetres (see Figure A.5 page 113). RH and temperature were both shown to have a significant effect on its tendency to curl; RH more significantly impacts its tendency to curl than temperature (see Figure 4.3 page 60 and Figure 4.4 page 61). Highest curl average values are reached for 50% and 60% RH whereas lowest ones are reached for 30% and 70% RH. The material was demonstrated to be sensitive to climate conditions.

Concerning the Alu/Pap/PE prototype laminate which tendency to curl has been identified as critical factor for its processability, deformation only occurs in the MD: the average curl value meets 3 millimetres (see Figure A.6 page 132). Whatever the environmental conditions, the CD-oriented curl parameter is always equal to zero millimetres. On whole, the laminate tendency to curl was shown to be low and not sensitive to climate conditions (see Figure 4.17 and Figure 4.18 page 72).

Considering the given material-specific results, recommendations for paper-based packaging do not seem to be applicable to paper-based laminates. Laminates behaviour may indeed depend on the different materials which are bound together and should not be boiled down to the behaviour of one single material. This kind of study should be conducted to assess RH and temperature effects on the tendency to curl of any single laminate (no extrapolation).

5.1.3 Materials climate-related risk assessment

The description of the two paper-based flexible laminates behaviour under fifteen different climates is certainly informative. Yet, materials behaviour cannot be separated from the nature of the filling processes the materials have to undergo. In this respect, specific critical limits need to be defined for each single material.

Despite its climate sensitivity, no curl-related problem has been encountered for the Pap/mPET/HSL laminate yet. Leaving aside the filling process, the material climate-related risk would probably be characterized as high. It was namely possible to observe and describe paper fibres hygroexpansion in the MD and the CD under various RH. Considering the given laminate structure, paper may directly exchange water with the environment (no barrier).

The Alu/Pap/PE prototype laminate which tendency to curl has been identified as critical factor was shown not to be sensitive to climate conditions. This result may be explained by the laminate structure: paper is sandwiched between aluminium and PE which probably protect it i.e. prevent it from absorbing water. Leaving aside the filling process, the material climate-related risk could be described as low, namely compared to the Pap/mPET/HSL laminate.

5.1.4 Solution to reduce paper-based laminates tendency to curl

Practices which have been suggested in order to avoid paper-based packaging materials curl were described (see Table 2.5 page 37). It was decided to more specifically assess the effect of PE wrapping.

In the case of the Pap/mPET/HSL laminate, PE wrapping limits the absolute variation in curl but eventually leads to a stronger tendency to curl: it cannot be considered as protective.

In the case of the Alu/Pap/PE laminate, PE wrapping leads to lower curl averages in the MD while enabling relatively high absolute variations in curl: it can be considered as protective.

These opposite observations make the PE wrapping effect complex to analyse. Considering the given material-specific results, it may not be possible to draw a single general conclusion for all the paper-based flexible laminates.

5.2 Further research recommendations

5.2.1 Limitations of the study

Concerning the limits of this study, it can firstly be highlighted that sampling could be improved by ordering rolls of the same dimensions. Samples location on the web may namely affect the results. Furthermore, the number of replications for each test should be set to twelve rather than six - when possible. This would for instance decrease the series standard deviations and thus make the average values more accurate. It would even be better to define a required minimum number of repetitions (the number of repetitions which enables normal statistical distributions?). Besides, as the fifteen climates of interest were mainly defined based on the manufacturing environmental conditions for January 2018, the relevance of the experimental plan should be validated by collecting RH and temperature data for one entire year. A more detailed picture of the seasonality effect would be interesting. In relation to the pre-conditioning phase or calibration, the influence of the room climate conditions may be questioned. Ideally, measures should be done in the same conditions as the climate chamber conditions to avoid “noise”. Finally, this study focuses on two specific paper-based flexible laminates which makes it non-exhaustive.

5.2.2 **Future research suggestions**

In this study, decision was made to implement the cross-cut method, one out of the three test methods which are suggested for the determination of packaging films tendency to curl (Deutsches Institut für Normung (DIN), 2014). It indeed showed the advantage to be quite simple, quick and affordable to carry out. Required materials include a weather station, a cutting mat, a specific cutting pattern, a cutter, a ruler and tape. Furthermore, both low and high tendency to curl can be characterized. By assessing the tendency to curl in the MD and the CD, this method eventually appears to be very informative. Therefore, stakeholders such as packaging developers and packaging suppliers are encouraged to apply the cross-cut method as standard quality control test method; researchers are encouraged to apply it in further studies which goal would be to assess the effect of certain factors on packaging materials tendency to curl

One tricky aspect when implementing this test method deals with the change in measurement method for low and high tendency to curl (see Figure 3.4 page 42). According to the German DIN 55403 standard (2014), in the case of low tendency to curl (i.e. when distances between the material edges are lower than 5 millimetres), curl parameters should be measured as the shortest heights between the table and the material edges; in the case of high tendency to curl (i.e. when distances between the material edges are higher than 5 millimetres), they should be measured as the shortest distances between the material edges. It can sometimes happen that curl values range from zero to ten millimetres for a given sample. How to proceed then? Well, considering the fact that from a data analysis perspective it is not relevant to mix "heights" and "lengths", the best fitted measurement method has to be identified right from the beginning.

In the case of quality controls which goal is to determine if the materials meet the requirements defined in packaging materials specifications, it would be advised to always measure curl parameters as the shortest distances between the material edges (simplified method to make sure the same consistent procedure is always applied). Consequently, a curl value lower than or equal to 5 millimetres would always be considered as small.

For future researches which goal would be to study the effect of certain factors on packaging materials tendency to curl, Table 5.1 can be used as a decision tool. If it is still unclear which kind of data is required, it would be advised to measure both heights and lengths for every single sample so as to collect as many data as possible in one go and thus avoid repeating experiments.

Table 5.1 Suggested matrix to help choose the most relevant measurement method for the cross-cut method - Case of further scientific researches.

	Low CD-oriented tendency to curl	**High CD-oriented tendency to curl**
Low MD-oriented tendency to curl	Measure heights for both curl parameters	Do you want to compare/relate curl parameters? - If yes, need for a single measurement method → Measure lengths for both parameters - If no, measure heights in the MD and lengths in the CD
High MD-oriented tendency to curl	Do you want to compare/relate curl parameters? - If yes, need for a single measurement method → Measure lengths for both parameters - If no, measure lengths in the MD and heights in the CD	Measure lengths for both curl parameters

These experimental procedure details apart, three areas of improvement or research can be suggested.

Firstly, in order to exactly describe the effect of the single paper layer, it would have been better to define a reference laminate for each of the two studied materials. A reference laminate would actually correspond to the test laminate without the paper layer i.e. mPET/HSL for the Pap/mPET/HSL material and Alu/Pap for the Alu/Pap/PE material.

Secondly, it could be interesting to conduct a similar study at the roll scale (as opposed to the sheet scale) which is more meaningful from the industry perspective. Differences in curl might indeed be expected depending on the location of the samples towards the roll core. Besides, considering the difference in the studied packaging materials exposure durations (see Table 3.3 and Table 3.4 page 50), it would be useful to define accurate rolls acclimatization durations for every single laminate (see Table 2.6 page 37).

Finally, in order to hierarchize the different factors leading to curl and thereby define effective solutions to this problem, it could be useful to undertake a study comparing the effect of the lamination process nature (see 2.3 Paper-based laminates manufacturing processes page 27) to the one of the environmental conditions on paper-based flexible laminates tendency to curl. Changing the lamination process could indeed reduce the tendency to curl much more than trying to protect packaging materials from "harmful" environmental conditions.

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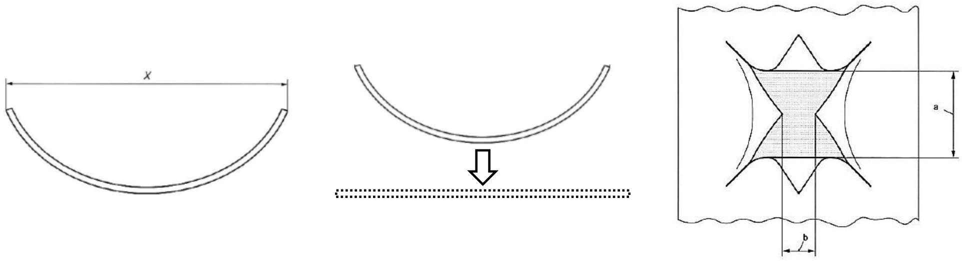
Appendix A Additional data

A.1 Test method selection

A.1.1 Comparison of the three test methods

Table A.1 Comparison of the three test methods described in the German standard DIN 55403.

Methods	*Advantages*	*Disadvantages*
Round sample method Cutting a circular sample and measuring the distance between the material edges (x parameter in mm) (see Figure A.1 and Figure 2.1).	- Relatively quick and simple - Affordable (weather station, cutting mat, circular sample cutter, ruler) - 1 parameter to characterize the tendency to curl = "easy to analyse"	- **Not applicable to very low and very high tendency to curl** (see Figure A.3) - Curl direction may be difficult to define (twist)
Flatness temperature Determining the temperature at which the material is flat (fixed RH) (see Figure A.1 and Figure A.4).	- Temperature which enables material processability	- Difficult to implement = time-consuming and moving materials (climate chamber airflow) - Expensive (climate chamber) **Can hardly be implemented by other stakeholders** - Flatness temperature determined for a specific RH = repeat the experiments for different RH values
Cross-cut method Cutting crosses in the packaging material web using a cutting pattern and measuring the shortest distance between the material edges, in the MD (a parameter in mm) and the CD (b parameter in mm) (see Figure A.1).	- Relatively quick and simple - Affordable (weather station, cutting mat, specific cutting pattern, cutter, ruler, tape) - Applicable to low and high tendency to curl - 2 parameters to characterize the tendency to curl = "more informative"	- Need to repeat measurements so as to get relevant results



„Round sample method“ „Flatness temperature“ „Cross-cut method“

Figure A.1 Overview of the three test methods described in the German standard DIN 55403.

A.1.2 Round sample method

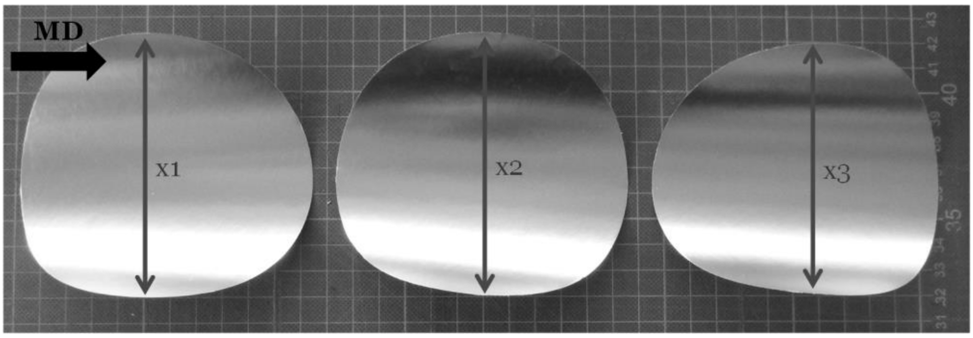


Figure A.2 Observations made for the Pap/mPET/HSL laminate at 50% RH and 23°C (low tendency to curl).

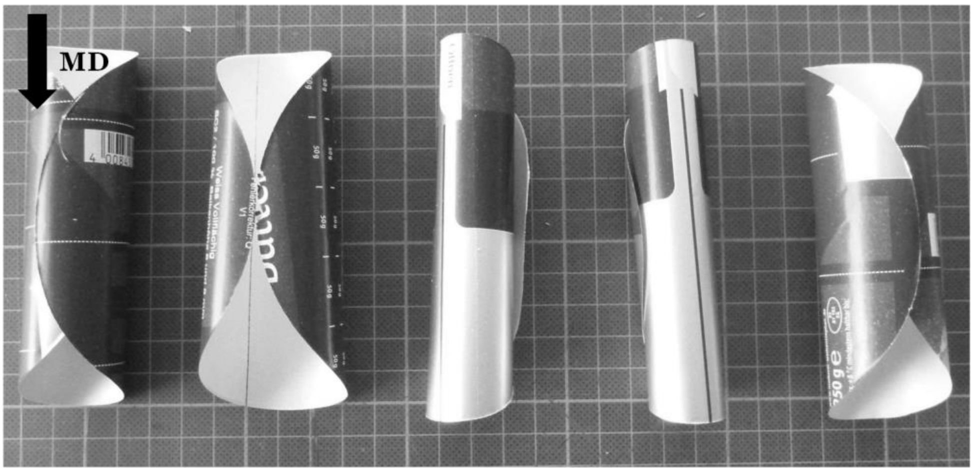


Figure A.3 Observations made for an Alu/Pap/HSL laminate at 50% RH and 23°C (high tendency to curl).

A.1.3 Determination of the flatness temperature

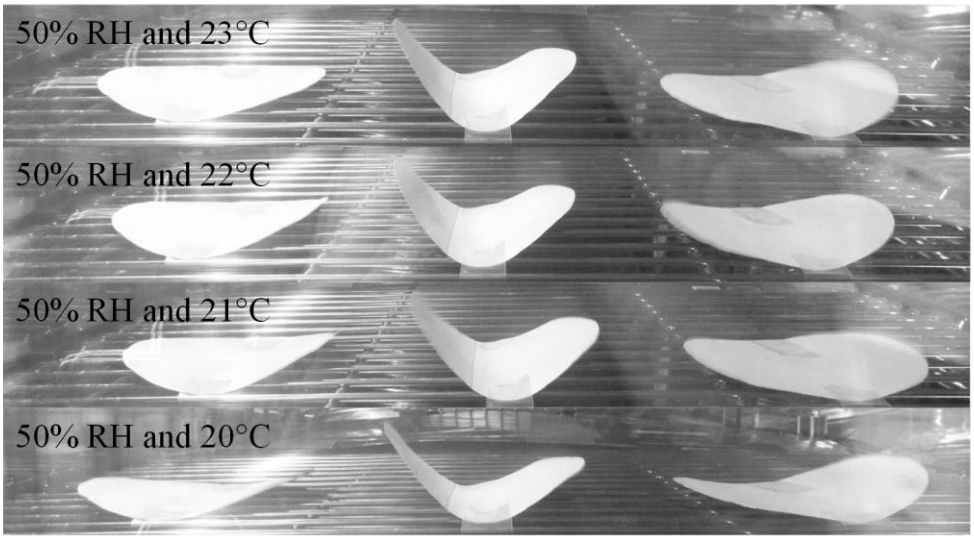


Figure A.4 Observations made for an Alu/Pap/HSL laminate.

A.2 Pap/mPET/HSL laminate

A.2.1 Raw data

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate.

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
1	21.0	33	23	50	No	52	15	-
						60	20	-
						53	25	-
						50	20	-
						53	7	-
						51	11	-
	21.6	33	20	70	No	15	4	-
						19	3	-
						19	5	-
						16	5	-
						15	4	-
						15	6	-
						11	3	-
						9	3	-
						9	2	-
						9	2	-
						12	1	-
						7	2	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	**T_r (°C)**	**RH_r (%)**	**T_{cc} (°C)**	**RH_{cc} (%)**	**PE wrapping**	**b (mm)**	**a (mm)**	**a* (mm)**
1	22.0	32	20	70	Yes	45	14	-
						47	18	-
						45	11	-
						44	13	-
						45	9	-
						32	17	-
2	20.8	35	23	50	No	50	23	-
						48	25	-
						49	24	-
						48	27	-
						51	21	-
						49	27	-
	20.5	36	20	60	No	46	34	-
						48	33	-
						47	34	-
						49	36	-
						50	33	-
						48	30	-
						46	27	-
						44	26	-
						45	27	-
						47	30	-
						47	30	-
						44	26	-
	20.5	36	20	60	Yes	42	16	-
						45	20	-
						42	14	-
						44	15	-
						44	12	-
						49	16	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
3	21.8	34	23	50	No	51	25	-
						48	25	-
						50	23	-
						49	25	-
						50	22	-
						52	28	-
	21.6	34	20	50	No	44	19	-
						46	20	-
						43	21	-
						45	20	-
						47	19	-
						45	21	-
						43	18	-
						43	18	-
						46	18	-
						45	21	-
						46	18	-
						44	16	-
	21.6	34	20	50	Yes	43	12	-
						45	16	-
						44	5	-
						44	17	-
						44	5	-
						45	22	-
4	21.7	34	23	50	No	51	27	-
						51	24	-
						49	23	-
						50	29	-
						49	28	-
						49	29	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
4	22.0	33	20	40	No	36	5	-
						41	6	-
						38	5	-
						39	9	-
						41	4	-
						42	9	-
						38	10	-
						37	7	-
						37	8	-
						41	9	-
						36	4	-
						40	5	-
	21.9	33	20	40	Yes	43	14	-
						44	20	-
						42	14	-
						43	19	-
						50	12	-
						49	18	-
5	15.6	24	23	50	No	28	6	-
						34	6	-
						32	9	-
						36	11	-
						30	7	-
						35	7	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
5	20.2	22	20	30	No	14	0	-
						14	1	-
						15	0	-
						12	0	-
						9	0	-
						12	1	-
						18	0	-
						17	0	-
						15	0	-
						18	0	-
						16	1	-
						15	1	-
	19.5	22	20	30	Yes	26	5	-
						25	5	-
						24	5	-
						17	6	-
						26	4	-
						26	7	-
6	21.0	33	23	50	No	51	31	-
						50	27	-
						50	35	-
						51	35	-
						54	32	-
						51	28	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
6	22.0	32	25	70	No	12	5	-
						16	5	-
						13	4	-
						13	4	-
						16	4	-
						16	6	-
						13	7	-
						12	7	-
						14	8	-
						13	9	-
						14	6	-
						13	8	-
	21.6	33	25	70	Yes	49	17	-
						44	16	-
						47	20	-
						44	17	-
						50	25	-
						47	16	-
7	20.8	35	23	50	No	54	21	-
						53	30	-
						52	26	-
						47	30	-
						53	25	-
						53	33	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
7	20.5	36	25	60	No	43	24	-
						43	27	-
						40	24	-
						40	25	-
						41	21	-
						41	27	-
						44	21	-
						42	24	-
						31	22	-
						43	21	-
						42	24	-
						41	20	-
	20.5	36	25	60	Yes	48	23	-
						44	26	-
						47	23	-
						42	31	-
						49	23	-
						52	31	-
8	22.5	31	23	50	No	53	14	-
						52	14	-
						50	15	-
						51	19	-
						52	15	-
						49	13	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
8	22.5	31	25	50	No	46	11	-
						46	12	-
						46	9	-
						47	14	-
						49	10	-
						48	13	-
						47	8	-
						47	13	-
						46	14	-
						47	17	-
						45	12	-
						47	16	-
	22.5	31	25	50	Yes	45	8	-
						48	9	-
						46	11	-
						46	8	-
						47	8	-
						47	15	-
9	21.7	31	23	50	No	52	12	-
						49	16	-
						52	17	-
						46	26	-
						53	15	-
						52	10	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
9	21.7	31	25	40	No	41	5	-
						43	3	-
						39	5	-
						44	6	-
						37	5	-
						43	6	-
						40	3	-
						41	4	-
						43	3	-
						41	6	-
						44	3	-
						44	4	-
	21.7	31	25	40	Yes	48	8	-
						46	6	-
						48	12	-
						47	11	-
						47	9	-
						46	13	-
10	20.8	22	23	50	No	45	7	-
						40	7	-
						44	6	-
						38	7	-
						45	6	-
						40	10	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
10	21.6	21	25	30	No	16	1	-
						12	0	-
						16	0	-
						15	0	-
						16	0	-
						16	0	-
						15	0	-
						17	0	-
						18	1	-
						15	0	-
						20	0	-
						16	0	-
	21.3	21	25	30	Yes	26	3	-
						23	3	-
						25	2	-
						27	2	-
						27	2	-
						29	2	-
11	22.6	32	23	50	No	48	11	-
						45	17	-
						49	18	-
						48	23	-
						51	17	-
						50	32	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
11	22.2	33	30	70	No	5	2	-
						2	0	-
						5	0	-
						4	1	-
						3	1	-
						4	1	-
						1	0	-
						1	0	-
						0	0	-
						1	0	-
						0	0	-
						0	0	-
	22.2	34	30	70	Yes	43	16	-
						47	21	-
						49	19	-
						46	21	-
						44	20	-
						47	22	-
12	22.6	32	23	50	No	51	17	-
						51	13	-
						48	22	-
						49	23	-
						55	17	-
						53	20	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
12	22.6	32	30	60	No	30	11	-
						31	11	-
						26	10	-
						33	11	-
						26	6	-
						31	9	-
						30	8	-
						29	11	-
						31	9	-
						28	12	-
						29	9	-
						27	11	-
	22.6	32	30	60	Yes	41	10	-
						42	8	-
						47	12	-
						43	16	-
						48	5	-
						47	10	-
13	22.3	35	23	50	No	51	16	-
						48	21	-
						50	20	-
						48	24	-
						52	23	-
						51	22	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
13	21.9	36	30	50	No	43	18	-
						43	20	-
						43	18	-
						44	17	-
						42	15	-
						45	18	-
						44	16	-
						43	14	-
						42	18	-
						41	13	-
						44	21	-
						44	18	-
	21.9	36	30	50	Yes	49	19	-
						52	23	-
						48	19	-
						50	24	-
						45	17	-
						48	25	-
14	22.1	34	23	50	No	51	16	-
						51	19	-
						49	21	-
						50	19	-
						50	9	-
						51	11	-

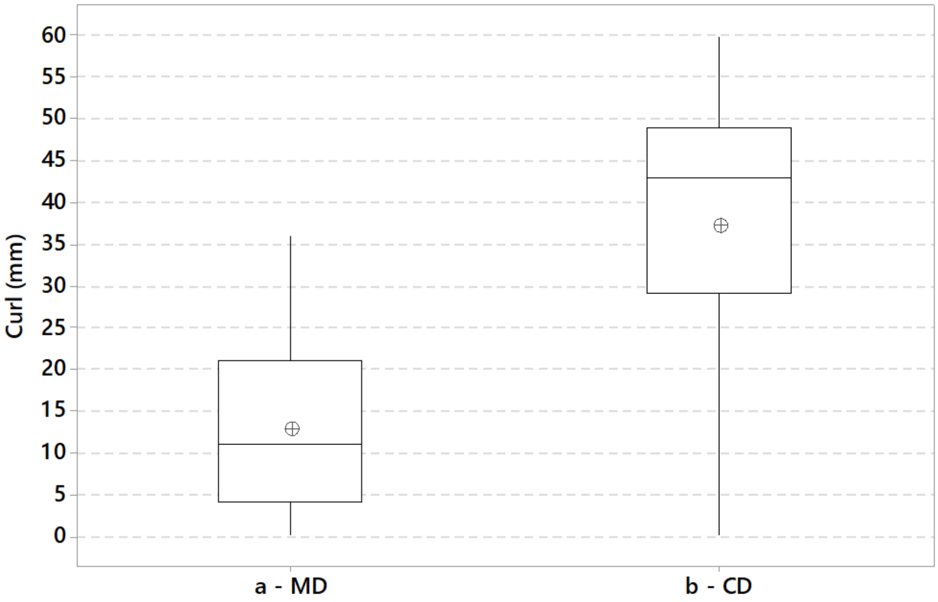
Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
14	22.6	35	30	40	No	40	3	-
						39	3	-
						18	4	-
						39	6	-
						41	4	-
						41	5	-
						41	1	-
						42	8	-
						42	1	-
						41	5	-
						43	5	-
						42	5	-
	22.4	34	30	40	Yes	45	7	-
						47	11	-
						43	8	-
						43	11	-
						42	6	-
						49	6	-
15	21.7	36	23	50	No	53	27	-
						49	22	-
						52	22	-
						48	24	-
						53	20	-
						51	23	-

Table A.2 Detailed measurements for the Pap/mPET/HSL laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
15	22.1	37	30	30	No	37	2	-
						37	2	-
						36	1	-
						40	3	-
						39	0	-
						43	1	-
						35	2	-
						35	5	-
						37	3	-
						40	5	-
						42	4	-
						41	5	-
	22.2	37	30	30	Yes	45	10	-
						45	9	-
						49	14	-
						47	6	-
						46	10	-
						47	12	-

A.2.2 Overall laminate tendency to curl



Note: All the results obtained for the pre-conditioning phase and the test without PE wrapping were aggregated (temperature: 20°C, 23°C, 25°C, 30°C / RH: 30%, 40%, 50%, 60%, 70%).

Figure A.5 Box plot describing the overall Pap/mPET/HSL laminate tendency to curl.

A.3 Alu/Pap/PE laminate

A.3.1 Raw data

Table A.3 Detailed measurements for the Alu/Pap/PE laminate.

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
1	22.0	38	23	50	No	0	7	-
						0	7	-
						0	3	-
						0	2	-
						0	2	-
						0	3	-
	23.0	44	23	50	No	0	4	10
						0	4	4
						0	4	13
						0	6	8
						0	3	17
						0	11	10
	22.0	38	20	70	No	0	2	-
						0	5	-
						0	1	-
						0	2	-
						0	4	-
						0	4	-
						0	1	-
						0	1	-
						0	1	-
						0	1	-
						0	1	-
						0	2	-

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
1	24.3	40	20	70	No	0	2	7
						0	3	10
						0	1	4
						0	1	3
						0	1	5
						0	6	12
						0	2	8
						0	1	3
						0	1	2
						0	1	4
						0	1	4
						0	2	7
	22.0	38	20	70	Yes	0	4	-
						0	3	-
						0	1	-
						0	1	-
						0	1	-
						0	3	-
	23.9	40	20	70	Yes	0	4	9
						0	2	5
						0	1	2
						0	1	1
						0	2	2
						0	3	10
2	24.4	33	23	50	No	0	8	-
						0	6	-
						0	3	-
						0	3	-
						0	5	-
						0	4	-

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
2	25.3	40	23	50	No	0	1	2
						0	1	1
						0	5	0
						0	0	0
						0	1	3
						0	1	1
	24.6	33	20	60	No	0	2	-
						0	4	-
						0	1	-
						0	2	-
						0	0	-
						0	3	-
						0	1	-
						0	2	-
						0	1	-
						0	1	-
						0	2	-
						0	3	-
	25.8	37	20	60	No	0	1	4
						0	1	1
						0	1	4
						0	0	0
						0	1	2
						0	1	5
						0	2	5
						0	1	3
						0	3	9
						0	0	0
						0	4	5
						0	1	12

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
2	24.6	33	20	60	Yes	0	0	-
						0	0	-
						0	2	-
						0	1	-
						0	2	-
						0	1	-
	26.0	38	20	60	Yes	0	4	1
						0	0	0
						0	0	0
						0	0	0
						0	7	1
						0	0	0
3	22.5	35	23	50	No	0	3	-
						0	1	-
						0	2	-
						0	1	-
						0	1	-
						0	2	-
	21.1	50	23	50	No	0	10	19
						0	7	13
						0	1	2
						0	2	6
						0	5	8
						0	3	15

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
3	22.5	35	20	50	No	0	1	-
						0	0	-
						0	1	-
						0	1	-
						0	3	-
						0	5	-
						0	4	-
						0	2	-
						0	2	-
						0	4	-
						0	2	-
	22.1	48	20	50	No	0	4	9
						0	5	14
						0	3	6
						0	4	9
						0	4	14
						0	7	14
						0	4	12
						0	3	6
						0	3	9
						0	3	6
						0	11	20
						0	6	15
	22.5	35	20	50	Yes	0	1	-
						0	2	-
						0	2	-
						0	0	-
						0	1	-
						0	2	-

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
3	21.7	49	20	50	Yes	0	6	14
						0	3	7
						0	2	9
						0	2	2
						0	3	11
						0	4	3
4	21.2	40	23	50	No	0	5	-
						0	9	-
						0	1	-
						0	5	-
						0	4	-
						0	1	-
	26.2	34	23	50	No	0	1	1
						0	2	7
						0	3	15
						0	3	9
						0	2	15
						0	2	10
	21.4	38	20	40	No	0	8	-
						0	6	-
						0	2	-
						0	3	-
						0	10	-
						0	5	-
						0	5	-
						0	3	-
						0	1	-
						0	0	-
						0	3	-
						0	2	-

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
4	26.8	32	20	40	No	0	1	5
						0	1	5
						0	1	7
						0	2	8
						0	2	8
						0	3	4
						0	1	3
						0	1	6
						0	1	8
						0	2	8
						0	2	8
						0	4	3
	21.5	37	20	40	Yes	0	5	-
						0	2	-
						0	1	-
						0	2	-
						0	1	-
						0	3	-
	26.8	32	20	40	Yes	0	0	0
						0	0	0
						0	0	0
						0	0	0
						0	0	0
						0	0	0
5	23.4	35	23	50	No	0	4	-
						0	6	-
						0	0	-
						0	0	-
						0	1	-
						0	2	-

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
5	24.1	43	23	50	No	0	1	3
						0	1	6
						0	3	8
						0	5	10
						0	4	10
						0	6	7
	24.2	35	20	30	No	0	5	-
						0	4	-
						0	2	-
						0	1	-
						0	3	-
						0	4	-
						0	4	-
						0	5	-
						0	1	-
						0	1	-
						0	2	-
						0	3	-
	26	39	20	30	No	0	1	5
						0	1	5
						0	1	3
						0	4	9
						0	2	10
						0	3	7
						0	2	3
						0	0	0
						0	0	0
						0	3	6
						0	1	7
						0	4	11

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
5	24.3	35	20	30	Yes	0	3	-
						0	2	-
						0	0	-
						0	3	-
						0	3	-
						0	4	-
	25.3	40	20	30	Yes	0	2	9
						0	1	6
						0	1	6
						0	1	2
						0	1	3
						0	1	0
6	24.2	34	23	50	No	0	3	12
						0	5	11
						0	1	4
						0	2	7
						0	10	18
						0	7	17
	24.3	34	25	70	No	0	1	4
						0	2	9
						0	0	0
						0	1	3
						0	1	5
						0	1	6
						0	1	4
						0	1	3
						0	1	2
						0	0	0
						0	1	4
						0	1	6

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
6	24.3	34	25	70	Yes	0	3	10
						0	2	0
						0	0	0
						0	1	1
						0	3	8
						0	1	5
7	22.1	40	23	50	No	0	9	15
						0	7	16
						0	6	11
						0	2	10
						0	7	17
						0	5	14
	22.8	37	25	60	No	0	3	10
						0	1	6
						0	2	4
						0	2	7
						0	1	6
						0	5	14
						0	5	13
						0	6	15
						0	2	7
						0	2	8
						0	3	12
						0	3	12
	23.1	36	25	60	Yes	0	3	11
						0	3	10
						0	1	3
						0	1	4
						0	4	7
						0	1	3

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
8	25.2	31	23	50	No	0	1	3
						0	3	9
						0	1	3
						0	2	4
						0	4	13
						0	3	10
	25.3	31	25	50	No	0	1	2
						0	3	11
						0	1	3
						0	1	4
						0	1	6
						0	3	9
						0	1	3
						0	2	7
						0	1	1
						0	1	5
						0	3	9
						0	4	11
	25.3	31	25	50	Yes	0	1	4
						0	1	5
						0	1	2
						0	0	0
						0	1	4
						0	1	1
9	23.1	38	23	50	No	0	7	14
						0	3	5
						0	3	9
						0	2	5
						0	4	13
						0	1	13

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
9	24.4	35	25	40	No	0	5	12
						0	4	13
						0	8	15
						0	5	12
						0	5	19
						0	6	16
						0	4	14
						0	4	12
						0	1	6
						0	2	7
						0	2	7
						0	2	10
	24.4	35	25	40	Yes	0	2	7
						0	1	4
						0	3	8
						0	1	1
						0	1	8
						0	1	2
10	21.0	45	23	50	No	0	8	14
						0	8	15
						0	5	13
						0	4	12
						0	12	14
						0	6	16

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
10	22.1	42	25	30	No	0	3	10
						0	5	13
						0	2	8
						0	1	5
						0	1	3
						0	2	3
						0	10	19
						0	7	11
						0	8	15
						0	8	17
						0	8	18
						0	10	18
	22.0	43	25	30	Yes	0	1	0
						0	1	4
						0	0	0
						0	0	0
						0	1	6
						0	3	7
11	22.4	37	23	50	No	0	2	4
						0	1	7
						0	1	2
						0	2	8
						0	7	17
						0	2	7

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
11	22.9	36	30	70	No	0	2	4
						0	5	11
						0	2	8
						0	2	7
						0	1	8
						0	2	6
						0	5	14
						0	2	0
						0	1	4
						0	1	0
						0	4	11
						0	1	4
	23.2	36	30	70	Yes	0	0	0
						0	2	10
						0	1	2
						0	1	6
						0	1	5
						0	1	0
12	24.0	32	23	50	No	0	3	12
						0	2	8
						0	1	6
						0	1	4
						0	3	11
						0	5	14

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
12	24.5	28	30	60	No	0	1	4
						0	2	6
						0	1	3
						0	1	2
						0	1	3
						0	2	3
						0	1	2
						0	1	5
						0	1	1
						0	1	2
						0	1	3
						0	1	6
	24.5	28	30	60	Yes	0	1	3
						0	1	3
						0	2	3
						0	1	3
						0	2	2
						0	1	6
13	22.4	34	23	50	No	0	3	10
						0	2	5
						0	1	5
						0	1	5
						0	4	13
						0	3	11

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
13	23	33	30	50	No	0	2	5
						0	2	5
						0	2	5
						0	1	1
						0	3	7
						0	3	10
						0	2	6
						0	2	9
						0	1	2
						0	1	2
						0	2	2
						0	1	2
	23.0	33	30	50	Yes	0	2	7
						0	1	3
						0	1	2
						0	1	3
						0	1	6
						0	1	5
14	26.4	24	23	50	No	0	1	5
						0	1	5
						0	1	0
						0	1	3
						0	2	7
						0	3	11

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	**T_r (°C)**	**RH_r (%)**	**T_{cc} (°C)**	**RH_{cc} (%)**	**PE wrapping**	**b (mm)**	**a (mm)**	**a* (mm)**
14	26.4	24	30	40	No	0	1	6
						0	1	2
						0	1	3
						0	1	2
						0	1	6
						0	1	2
						0	3	7
						0	3	10
						0	1	6
						0	2	4
						0	3	9
						0	3	10
	26.5	23	30	40	Yes	0	1	3
						0	2	3
						0	1	1
						0	1	0
						0	1	4
						0	1	4
15	24	32	23	50	No	0	3	10
						0	1	6
						0	1	3
						0	2	6
						0	5	13
						0	4	10

Table A.3 Detailed measurements for the Alu/Pap/PE laminate (continued).

Experiment number	T_r (°C)	RH_r (%)	T_{cc} (°C)	RH_{cc} (%)	PE wrapping	b (mm)	a (mm)	a* (mm)
15	24.8	31	30	30	No	0	2	8
						0	3	10
						0	1	4
						0	3	9
						0	3	9
						0	2	8
						0	6	15
						0	5	15
						0	1	3
						0	3	10
						0	6	14
						0	7	17
	25.0	30	30	30	Yes	0	3	9
						0	2	10
						0	1	3
						0	1	2
						0	4	8
						0	2	5

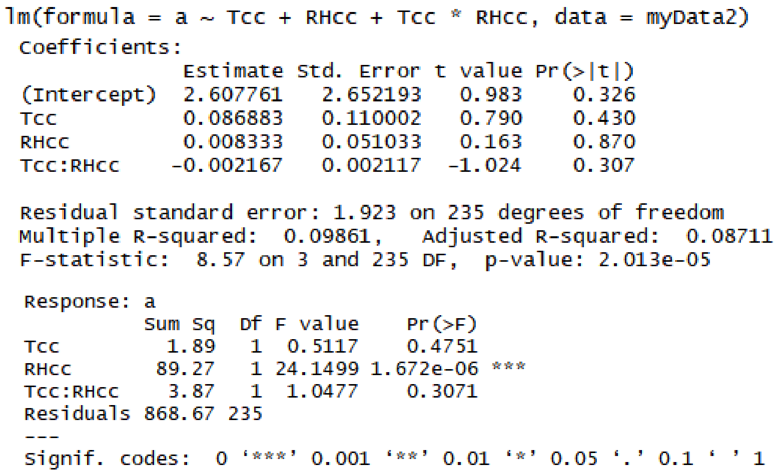
A.3.2 Overall laminate tendency to curl

Note: All the results obtained for the pre-conditioning phase and the test without PE wrapping were aggregated (temperature: 20°C, 23°C, 25°C, 30°C / RH: 30%, 40%, 50%, 60%, 70%).

Figure A.6 Box plot describing the overall Alu/Pap/PE laminate tendency to curl.

A.3.3 Predict curl as a function of climate chamber and room conditions

A.3.3.1 Case of the a parameter



```
lm(formula = a ~ Tcc + RHcc + Tcc * RHcc, data = myData2)
 Coefficients:
              Estimate Std. Error t value Pr(>|t|)
 (Intercept)  2.607761   2.652193   0.983    0.326
 Tcc          0.086883   0.110002   0.790    0.430
 RHcc         0.008333   0.051033   0.163    0.870
 Tcc:RHcc    -0.002167   0.002117  -1.024    0.307

 Residual standard error: 1.923 on 235 degrees of freedom
 Multiple R-squared:  0.09861,   Adjusted R-squared:  0.08711
 F-statistic:  8.57 on 3 and 235 DF,  p-value: 2.013e-05

 Response: a
           Sum Sq  Df F value    Pr(>F)
 Tcc         1.89   1  0.5117    0.4751
 RHcc       89.27   1 24.1499 1.672e-06 ***
 Tcc:RHcc    3.87   1  1.0477    0.3071
 Residuals 868.67 235
 ---
 Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1
```

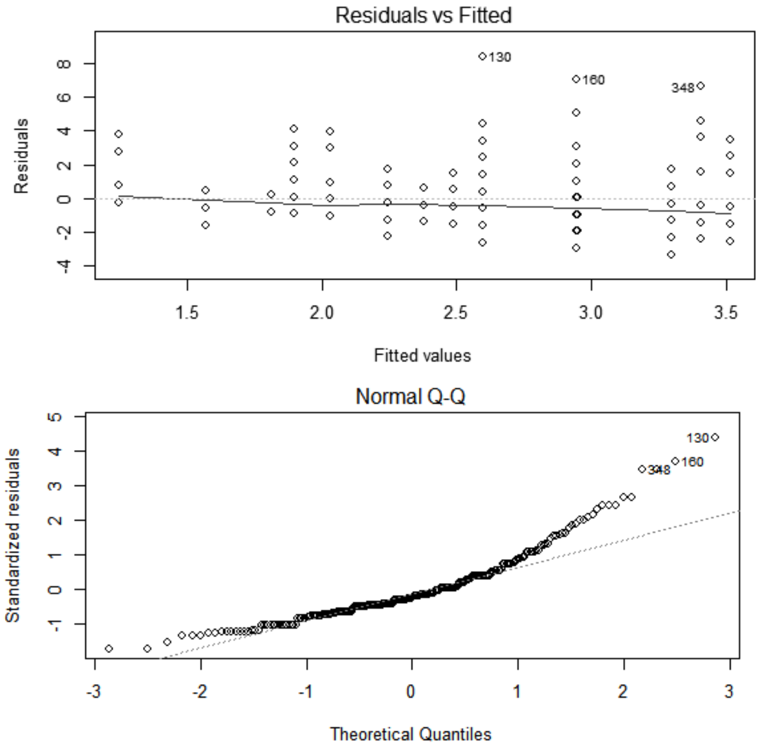
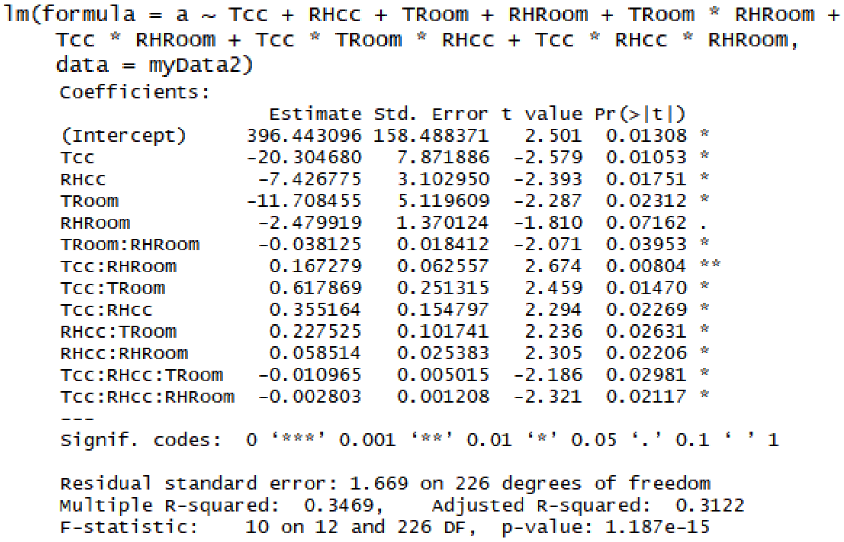


Figure A.7 Linear regression to predict the a parameter as a function of climate chamber conditions ($A_i = c_0 + c_1 * T_{cci} + c_2 * RH_{cci} + c_3 * T_{cci} * RH_{cci} + E_i \ for \ i = 1{:}239$).



```
lm(formula = a ~ Tcc + RHcc + TRoom + RHRoom + TRoom * RHRoom +
    Tcc * RHRoom + Tcc * TRoom * RHcc + Tcc * RHcc * RHRoom,
    data = myData2)
    Coefficients:
                     Estimate Std. Error t value Pr(>|t|)
    (Intercept)     396.443096 158.488371   2.501  0.01308 *
    Tcc             -20.304680   7.871886  -2.579  0.01053 *
    RHcc             -7.426775   3.102950  -2.393  0.01751 *
    TRoom           -11.708455   5.119609  -2.287  0.02312 *
    RHRoom           -2.479919   1.370124  -1.810  0.07162 .
    TRoom:RHRoom     -0.038125   0.018412  -2.071  0.03953 *
    Tcc:RHRoom        0.167279   0.062557   2.674  0.00804 **
    Tcc:TRoom         0.617869   0.251315   2.459  0.01470 *
    Tcc:RHcc          0.355164   0.154797   2.294  0.02269 *
    RHcc:TRoom        0.227525   0.101741   2.236  0.02631 *
    RHcc:RHRoom       0.058514   0.025383   2.305  0.02206 *
    Tcc:RHcc:TRoom   -0.010965   0.005015  -2.186  0.02981 *
    Tcc:RHcc:RHRoom  -0.002803   0.001208  -2.321  0.02117 *
    ---
    Signif. codes:  0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1 ‘ ’ 1

    Residual standard error: 1.669 on 226 degrees of freedom
    Multiple R-squared:  0.3469,    Adjusted R-squared:  0.3122
    F-statistic:    10 on 12 and 226 DF,  p-value: 1.187e-15
```

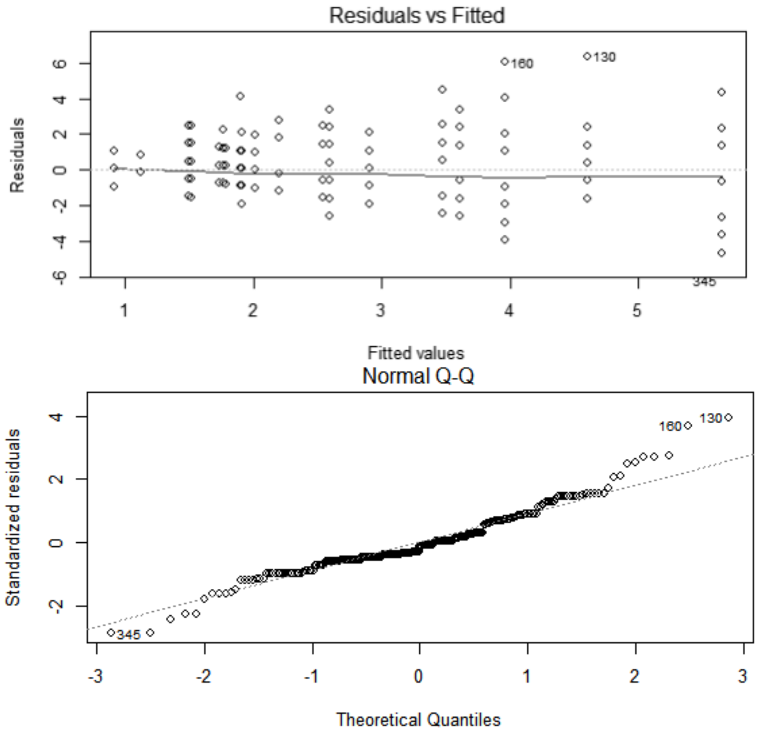
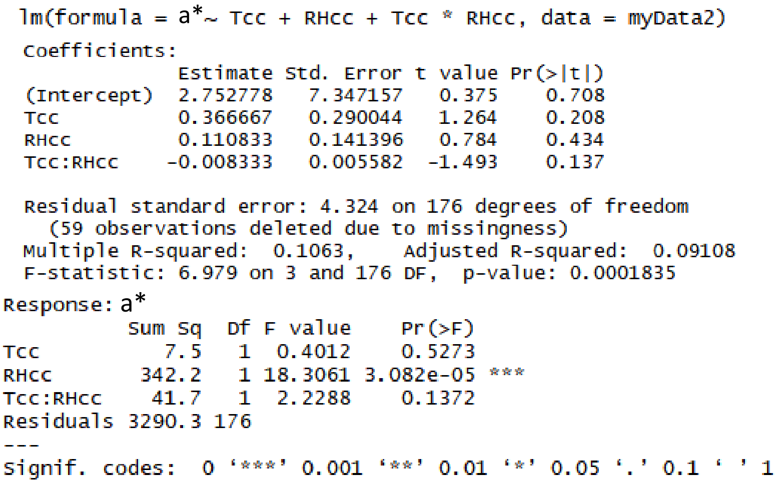


Figure A.8 Linear regression to predict the a parameter as a function of climate chamber and room conditions ($A_i = c_{10} + c_{11} * T_{cci} + c_{12} * RH_{cci} + c_{13} * T_{ri} + c_{14} * RH_{ri} + c_{15} * T_{ri} * RH_{ri} + c_{16} * T_{cci} * RH_{ri} + c_{17} * T_{cci} * T_{ri} * RH_{cci} + b_{24} * T_{cci} * RH_{cci} * RH_{ri} + E_i \ for \ i = 1{:}239$)

A.3.3.2 Case of the a parameter*



```
lm(formula = a*~ Tcc + RHcc + Tcc * RHcc, data = myData2)
Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  2.752778   7.347157   0.375    0.708
Tcc          0.366667   0.290044   1.264    0.208
RHcc         0.110833   0.141396   0.784    0.434
Tcc:RHcc    -0.008333   0.005582  -1.493    0.137

Residual standard error: 4.324 on 176 degrees of freedom
  (59 observations deleted due to missingness)
Multiple R-squared:  0.1063,    Adjusted R-squared:  0.09108
F-statistic: 6.979 on 3 and 176 DF,  p-value: 0.0001835
Response: a*
          Sum Sq  Df F value    Pr(>F)
Tcc          7.5   1  0.4012    0.5273
RHcc       342.2   1 18.3061 3.082e-05 ***
Tcc:RHcc    41.7   1  2.2288    0.1372
Residuals 3290.3 176
---
Signif. codes:  0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1 ‘ ’ 1
```

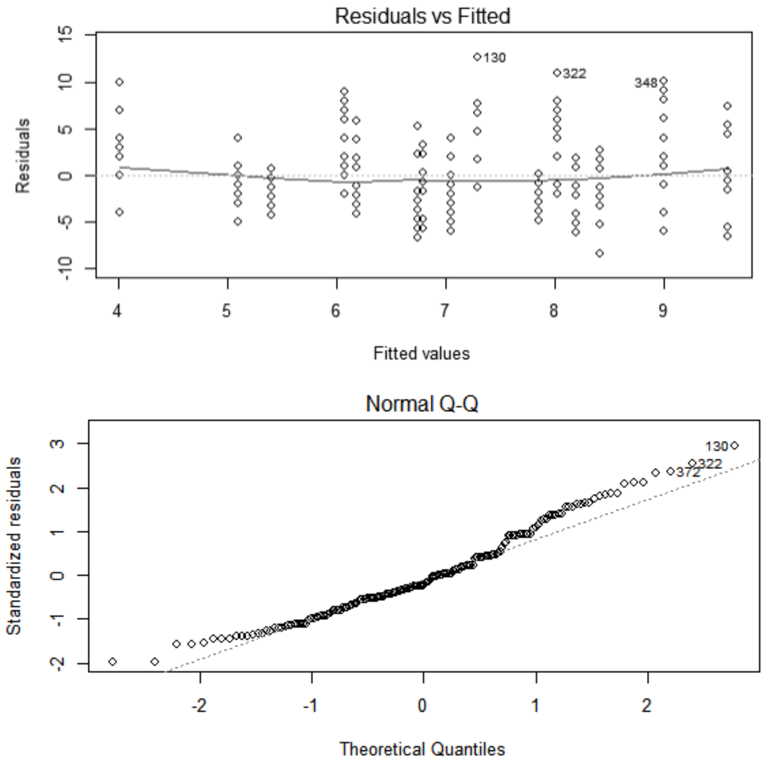
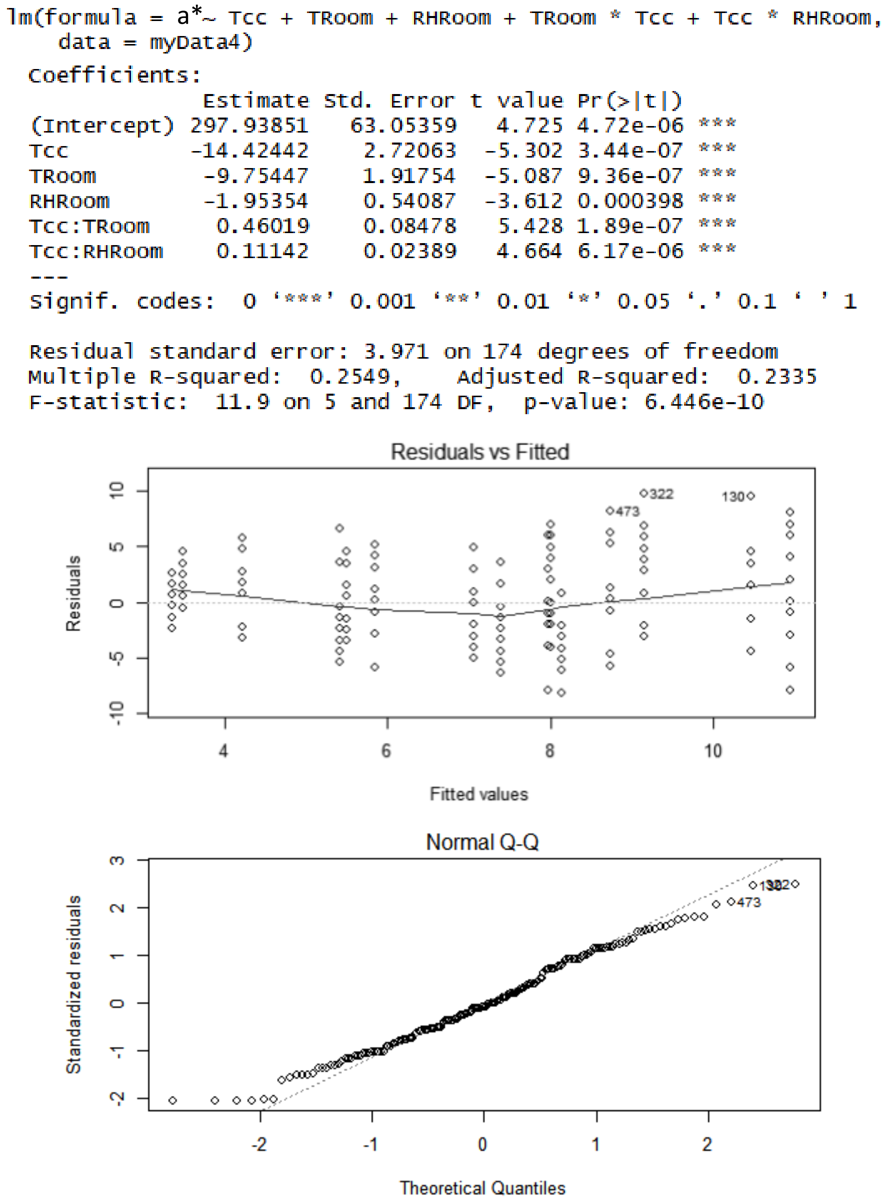


Figure A.9 Linear regression to predict the a* parameter as a function of climate chamber conditions ($A^*_i = d_0 + d_1 * T_{cci} + d_2 * RH_{cci} + d_3 * T_{cci} * RH_{cci} + E_i \;\; for \; i = 1{:}\,180$).



```
lm(formula = a*~ Tcc + TRoom + RHRoom + TRoom * Tcc + Tcc * RHRoom,
    data = myData4)
 Coefficients:
               Estimate Std. Error t value Pr(>|t|)    
 (Intercept) 297.93851   63.05359   4.725 4.72e-06 ***
 Tcc         -14.42442    2.72063  -5.302 3.44e-07 ***
 TRoom        -9.75447    1.91754  -5.087 9.36e-07 ***
 RHRoom       -1.95354    0.54087  -3.612 0.000398 ***
 Tcc:TRoom     0.46019    0.08478   5.428 1.89e-07 ***
 Tcc:RHRoom    0.11142    0.02389   4.664 6.17e-06 ***
 ---
 Signif. codes:  0 ‘***’ 0.001 ‘**’ 0.01 ‘*’ 0.05 ‘.’ 0.1 ‘ ’ 1

 Residual standard error: 3.971 on 174 degrees of freedom
 Multiple R-squared:  0.2549,    Adjusted R-squared:  0.2335 
 F-statistic:  11.9 on 5 and 174 DF,  p-value: 6.446e-10
```

Figure A.10 Linear regression to predict the a* parameter as a function of climate chamber and room conditions ($A^*_i = d_{10} + d_{11} * T_{cci} + d_{12} * T_{ri} + b_{13} * RH_{ri} + d_{14} * T_{cci} * T_{ri} + d_{15} * T_{cci} * RH_{ri} + E_i \;\; for\ i = 1{:}180$).