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Recyclability of Release Liners

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ABSTRACT

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REISINGER MINNA: Recyclability of Release Liners

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Recyclability is one of the most important sustainability goals in many industry fields nowadays. Paper recycling is an old and well-known process in Europe and there are set standards as to which paper based products are classified as recyclable in a standard recycling paper mill. Siliconized paper based products, release liners, are defined to the not recyclable paper products.

The main target in this study was to test the recyclability of release liners to see whether the industrial waste can be recycled. It was assumed that some release liners can be recyclable in a standard paper recycling mill when taking into account the used base paper. Release liners are coated with silicone on specialized base papers depending on the end application. The main used base papers for siliconization are glassines and precoated kraft papers. The silicone as release coating itself does not automatically make the paper product unfit for recycling.

The most common release liner products were tested in the laboratory based on the Cepi recyclability laboratory method (version 2) on paper and board based products. The method is created to test the recyclability on a laboratory scale based on the process of standard paper recycling mill.

Based on the recyclability tests carried out on release liners it can be said that there is a variety of factors, which affect the recyclability level. It was found that the recyclability level on glassine samples differs between the base paper suppliers. Release liners on clay-coated kraft (CCK) papers and machine finished (MF) papers are recyclable. Machine glazed (MG) liners are recyclable when precoated. PE-coated kraft papers are limited recyclable which means that after some minor changes in process parameters or product improvement they could be recyclable on standard paper recycling mill.

Silicone release coated papers do not need to be automatically categorized as unrecyclable products. The recyclability level of release liner depends mainly on the used base paper. An interesting for further research would be, what happens to the silicone in the recycling process and which process parameters of base paper supplier or base paper properties affect to the difference recyclability level.

Key words: recyclability, release liner, silicone, Cepi

TIIVISTELMÄ

Tampereen ammattikorkeakoulu Risk Management and Circular Economy

REISINGER MINNA: Tarrantaustapapereiden kierrätettävyys

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Teollisuuden aloilla kestävän kehityksen yksi tärkeimmistä tavoitteista on tuotteiden kierrätettävyys. Paperin kierrätys on perinteinen prosessi Euroopassa, ja paperipohjaiset tuotteet on standardoitu joko kierrätettäviin tai ei-kierrätettäviin tuotteisiin. Silikonoidut paperipohjaiset tuotteet, tarrantaustapaperit on kategorisoitu ei-kierrätettäviin tuotteisiin.

Tämän työn päätavoite oli testata tarrantaustapapereiden kierrätettävyyttä. Tarkoitus oli tutkia, voidaanko tuotantoprosessista tulevaa tuotantojätettä kierrättää perinteisessä paperin kierrätysprosessissa. Oletuksena oli, että jotkut tarrantaustapaperituotteet ovat kierrätettävissä riippuen käytetystä käytetään silikonointiin Pohjapapereina tarkoitettuja pohjapaperista. erikoispapereita, ja eniten käytetyt pohjapaperit silikonoinnissa ovat erilaiset esipäällystetyt voimapaperit ja pintakäsitellyt, superkalanteroidut glassiinipaperit. Oletuksena oli, että silikonointi ei tee tarrantaustapapereista automaattisesti eikierrätettäviä tuotteita. Tässä opinnäytetyössä yleisimpien tarrantaustapapereiden kierrätettävyyttä tutkittiin laboratoriossa pohjautuen Cepin julkaisemaan mittausmenetelmään (versio 2), kierrätettävyystestiin laboratoriassa paperi- ja kartonkipohjaisille tuotteille. Tämä mittausmenetelmä luotu testaamaan paperipohjaisten tuotteiden kierrätettävyyttä on standardoituun laboratoriomittakaavassa pohjautuen paperin kierrätystuotantoprosessiin.

Tehtyihin kierrätettävyystesteihin perustuen voidaan sanoa, että erilaiset tekijät vaikuttavat tarrantaustapapereiden kierrätettävyyteen. Silikonoitujen glassiinipapereiden kierrätettävyysaste riippuu pohjapaperin toimittajasta. Kun tarrantaustapohjapaperina on käytetty esimerkiksi kaoliiniesipäällystettyä voimapaperia, voidaan sanoa, että tuote on kierrätettävissä. Myös muut testatut, silikonoidut ja pintaliimoilla esipäällystetyt paperit ovat kierrätettävissä. Kun pohjapaperi on päällystetty polyeteenillä, kierrätettävyysaste on rajallinen. Se tarkoittaa, että pienillä tuotantoparametrien muutoksilla kierrätysprosessissa tai tuotteen tuotekehityksessä tehtävät parannukset voivat edistää tuotteen kierrätettävyysastetta.

Silikonoidut tarrantaustapaperit ovat tietyillä pohjapapereilla kierrätettävissä, kun kierrätettävyyttä on testattu Cepin mittausmenetelmään pohjautuen. Jatkossa voitaisiin tutkia, mitä silikonille kierrätysprosessissa tapahtuu ja mitkä tuotantoparametrit paperinvalmistuksessa ja mitkä paperin ominaisuudet vaikuttavat kierrätettävyysasteeseen.

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ABBREVIATIONS

Сері	Confederation of European paper industry	
EN643	Standard list of European paper and board products for	
	recycling	
Release Liner	Silicone release coated substrate	
g/m²	Unit of basis weight	
PVA	Polyvinyl alkohol	
Reject	Substance, which does not pass the screening	
Accept	Substance, which passes the screening	
Glassine	Supercalandered kraft paper	
CCK	Clay-coated kraft paper	
PE	Polyethylene	
PCK	PE-coated kraft paper	
MF	Machine finished kraft paper	
MG	Machine glazed kraft paper	
XRFA	X-ray fluoroscence analysis	

1 INTRODUCTION

Recyclability is one of the most important sustainability goals in many industry fields. Standard paper recycling is already well-known and largely used method in Europe but when the paper is converted and different kind of chemical additives used, the standard recycling process can be disturbed. For standard paper recycling mill it is important to get high yield of the recycled material with fluent processability without unwanted impurities. Good recyclability level means that there is coming as less reject as possible when the product is recycled in standard process and when the recycled paper is produced there is not too many visual impurities or stickies on the paper.

Cepi, the confederation of European paper industry, has created a laboratory method to test the recyclability of paper and board products for standard paper recycling mill. The laboratory method is enabling to evaluate the most important process parameters for recycling process, reject amount, visual impurities and stickies. The new version of Cepi laboratory method has been published in summer 2022 but some parts of it are still pending.

Cepi has created EN643 standard, which is listing the standard European paper and board products for recycling. It is a standard to advice paper stakeholders about the recyclability of different paper and board qualities. Classification is done by different qualities to five groups. Groups 1 to 4 in EN643 standard are recyclable paper grades in standard recycling mill and group number 5 paper grades, which are recyclable in specialized recycling mill. (Ervasti, 2010, 14-15) To the group 5 belongs for example release liners which are used for self-adhesive labels and other laminated products.

Release liners are mostly paper based products, which are converted by siliconization on-line or off-line processes. Release liner is used in laminated products to protect the adhesive layer of the end-product. Release liner is one part of the end-product and when the end product is used, the release liner is peeled and thrown away. For release liner converters it is important to reach the sustainability targets and be able to recycle products they are producing. EN643 standard list of paper and board products qualifies release liners to the group of paper grades, which are difficult to recycle because these products are siliconized. In this study the main target is to test the recyclability of different release liner products and to be able to do classification on recyclability level based on used base paper. Silicone as a release coating is insoluble in water and therefore may cause issues on the recycling process. It is assumed that the recyclability can vary between the release liner applications. For example liners used in label and tape applications are produced on high dense base paper, glassine. The presume, is that the high dense papers are more difficult to repulp in standard recycling process than for example liners on clay coated kraft papers used in hygiene and envelope applications.

In this study the most common release liner products are tested according to the new version of Cepi laboratory recyclability method. Tested liners are divided to the groups based on used base papers. To be able to have a full picture of different release liners, each base paper group was further divided to three different basis weight ranges, low, medium and high. Release liners can be one or two side siliconized and the silicone amount varies based on the application. In this study also the effect of one or two sidedness siliconization as well the relevant amount of silicone on tested product is considered.

Cepi recyclability laboratory method is a good method for different paper producers and its stakeholders to test the recyclability of paper products based on the process of standard recycling paper mill. In Cepi method is at a time tested the recyclability of one product and it needs to be observed that in recycling mill the recycled waste paper contains various grades and the amount of release liners from total paper waste would be minimal. The main amount of paper based release liner waste is coming from industrial side from the release liner producers and its converters. The household waste of release liners is not considered in this study. In this report is described first the process of standard paper recycling mill and later on the used Cepi recycling laboratory method. Beside the recycling processes, the release liner and most common paper based liner products are introduced. Results of recyclability tests done on release liner products and conclusions are included to the end part of the report.

2 PAPER RECYCLING PROCESS

Paper recycling is an old and well-known process where good quality paper of recycled fibers can be produced. The principle of paper recycling process is to repulp the paper, remove the contaminants and bleach the fibers. Recycled papers are mainly used for producing newspaper, tissue paper or carton board. (Ek, Gellerstedt, Henriksson, 2009, 392) In figure 1. can be seen the amount of used paper for recycling in Europe from the year 1991 to 2021. First ten years the utilisation was growing linearly and later on the amount of used paper for recycling was stabilized. From 2020 the use of paper has grown by 5.3 % to its highest level, over 50 million. (Cepi, 2021)

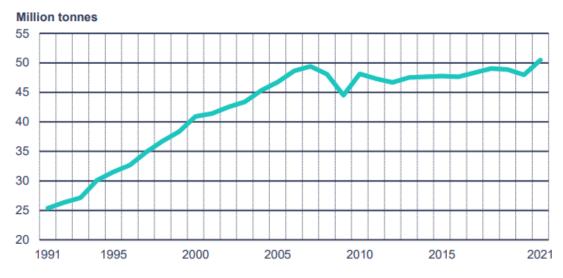


FIGURE 1. Usage of paper for recycling in Europe. (Cepi, 2021)

In the Europe around half of the paper material for recycling comes from the industrial side, 40 % from the households and rest from the offices. (Bajbai, 2013, 274) From the circular economy point of view it is important to reduce the amount of waste and the amount of raw material use. Paper recycling process decreases not only the use of wood material but also the used energy and amount of used water. (Bajbai, 2013, 272)

Recycling of paper is reducing the need of virgin fibers and therefore trees are not needed to be cut that much from the forests. Recycling process requires less energy than a process to make paper from wood fibers, which is good for the environment. To be able to produce more recycled paper instead of virgin paper, it is needed to increase the amount of available paper and board based grades for recycling. (Bajbai, 2013, 273)

The process of recycled paper depends on the end use application as well as the available raw material. For the process it is important to minimize and get rid of unwanted substances like for example plastics, printing inks and adhesives. Adhesives may cause in the papermaking process of recycled fiber stickies and reduce the runnability of paper machine. (Ek, Gellerstedt, Henriksson, 2009, 400-402)

2.1 Repulping

The main target of repulping is to dissolve the paper to the detached fibers and separate the inks or coating colors from the surface of the paper. Repulping is normally done in high or low consistency process. Untreated paper is very hydrophilic and easy to suspense in water but different kind of treatments can increase the resistant to water. For example wet-strength additives in tissue papers are reducing repulpability but the repulping time can be increased to reach required level. Beside the repulping time, the level of repulping can be affected by the temperature and pH of the suspension. The standard repulping time is 20 minutes, which is enough for breaking up the hydrogen bonds between the fibres. (Ek, Gellerstedt, Henriksson, 2009, 415)

2.2 Screening

The main purpose of screening in the recycling process is to minimise the amount of unwanted substances and contaminants. The target is that the recycled pulp for papermaking is clean enough and contains as much recycled fibers as possible and therefore the size of screening slots needs to be suitable for fibers going through and improve the yield of recycling process. (Ek, Gellerstedt, Henriksson, 2009, 416-417)

The repulped suspension is screened in the standard process in two sections. First screening section is called coarse screening where the holes or slots in the screeners are bigger than in second screening section, called fine screening. In the first section the main target is to remove the biggest unwanted particles, like for example plastic and metal. (Ek, Gellerstedt, Henriksson, 2009, 416-417)

Fractionating stage between in these two screening sections is enabling the separation of short and long fibers. Beside the fibers, also the fines and fillers are able to be separated in fractionating stage and processed further. (Ek, Gellerstedt, Henriksson, 2009, 416-417)

2.3 Deinking

The waste paper contains all kind of recyclable paper grades and mostly includes papers, which are printed. The most ink is already removed from the paper surface in the repulping stage but in the deinking process the target is to get rid of remaining ink particles. Deinking can be done by flotation or washing process. (Ek, Gellerstedt, Henriksson, 2009, 408-410)

When the deinking is done in the flotation process, the ink residuals from the fibers are first released by using chemical additives, then agglomerated with specialized chemical and after that, flotated with air bubbles up to be able to collect them. In the washing process, the ink residuals are removed from the fibers by chemical additives and afterwards screened by using the advantage of size difference between fibers and ink particles. (Ek, Gellerstedt, Henriksson, 2009, 408-410)

2.4 Bleaching

After deinking process the repulped paper fibers are bleached to the required level of whiteness. The brightness of recycled paper can already be reached when the ink is removed but when the high brightness is needed, also chemical bleaching treatment is required. It depends on the recycling process whether bleaching is needed. (Bajbai, 2013, 155)

The most used bleaching chemical in recycling process is nowadays chlorinefree hydrogen peroxide. It is more environmental friendly bleaching chemical than chlorine. Beside the used chemical and dosage of it, to the bleaching level can be affected by reaction time, temperature and stock consistency. The higher level of brightness on recycled paper enables its wider end-use possibilities. (Bajbai, 2013, 157)

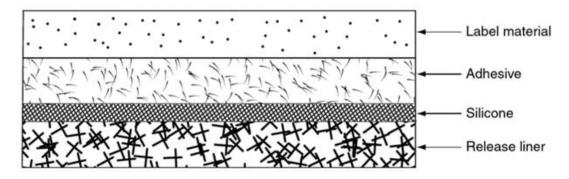
2.5 Dispersing

When the repulped suspension is cleaned via previous mentioned stages it can be optionally refined or dispersed to be able to improve the further fiber bonding in papermaking process. (Bajbai, 2013, 182)

The main target in paper recycling process of the refining is to refine recycled fibers so that the mechanical properties of the produced paper can be stabilized. Repulped suspension can include the fibers, which are refined on different levels on the virgin papermaking process and therefore post-refining in recycling process is required. Refining process needs a lot of energy and may reduce some paper properties and therefore more gentle process, dispersing can be used as well. Refining or dispersing is normally the last recycling process step before the pulp will be storagde or recycled paper made out of it. Refining and dispersing can be done on low- or high consistency depending on the required properties of repulp. (Bajbai, 2013, 181)

3 RELEASE LINER

Release liner is a silicone-coated substrate designed for pressure-sensitive adhesive solutions such as labels, envelopes, graphic arts and hygiene products. Release liner is used in the laminated product where its function is to protect the adhesive layer of the product and stick to it but same time to be able to peel off or release easily. (Meinander, 2000, 116-117) In picture 1. can be seen the separate layers of the laminated product.



PCTURE 1. An example of separate layers in laminated product. (Kirwan, 2013, 132)

Release liner consist of the base material and a uniform thin layer of silicone coating. Used base materials are specialized for siliconization because a good hold out of coating is needed to perform needed release behaviour. Base materials can be polymer films or special base papers with or without pre-coat. In this thesis only the paper based materials are considered.

3.1 Release liner base papers

The main used base papers for release liners are special surface treated and precoated kraft papers. The grammage of base papers varies between 35 g/m² and 135 g/m² depending on the end use application. Low grammage papers are used for liners for label, envelope and hygiene applications and higher grammages are used for tape and graphic art applications. For siliconization, it is important that the surface of raw paper is closed and smooth, that silicone stays on the surface of coated material and does not absorb into the paper.

There are different ways to pre-treat the surface of the base paper. Base papers can be surface sized or precoated on the paper machine, supercalandered, or precoated with different applications offline.

3.1.1 Glassine

Glassine is used as a base paper for label and tape release liners and it is one of the most used base papers for siliconization. Glassine is supercalandered to reach high smoothness, to close the surface of the paper, to minimize the thickness variation and to have slight transparency for the end-use processes. These properties are important for good siliconizability.

Glassine papers are having normally basis weight from low grammage 35 g/m² to high grammages 120 g/m². Glassine can be bleached or unbleached and is available in different colours, like for example in white, havanna, yellow and brown. Depending on the end applications, glassine base papers can be one or two side siliconized with different silicone amounts. In the picture 2. is shown an example of label product, which is a typical end-use for glassine based release liners.



PICTURE 2. Low grammage glassine papers are used in label application. (Mondigroup, n.d.)

In Europe the biggest glassine suppliers are producing the paper without wet strength additives which is good for recyclability. In the paper making process of glassine paper, the base is surface sized one or two sided. The most used surface sizing chemicals are starch or polyvinyl alkohol (PVA). Glassine is a high dense paper due the supercalandering and the high density may influence to the recyclability. There are different kind of supercalanderes with diverse amount and material of rollers. Also the level of calandering can vary between the suppliers. It is possible to calander the paper more one sided or to calander both sides. In the table 1. can be seen an example of typical densities of different glassine base papers from two different European paper suppliers. It can be seen that the density of glassine papers between two suppliers is not varying notable.

TABLE 1. Typical densities of glassine base papers from two different base paper suppliers.

	kg/m³		
Supplier	Low g/m ²	Medium g/m²	High g/m²
Х	1132	1189	1153
Y	1132	1173	1188

As an comparison, in table 2. can be seen the typical densities of clay-coated kraft papers in different basis weight groups. When compared to glassine papers, it can be said the density of clay-coated kraft paper is under 1000 kg/m³ and glassines have density around 200 kg/m³ more than CCK base papers.

TABLE 2. Example of densities on CCK-base papers.

	kg/m³		
	Low g/m²	Medium g/m²	High g/m²
ССК	911	951	971

When the density of low grammage of machine glazed and – finished papers is considered, it can be said that the density is on the same level or even lower than CCK-papers. In table 3. is shown the typical density of both base paper grades.

TABLE 3. Example of density of MG and MF- base papers

	kg/m³	
	Low	
	g/m²	
MG	754	
MF	972	

Glassines are produced for one side or for two side siliconization and standard silicone amount varies from low amount on low grades to higher silicone amount on higher grades. For example for the low grammage glassines the standard silicone amount is around 1 g/m² and for the higher grammages the silicone amount is around 2 g/m². When the glassine is siliconized two sided, it has mostly on both sides the same amount of silicone but often other side has controlled release behaviour.

3.1.2 Clay-coated kraft paper

Clay-coated kraft paper (CCK) is the second most used base paper for siliconization. It is used for liner applications in envelopes, hygiene products and graphic arts. In the picture 3. can be seen an example of envelope product, which is typical application for CCK based liners. The base paper grammages of clay coated kraft papers are from 40 g/m² to 135 g/m².



PICTURE 3. Clay coated kraft papers used typically in envelope applications. (Mondigroup, n.d.)

The surface of the base is precoated with clay in the paper making process. The amount of clay coating varies from low grammage papers of 5 g/m² to high grammages of 25 g/m². At the end of the paper making process, the clay coated kraft paper is calandered to reach the required smoothness level. CCK-papers are

available in white color and have good printability properties. (Mondi Release Liner, 2022)

3.1.3 PE-coated kraft papers

Polyethylene -coated (PCK) siliconized papers are used mainly for label, tape and graphic art applications. PE-layer is mainly coated to the base paper by extrusion process offline. PE-extruded papers can be kraft or fine-papers, depending on the required quality of the end-product and the extrusion can be done one or two sided of the base paper. Normally the base paper grammage is between 60 g/m² and 120 g/m² and the amount of extruded PE varies typically from 5 g/m² to 25 g/m². (Mondi Release Liner, 2022)



PICTURE 4. An example of typical end-use product of PCK. (Mondigroup, n.d.)

The silicone amount in PE-coated papers can be kept low as the PE-layer makes the surface of paper very smooth and easy to siliconize. PE is a polyolefin, which is unwanted material in recycling process but if it is easily removable at the beginning of recycling process it should not cause any problems in the process.

3.1.4 Machine finished and machine glazed kraft papers

Machine glazed (MG) and machine finished (MF) papers are uncoated kraft papers. MG and MF papers are used typically for release liner applications in hygiene, envelope and label products. Typical grammages for MG and MF papers are from low grammages to medium grammages, from 30 g/m² up to 100 g/m². In the picture 5. can be seen the typical end use application of these release liners.



PICTURE 5. An example of typical end use product hygiene release liners. (Mondi Release Liner, 2022)

The main difference between these two grades is the smoothness of the paper. Machine glazed paper is one side smooth and other side rough. Machine finished paper has both sides smooth. These base papers need to be precoated before siliconization as the surface of the paper is not closed in the paper making process. (Mondi Release Liner, 2022)

3.2 Silicone

Silicone is a polymer, which is a compound of silicon, carbon, oxygen and hydrogen. The structure of silicone is done from polyorganosiloxanes where atoms of silicon are combined to oxygen to form the siloxane bond and the rest of the silicon valences are combined to methyl groups. In the picture 6. is shown a silicone polymer. Silicone is synthetic and static and it appears in many different forms, as for example as resin, oil or rubber form. (Elkem, n.d.)



PICTURE 6. Silicone polymer. (Charati, Stern, 1998)

Silicone is thermal stabile and has a low surface energy. Silicone is wetting well on several substrates and it is easy to process in coating stages. Its properties are suitable for food contact and different kind of medical applications. Depends on the end use application, it gives good release or adhesion properties. Silicone is insoluble in all solvents, including the water. (Elkem, n.d.)

In release liner application the silicone is used to give for laminated end product certain release properties and to protect the adhesive layer of the lamination. The chemistry of silicone defines if it can be cured thermally or by radiation. Silicone can be applied to the paper or film surface using several techniques in different forms, like in solvent, solvent less or emulsion. (Orlych, 2004) The coat weight of silicone is normally kept low, as it is an expensive coating substance.

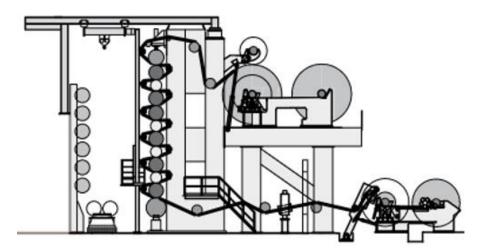
The release properties of the release liner differs from the application and the used adhesive by the customer. The level of release can be adjusted with the combination of crosslinker, catalyst and controlled-release additive added to the silicone coating. Crosslinker is used beside the speed of coating machine and temperature of siliconization process to improve the curing of silicone on used substrate. (Orlych, 2004) Controlled release additive is used as release modifier and it is typically used in two side siliconized products.

Beside the additives, the required release level can be adjusted by the amount of coat weight and by the coverage of the silicone. The less silicone, the higher release value is reached as the silicone is not covering the surface of liner uniformly and therefore the adhesive sticks to the uneven silicone coated areas.

3.3 Supercalandering

Glassine base papers used for siliconization are supercalandered. Calandering is a finishing unit in the paper making process where to the produced paper is applied compression forces between multiple rollers. The used compression force between the rollers makes the glassine paper dense. The main target of calandering is to improve the smoothness of the base paper but also the stability of paper thickness. Also the porosity and slight transparency of paper can be improved. (Ehrola, Hernesnniemi, Kuosa, Kyytsönen, Linnonmaa, Mäenpää, Pietikäinen, Stapels, Tani, Vuorikari, Rautiainen, 2010, 48-49)

Supercalandering differs from the standard calandering process by the number of used rollers. In supercalandering is used the mixture of hard and soft rollers to be able to reach the required smoothness level. Beside the hardness of the used roller, the required smoothness level in calendaring process can be achieved by heat, nip pressure, steam and speed of the machine. In the picture 7. can be seen the typical supercalander machine. (Ehrola, Hernesnniemi, Kuosa, Kyytsönen, Linnonmaa, Mäenpää, Pietikäinen, Stapels, Tani, Vuorikari, Rautiainen, 2010, 48-49)



PICTURE 7. Example of supercalander. (Ehrola, Hernesnniemi, Kuosa, Kyytsönen, Linnonmaa, Mäenpää, Pietikäinen, Stapels, Tani, Vuorikari, Rautiainen, 2010, 48-49)

Glassine papers are supercalandered one or two sided depending of the sides, which will be siliconized. The level of calandering depends on the used settings and existing machines by paper supplier. The level of calandering affects to the density of base paper, which can make the paper more difficult to repulp in recycling process.

4 CONFEDERATION OF EUROPEAN PAPER INDUSTRY

Confederation of European paper industry is called Cepi and it represents the total pulp and paper industry in Europe. Cepi is a non-profit organization, which co-operates closely with other stakeholders in entire paper chain. The main target of Cepi is to assure the competitiveness towards policy makers in pulp and paper industries and prove that sustainability can concur with competitiveness. (Cepi, n.d.)

Cepi consists of board members, partners and a number of networks. The board of Cepi has members from national associations like for example Austropaper from Austria, Finish forest industries from Finland and Die Papierindustrie from Germany. Partners of Cepi are stakeholders in pulp and paper industries, specifically machine and chemical suppliers like Omya, Voith, Valmet, Pöyry and Buckmann. (Cepi, n.d.)

The paper industry in Europe is one of the most sustainable industry fields where lot of efforts are done to create more sustainable and circular model. The sustainability comes mainly from the raw material where pulp is done from sustainably managed forests and paper is made in process where own made electricity and self-cleaned water can be used. One of the Cepi's initiatives is to improve the circularity of paper-based products after its use by increasing the knowhow and level of recycling process. Together with Cepi's co-operators and one European alliance called, 4evergreen, Cepi has developed a protocol for evaluating the recyclability of paper and board products in laboratory scale. (Cepi, n.d.)

The Cepi laboratory method for recycling of paper and board based products is done to be able to assess the level of recyclability by simulating the most relevant stages of standard paper recycling mill. The major recycling stages are pulping, screening and sheet formation. It is important to be able to simulate the standard process of recycling mill to make sure that the most used paper and board grades can be recycled easily with the existing equipment as well as to ensure the quality of formed sheet of re-used fibers. Beside the used equipment and the quality of formed sheet, it is important to know the level of coarse and fine reject to be able to get a good yield of recycling process. (Cepi, 2022) Cepi has published the first version of laboratory recycling method in beginning of 2021 and the second version in the summer 2022. In this study is concentrated to the revised version of the laboratory method but all parts of the new version were not able to be considered as laboratory tests were started before new version was published. (Cepi, 2022) In this study, for example the possibility to increase the repulping time to double, if the reject amount in the first stage is too high, was not considered as second version of the method was not yet published when the measurement were started. On the other hand, in the second version available score calculation for recyclability level was not completely published till this report was written and therefore not detailed considered in the report.

5 RESEARCH METHODS

5.1 Cepi recyclability laboratory test method

In this thesis the European laboratory test method to assess the recyclability of paper based products is used. Cepi has created the harmonized laboratory method together with 4evergreen and other its members. The main parts of the test method are repulping, coarse screening and fine screening as well as the evaluating of sheet adhesion and visual impurities of recycled material. The test procedure used in this study is explained in this chapter and the process flow of Cepi recyclability test method version 2, is shown in appendix 1. (Cepi 2022)

5.1.1 Sample preparation

The amount of sample, which is needed for Cepi laboratory recycling test, is 50 g of dry paper sample. To be able to get the correct amount of sample, the moisture content of paper need to be measured and the total paper weight needs to be recalculated. The sample needs to be cut to the size of 3 cm*3 cm (\pm 0,5 cm) as shown in the picture 8.



PICTURE 8. Sample preparation. (Reisinger, 2022)

5.1.2 Pulping stage

After sample preparation, the sample is repulped together with tap water in temperature of 40 $^{\circ}$ C (±1 $^{\circ}$ C) and pH of 7 ±1. In the picture 9. is shown the used disintegrator for repulping.



PICTURE 9. Disintegrator for re-pulping. (Reisinger, 2022)

To the measured 50 g of dry paper sample is added tap water in correct temperature until it reaches 2000 g to be able to reach required stock consistency. In picture 10. can be seen the sample with correct amount of water before disintegrating.



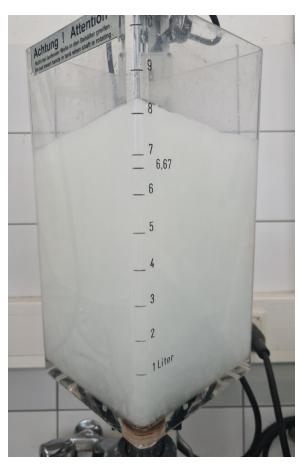
PICTURE 10. Prepared sample together with water ready to be repulped. (Reisinger, 2022)

Sample is repulped in disintegrator for 10 minutes with 30 000 revolutions. In the picture 11. can be seen sample after re-pulping. Disintegrator needs to be cleaned well and all remain stock of the sample needs to be collected and transferred to the distributor.



PICTURE 11. Sample after repulping. (Reisinger, 2022)

Total stock will be transferred to the distributor after repulping. Like in picture 12. is seen, the total stock will be diluted further and homogenized in the distributer for 10 minutes.



PICTURE 12. Distributing the total stock. (Reisinger, 2022)

Before the suspension of total stock is screened for the first time, two hand sheets will be done for the later evaluation. Each hand sheet should have the determinate basis weight by Cepi, which is 60 g/m^2 (±2) to be able to have equal analyse for the optical properties. When hand sheet has a weight of 1.8 g, it has the required basis weight. To reach the correct weight for the hand sheet, it is needed to take 300 ml of total stock suspension for making one hand sheet. In the picture 13. is shown the used hand sheet maker where the suspension is added.



PICTURE 13. Handsheet maker. (Reisinger, 2022)

In Cepi recycling test it is required to make pictures from each hand sheet and steps which can be later on analysed and included to final test report. In the picture 14. is seen the example of two hand sheets done from the total stock.



PICTURE 14. Example of wet hand sheets of total stock. (Reisinger, 2022)

The wet hand sheets will be dried under the standard conditions in the dryer like shown in the picture 15.



PICTURE 15. Hand sheet dryer. (Reisinger, 2022)

5.1.3 Screening

When the total stock is distributed well and hand sheets are done, the suspension will be screened in fractionator in two separate stages. First stage is a coarse screening with the screening plate with holes of diameter of 5 mm and second stage is fine screening with the plate with slots in size of 150 μ m. In the picture 16. is shown the used fractionator.



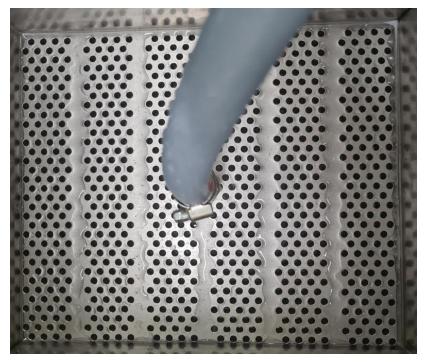
PICTURE 16. Used fractionator based on Cepi method. (Reisinger, 2022)

Coarse screening is lasting 5 minutes and fine screening 20 minutes and in both stages the amount of water which flows in the fractionator is 8,6 litres per minute. In the picture 17. is an example how screening looks in fractionator together with repulped sample and flowing water.



PICTURE 17. An example of screening in fractionator. (Reisinger, 2022)

After fractionating 5 minutes, accept which goes through from the coarse filter will be collected, diluted and distributed again to be able to make coarse-accept hand sheets, same way like total stock hand sheets. In the picture 18. is shown the coarse screening plate with the correct size of holes.



PICTURE 18. Coarse screening plate. (Reisinger, 2022)

The reject, which is not going through the screening plate will stay on the screening plate and will be photographed, collected carefully and after drying, weighted. In the picture 19. is shown the example of coarse reject after fractionating. This dry amount of reject is called as coarse reject and the amount in percentages will be calculated for the end result which is part of the total recyclability level. If the coarse reject is higher than 20 %, it is recommended in the new Cepi method version 2, to do disintegrating, instead of 10 minutes, 20 minutes. In this thesis this new setting was not used as laboratory tests were started before the new version was published.

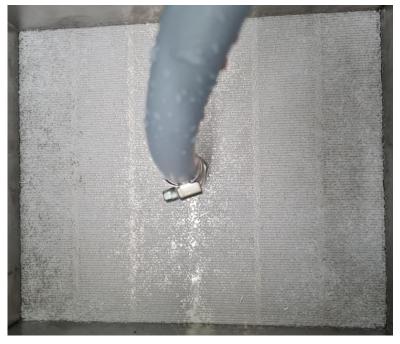


PICTURE 19. An example of coarse reject on the filter after fractionating. (Reisinger, 2022)

The rest of the suspension will be fractionated with fine screening plate where the slots are size of 150 μ m. The fine screening plate can be seen in picture 20. The accept of the fine screening will be collected, diluted and homogenized in dirstributor to be able to make hand sheets like for total stock and coarse accept is done. The residual of screened suspension which is not going through the fine filter is called fine reject. In picture 21. can be seen an example of reject on fine screening plate. After fractionating the fine reject will be photographed, collected carefully and dryed. The dryed fine reject will be weighted and the amount will be calculated as percentages which will affect to the final recyclability level together with coarse reject amount.



PICTURE 20. Fine screening plate with slots in size of 150 µm. (Reisinger, 2022)



PICTURE 21. An example of fine reject on the filter after fractionating. (Reisinger, 2022)

5.1.4 Rating the recyclability level according to Cepi method

The new Cepi method is based on scores, which are calculated of the combination of coarse and fine rejects, visual impurity level and the level of adhesion on hand sheets. The scorecard is not yet published but it has been used for evaluating the recyclability level of tested sample on this study. All samples which have scores from 0 to 100 are recyclable on standard recycling paper mill and the samples which have scores less than 0 are mainly not recyclable. Samples which are having scores slightly under 0, could be recyclable after slight changes in the recycling process, like for example, improving the repulping time to 20 minutes when the coarse reject is too high. (Cepi, 2022)

If the recycled sample has the coarse reject less than around 20%, the made hand sheet has no adhesion to its cover board or cover sheet, and the level of impurities is good, the sample is recyclable. The level of impurities and sheet adhesion is analysed from the hand sheet, which is done from the last phase of recycling laboratory test, from fine accept suspension.

Visual impurities by Cepi method are analysed first by the possible material of the visual impurity. Metal particles and wax stains are not accepted in the recycling process and therefore the sample with these kind of residuals are automatically on high level of visual impurities on analyse and therefore decreases the points in the end calculation for scores. Other materials, which Cepi has described as possible visual impurities are for example ink residuals and pigment coating particles. Samples, which are tested in this study, do not include any of these particles and according to Cepi method, visual impurities on tested samples belongs to material group, called, others. (Cepi, 2022)

After the material of impurity is clarified, the amount and size of impurities will be defined. When the amount and size of visual impurities are minimal, the analysed sample has visual impurities on level 1. If the amount or size of the impurities gets more, then the level gets higher and decreases points. In the table 4. is shown how to analyse the amount and size of the visual impurities. (Cepi, 2022)

Visual impurities				
Amount	Few	Some	Many	
Amount	< 10	10 - 100	> 100	
	Small	Big	Combination	
Size	Size < 1mm	>= 1mm	Consider the worst case level	

TABLE 4. Amount and size evaluation of visual impurities. (Cepi 2022)

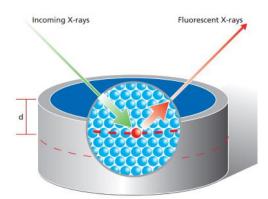
For score calculation in Cepi method the level of impurities is not affecting to the end scores as much as the level of total reject. If the level of visual impurities is for example 2, in total points there will be only few minus points. In table 5. is explained the levels of visual impurities.

TABLE 5. Levels of visual impurities. (Cepi 2022)

Level 1	No visual quality issues			
Level	Minor visual quality			
2	issues			
Level	Some visual quality			
3	issues			
Level	Significant visual quality			
4	issues			

5.1.5 X-ray fluorescense analysis

Electromacnetic waves with different kind of wavelengths can be called X-rays. When these X-rays are radiated to the sample, elements on its surface are emitting fluorescent X-ray radiation on certain level depending on the characteristics of each element. (Brouwer, 2010, 10) In the picture 22. is shown an example how the incoming X-rays are absorbed to the sample surface and how elements on it reflect fluorescent X-rays.



PICTURE 22. Example of incoming and outcoming x-rays on the sample surface. (Brouwer, 2010, 15)

With X-ray fluorescence analysis it is possible to measure the amount of an element for example on the paper surface. From the release liners the amount of silicone can be measured with this method.

6 TEST RESULTS

Release liners are typically categorized to the paper based products, which are not recyclable due the silicone coating. At the beginning of this study, the main assumption was that the recyclability level is not depended only to the silicone coating but the characteristics of used base paper. Glassine is supercalandered paper and denser than for example clay-coated base paper in siliconization process. Higher density of the paper could affect to the repulping level and therefore increase the reject on screening stage and reduce the recyclability level.

Release liner product range is wide with all used applications from graphic arts and labels to hygiene products and envelopes. For this study, it was decided to divide products to five groups by the used base papers of the most common release liners. The base paper groups are glassines, clay-coated, MG- and MFpapers as well as PE-extruded base papers.

Each base paper group was divided further to three different basis weight groups to be able to cover all products without a need to measure the recyclability of every separate basis weight. Three basis weight groups were low grammage products from 35 g/m² to 69 g/m², medium grammage products from 70 g/m² to 99 g/m² and high grammage products from 100 g/m² to 135 g/m². In this study the recyclability of one side siliconized products and two siliconized products were considered to able to see if the silicone amount has an effect to the recyclability level. In the table 6. can be seen the first testing plan for recyclability level of release liners.

Glassine based liner	Clay-coated kraft based liners	PE-coated based liners	Machine glazed based liners	Machine finished based liners
GL 1s Low	CC 1s Low	Kraft/PE 1s Low	MG 1s Low	MF 1s Low precoat A
GL 2s Low	CC 1s Medium	Fine/PE 1s Low	MG 2s Low precoat B	MF 1s Low precoat B
GL 1s Medium	CC 2s Medium	Fine/PE 1s Medium		
GL 2s Medium	CC 1s High	Fine/PE 1s High		
GL 1s High				
GL 2s High				

TABLE 6. First testing plan for recyclability tests of most comm	on release liners.
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6.1 Recyclability tests on glassine release liners

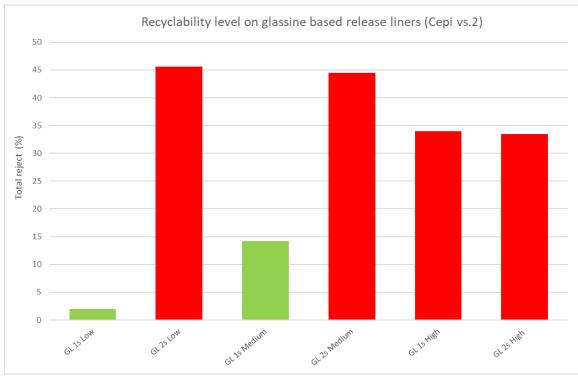
In the table 7. is shown the plan for testing the recyclability level on glassine based liners. The target was to see the difference on recyclability levels between

base paper grammage groups and when the glassine based liner is one and two side siliconized.

siliconized	g/m²	basis weight group
1 side	35-69	low
2 side	35-69	low
1 side	70-99	medium
2 side	70-99	medium
1 side	100-135	high
2 side	100-135	high

TABLE 7. First plan for the recyclability tests on glassine based release liners

When the recyclability of glassine samples was tested, it was seen that there is a big difference in recyclability level between the tested glassine products. In the figure 2. is seen the results on tested glassine products by one and two side siliconization and by each grammage group. Green colour in the figure 2. means that the product is recyclable in standard paper recycling mill based on Cepi method. The red colour means that the product is not recyclable on standard paper mill. In the vertical axis is presented the total reject amount of coarse and fine screening in percentages.



Recyclable
Limited recyclability
Not recyclable

FIGURE 2. Recyclability levels of tested glassine based release liners

From the tested glassine products, only two products were recyclable, low and medium grammage glassines, when only one side is siliconized. As seen from the figure 2. there is a big difference on the total reject amount, for example, when one side siliconized low grammage and two side siliconized low grammage products are compared.

The recyclability level of one side and two side siliconized high grammage glassines were on the same level on total reject amount, which was not expected. It is also unexpected that the level of reject is higher on low and medium grammage two side siliconized products then on two side siliconized high grammage product. Due these unexpected high reject amounts, more tests were needed and in the table 8. is shown the plan for recyclability tests on glassine products to be able to explain the unexpected reject amounts on presented products above.

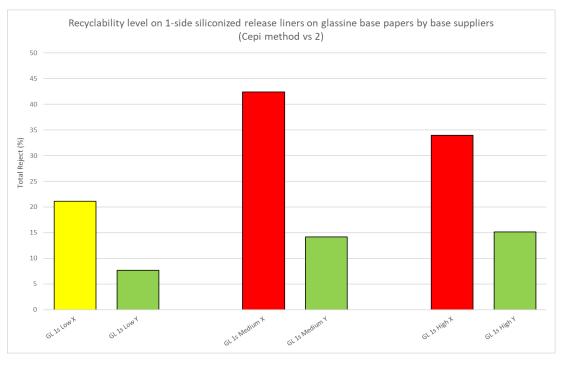
		Basis weight	
siliconized	g/m²	group	supplier
1 side	35-69	low	Х
1 side	35-69	low	Y
	-	-	
2 side	35-69	low	Х
2 side	35-69	low	Y
1 side	70-99	medium	Х
1 side	70-99	medium	Y
2 side	70-99	medium	Х
2 side	70-99	medium	Y
1 side	100-135	high	Х
1 side	100-135	high	Y
2 side	100-135	high	Х
2 side	100-135	high	Y

TABLE 8. The second recyclability test plan for glassine based release liners.

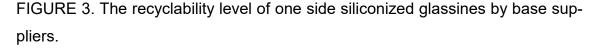
As seen in the table 8. it was decided to test the recyclability level of glassine products from two different base paper suppliers to be able to see where the unexpected reject amounts could come from. In the next paragraphs is presented the results of one side and two side siliconized glassines to be able to explain results better.

6.1.1 One side siliconized glassines

The one side siliconized glassine samples, which were tested on a first stage were having unexpected results. Low and medium grammage products were recyclable but high grammage not, as the total reject amount was too high. In the figure 3. is shown the recyclability level of one side siliconized glassines when two different base paper suppliers were tested.



Recyclable
Limited recyclability
Not recyclable

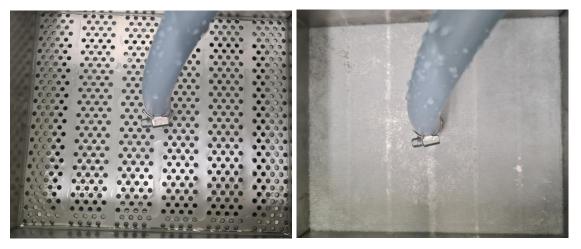


In the figure 3. is each grammage group presented by base paper supplier X and supplier Y. It can be seen that when the release liners on glassine base paper from supplier X are tested, the products are not recyclable. The yellow colour in figure 3. means that the sample is limited recyclable and based on Cepi method (version 2), if some process parameters would be changed, the product could be recyclable. As seen in the figure 3. the one side siliconized glassines are recyclable when the base paper is used from the supplier Y.

The relative silicone amount on the low grammage one side siliconized liners is above two percentages from the total weight of the liner. The high grammage products have relative silicone amount under two percentages and the relative silicone amount is same when the different base paper suppliers are compared. The relative silicone amount is not explaining the difference on reject amounts between the paper suppliers. Even the relative silicone amount on the low grammage glassine liners is higher than on high grammage glassine products, the total reject amount is less and the low grammage glassine liners are better recyclable than high grammage glassine liners. The relative silicone amount is not affecting to the recyclability level on one side siliconized glassines. Based on tested recyclability levels, it can be seen that there are differences on paper making processes of glassine suppliers. At the beginning, the assumption was that glassine based release liners are difficult to recycle due the high density of the paper. When the density of different suppliers is considered, there is not any prominent differences seen. In the paper making process of glassine paper is not any wet strength additives used, which could explain the bad recyclability level by supplier X.

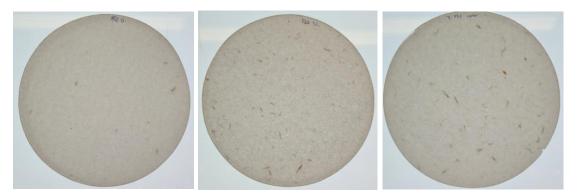
Glassine papers are normally surface treated by one or two sided with starch or PVA but the content of surface treatment chemicals in total grammage of the base paper are minimal and therefore not affecting negatively to the recyclability level. The smoothness and transparency of glassine base paper is reached by supercalandering and the level of calandering and available equipment can vary by different suppliers. Supercalandering can be done for one side or for both sides of the base paper with variable machine settings, which could cause the difference between the glassine papers from the different suppliers. In this study the effect of differences on calandering or paper making process between the glassine suppliers was not considered deeper.

Below in the picture 23. is shown the typical coarse and fine rejects for one side siliconized products. During the recyclability tests, it was seen that normally there is not any coarse reject after fractionating on the one side siliconized products and all suspension has went through the coarse filter. In the picture 23. on the right hand can be seen the reject of fine screening which was varying depending on the tested sample.



PICTURE 23. Example of coarse and fine reject typical for one side siliconized glassines. (Reisinger, 2022)

Beside the total reject amount, to the recyclability level affect also the sheet adhesion and visual impurities of hand sheet made from accept of second screening stage, fine screening. In the picture 24. is shown examples of the visual impurities on fine accept hand sheets on the recycled one side siliconized glassines. First sheet in the picture 24. is evaluated in score calculation according to Cepi method, to have visual impurities as level 1, middle sheet to have impurities as level 2 and the third sheet, level 3. Recycled glassine liners were not having any sheet adhesion and therefore not affected to the level of recyclability. The effect of visual impurities and sheet adhesion are included in the recyclability levels presented in figures 2. and 3.



PICTURE 24. Example of visual impurities on recycled one side siliconized glassines in the laboratory recycling tests. (Reisinger, 2022)

Based on the recyclability tests done according to Cepi method, it can be said that the one side siliconized glassine release liners are recyclable on standard paper recycling mill depending on the base paper supplier and the paper making process. If the base paper supplier X with the current references is used, the total reject amount is too high and these products are therefore not recyclable. It is possible to say that in spite of high density, the glassine products are recyclable and silicone release coating on one side does not cause issues in the standard recycling process.

6.1.2 Two side siliconized glassines

Like one side siliconized glassines, also two side siliconized glassines were decided to test by two different base paper suppliers to be able to explain unexpected recyclability results in first stage. In the figure 4. is shown tested two side siliconized release liners in each grammage group by base paper suppliers X and Y. It can be seen that any of tested two side siliconized glassines are not recyclable but the supplier Y has always lower reject amount than supplier X.

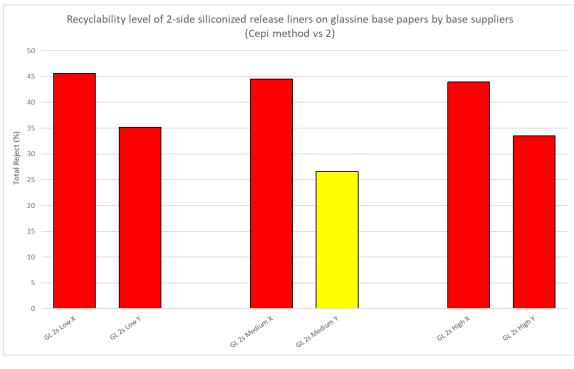


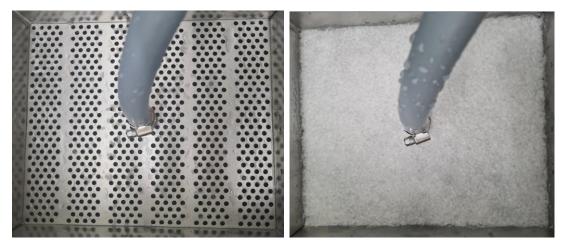


FIGURE 4. Recyclability level of two side siliconized glassines by base suppliers.

The low grammage two side siliconized release liners on glassine base have a relative silicone amount over four percentages. The medium grammage group has a relative silicone amount around four percentages and high grammage glassine liners under four percentages. When the recyclability levels in figure 4.

are considered based on relatively silicone amount, it can be seen that the low grammage products have slightly higher total reject amount than medium or high grammage products probably due the higher relative silicone amount. When considered the results of one and two side siliconized glassine liners, it can be said that the relative silicone amount is not a key factor, which reduces the recyclability level but the siliconization two sided closes the surface of the glassine paper and decrease therefore the level of repulpability.

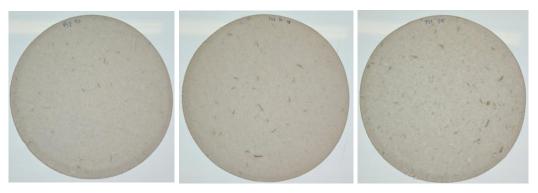
When the fractionating was done for two side siliconized glassine papers, the coarse filter and fine filter were typically looking like shown in picture 25. The coarse reject amount was low but it was more than on the one side siliconized products. The reject of fine screening was high and the difference on one side siliconized samples is notable.



PICTURE 25. Example of coarse and fine reject typical for two side siliconized glassines. (Reisinger, 2022)

In the picture 26. can be seen the visual impurities of hand sheets done from fine accept. First hand sheet on the left was evaluated according to Cepi method to have level 1, middle hand sheet level 2 and right hand sheet level 3. These impurities were seen often on recycled glassine papers and it was recognized that these fiber accumulation, which are visual are coming from the collecting of fine accept. In the lab the accept is collected from the really fine sieve by hand which cause the accumulations. Before doing the fine accept hand sheet the suspension is only distributed which is not strong enough treatment for destroying the accumulations. In standard recycling paper mill the accept will be pumped and

therefore the visual impurities are for sure getting less and improving the quality of recycled glassine release liners.



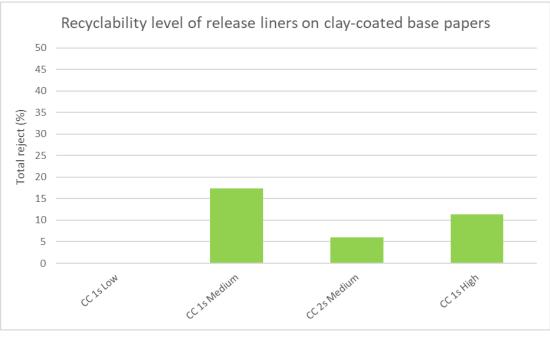
PICTURE 26. Example of visual impurities on two side siliconized recycled glassine paper in the laboratory recycling tests. (Reisinger, 2022)

Based on the recyclability tests done according to Cepi method, it can be said that two side siliconized glassine release liners are not recyclable on standard paper recycling mill. As silicone is not water soluble it can be that two side siliconization does not enable needed level of repulping together with high density properties of glassine base paper. It can be that silicone closes the surface of glassine paper and therefore the liner does not disintegrate well enough and the reject amounts in recyclability process stay in coarse and fine screening too high. It would be interesting to test how the level of recyclability improves when the repulping time is doubled or is the two side siliconization still an issue in the recycling process.

6.2 Recyclability tests on CCK release liners

Clay coated kraft papers are mainly used for one side siliconization and therefore tests were done for low, medium and high grammage kraft papers, siliconized one sided. On this study was used one sample with two side siliconization to see if the behaviour is the same than on glassine samples.

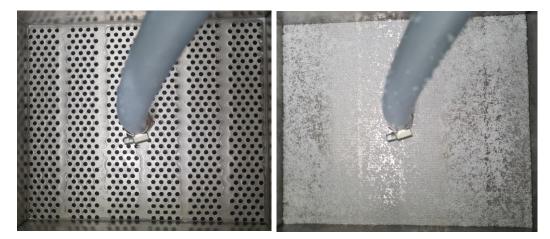
In the figure 5. is shown the recyclability level of tested clay coated kraft paper samples. It can be seen that all products are recyclable and reject levels lower than on glassine samples. Two side siliconized medium grammage product is also recyclable and reject level low.



Recyclable
Limited recyclability
Not recyclable

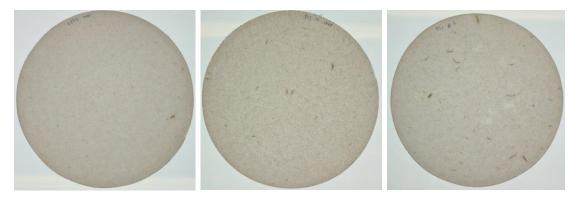
FIGURE 5. Recyclability level of tested release liners on CCK.

In the picture 27. can be seen the typical rejects on clay coated kraft papers on coarse and fine screening. When the fractionating was done on coarse screening there was not any reject left. On the fine screening therefore was some reject left.



PICTURE 27. Examples of coarse and fine rejects on tested CCK liners. (Reisinger, 2022)

In picture 28. is seen typical visual impurities on clay coated kraft papers when the recyclability was tested in the laboratory. The formation of the hand sheets made of fine accept is looking homogenous and only few visual impurities can be seen. According to Cepi method the first hand sheet is evaluated on visual impurities to be on level 1, middle on to be on level 2 and sheet on the right to be on level 3.

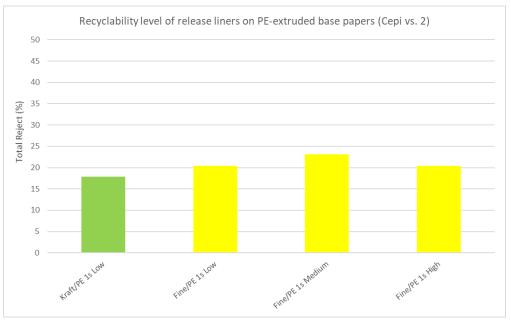


PICTURE 28. Examples of visual impurities on recycled CCK release liners in the laboratory recycling tests. (Reisinger, 2022)

Based on the tests done in the laboratory according to Cepi method, it can be said that release liners on clay coated kraft base papers are recyclable on standard paper recycling mill. The release silicone coating does not cause any issues on the recycling process even the CCK would be two side siliconized.

6.3 Recyclability tests on PE-coated release liners

Recyclability was tested on PCK release liners by each grammage group and on one side siliconized products. From the figure 6. can be seen that tested liners are mostly limited recyclable on standard paper recycling mill and the reject amount is around 20%.



Recyclable
Limited recyclability
Not recyclable

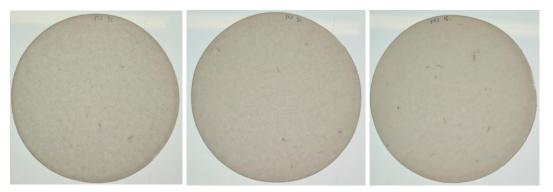
FIGURE 6. Recyclability level on tested PE-coated release liners.

In the picture 29. can be seen the example pictures of coarse screening plate and fine screening plate after fractionating PE-coated release liner. It is possible to see that the PE-layer of the liner is removing from the paper already in a first screening stage. It means that the unwanted polyolefin-material in recycling process is possible to be removed and the paper fibers, which are going through the coarse and fine screening plates, can be further reused.



PICTURE 29. Examples of coarse and fine rejects on tested PE-liners. (Reisinger, 2022)

In the picture 30. below is shown the typical sheet formation and visual impurities on recycled PE-coated release liners. The formation looks even and there are not many visual impurities.



PICTURE 30. Examples of visual impurities on recycled PE-coated release liners in the laboratory recycling tests. (Reisinger, 2022)

Based on the recyclability tests done on PCK one side siliconized liners, it can be said that these products are limited recyclable on standard recycling paper mill, which means if some slight process changes or product improvement are done, liners could be recyclable. Based on the results, it can be seen that the unwanted PE-material is easily removed on the early stage of recycling process and the paper content of liner can be recycled to end of the process.

6.4 Recyclability tests on MF- liners

The recyclability of machine finished kraft paper based release liners were tested and the results can be seen in the figure 7. Two different low grammage products were tested with two different precoatings done before siliconization. It can be seen that both tested liners are recyclable and the total reject amount is low. MFliner with precoat A, has a bit higher reject amount than MF-liner with precoat B.

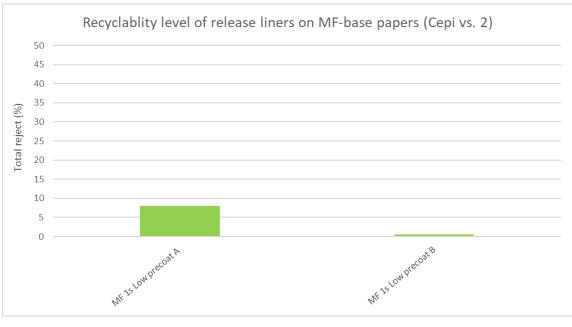




FIGURE 7. Recyclability level of two different tested MF-release liners.

In the picture 31. below can be seen the typical coarse and fine screening plates after fractionating the MF-liners. The total reject amount on tested samples was coming from the fine screening stage but as seen, the amount of reject is still low.



PICTURE 31. Examples of coarse and fine rejects on tested MF-liners. (Reisinger, 2022)

When the hand sheets of recycled MF-liners were analysed there was one sample without visual impurities as seen in the picture 32. on the left side. On the right side in the picture 32. can be seen the second tested samples where are more impurities visible.

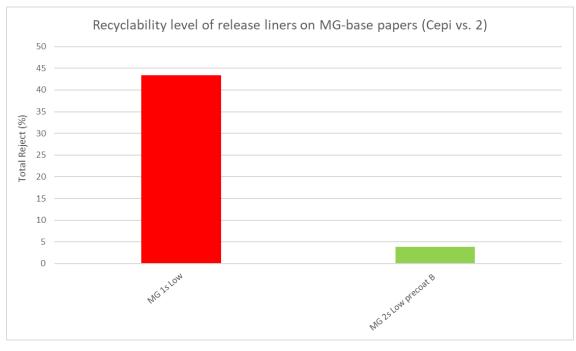


PICTURE 32. Examples of visual impurities on recycled MF-liners in the laboratory recycling tests. (Reisinger, 2022)

Based on the tests done according to Cepi method for silicone coated machine finished papers it can be said that these products are recyclable in standard paper recycling mill and the silicone does not cause issues in the process.

6.5 Recyclability tests on MG-liners

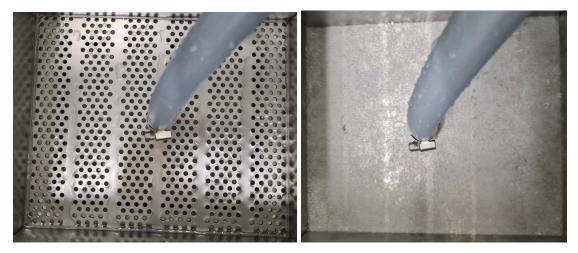
The recyclability of two different release liners on machine glazed base paper was tested. It is interesting to see in the figure 8. that when the base paper is not precoated, the reject amount is high. When the MG paper was precoated and siliconized on two sides, it can be seen that the reject amount is low and MG-liner recyclable.



Recyclable
Limited recyclability
Not recyclable

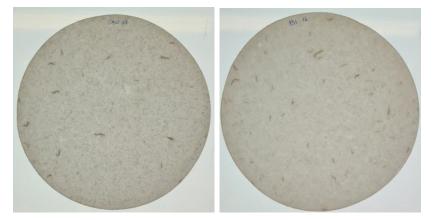
FIGURE 8. Recyclability level of two different kind of MG-liners.

When the machine glazed base paper is not precoated before siliconization, the paper absorbs the silicone inside to the paper and the silicone sticks to the paper fibers. When the fibers are covered with the silicone, the repulpability level is not good enough and the reject amount on screening section stays too high. From the picture 33. can be seen examples of coarse and fine reject on precoated MG-paper sample after fractionating.



PICTURE 33. Example of coarse and fine rejects on MG-liner when base paper precoated before siliconization. (Reisinger, 2022)

When the visual impurities of recycled MG-liners were considered, it was seen that there are some impurities visible on the hand sheets. In the picture 34. can be seen examples of the visual impurities on recycled MG-liners. According to Cepi method the both hand sheets were evaluated on visual impurities to be on level 2.



PICTURE 34. Examples of visual impurities on recycled MG-liners in the laboratory recycling tests. (Reisinger, 2022)

6.6 Silicone reduction on recycled glassine liners

The recyclability of different release liner products was tested and it is also interesting to know where the silicone residuals can be seen in the laboratory recycling process. The silicone amount has been tested for one side and for two side siliconized glassine liners by X-ray fluorescence analysis first from the liner itself before recycling process. In the figure 9. is shown the reduction of silicone by tested glassine products after each recycling step. XRF is able to measure only the silicone amount of the surface of the sample and therefore, the silicone amount was measured from the each hand sheet from the both sides and the sum was compared to the silicone amount of liner before recycling.

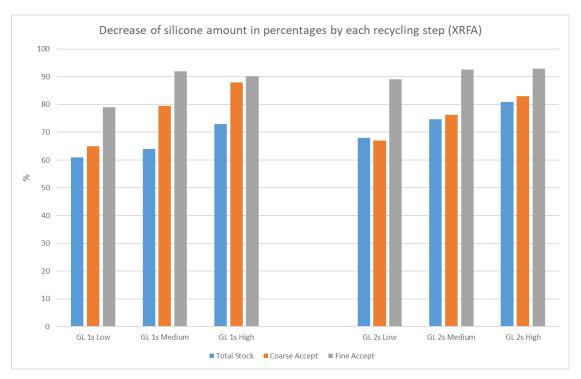


FIGURE 9. Decrease of silicone amount by separate recycling steps measured from the made hand sheets.

From the figure 9. can be seen that there is not differences on the silicone amount reductions when recycling processes of one and two side siliconized products are compared. After first stage of the laboratory recycling process, repulping, the silicone amount of the hand sheet surfaces are more than 60 % less when compared to the liners before recycling. When the silicone amount is calculated from the hand sheets after coarse screening, it can be seen that it stays almost on the same level like after repulping. At the end of the recycling process it can be said that the level of silicone reduction is from 80 % to bit over 90 % when compared to the liner before recycling.

Beside the silicone reduction on the made hand sheets, more interesting would be to test the silicone amount of the reject after coarse and fine screening but in this study it was not possible to be considered. It would be also interesting to know if the wastewater of the recycling process includes some silicone particles after recycling release liners.

7 CONCLUSIONS AND DISCUSSION

The main target of this study was to test the recyclability of paper based silicone coated release liners. Release liners are used in laminated products in different kind of applications to protect the adhesive layer of the product and stick to it but same time to be able to peeled off easily. EN643 is a standardized list of standard paper and board grades for recycling, which lists release liners to the group number 5, meaning that they are not recyclable on standard recycling paper mill. Silicone is not soluble to any solvents including the water and therefore siliconized release coated papers are listed to the not recyclable products. Due the properties of silicone, it could cause issues in the standard paper recycling process.

The confederation of European paper industry, Cepi, has created the harmonized recycling laboratory method to be able to test the recyclability of paper and board based products in the laboratory scale based on to the process of standard paper recycling mill. The main process steps in Cepi laboratory method as well as in standard recycling mill are, repulping, coarse screening and fine screening. To the level of recyclability affects the total reject amount of screening stages as well as the visual impurities and sheet adhesion of made hand sheets. In this study the newest version of Cepi laboratory method was used for determination the recyclability level.

The tested paper based release liners were divided to the groups based on the used base papers. The main used and for siliconization specialized base papers are glassine, clay-coated kraft papers, PE-coated kraft papers as well as machine glazed and machine finished kraft papers. It was assumed that the recyclability level of different release liners is mainly based on the properties of the used base papers. Glassine, for example, is a high dense paper due its finishing process, supercalandering, and may not be that easily recyclable than other base papers used in release liners.

To be able to get the full picture of recyclability of the most typical release liner products, it was decided to divide siliconized products beside the used base paper, also to groups based on the typical basis weights of the product. The groups based on the grammages in this study were low, medium and high grammage groups. For some release liner applications it is typical to have instead of one side siliconization, two side siliconized release coating. The difference on recyclability level on one side and two side siliconized products was also considered in this study.

Glassine based release liners are typically used in release liners for label and tape applications. Glassine papers are supercalandered to reach closed and smooth surface for siliconization. The calandering process and its level can vary depending on the base paper supplier. When the recyclability of glassine based release liners were tested it was seen that the results are not as expected and for example the difference on the reject amounts, between low and medium grammage products on one side siliconized glassine liners was remarkable.

To be able to have an explanation to the unexpected results, it was decided to test the recyclability levels on glassine based release liners from two different base paper suppliers. When the one side siliconized glassine liners were tested, it was seen that these products in each grammage group were recyclable when the base paper supplier Y was used. When the recyclability level was tested on base paper supplier X, it was seen that the one side siliconized glassine based release liners are not recyclable.

The paper making process between base paper suppliers can vary and for example different kind of chemical additives for surface sizing or precoating can be used. There are also different kind of supercalanders and levels of calandering, which could cause the differences on recyclability levels between glassine base paper suppliers.

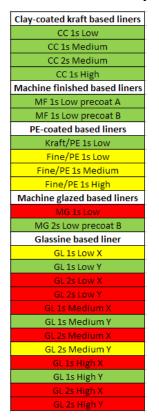
The recyclability of two side siliconized glassines was tested and there was seen as well differences on the reject amounts between two base paper suppliers. After recyclability tests, it can be said that despite the paper supplier, two side siliconized glassine products are not recyclable. Glassine paper itself is a high dense paper and when it is two side siliconized, the silicone both sides closes the surface of the paper and reduce the repulpability of the liner and therefore increase the reject amount. From the one and two side siliconized glassine liners was also the reduction of silicone amount by each recycling step tested. When the silicone amount of recycled glassine liner was compared to the silicone amount of release liner before recycling, it can be said that the reduction of silicone can be up to 90 % during the recycling process. The measured silicone amount is measured from the surface of both sides of the hand sheet and therefore it is not telling the reality about the total silicone amount which can be inside the recycled paper.

PE-coated kraft papers are typically used in release liners for tape and graphic art applications. Plastic particles generally are unwanted in the standard paper recycling process and by PCK products, the PE-layer can be harmful for the process. When the recyclability on PCK based and one side siliconized liners was tested the level of recyclability was limited which means that based on Cepi method, after some slight process changes it could be recyclable on standard paper recycling mill. It was seen that the PE-layer of the base paper stays on the coarse screening stage and most of the fiber-based material was able to reused.

The recyclability level of other tested kraft based release liners were recyclable when they were precoated. One and two side siliconized clay-coated kraft papers were having low total reject amounts and good recyclability level. Also the MF and MG liners were having good recyclability levels when precoating colors A and B were used. When the MG-liner without precoat was tested, it was seen that the silicone was absorbed to inside the paper instead of staying on the paper surface and stuck to the fibers. Silicone around the paper fibers decreases the level of repulpability and therefore increases the reject amount in the process.

In the table 9. is shown a summary of recyclability levels on tested release liners. Green color means that based on the Cepi laboratory method, the tested product is recyclable in the standard paper recycling mill. Yellow color in the table 9. means that the product is limited recyclable and red color means not recyclable when Cepi laboratory method is used.

TABLE 9. Summary table of recyclability levels of tested release liners.



Based on the recyclability tests done for release liners, it can be said that only due the silicone release coating, release liners does not need to be categorized as not recyclable products. When release liners are produced on CCK or MG and MF base papers with precoating before siliconization, it can be said that these release liners are recyclable and the categorizing in EN643 standard list could be revised. From the circular economy point of view, it is important to be able to increase the amount of recycled material instead of use natural resources. In this case it can be said that the industrial waste of release liners used in hygiene applications can be recycled and increase the resources for paper recycling process.

In the future it could be studied how much there is silicone in total inside the recycled paper as well as how much silicone is on reject got from coarse or fine screening. Cepi method version 2, allows higher repulping time if the coarse reject is too high but unfortunately it was not tested on this study as the new version was published after some tests were already done based on Cepi's first version of recyclability tests in laboratory. It would be interesting to test how much the

total reject amount decreases for example on two side siliconized glassine products when repulping time is doubled. It could be also studied in the future, which are the properties, which makes the differences on recyclability levels between two base paper suppliers on glassine products. When the standard paper recycling process is considered, it would be interesting to know if the silicone from the repulped release liner waste would be somehow possible to be removed or collected with suitable chemical additives like the printing inks in deinking process.

REFERENCES

Bajpai, P. 2013. Recycling and Deinking of Recovered Paper. London: Elsevier.

Brouwer, P. 2010. Theory of XRF. Edition 3. Almelo: PANanalytical B.V. Read on 11.12.2022. <u>https://www.iotcco.com/uploads/VirtualTeaching/Articles/PANanalyti-</u> cal/PANanalytical%20XRF%20theory.pdf

Cepi. N.d. About Cepi, members. Read on 24.10.2022 https://www.cepi.org/about-cepi/members/

Cepi. N.d. About Cepi, organization. Read on 24.10.2022 https://www.cepi.org/about-cepi/organisation/

Cepi. 2021. Preliminary Statistic 2021. Read on 05.12.2022 <u>https://www.cepi.org/wp-content/uploads/2022/02/Cepi_Preliminary-_2021_Report.pdf</u>

Cepi. 2022. Cepi recyclability test method, version 2. Read on 24.10.2022 <u>https://www.cepi.org/cepi-recyclability-test-method-version-2/</u>

Charati, S. Stern, S. 1998. Diffusion of Gases in Silicone Polymers. New York: American Chemical Society. Read on 14.12.2022 <u>https://pubs-acs-org.libproxy.tuni.fi/doi/pdf/10.1021/ma980387e</u>

Ehrola, J. Hernesnniemi, A. Kuosa, H. Kyytsönen, M. Linnonmaa, P. Mäenpää, T. Pietikäinen, R. Stapels, R. Tani, M. Vuorikari H. Rautiainen, P. 2010. Calendering. In Rautiainen, P (ed.). Paper making Science and technology. Book 10, Papermaking part 3, finishing. Helsinki: Fapet Oy, 14-167.

Ek, M. Gellerstedt, G. Henriksson, G. 2009. Pulping Chemistry and Technology. Stockholm: KTH.

Elkem. N.d. Silicones, blog. Read on 05.10.2022. https://www.elkem.com/silicones/blog/faq/

Ervasti, I. 2010. Recovered paper statistics and definitions. In Höke, U. Schabel S. (ed.). Paper making Science and technology. Book 7. Recycled fiber and deinking. Helsinki: Fapet Oy, 12-24.

Kirwan, M. 2013. Handbook of Paper and Paperboard Packaging Technology. 2nd edition. London: John Wiley & Sons, Ltd.

Meinander, O. 2000. Specialty Papers. In Paulapuro, H (ed). Paper making Science and technology. Book 18. Paper and Board Grades. Helsinki: Fapet Oy, 101-130.

Mondigroup. N.d. Products and solutions, envelope release liners. Read on 19.11.2022.

https://www.mondigroup.com/en/products-and-solutions/release-liners/release-liners-products/envelope-release-liner/

Mondigroup. N.d. Products and solutions, release liners for labels. Read on 19.11.2022.

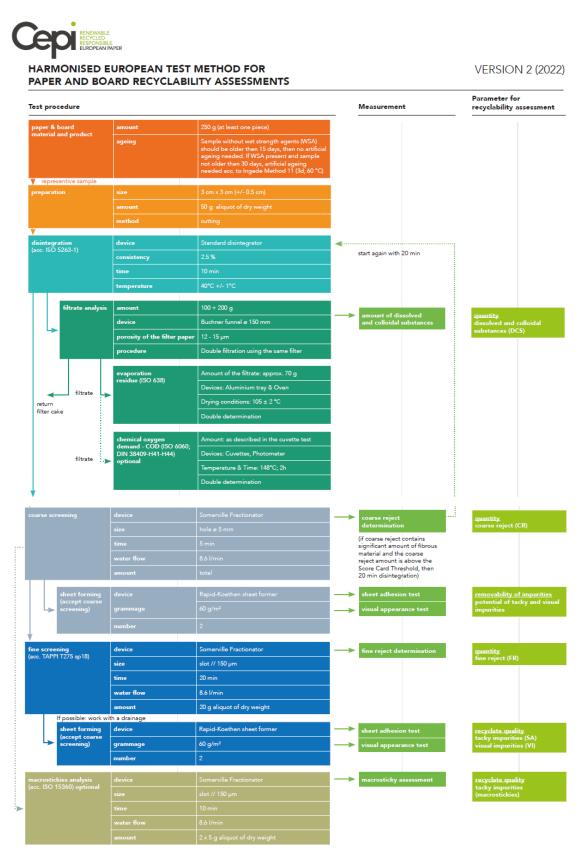
https://www.mondigroup.com/en/products-and-solutions/release-liners/release-liners-products/release-liners-for-labels/

Mondi Release Liner. 2022. Internal Product and plant training.

Orlych, M. 2004. Silicone-Adhesive interactions in release liner applications. Read on 14.12.2022. <u>https://go-gale-com.libproxy.tuni.fi/ps/i.do?p=ITOF&u=tam-pere&id=GALE|A116796145&v=2.1&it=r</u>

APPENDICES

Appendix 1. Process flow chart of Cepi recyclability test



(Cepi, 2022)