



BRNO UNIVERSITY OF TECHNOLOGY

VYSOKÉ UČENÍ TECHNICKÉ V BRNĚ

FACULTY OF CHEMISTRY

FAKULTA CHEMICKÁ

INSTITUTE OF CHEMISTRY AND TECHNOLOGY OF ENVIRONMENTAL PROTECTION

ÚSTAV CHEMIE A TECHNOLOGIE OCHRANY ŽIVOTNÍHO PROSTŘEDÍ

EVALUATION OF THE QUALITY OF POLETHYLENE TEREPHTHALATE FLAKES FOR THE PRODUCTION OF FOOD GRADE RECYCLED REGRANULATE

HODNOCENÍ KVALITY VSTUPNÍHO POLYETHYLENTERFTALÁTU PRO VÝROBU RECYKLOVANÉHO REGRANULÁTU POTRAVINÁŘSKÉ KVALITY

MASTER'S THESIS

DIPLOMOVÁ PRÁCE

AUTHOR

AUTOR PRÁCE

Bc. Martina Snopková

SUPERVISOR

VEDOUCÍ PRÁCE

prof. Ing. Jiří Kučerík, Ph.D.

BRNO 2023

Assignment Master's Thesis

Project no.: FCH-DIP1767/2022 Academic year: 2022/23
Department: Institute of Chemistry and Technology
of Environmental Protection
Student: **Bc. Martina Snopková**
Study programme: Environmental Sciences and
Engineering
Field of study: no specialisation
Head of thesis: **prof. Ing. Jiří Kučerík, Ph.D.**

Title of Master's Thesis:

Evaluation of the quality of polyethylene terephthalate flakes for the production of food grade recycled regranulate

Master's Thesis:

1. Literature review on the PET recycling and regranulation processes. Types of contamination and their consequences on the quality of PET recycle.
2. Evaluation the quality of PET flakes from different suppliers based on the analysis of various physicochemical properties by using a variety of analytical methods.
3. Data evaluation and discussion. Evaluation of the quality of individual suppliers based on the obtained data.

Deadline for Master's Thesis delivery: 8.5.2023:

Master's Thesis should be submitted to the institute's secretariat in a number of copies as set by the dean This specification is part of Master's Thesis

Bc. Martina Snopková
student

prof. Ing. Jiří Kučerík, Ph.D.
Head of thesis

Head of department

In Brno dated 1.2.2023

prof. Ing. Michal Veselý, CSc.
Dean

ABSTRACT

Polyethylene terephthalate (PET) is a widely used plastic, especially to produce PET bottles. However, waste PET bottles pose an environmental risk if they are not well managed. Recycling is an appropriate way to treat PET bottles, ideally by recycling them back into PET bottles. Nevertheless, recycling PET into food-grade recycled PET is challenging and the resulting quality is heavily dependent on the quality of the feedstock, i.e., the PET flakes. This thesis aimed to evaluate the quality of food-grade PET flakes on the EU market. Although the quality of PET flakes has already been the subject of several scientific studies, the analysis of the qualitative parameters of PET flakes has never been applied to such a large area as is the EU market. However, extensive analysis is essential for trend mapping and possible quality optimisation. For the analysis, material from different areas of the EU collected over a period of two years was used. The quality assessment included the quality control methods set out in EN ISO standards and standard operating procedures (SOPs), as well as spectrophotometry, Fourier-transform infrared spectroscopy – Attenuated total reflection (FTIR-ATR), and measurement of intrinsic viscosity. The suitability of each supplier and trends across the batches delivered were assessed. The most important findings included (1) other plastics content is the most problematic quality parameter of PET flakes, (2) other plastics content originates mainly from bottles and is independent of the collection system, (3) a higher degradation rate of PET flakes supplied by Western producers compared to the material from producers located in the East of the EU. A comprehensive analysis of the EU market and quality trends in PET bottle recycling by geographical area and type of collection system were provided. These results will not only be useful for practice, but they could also provide an incentive to improve legislation on the assessment of plastic contaminants or the introduction of more appropriate PET bottle design.

ABSTRAKT

Polyethylentereftalát (PET) je široce používaný plast, zejména pro výrobu PET lahví. Odpadní PET lahve však představují riziko pro životní prostředí, pokud se s nimi dobře nenakládá. Vhodným způsobem nakládání s PET lahvemi je recyklace, ideálně jejich zpětná recyklace na PET lahve. Recyklace PET na recyklovaný PET potravinářské kvality se však může potýkat s několika problémy a výsledná kvalita je do značné míry závislá na kvalitě vstupní suroviny: PET vloček. Cílem této práce bylo zhodnotit kvalitu potravinářských PET vloček na trhu EU. Ačkoli kvalita PET vloček již byla předmětem několika vědeckých studií, analýza kvalitativních parametrů PET vloček nebyla nikdy aplikována na tak rozsáhlou oblast, jakou je trh v rámci EU. Rozsáhlá analýza je však nezbytná pro zmapování trendů a případnou optimalizaci kvality. Pro analýzu byl použit materiál z různých oblastí EU shromážděný v průběhu dvou let. Při hodnocení kvality byly použity metody kontroly kvality stanovené v normách EN ISO a interních normách, jakož i spektrofotometrie, infračervená spektroskopie s Fourierovou transformací metodou zeslabeného úplného odrazu (FTIR-ATR) a měření vnitřní viskozity. Hodnotila se vhodnost jednotlivých dodavatelů a trendy napříč dodanými šaržemi. Mezi nejdůležitější zjištění patří (1) obsah ostatních plastů je nejproblematičtějším parametrem kvality PET vloček, (2) ostatní plasty pochází především z lahví a jejich obsah není závislý na systému sběru, (3) vyšší míra degradace PET vloček dodávaných západními výrobci ve srovnání s materiálem od výrobců nacházejících se na východě EU. Byla poskytnuta komplexní analýza trhu EU a kvalitativních trendů v recyklaci PET lahví v závislosti na zeměpisném umístění a typu sběrného systému. Tyto výsledky budou užitečné nejen pro praxi, ale mohly by být také podnětem pro zlepšení legislativy v oblasti vyhodnocování plastových kontaminantů nebo pro zavádění vhodnějšího designu PET lahví.

KEY WORDS

PET recycling, food-grade rPET, contamination, quality, PET flakes

KLÍČOVÁ SLOVA

Recyklace PET, potravinářský rPET, kontaminace, kvalita, PET vločky

CITATION OF THE PRINTED VERSION

SNOPKOVÁ, Martina. Hodnocení kvality vstupního polyethylterftalátu pro výrobu recyklovaného regranulátu potravinářské kvality. Brno, 2023. Dostupné také z: <https://www.vut.cz/studenti/zav-prace/detail/148944>. Diplomová práce. Vysoké učení technické v Brně, Fakulta chemická, Ústav chemie a technologie ochrany životního prostředí. Vedoucí práce Jiří Kučerík.

CITATION OF THE ELECTRONIC VERSION

SNOPKOVÁ, Martina. Hodnocení kvality vstupního polyethylterftalátu pro výrobu recyklovaného regranulátu potravinářské kvality [online]. Brno, 2023 [cit. 2023-04-11]. Dostupné z: <https://www.vut.cz/studenti/zav-prace/detail/148944>. Diplomová práce. Vysoké učení technické v Brně, Fakulta chemická, Ústav chemie a technologie ochrany životního prostředí. Vedoucí práce Jiří Kučerík.

DECLARATION

I declare that the diploma thesis has been worked out by myself and that all the quotations from the used literary sources are accurate and complete. The content of the diploma/bachelor thesis is the property of the Faculty of Chemistry of Brno University of Technology, and all commercial uses are allowed only if approved by both the supervisor and the dean of the Faculty of Chemistry, BUT.

.....
student's signature

ACKNOWLEDGEMENT

I would like to thank the consultant Ing. Lucie Šudomová for providing valuable information on the topic and for the major help during writing the thesis. I also want to thank the supervisor of the thesis, prof. Ing. Jiří Kučerík, Ph.D., and the co-supervisor for RPTU, Dr. Zacharias Steinmetz, for their professional supervision and the helpful comments during the development of the thesis. I am also grateful to the management of the company rPET InWaste s.r.o. for providing samples and facilities without which this thesis could never have been written.

TABLE OF CONTENTS

Table of contents	5
1 Introduction	7
2 Theoretical part.....	8
2.1 Polyethylene terephthalate.....	8
2.1.1 Recycling of PET	8
2.1.2 Carbon footprint and life cycle analysis of recycled PET	9
2.2 Mechanical recycling: collection, contamination, and its separation	10
2.2.1 Collection systems.....	10
2.2.2 Sorting technologies	12
2.3 Food-grade recycled PET production and contamination	16
2.3.1 Quality specification of food-grade recycled PET	16
2.3.2 Contamination in food-grade recycled PET	17
2.3.3 Extrusion and regranulation	19
2.4 Contamination of PET.....	22
2.4.1 PET and its modifications	22
2.4.2 Other polymers	25
2.4.3 Metal and non-metal non-polymer contaminants.....	26
2.4.4 Moisture content.....	27
2.4.5 Other parameters	28
2.5 Theory on the methods	30
2.5.1 Spectrophotometry: CIELAB method.....	30
2.5.2 Fourier transform infrared spectroscopy with attenuated total reflection	30
2.5.3 Intrinsic viscosity and plastometers.....	30
2.5.4 Correlation visualisation with ggpairs() function and Pearson coefficient.....	31
2.5.5 Hypotheses summary.....	31
3 Experimental part	32
3.1 Materials.....	32
3.2 Methods.....	33
3.2.1 Entrance quality control analysis parameters	33
3.2.2 Spectrophotometry (L^* , a^* , b^*).....	34
3.2.3 Determination of intrinsic viscosity	34
3.2.4 Identification of other polymers and other contaminants	34
3.3 Results and discussions	36
3.3.1 Entrance quality control analysis parameters	36
3.3.2 Spectrophotometry (L^* , a^* , b^*).....	57

3.3.3	Intrinsic viscosity.....	61
3.3.4	Other polymers and other contaminants.....	62
3.3.5	Correlation analysis.....	69
4	Conclusion.....	73
	Bibliography.....	74
	List of abbreviations.....	90

1 INTRODUCTION

Plastics have become a popular material among manufacturers and consumers as they offer a wide range of properties and possibilities of application. However, despite their advantages, the excessive use of plastics and the poor management of plastic waste introduces severe problems and environmental burdens. For this reason, it is essential to provide and implement optimal solutions for plastic waste treatment.

Recycling is often considered an appropriate solution. One of the most and best recyclable plastics is polyethylene terephthalate (PET). PET bottles are an intensively recycled plastic application, even for food reuse. For food applications, extrusion (melting) takes place to produce pellets of recycled PET. To ensure the safety of the recycling process, a high quality of the supplied material – PET flakes – is required [1].

To be used in food-grade applications, the quality of PET is determined by several parameters [2]. The final quality of recycled PET depends on various factors such as the material supplied or the processing technology. As a result, the quality of PET flakes can vary from one supplier to another [1, 2]. This can cause complications for PET flake processors.

Although the quality parameters of PET flakes have been investigated and described in many papers, including local market research [3–5], it was concluded that there is a gap in the scientific literature: none of the papers addressed quality parameters at a scale beyond the local market, let alone the market within the European Union (EU). It was assumed it would be appropriate to fill this gap, as this can provide valuable information for industry and research. Therefore, the aim is to assess the quality of PET flakes in the EU market based on a comprehensive analysis of material from suppliers operating in different EU countries.

The theoretical part will provide the necessary information for understanding mechanical recycling technologies and their connection with quality parameters. Furthermore, the quality parameters will be discussed to understand better the origin or causes of these parameters, the associated problems, and possible improvements.

For the experimental part, a cooperation existed with a pellet producer who processes PET flakes to study a range of PET flake samples from many manufacturers operating in different EU countries. Various parameters in these PET flake samples were measured and evaluated in relation to the individual manufacturers. Quality control parameters (e.g., contamination, bulk density), colour, and intrinsic viscosity were the subjects of the analyses. Trends were sought, as well as the drivers of these trends and the resulting recommendations for PET flakes processors. Relationships between the different parameters were also investigated. In addition, new measurement methodologies were proposed to improve the quality assessment of PET flakes.

It is important to note that the topic of this thesis is closely related to and intended to serve practice. Therefore, some specific information may not be supported by literature sources but is confirmed by practice. This type of information will be used most in Chapters 2.4 and 3.3, and for each such statement, the origin from practice will be mentioned.

2 THEORETICAL PART

2.1 Polyethylene terephthalate

PET is a thermoplastic that belongs to the most widely produced polymers. PET properties and other indicators justify its preference over other materials [1, 6, 7]. PET is suitable for use in many applications: non-food (straps, fibres) and food, from which packaging materials and PET bottles are especially important [7]. Of all PET applications, PET bottles are the most consumer-relevant. PET has several exceptional properties that make it an ideal material for beverage packaging, such as low weight or low breakability [1].

In general, the preferential use of PET leads to its intensive production [8] and, consequently, to PET waste generation. Nonetheless, its high resistance to biodegradation poses a challenge [9]. Thus, PET waste should be managed optimally. Once PET waste is not handled well, it introduces many problems such as littering [1], pollution, microplastic formation [9], and a worse carbon footprint [1]. Recycling is one of the best options for plastic waste management [10]. It represents an economically optimal way to reduce discarded PET [6]. Moreover, optimized recycling processes can save materials and resources [10].

Particularly PET bottles are products that exhibit good properties for recycling [1]. Specifically, their specific weight, mechanical strength, low price, and well-established collection infrastructure facilitate the collection and subsequent recycling of PET [7]. Currently, recyclate made from PET bottles is not only used for non-food and general food applications; PET bottle-to-bottle recycling is developing intensively. Even though PET bottle-to-bottle recycling is now operating on a relatively large scale, recycling still faces several challenges today. Efforts are therefore being made to optimise it [1, 7, 11].

2.1.1 Recycling of PET

PET recycling is widely promoted, especially for environmental and sustainability reasons [7]. In general, four types of plastic recycling exist: primary, secondary, tertiary, and quaternary. While primary recycling refers to the recycling of homogeneous industrial waste, others refer to the recycling of the postconsumer wastes [12]. Mechanical (secondary) recycling is based on reprocessing wastes mechanically [1, 12] and will be elaborated further in the upcoming chapters. Tertiary (chemical) recycling is a process that leads to the intended chemical cleavage of PET chains into high-quality monomers or oligomers that can be resynthesized into various applications [12, 13]. Quaternary recycling refers to the incineration of wastes, generating energy that can be further used [12]. This thesis will deal with mechanical recycling for several reasons. Firstly, analysed samples – PET flakes – were obtained via mechanical recycling. Secondly, this type of recycling is very common [13]. Thirdly, mechanical recycling is the most economically and environmentally friendly when compared to other recycling technologies [14, 15].

Nowadays, the recycling is widely applied mainly in the case of PET bottles. There are many reasons, for example, the distinguishability of PET bottles from other plastic waste, accessibility [1, 16], and low use of additives [1]. A significant amount of produced recycled PET (rPET) is used in non-food applications such as straps or fibres because these applications do not require high-quality material and tolerate a higher degree of contamination of rPET when compared to food-grade rPET¹. When such rPET is used in food applications, the possibility is to use multilayer containers where the rPET layer is not in contact with food [1]. rPET can be even mixed with other polymers using compatibilizers [17].

However, as sophisticated decontamination technologies exist, rPET can match the quality of virgin PET and can therefore be used in the food industry, especially to produce PET bottles or trays. For recycling a discarded PET bottle into a new PET bottle, the term bottle-to-bottle recycling is used [1].

¹ Refer to Figure 7 to see an example for quality requirements for PET flakes for food-grade applications.

The blend of rPET with virgin PET is usually used in production of new PET bottles to ensure the quality requirements [17]. Recycling PET bottles is becoming a necessity in some countries as it has recently been enshrined in their legislation. In the EU, PET bottle manufacturers will be required to use at least 25% rPET in produced bottles from 2025. This minimal limit will rise to 30% in 2030 [18]. As a result, recycling rates will need to be significantly increased along with continuous optimisation of recycling processes.

Despite all the benefits that PET recycling brings, it is also crucial to ensure that the whole operation is financially viable for the manufacturer [7]. Unlike many other polymers, recycled PET has a significant market value, plus many other benefits [7, 19]. One of these advantages relates to carbon footprint and life cycle assessment (LCA).

2.1.2 Carbon footprint and life cycle analysis of recycled PET

With climate change being one of the most debated environmental issues, measuring carbon footprint has become an important indicator for various processes and products. The carbon footprint is defined as the amount of greenhouse gases emissions that directly result from the phenomenon under study throughout its life cycle [20]. Data provided by the Austrian recycling company PRT (PET Recycling Team GmbH) has shown that replacing virgin PET with rPET can reduce the associated greenhouse gas emissions by up to 80%. More specifically, the carbon footprint of rPET was estimated to be 0.45 kg carbon dioxide per kg of recycled material, whereas the carbon footprint of virgin PET is 2.15 kg per kg of virgin material. In other words, carbon footprint of virgin PET is 1.7 kg higher per 1 kg of material compared to rPET. All operations related to rPET production (such as collection, sorting, pelletising, and so on) were factored into the study [21]. Another study showed that producing rPET instead of virgin PET can save 2.28 kg/kg of carbon dioxide emissions. Moreover, the production of rPET saves 87.6% of energy (including local transport) compared to virgin PET [22].

In addition to the carbon footprint, another informative tool that describes the environmental impact of recycling is the LCA. LCA assesses the environmental burden brought by all operations during the whole life cycle of an object [23]. Using LCA, it was found that PET bottles (made of virgin PET) could be replaced by either 100% recycled cans for carbonated drinks to reduce the impact, while beverage cartons were found to be more suitable for juices and milk. Glass has always had worse results than PET, even 100% recycled, but returnable bottles were not considered [24]. Once included in the study, returnable glass bottles either performed slightly worse (in the case of non-carbonated beverages) or had similar results to 100% virgin PET bottles [25]. In another study where the recycled content of PET bottles was included, the results were by far the most favourable for PET bottles. At the point where 50% recycled content is used in the PET bottle, returnable glass bottles may still represent a more optimal solution. However, PET bottles with 100% recycled content outperformed all other types of beverage packaging [26].²

Additionally, rPET does not outperform virgin PET only in the case of carbon footprint. Using recycled resin also reduces other negative phenomena, e.g., acidification, eutrophication, and smog impact. It has been found that the use of rPET saves energy. However, rPET is associated with higher water consumption [27].

The previous chapters have aimed to justify the recycling of PET. What needs to be done as a first step to produce rPET is to collect PET waste from households and other waste producers. As this step is crucial for further processing and the resulting quality, it will be addressed in the next chapter.

² It is important to draw the reader's attention to the issue of comparing and generalising LCA studies. The results are highly dependent on individual indicators (e.g. transport, energy sources) [24]. It is therefore advisable to consider these results as indicative but not to apply them generally and in absolute terms.

2.2 Mechanical recycling: collection, contamination, and its separation

In theory, mechanical recycling is a process in which the polymer is reprocessed by physical means [28], without changes in chemical and polymeric structure [13, 17]. However, in practice, polymer chains can be cleaved in the process, which is an unwanted degradation due to contamination and mechanical stress [6, 13, 28, 29]. The degradation will be described in detail in the following chapters.

In the case of PET bottles, the process includes the following steps: washing, grinding, drying, and sorting (in varying order). Small fragments of PET bottles – PET flakes – are produced. Subsequently, PET flakes are subjected to extrusion (melting) and possibly to further processing – for example, to regranulation and pelletising which result in the production of granules or pellets [1, 6] – see Figure 1. During regranulation, additional steps can be employed to enhance the quality of the recycled material up to food grade, such as solid-state polycondensation – SSP [1] or liquid-state polycondensation LSP [30].

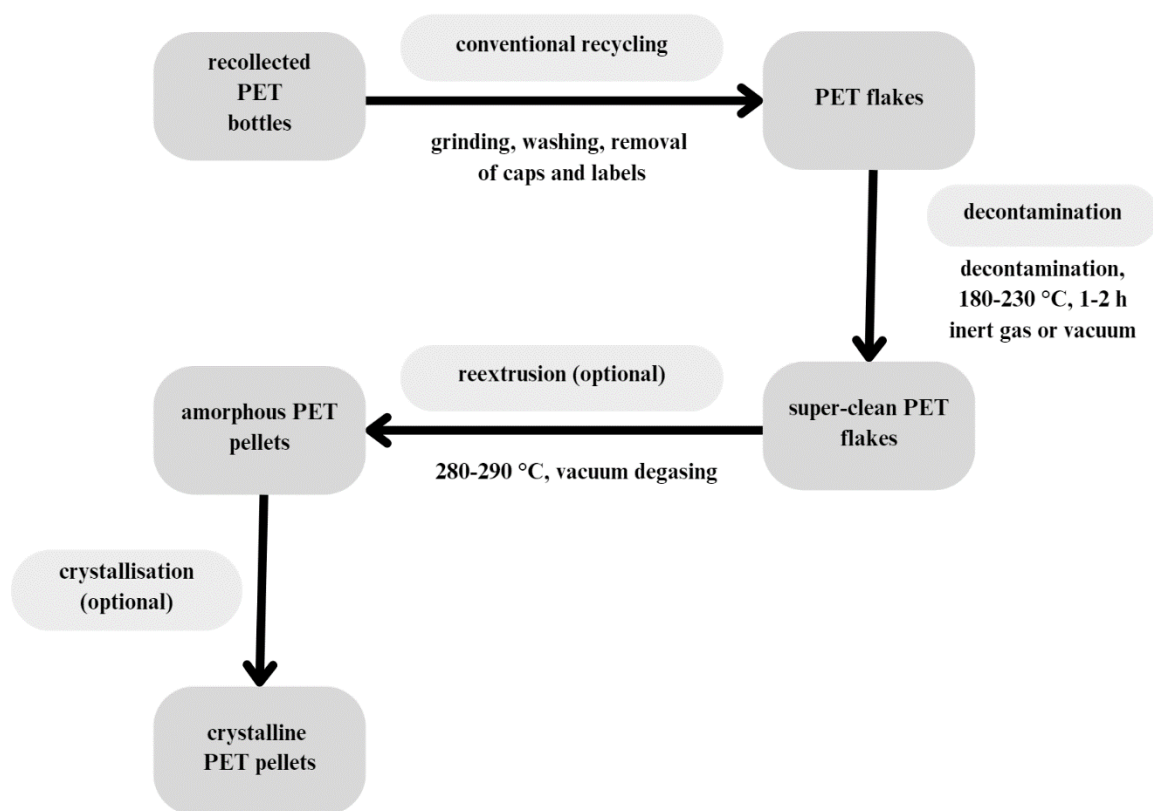


Figure 1: The example of PET mechanical recycling process. Modified from source: [1], pg. 871.

The contamination of postconsumer PET significantly affects the quality of rPET [6]. Mechanical recycling is particularly susceptible to contamination in the recycled material [28]. In this regard, the type of collection system seems crucial as different contamination levels were identified in rPET produced from various collection systems [31]. Therefore, the following section will describe the types of collection systems.

2.2.1 Collection systems

PET can be collected in different ways. A strategy that is considered to yield lower contaminant levels is the mono-collection system, mainly implemented through deposit-refund systems (DRS) [31]. DRS is based on the introduction of a refundable fee for packaging material. When buying a beverage in a PET bottle, the consumer must pay an additional cost (deposit) for the bottle itself. This deposit is

refunded to the consumer when the emptied bottle is handed in at the collection point [32]. This collection strategy is in place in many European countries (see Figure 2).

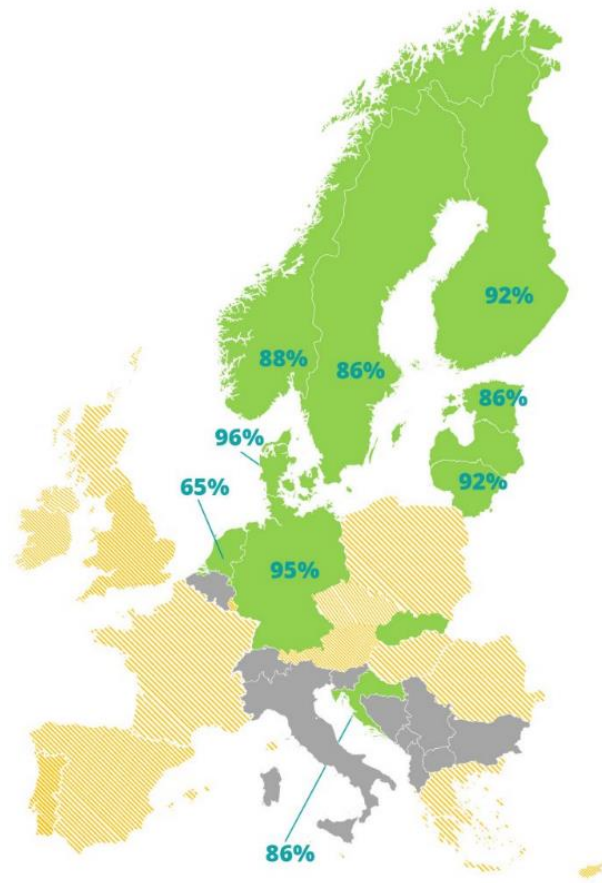


Figure 2: European countries with deposit systems for PET bottles (green) together with collection rates.

Source: [33].

However, mainly co-mingled separation strategy is implemented in the EU for plastics in general [34]. This system is founded on the collection together with other materials with consequent separation in the sorting facilities. Plastics can be collected with metals [31] or beverage cartons. These combinations can be found in many European countries, for example, Belgium, France [34], and the Czech Republic [35]. A more heterogeneous collection system exists in the UK, based on mixed collection of the mentioned materials with glass and paper from which plastics are separated in the sorting facilities [31].

In non-EU countries, the waste management may differ. In general, the waste management in developing countries (such as Brazil or India) may be realised in insufficient, improper ways [36–38]. An effort to implement functioning waste management policies (or already a realization of a policy) is observable in many non-EU regions, such as South America or South Asia [13]. Still, the organized recycling systems can be absent [36]. Material and PET flakes with an origin outside the EU will not be examined in this thesis.

The waste collection is considered to have an impact on rPET contamination. Mono-collection systems of PET bottles are regarded to yield less contaminated rPET [31]. But the co-mingled separation systems are more frequently implemented in many countries [34], including the Czech Republic [35]. However, it is not only the waste stream that is a source of contamination – but the materials also used to produce PET bottles and their components often pose problems as well. Therefore, not only the collection strategy, but also the technology and separation equipment used are crucial for the resulting quality of rPET [31]. Some authors find these technologies even more important than the collection systems [11].

Some researchers support this claim in cases where the design of PET bottles is inappropriate in a certain area and more contamination comes from bottle components than from faulty sorted items [5]. Thus, not only the collection scheme but also the technology used in reprocessing and the bottle design are critical for the final quality of rPET. In any case, sorting is an important step, and many technologies are used to reduce contamination as much as possible. These technologies will be a subject of the next chapter.

2.2.2 Sorting technologies

Separation of contaminants is essential to achieve a suitable quality of recyclate. To prevent the contaminants to enter the final product, various separation technologies must be introduced [1, 6, 11]. In general, separation is based on finding a property that differs between PET and a given contaminant – for example, melting point, density, or NIR (near-infrared) spectrum. Identification and separation are subsequently done based on these differences [11].

In PET flakes production, sorting is introduced before the waste enters the recycling technology and usually, the final product (PET flakes) is re-sorted to achieve higher quality [11]. Both manual and automatic sorting can be used for the input material. Thus, the producer can choose to use either manual or automatic sorting, each of which has advantages and disadvantages [6, 11, 39].

2.2.2.1 Manual sorting

The first option is to introduce manual sorting. Before entering the recycling line, the waste is moved onto a conveyor belt. Workers are positioned around the conveyor belt to collect and remove objects that should not enter the technology [39]. The disadvantages of this type of sorting are the costs [6] and the need for trained workers [40]. Moreover, it is often inefficient concerning contaminant removal [41] and sorting capacity [39]. The fact that the working conditions are not appealing to people cannot be overlooked [42]. But based on the practical experience, there is an important advantage: workers can sort waste that cannot be sorted using automatic sorting systems such as NIR [43].

Compared to manual sorting, automatic sorting is considered consistent, more efficient, and able to manage the high throughput of waste. The initial high investment should be recouped in the future if the recycling or sorting facilities handle a larger volume of material. However, this may not be met for some smaller plants. Different systems can be used, e.g., machine vision or NIR sorting [39]. However, once the contaminant has similar characteristics that are difficult to distinguish from PET bottles, it is not recognised and separated by some automatic sorting systems [44]. For this reason, and based on the practical experience, combining manual and automatic sorting seems to be appropriate option [43].

2.2.2.2 Flotation

Before the automatic sorting technologies are described in more detail, it is necessary to focus on flotation process. Flotation is a widely used, simple, and low-cost separation technology [45] based on the principle of differences in densities. The ground material is fed into the tank filled with a flotation medium. The plastics with a density higher than the density of the medium will sink to the bottom and vice versa. Water is usually used as a flotation medium [11]. In such a case, the separation threshold is $1.00 \text{ g}\cdot\text{cm}^{-3}$; see Figure 3: once the position of a red bar is to the left of the density of water (blue line), the material will float during flotation. Conversely, materials with the bars positioned to the right will sink and not be separated. Flotation using water as a medium is environment friendly [45, 46]. Nevertheless, flotation is not able to separate two polymers with a density higher than $1.00 \text{ g}\cdot\text{cm}^{-3}$ (such as polyvinylchloride – PVC – and PET). Moreover, phenomena such as attached bubbles, particle shape or size, and feedstock dosage can influence this type of separation [45].

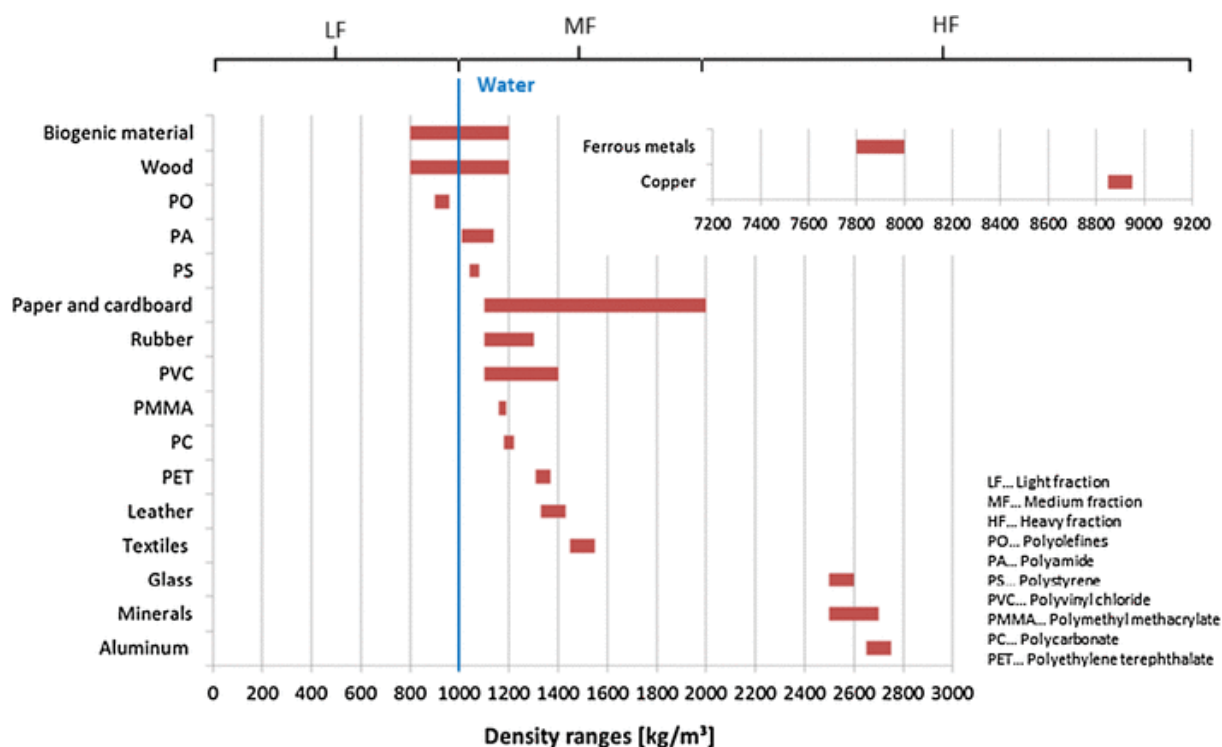


Figure 3: Densities of commonly occurring plastics and non-plastic materials compared to density of water.
Source: [45], pg. 1785.

Other types of flotation media working on different principles may be used to separate such polymers during flotation, but they can introduce additional problems, such as additional costs or the production of hazardous waste [47, 48].

2.2.2.3 Automatic sorting systems

NIR sorting systems are widely used for whole bottles and ground PET flakes [11]. The reason is that NIR sensors can detect a sequence of different polymers fast and reliably enough, also for higher throughputs. However, NIR does not distinguish between different applications made of the same polymer (such as PET bottles and PET trays) [42] and polymers with the resemblance in spectra, such as PET and polyethylene terephthalate glycol (PETG) [44]. Also, the exception exists for black-pigmented plastics, as they cannot be recognized by NIR [49, 50] due to their high absorbance [49]. The issue of black-pigmented plastics can be tackled by MIR (mid-infrared) and other types of detectors [49, 50] or colour detectors, that can be programmed to automatically separate all black particles [51]. The fact that colour sensors remove black particles non-selectively (concerning polymeric composition) does not pose a problem for production of clear food-grade rPET. In such a case, black particles represent either polymer contamination or darkly-coloured PET flakes which are undesirable in the material because of their effect on the final colour and quality of the recyclate [5]. The separation of products covered with labels is also problematic [42].

Machine vision is an automatic sorting method that has the potential to bridge the shortcomings of NIR sorting. Using algorithms, AI (artificial intelligence), and RGB (red-green-blue) cameras, the equipment can be programmed to sort incoming waste based on visual characteristics. This can solve problems with dark coloured plastics and contaminants, applications made from the same polymer, or labelled products [42]. Moreover, these devices are claimed to be economical and universal in use [52]. Theoretically, they could replace manual sorting. Still, researchers reported several disadvantages concerning machine vision sorting. First, machine vision should be used in combination with different sorting methods, as this approach is ineffective alone. Second, since machine vision is based on deep learning, large datasets

need to be created to achieve highly effective separation [42]. However, this is also a problem in manual sorting [40].

Colour separators can distinguish between bottles or flakes of different colours. Colour separation is necessary as differently coloured flakes or bottles are undesirable. First, they have different economical values [51], but also may influence the colour of final rPET pellets [53]. Manual sorting can eliminate a considerable amount of differently coloured bottles, but automatic sorters can also perform colour sorting of flakes [51]. Camera sensors based on visible spectrometry or prism-coupling are used to identify unwanted particles [19]. Unlike machine vision, colour separators can sort particles only based on colour, but not based on shape [19, 51].

Magnetic and eddy current separators are usually used for the separation of metals. Magnetic separators are relatively simple, using magnetic forces to separate ferrous metals from the waste stream. Eddy current separators are used to also separate non-ferrous (i.e., non-magnetic) metals such as aluminium. In an eddy current separator, a magnetic rotor generates an electric field and, consequently, a magnetic field. As a result, metal particles are separated due to repulsion [54], see Figure 4.

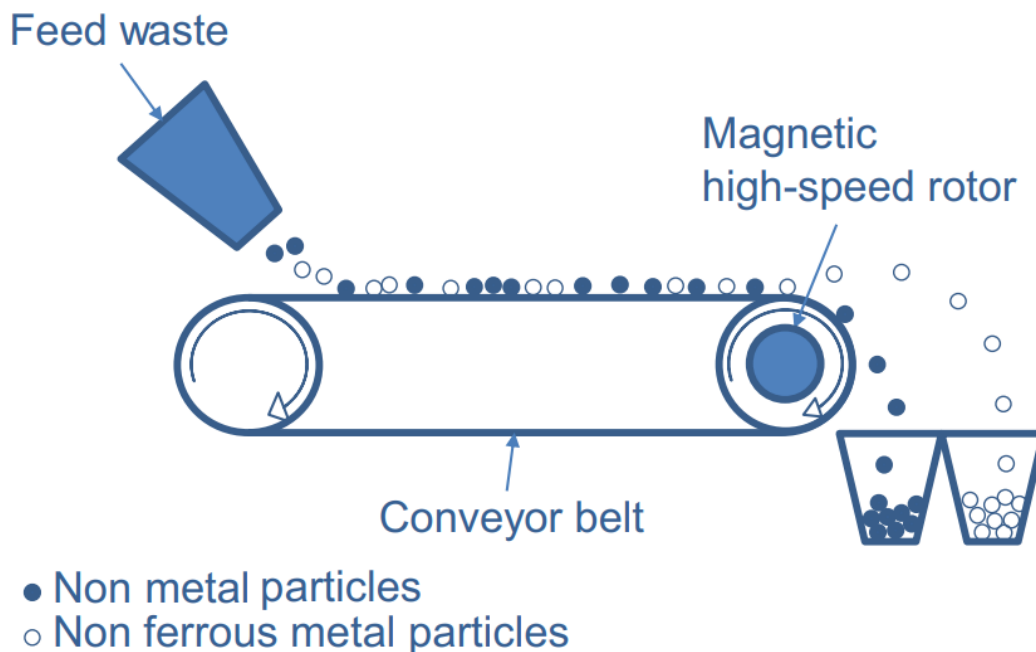


Figure 4: Eddy current separation scheme. Source: [54], pg. 27.

Another possible way of separating metal particles is to employ X-ray detectors. If X-ray detection is connected to the atomic mass analysis, heavy atoms such as chlorine can be detected in a waste stream. This attribute can be useful for detecting chlorinated polymers, such as PVC and other halogenated plastics [55].

There are several types of separation technologies, but PET manufacturers typically use a combination of metal, colour, and NIR separators in their technology [11]. As a result, sorting machine manufacturers can combine multiple sensor types in a single sorting machine [56, 57]. For example, VARISORT+ by company Sesotec combines NIR, colour, and metal sensors [57] as well as FLAKE PURIFIER+ by the same producer [56]. VARISORT is described in Figure 5.

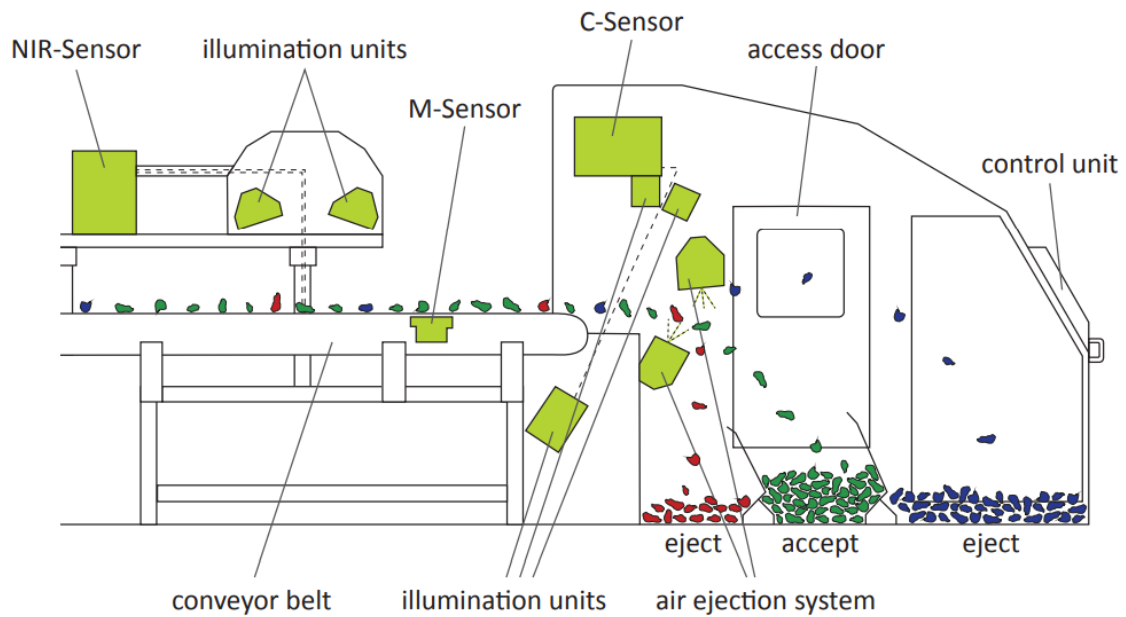


Figure 5: VARISORT sorting system, where three sensors (colour, metal, and NIR) are employed simultaneously. Source: [58], pg. 2.

Despite all the options, it is important to acknowledge that the separation of contamination is a trade-off between the resulting quality and quantitative losses. Due to the introduction of final flake sorting, up to 10% of the material can be lost [5]. Higher sensitivity results in higher losses and thus must be well considered [59].

After the PET flakes are free of contamination and have gone through the entire production process, they are ready for direct use or extrusion. The problem with extrusion is that the material must be treated by sophisticated technologies to obtain pellets of a certain quality after processing [1]. Why this is the case, what happens during extrusion, and why the presence of contamination in this step is problematic will be the subject of the next chapter.

2.3 Food-grade recycled PET production and contamination

2.3.1 Quality specification of food-grade recycled PET

The quality requirements, especially concerning contamination levels, are more stringent for food-grade rPET than for rPET used in non-food applications [1]. Therefore, additional requirements are posed on food-grade rPET, especially in terms of molecular contamination. The significant difference in these two groups of rPET applications also affects the recycled feedstock. Even though made of the same polymer, non-food PET waste can be a source of contamination. As a result, the amount of non-food PET products in the recycled waste stream needs to be carefully monitored and may only represent 5% of the total amount of material to be recycled in food-grade rPET production [2]. The reason is that this level was assessed to have no adverse effects on human health [2, 60].

Among others, these requirements have been adopted by legislation in recent years. According to Regulation (EC) No 282/2008 and – more recently – Regulation (EU) 2022/1616, producers planning to produce food-grade rPET must first obtain permission from the European Food Safety Agency (EFSA) [61, 62]. Although mechanical recycling has already been declared as the technology that can produce food-grade rPET, every producer must undergo EFSA testing and certification [62]. Besides the EFSA certification, the producer has to comply with good manufacturing practices [11] and, of course, comply with the requirements of the final buyers of rPET pellets, which go beyond EFSA and other regulations [43].

At the end of the technology testing, EFSA will publish an assessment for each manufacturer summarising the technology used, the test results, and the final opinion. The opinion sets out the decontamination conditions that must be met for the food-grade rPET production. The final provisions specify, for example, what proportion of recyclate can be used in food applications or whether recycled products can be filled with hot contents. These provisions may vary from manufacturer to manufacturer [63–66] – see Figure 6.

Company	Technology	Maximal rPET content*	Hot fill	Source
RE-PET	Erema Basic	70%	×	[63]
Omorika Recycling	PET direct iV+	100%	✓	[64]
Greenpet	Starlinger iV+	100%	✓	[65]
rPET InWaste	NGR	100%	✓	[66]

* Maximal content of rPET that can be used for PET drinking bottles production

Figure 6: Examples of EFSA evaluation for selected companies. Sources: [63–66].

Even though challenge tests focus mainly on volatile contaminants, pellet producers also have special requirements for the quality of incoming PET flakes [63–66]. An example of rPET InWaste company is shown in Figure 7.

Parameter	Maximal limit [mg·kg ⁻¹]
Moisture	10 000
PVC content	50
Glue max. PET flakes with glue content	500
Polyolefins content	100
Polyamide content	50
Metals	50
Wood, paper, cellulose	50

Figure 7: Technical parameters of PET flakes reprocessed in rPET InWaste. Modified from source: [66], pg. 10.

The technical parameters of the inputs (PET flakes) may differ from one rPET pellet manufacturer to another. This depends mainly on the process equipment and its ability to cope with contamination in flakes [63–66] as well as the robustness of machinery [43]. In general, significant progress has been made in contaminant removal during the mechanical recycling of PET [1]. Nevertheless, some types of contaminants exist that can introduce technological, qualitative, or health issues – be it solid or volatile contamination.

2.3.2 Contamination in food-grade recycled PET

2.3.2.1 Volatile contamination

Volatile contaminants can enter rPET together with solid contamination (e.g., as residues from cleaning products and other non-food plastics) [67] or can be formed as by-products of the degradation of PET or other contaminants [6, 68]. Volatile contaminants are especially important in final food-grade rPET pellets [2], see Figure 8. The main concern is that the contaminants and their residues may migrate into food or beverages and subsequently cause health risks. The health risks become particularly significant in the case of infants who may drink beverages from PET bottles containing some recycled material. As a result, the requirements have been set with a high degree of caution [2, 69]. In practice, manufacturers must use technologies that remove between 90% to 97% of each type of contaminant [69], but the efficiencies can be even higher than 99.9% in some cases. The efficiencies are evaluated in challenge tests done according to EFSA [66].

The testing is done using EFSA’s approved challenge tests. At the beginning of the testing, the PET flakes are artificially contaminated. The flakes are then subjected to a technological process. The residual concentrations of the introduced contaminants are then measured to evaluate the effectiveness and applicability of the tested technology. Challenge tests map the contamination that remains in food-grade rPET after processing, as well as the ability of contaminants to migrate from food-grade rPET to food [2, 60].

To ensure that the recycling process is assessed as not posing health risks, the residual contamination of observed contaminants in final rPET pellets (c_{res}) must be below the model cut-off concentration (c_{mod}). c_{mod} is determined based on the ability of the contaminant to migrate into the food and subsequently into the exposed person; the default exposed person is usually an infant as these limits are the lowest [2]. See the monitored volatile contaminants and limits in Figure 8.

Surrogate	c_{mod} [mg·kg ⁻¹ PET]
Toluene	0.09
Chlorobenzene	0.09
Methyl salicylate	0.13
Phenylcyclohexane	0.14
Benzophenone	0.16
Lindane	0.31
Methyl stearate	0.32

Figure 8: Observed volatiles in rPET, their molecular weights, and determined model concentration by EFSA.

Modified from source: [2], pg. 24.

Acetaldehyde, for instance, is not the subject of the challenge tests, even though it is the most important molecular contaminant in food-grade rPET because it is formed during PET degradation [70]. Although the migration concentration limit (6.00 mg·l⁻¹) is so high that it cannot be exceeded in the case of bottle recycling (so it does not introduce any health risk), the acetaldehyde content is closely observed by the producers due to product quality [43, 70]. The reason is that relatively low concentrations of residual acetaldehyde (10-25 µg·l⁻¹) lead to a significant defect in the organoleptic properties of a product, especially mineral waters [70].

In the sections above, the volatile compounds impact was discussed. In conclusion, the health risks and hazards of some volatiles are the most severe problems. In this context, the amount of volatiles in the final recyclate determines whether rPET can be used for food-grade [2]. Less threatening, but still crucial for food-grade rPET producers, is the impact on the organoleptic quality of the contents of PET bottles [70].

2.3.2.2 Solid contamination

Solid contamination can be caused mainly by faulty sorted materials or bottle components, such as caps or labels. Solid contaminants are other polymers than PET plus various non-plastic materials [31, 71, 72].

Solid contamination is associated with several impacts, which will be described in more detail in Chapter 2.4. Their presence results in severe degradation, manifested by a reduction in intrinsic viscosity (IV) and molecular weight, and consequently, a deterioration in the quality of rPET [6, 72]. This is because IV is closely related to the material's mechanical properties [73]. While on a molecular scale, PET flakes do not exhibit different properties than the original bottles, extrusion conditions trigger processes that lead to a deterioration in the quality of the resulting granulate [3].

The decrease in IV and the molecular weight is closely connected to polymer chain degradation [72]. This is because PET is formed by a reversible condensation reaction [74]. The reaction can be reversed under specific conditions, such as the presence of moisture, acidic and alkaline contaminants or their acidic and alkaline degradation products, and heat. Consequent depolymerization cleaves the PET chains, thus reducing polymer IV and molecular weight [3, 72, 74].

IV reduction is problematic, especially when it comes to the intended use of rPET. Specific applications require a defined IV range [6]. For virgin PET with an IV of around 0.78 dl·g⁻¹ [75], this is not a problem as its IV is high enough to be used in various applications. However, while the IV of rPET used in filaments can be only 0.65 dl·g⁻¹, the requirements for use in bottles are more strict (0.73–0.80 dl·g⁻¹) [6]. As the IV of the recycled extrusion feedstocks (flakes from PET bottles originally made from virgin PET) is estimated to be approximately 0.72–0.80 dl·g⁻¹, its IV after conventional extrusion would be lower – possibly below the minimal threshold for the use in food-grade rPET. Therefore, the extruded rPET may be of inferior quality compared to the original virgin PET [72].

The screens are another essential step used for melted PET. Usually, screens are used to separate non-melted contamination from the melt [76]. Once not separated, contaminants can form blackspots. Black spots are charred contaminants that passed through melt filtration [5, 76]. They are particularly visible in amorphous pellets and in final PET bottles, as can be seen in Figure 9. The problems with solid contamination will be discussed in detail in Chapter 2.4.



Figure 9: Black spots in amorphous PET pellets. Source: own source.

2.3.3 Extrusion and regranulation

The separation technologies, discussed in Chapter 2.2.2, should deal with the contamination of PET flakes before their reprocessing. Nevertheless, flakes may still contain a certain quantity of solid impurities that do not allow these flakes to be used in food applications. Moreover, it is crucial to address volatile contamination. Thus, additional steps must be employed for decontamination of food-grade rPET [1, 6, 11]. For these reasons, PET flakes are often subjected to extrusion and further steps after extrusion, so-called regranulation. These additional steps involve many technologies [77]. In this thesis, the focus will be laid on SSP and LSP.

2.3.3.1 Extrusion and melt filtration

PET flakes are obtained by conventional mechanical recycling, but they are often not a final product. Flakes can undergo melt reprocessing, i.e., extrusion. Extrusion needs to be introduced for rPET to remove the remaining contamination, especially in the case of food-grade rPET, particularly PET bottles [1, 60]. Even though a considerable volume of contaminants is removed by washing and sorting, these two steps cannot deal with the organic contaminants embedded within the polymer structure. Moreover, PET flakes are reprocessed into granules that are easier to handle during extrusion [1, 6]. Processing into small, homogeneous granules or pellets also results in homogeneous distribution of contaminants, thereby diluting the contamination [1].

Melt filtration can be employed during extrusion. Melt filtration was proven to be important for the rPET quality [1, 78] as it allows the removal of contaminants with higher melting temperatures than PET – especially various metals. The PET melt passes through the screens while solid impurities are filtered out [76, 79]. A screen after use is shown in Figure 10.



Figure 10: Used filtration screen covered with aluminium and metal as well as some charred particles. Source: own source.

Two general categories of screen changers exist based on the screen change method: discontinuous and continuous screen-changing devices. The discontinuous screen change requires an operator to change the screens whenever needed. This arrangement is advantageous in terms of lower price. Nevertheless, the screen change, especially when it must be done often in case of more contaminated flakes batch, can significantly interrupt the whole production process and can lead to the formation of black spots [76]. Thus, continuous screen change was recommended by scientists as it reduces the number of process interruptions [78]. Screens are usually situated behind the extruder [77]. If LSP is included, screens are placed after the LSP reactors [80].

To prevent the degradation of rPET polymeric chains and decrease of IV during the extrusion and melt filtration process, several technologies were designed and some of them are implemented in practice [1, 77]. Moreover, technology that eliminates volatile contamination is needed: for example, LSP [30] and SSP [1].

2.3.3.2 SSP and LSP

SSP is performed with solid rPET pellets after extrusion. Pellets are exposed to temperatures between 200 and 240 °C, which is a temperature above PET glass transition, but below its melting point (thus, pellets remain unmelted). Due to the applied conditions, thermal degradation is prevented [81] and the condensation of the polymer takes place. Condensation results in longer PET chains formation and an increase of IV. Usually, a nitrogen atmosphere or vacuum is employed in SSP to remove the condensation by-products of the process and to prevent oxidation [82, 83]. Moreover, the remaining molecular contamination absorbed into PET (limonene, acetaldehyde, etc.) is removed [60].

LSP, unlike SSP, occurs during the melt phase before pelletising. PET melt is under a high vacuum, which improves the condensation reaction. By the developers, NGR, LSP is claimed to improve IV quickly, with lesser energy demand, and thus to be economical-friendly for rPET pellets producers [80]. These claims were supported by scientific literature [30]. LSP also eliminates molecular contaminants from material [30, 80].

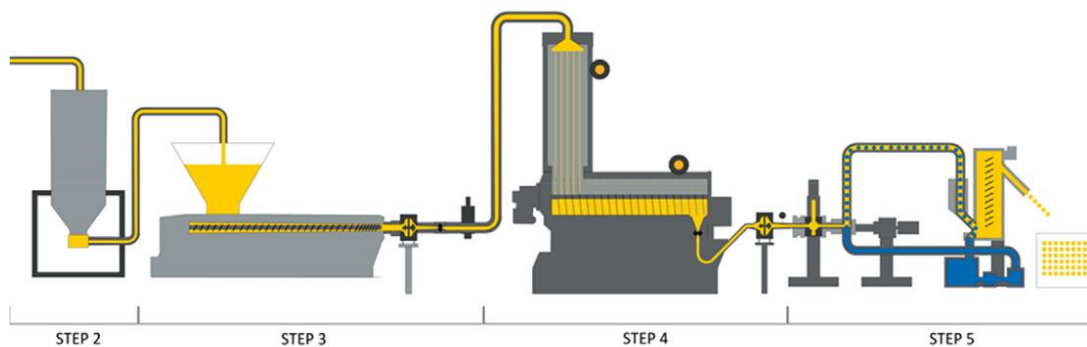


Figure 11: A scheme of rPET pellet production technology with LSP. Source: [66], pg. 6.

Figure 11 shows a possibility of how reprocessing of PET flakes can be arranged. In step 2, the flakes are dried and delivered to step 3: extrusion, where the flakes are melted. The melt is delivered in LSP (step 4). Finally, during pelletising (step 5), melt is cooled and pellets are produced [66].

2.4 Contamination of PET

Every material other than PET is considered as contamination – whether it concerns plastics (PETG, PVC), various materials such as paper [72], metals [71], or volatile compounds [2]. This chapter will focus on larger particle impurities because the molecular contamination was discussed in Chapter 2.3.2.1.

Contaminants influence many properties of rPET. In general, polymeric and non-polymeric particle contaminants cause haziness, i.e., a decrease in transparency of rPET [5, 75] and are a reason for the darkening of a material [5]. Contaminants can appear as black spots [72, 84], higher opacity can be observed [84], or harmful substances can be formed [5, 71]. Also, IV reduction due to degradation is typical for many contaminants [68, 72, 85].

Especially, other polymers are challenging contaminants. Generally, they are remarkably diverse materials that differ from each other in terms of properties such as melting point, resistance towards heat and chemicals, etc. Different types of polymers are incompatible, and their presence in single-type plastic recyclate leads to quality deterioration [3, 11] and degradation [3]. Some of the polymeric contaminants together with antimony originating from antimony trioxide (a catalyst used in virgin PET production) cause grey discolouration of rPET [31]. On the other hand, some of the contaminants (such as polyamide – PA), cause yellowing [5].

Yellowish discolouration, occurring due to the non-homogeneous material feedstock, can be suppressed by colour additives [86]. Yellowing is also induced by the degradation of PET [87] and can be reduced by blue-coloured additives [75]. Moreover, the additives can improve other properties, such as IV, but it is crucial to consider the subsequent applicability of the recyclate. In other words, not all additives can be used in food-grade rPET [88]. Moreover, although additives suppress yellow colour, they make recyclate greyer. The greying can be indicated by spectrophotometry analysis [75].

As a result, contamination should be eliminated as effectively as possible in order to prevent its negative impacts. To make its separation more effective, it is useful to know the contaminants: their origin, the possibilities to separate them, and the effects on final rPET recyclate once the specific contaminant penetrates the process and final product. This chapter will describe the most common contaminants and how to manage them.

2.4.1 PET and its modifications

In some cases, PET itself can be a contaminant. This applies especially to PETG, PET flakes with pigments, or PET trays [11, 44, 89]. Their separation is complicated because separating two identical polymers is difficult as the properties can remain the same. All mentioned have a higher density than water which prevents them from separation by flotation [45]. Another technology, NIR separation, is not effective due to the similar material used [42].

2.4.1.1 PET with pigments and opaques

Ink is used for the best-before-date and batch number marking on a bottle. As a result, PET contains pigments and inks, which cause discolouration of rPET, in particular darkening [5]. Therefore, using lasers to engrave these details is recommended [90]. Moreover, ink can cause the scission of polymeric chains and reduce rPET IV [5]. However, this type of contamination cannot be avoided and should therefore be considered in a feedstock [43].

PET opaques are not transparent as they contain a high proportion of pigments. They are mainly used in milk bottles. They are not compatible to be recycled with transparent PET bottles [91]. In addition, they are often used for cleaning products and cosmetics packaging and are thus susceptible to molecular contamination [92]. Non-food packaging may only be present in a limited amount (5%) in the PET recycling stream [2].

2.4.1.2 PET trays

Besides PET bottles, PET is used to produce trays. However, recycling of PET trays is more complicated and not widespread. This is because the trays consist of multiple polymers in layers. Usually, polyethylene (PE) is used under the surface layer in PET trays. An ethylene vinyl alcohol (EVOH) layer can also be used. Since PE and EVOH are inseparable in mechanical recycling, trays would introduce PE and EVOH contamination into rPET [93].

Separation by NIR is hampered due to the fact there are multiple layers [94] and PET layer has the same polymeric spectrum [42, 43]. However, since the visual difference between trays and bottles is significant, the introduction of machine vision sorting seems to be a promising option for PET tray separation [42].

During regranulation, EVOH and PE are usually introduced as contamination with the penetrating trays. The effects of PE will be described in Chapter 2.4.2.2. The impacts of EVOH are the decrease of IV and the disruption of crystallization process [5]. Additionally, there is a problem with the IV of trays. It is estimated to be very low compared to the bottles, thus they are not suitable to be recycled into PET bottles [95].

2.4.1.3 Non-food PET bottles

Non-food PET bottles are collected with beverage PET bottles in collection systems other than PET bottle DRS. It was observed that material obtained from DRS contained significantly lower levels of non-food bottles than from the separate collection (0% and 17–24%, respectively). When faulty sorted, they can enter PET recycling stream [5].

As NIR sorting is not effective in this case [42, 43], other technologies must be introduced to separate non-food bottles. Usually, manual [5] or automated machine vision sorting can be the option [42].

Because non-food PET bottles are mainly used for detergents, house chemicals, drugs, or cosmetics, they are prone to contamination by hazardous compounds. These compounds, so-called non-intentionally added substances (NIAS) are harmful and can pose a health risk to consumers [67]. For food-grade rPET, EFSA allows only for a 5% presence of non-food PET in the feedstock [2].

2.4.1.4 Polyamide (PA) and barrier bottle flakes

In some cases, a barrier layer made of polyamide (PA) polymers is found within PET bottles. PA can occur as a separate interlayer or can be incorporated into PET [90]. This is because the barrier layer prevents the penetration of gas molecules more effectively than PET alone, thus preserves the quality of the beverage. Barriers are mainly found in beer [96], juices, or dairy bottles [5].

Based on practical experience, PA, once peeled off the flake, can be recognized by NIR and separated [43]. In the case of a barrier bottle flakes, barrier layer is embedded in a bottle, thus mechanically inseparable. However, manual or machine vision sorting can be used to separate the bottles of concern [43]. Even though the barrier material and PET spectra might differ, NIR separation is not reliable as only the PET layer can be scanned, and the particle would be evaluated as pure PET flake [94]. In laboratory analysis, the presence of barriers in bottles can be recognized. For the clear bottles, a spectrophotometric measurement can be used to identify bottles with and without a barrier [5].

Smaller quantities of barrier bottles are not problematic; the problem arises with higher amounts. The researchers observed more intense yellowing and a decrease in rPET IV already at 0.6% barrier flake content [5]. The problem is that as well as PA, a barrier is prone to yellowing once heated [5, 97]. Yellowing is shown in Figure 12: on the left, yellowing cannot be observed as those flakes were not exposed to heat. On the right, flakes from the same batch were exposed to heat. Heating led to the yellowing of barrier bottle flakes, highlighted by red arrows. The yellowing effect influences the colour of the final pellets greatly [5]. There is a possibility to solve the yellowish discoloration using blue-

coloured additive [98]. But blue additive dosage must be determined according to a barrier flake content within a batch, otherwise, the pellets tend to be yellower (higher b^*) and greenish (lower a^*). Based on experience from practice, this can lead to additional difficulties in pellet production as the additive dosing has to be frequently controlled and corrected several times – especially, once there is an inconsistency in barrier bottle flakes content amongst the batches [43]. Besides the yellowing effect, benzene can be formed due to the presence of some barrier compounds [68].

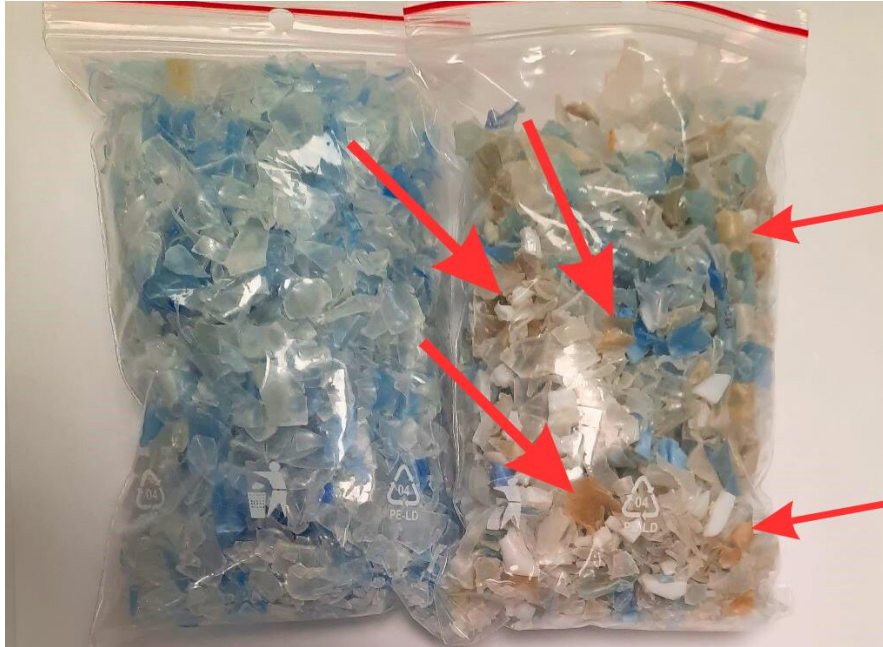


Figure 12: Barrier flakes before (left bag) and after heating (right bag). Source: own source.

2.4.1.5 Polyethylene Terephthalate Glycol (PETG)

PETG is PET modified with different types of glycols, such as 1,4-cyclohexanedimethanol [99]. PETG use is on the rise in 3D printing, but it is also used for many other applications [100], including the manufacture of containers [101]. In the case of PET bottles, it is preferred as a sleeve label due to its ability to shrink and its good printability. The sleeve is a specific type of label, which shrinks and thus replicates the surface of the bottle without the need for adhesives [89]. Moreover, by some sources, PETG labels are considered to be easily recycled together with PET as opposed to PVC shrink labels [102].

In practice, PETG is not compatible with PET in the recycling process and therefore should be eliminated from the input. However, more complications arise in this matter. PETG is difficult to separate from PET due to equivalent properties (density, NIR spectra). For these reasons, companies in the recycling industry encourage PET bottle producers to use other materials. However, PET bottle producers do not want to give up such a suitable material [44, 89].

Nonetheless, the presence of PETG causes serious problems during regranulation. Firstly, because of its lower melting temperature than PET, it can form a sticky mass during the drying and transfer process, thus blocking the pipes before extrusion. Another undesired effect is the yellowing of rPET [89]. Additionally, PETG consists of glycol [99]. Ethylene glycol is used in the chemical recycling of PET (glycolysis) for PET degradation, i.e., for breaking polymeric chains [14]. Based on the conditions, the same effect may occur once glycol is released during regranulation, resulting in a decrease of rPET IV.

2.4.2 Other polymers

Other polymers³ originate both from faulty-sorted objects as well as from bottle components [31]. Usually, those contaminants can be removed in mechanical recycling. Nevertheless, other polymers can also be blended with PET in a form of an inseparable composite. Polymeric composites are used to improve material properties as a layer or the bottle itself [90]. Bottles with components made of different polymers and polymeric blends have to be either separated from the waste stream in the beginning or they can enter recycling in very limited amounts [5, 31, 103]. Still, PET material will always contain a certain degree of polymeric contamination [50]. Many polymeric types can be expected in PET polymeric contamination. In the contamination analysis of PET flakes from PET bottles from Central Europe, 17 polymers and polymeric blends were identified [104]. Only those of particular importance for EFSA approval and regranulation process will be described more closely in this thesis.

2.4.2.1 Polyvinylchloride (PVC)

PVC is a widely used thermoplastic, especially because it is a resistant material despite its low price [105]. Therefore, it is one of the most produced and demanded plastics [8, 106] with a wide range of applications [8]. It is also used in bottles [72] and shrink labels [90], therefore a certain level of PVC content needs to be expected in plastics and bottle collection [29]. PVC separation is hindered due to the same density as PET. As a result, PET and PVC cannot be separated using only the conventional flotation method [72]. NIR separation technology proved to be efficient [48]. Separation efficiencies close to 100% lead to higher material and financial losses [59]. During mechanical recycling, contamination and complicated detection are not the only problems arising from sleeve labels made of inappropriate material (PVC, PS): PET can be hard to recognize through the sleeve label [42]. Moreover, sleeve labels are harder to remove than the conventional labels glued with adhesive [107]. Thus, such bottles are sorted out, resulting in decrease of PET bottles recycling rate [108].

PVC is of major interest in PET bottle regranulation because it causes severe degradation of rPET. Concerning molecular contamination, chlorine is formed during PVC thermal degradation and consequently, hydrogen chloride and hydrochloric acid are both mentioned as responsible for PET chain cleavage and IV reduction [5, 29]. Additionally, PVC is the main precursor for the formation of benzene – either via catalytic reaction between PVC degradation products and PET chains or via degradation of PVC itself; both occurring at temperatures used during PET reprocessing process. In other words, a positive correlation exists between PVC content and benzene concentration [68].

The second effect is the formation of cyclic oligomers. The discoloration effect can be observed even in the cases when a very small amount of PVC is present [72]. Moreover, PVC contamination is claimed to cause black spots in rPET granules [72, 84]. Black spots occur due to the infiltrated fine particles of PVC which turn black when exposed to heat [109]. Negative impacts on rPET quality, such as discolouration or degradation, can be caused by even a very low PVC amount (100 ppm) [29]. Other works state that even 1 ppm of PVC can become problematic [72].

2.4.2.2 Polyolefins

Polyolefins such as PE and polypropylene (PP) enter the recycling stream as materials used for labels and enclosures [72]. Also, PET trays can be a source of PE contamination as a multi-layer product [5]. Usually, their presence is not problematic, as they are easily separated by flotation. Nevertheless, heavy printing can result in increased density and negatively affect the flotation efficiency [72]. Additionally,

³ The term *other polymers* is not scientifically accurate, as these are purely plastic materials. The group *other contaminants* (see in the following pages) also includes polymers such as wood and rubber. However, this terminology is used in practice, and it is therefore important to point out to the reader that in the context of this thesis, the term *other polymers* refers exclusively to plastic materials; the term *other contaminants* to other non-plastic materials, including non-plastic polymers.

flotation efficiency can be lower due to overfeeding of the flotation tank, and is further influenced by particle size and shape [45]. Nevertheless, NIR, colour, or machine vision separators can remove penetrating polyolefin particles; a key factor being their visual characteristic and device sorting sensitivity [42, 51, 94, 110].

General adverse effects such as haze occur during regranulation [5]. It was found that the presence of PP reduces crystallinity in multiply extruded rPET [111]. Based on practical experience, polyolefins can form bulky particles during extrusion [43].

2.4.2.3 Polystyrene (PS)

Polystyrene (PS) is used in a variety of applications, such as household goods, building materials, and packaging [112]. It can also be found in the PET bottle label material or sleeves [89, 108].

PS entering the recycling line can contaminate both PET flakes and polyolefin streams. That depends on PS particle density [112] which determines whether the particle will sink or float in the flotation tank [89]. PS can be separated by NIR [110].

If the PET stream is contaminated, the presence of PS during regranulation can cause the yellowing of rPET and the general deterioration of its quality [89]. It also has a negative effect during SSP as it hinders the formation of longer PET chains [31]. In addition, the thermal degradation of PS produces styrene, which can migrate into food. Higher styrene concentrations are not typical for high-quality rPET but can be found in lower-quality rPET [68].

2.4.2.4 Polylactic acid (PLA)

Polylactic acid (PLA) is used in 3D printing [113] and as a packaging material [114]. It is a bioplastic produced from renewable sources regarded to be compostable. Due to the reasons mentioned, it is considered as an environment-friendly compostable alternative to conventional fossil-based plastics [115], although some papers argue such statements are misleading and not legitimate [59, 116, 117]. A frequently cited argument in the literature is that PLA will introduce additional contamination into the recycling streams of conventional plastics due to the problematic management of biodegradable plastic waste [59, 117, 118]. PLA has been already analysed in PET flakes analysis [104].

NIR separation is possible at efficiencies ranging from 97.5% [117, 119] to 99.6% [59]. Thus, the main concern is an increase of PLA in the PET bottle waste stream, as predictions expect increasing PLA use in the future. At a certain level of PLA in the PET stream, NIR separation may not be sufficient to prevent deterioration in the quality of the final rPET because even with the highest separation efficiency, the remaining PLA could still remain problematic [59, 117].

An undesired effect of PLA in rPET is an increase in material opacity and haziness [84]. The melting temperature of PLA is much lower than of PET. As a result, PLA causes degradation and yellowing. In addition, melted PLA can form sticky residues, leading to lump formation and associated technological problems, for example, during the drying of PET flakes [59]. Plus, PLA has a negative impact on the speed of IV recovery in SSP and affects crystallization, probably due to the formation of co-polymers between PLA and PET [5]. Adverse effects are evident when 1% PLA is present in the recyclate [117]; though other works reported lower levels (1000 ppm) to be problematic [90].

2.4.3 Metal and non-metal non-polymer contaminants

2.4.3.1 Paper

Paper can be found in bottle labels [72]. Larger pieces of paper can be separated by screens in melt separators [76]. The problem is that during the recycling process, the paper breaks down into small pieces that are difficult to separate and further merge with the positive flow [90].

Once paper is exposed to higher temperatures, especially during regranulation, acidic products are formed from the paper fibres, resulting in an IV decrease [72]. As a result, paper causes black spots formation in pellets [5].

2.4.3.2 Wood

Wood can occur in feedstock due to poor sorting or from pallets. For wood removal, manual or machine vision sorting can be employed. Additionally, during regranulation, wood can be separated in melt filtration. Based on practical experience, larger quantities of wood require more frequent screen changes; otherwise, the screens could clog [43]. As non-meltable in the process conditions, wood penetrating further with the melt causes black spots in pellets [5, 43].

2.4.3.3 Rubber

Usually, rubber particles originate from poorly-sorted waste material or, as was observed in practice, from the conveyor belts [43]. A significant proportion of rubber is dark-coloured, thus cannot be separated from the flakes by NIR sorting machines [49, 50]. Dark-coloured rubber particles can be detected either manually, by machine vision, or by colour-based separation [39, 42, 51].

As well as in the case of wood, rubber can be separated by melt filtration during regranulation [79], but based on practical experience, higher quantities can clog the screens [43]. Thus, more frequent screen change is required for higher wood contamination. Also, as a non-melting contaminant, penetrating particles can form black spots in rPET pellets [5, 43].

2.4.3.4 Metals

Metal contamination can stem from the sorting once not properly done. Nevertheless, PET bottles themselves can contain metal components, such as caps or labels [5]. Metal separation was described in Chapter 2.2.2.3. In addition to these technologies, melt filtration acts as another separation step due to the higher melting points than PET [76].

Once not separated, metals can negatively affect the resulting rPET quality. On a molecular scale, metals act as catalysts that accelerate the thermal degradation of PET during regranulation [85]. Besides that, metals in the fed material can also damage the technology itself, for example, the extruders [11]. Nevertheless, metal contamination is not linked only to faulty sorting – metals are also embedded in the polymer itself as residues of catalysts. Antimony is typical as a catalyst in PET, but cobalt or manganese can also be used [72]. Besides the origin from the catalytic process, some metals (palladium, cadmium, chromium) can occur in rPET due to cross-contamination from other wastes [120]. Aside from considerable health risks arising from the presence of metal contaminants in rPET [71], they also cause the non-homogeneity of rPET or change in melt properties [72].

2.4.4 Moisture content

Water remains in the flakes due to poor drying during production [121]. Moreover, as observed in practice, higher moisture content can also be a result of prolonged storage in inappropriate conditions, especially after exposure to weathering [43].

The moisture content in PET flakes is a controlled parameter because water can cleave the PET bonds and reduce polymer IV and molecular weight [72]. This degradation mechanism is called hydrolysis. The hydrolysis reaction occurs at higher temperatures (280 °C, the temperature used in the process) in PET in presence of water [6]. The detailed mechanism is shown in Figure 13.

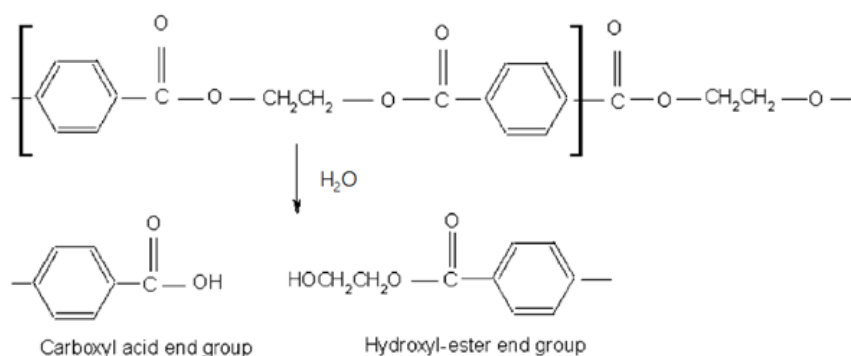


Figure 13: PET hydrolysis. Modified from source: [6], pg. 1460.

Moreover, based on practical experience, moisture in PET flakes causes weight loss during regranulation. When processing a bulky batch (e.g., one hundred tonnes of PET flakes) with 1% of moisture, one tonne of the total reprocessed volume will be just the water escaping the process without creating any value. It can also be deduced that the higher the moisture content, the more water needs to evaporate, and the energy consumption of the process increases with the moisture level [43]. Generally, it is essential to dry PET flakes thoroughly before reprocessing to a low moisture level (0.02%) to minimize IV reduction [6].

2.4.5 Other parameters

2.4.5.1 Adhesives

Adhesives are used to glue the labels. They should be removed in the PET flake washing step. Unfortunately, not all types of adhesives can be washed off easily, and adhesive residues may remain on the flake [72].

During the reprocessing, adhesive residuals and higher temperatures yield acidic products [72, 85] – mainly resin and abietic acid [14]. Subsequently, these products cause the hydrolysis of PET chains, reducing molecular weight and IV [72, 85]. The use of adhesives can be eliminated by the use of shrink labels [89]; nevertheless, those made of PVC, PS, and PETG are problematic, as stated above. Shrink sleeves are only recommended if they are made of suitable materials (PE), are easily removable, and should leave part of the PET bottle exposed so that the PET remains detectable by automatic sorting systems [90]. Flakes with adhesives are hard to separate, as only some part of the flake can be covered, and thus is hardly recognizable by NIR or different sorting systems [43].

2.4.5.2 PET dust

PET dust is a fraction of particles that pass through a fine mesh sieve. Higher PET dust content indicates poor dust separation in PET flake production. The dust particles can be of various origins, but usually, dust is rich in impurities [3] and polymeric contamination [5].

As a result, dust causes deterioration of rPET quality [3]. According to the practical experience, during regranulation, homogeneous heating of flakes becomes challenging for material with higher PET dust [43]. Thus, good dust removal from PET flakes can result in a better quality of rPET [3].

2.4.5.3 Bulk density

The bulk density depends on the size and thickness of the flakes. As the thickness of PET bottles has been decreasing in recent years, an increase in bulk density is possible mainly through a reduction in flake size [4]. In general, based on practice, smaller flakes (i.e., batches with higher bulk density) make the washing process more efficient because it is easier to wash smaller pieces [43].

As shown in Figure 11, the flakes are dried in a silo and then fed to the extruder in the regranulation process. However, silos have a limited volume; thus, if PET flakes are of lower bulk density, they can accommodate smaller flake weights [43]. Moreover, bulk density influences drying time, hence the cost of the energy required for drying [4]. Based on practice, the correct flake size also ensures the continuity of the feed to the extruder and the stability of the entire production [43].

2.5 Theory on the methods

For a better understanding of the method sections and measurement results, the principles will be described briefly in this short chapter.

2.5.1 Spectrophotometry: CIELAB method

Konica Minolta CM-5 spectrophotometer uses a standardized method CIELAB $L^*a^*b^*$ characteristics to determine a colour of a sample [122]; i.e., 3-dimensional space formed by three axes L^* , a^* , and b^* according to the standard EN ISO/CIE 11664-4:2019 [123]. L^* axis represents lightness, ranging from 0 to 100, while 0 represents black and 100 white colours. Parameters a^* and b^* specify the colour further: negative a^* means that the sample is green, and positive a^* represents a higher presence of red. b^* axis moves from blue to yellow, i.e., negative b^* is measured for blue samples, whereas positive b^* is obtained once the sample is yellow [124]. The trends can be also seen in Figure 14.

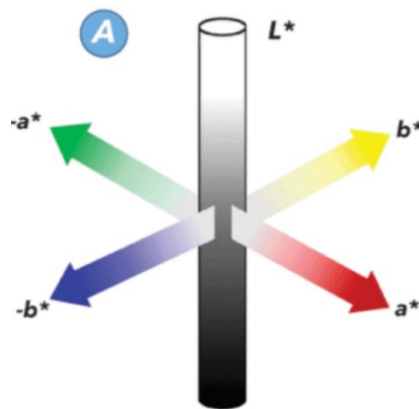


Figure 14: Graphical display of colour space using CIE $L^*a^*b^*$ method. Source: [125], pg. 19.

Still, measuring colour using spectrophotometers is problematic in terms of reproducibility. The spectrophotometers can use different methods than CIELAB, and even though two spectrophotometers use the same principle, the results may still differ and thus should be a matter of correlation [124].

2.5.2 Fourier transform infrared spectroscopy with attenuated total reflection

Infrared spectrometry is a spectroscopic method that works on the principle of molecular vibrations induced by a beam of IR radiation [126]. FTIR, i.e., Fourier transform infrared spectroscopy, guarantees a higher quality of the measurement and the results themselves by, e.g., reducing the signal-to-noise ratio [127]. FTIR is typical for high speed of measurements due to effective multiple wavelengths investigation [126]. The ATR method is specific in that it interacts with the sample only on the order of micrometres [127, 128]. Although this method cannot capture the complexity of the sample, if the sample is homogeneous or if the purpose is to measure surface layers, this limitation is not an obstacle [128]. In addition, ATR analyses can be performed relatively quickly without the need for sample preparation [129]. FTIR-ATR is also applicable to strongly absorbing dark samples [128].

2.5.3 Intrinsic viscosity and plastometers

IV is calculated based on measurement of two other rheological properties: mass-flow rate and volume-flow rate [43]. These two properties are determined using plastometer and a cylindrical form of a measured polymer. The plastic cylinder is melted in a chamber with elevated temperature and forced through a narrow nozzle with defined dimensions. As a result, plastic fibre leaves the nozzle. This fibre is cut several times using a uniform time interval. The cuttings are weighted, and the weights are used for all the calculations [43, 130].

2.5.4 Correlation visualisation with ggpairs() function and Pearson coefficient

The visualisation was performed in R using the ggpairs() function from the GGally package. This function can visually show the pairs of variables quickly and return Pearson's coefficient values [131].

With values ranging from -1 to 1, the Pearson correlation coefficient determines the linear relationship and slope of the variables. When the value is positive and close to 1, it is a positive correlation. Approaching -1 means a negative correlation. However, if the value is close to 0, it is not a correlation, i.e., no linear dependence [132].

2.5.5 Hypotheses summary

Because this thesis is associated with several hypotheses that will be discussed at various points in the thesis, it was deemed appropriate to summarise these hypotheses in one place. The following hypotheses arose regarding suppliers, collection schemes, and type of market:

- CM (customer material, see the explanation in Chapter 3.1) suppliers may not comply with some internal parameters, as they follow only EFSA parameters,
- some of the measured characteristics will depend on the type of collection system,
- a clear differentiation of the results of some characteristics based on the market type.

In addition, hypotheses have arisen in relation to the measurement methods:

- the use of a plastometer for IV measurement will be a more appropriate than dilute solution viscosity method,
- replacing the current method of visual evaluation using a different parameter.

Finally, a hypothetical dependency between the following parameters was assumed:

- PVC, other polymers, other contaminants, PA, PET trays (correlation between all parameters with each other),
- light blue flakes individually with visual evaluation, light-coloured flakes, dark-coloured flakes, and barrier flakes,
- barrier flakes with PA and visual evaluation,
- spectrophotometric parameters (L^* , a^* , b^*) with visual evaluation (assuming the highest correlation coefficient with the L^* parameter),
- spectrophotometric parameters (L^* , a^* , b^*) with barrier flakes (assuming highest correlation coefficient with parameter b^*),
- spectrophotometric parameters (L^* , a^* , b^*) with light blue flakes (assuming the highest correlation coefficient with parameter b^*),
- PET dust and bulk density.

The main aim of this thesis is to evaluate the quality of flakes on the EU market, to offer processors the missing information, and to hypothetically contribute to the optimization of the PET bottle recycling industry.

3 EXPERIMENTAL PART

3.1 Materials

PET flake batches from 18 suppliers were used as materials. Batches of interest were selected from the total volume of batches that were delivered within two-year period. The number of batches per supplier ranged from one to 48. A total of 158 batches were analysed. The entire population, i.e., 158 batches, was measured at the initial QC inspection and analysed for contamination. In spectrophotometry, 145 batches were measured due to a lack of material for some samples. For the viscosity measurements, one measurement each was taken for single-batch samples, two measurements each for two-batch samples, and three or more measurements each for multi-batch samples.

To maintain anonymity of suppliers, letter codes were used to identify them. Some letters are suffixed with the abbreviation CM, meaning "customer material". This material meets the parameters set by EFSA but may not meet the processor's requirements. It is then up to the processor's judgment whether the deteriorated properties may damage the technology or interfere with the conventional production of rPET pellets. In general, the customer's material is expected to be of inferior in some respect when compared to material purchased by the processor (i.e., without the CM abbreviation).

A detailed list of suppliers and relevant batches is given in Figure 15. Since it was hypothesized that some of the measured characteristics are related to the collection system, information on the type of collection system from which the recycled material comes is also provided. Furthermore, it was decided to split the suppliers into those from the Eastern and Western parts of the EU. The reason is that the input quality and used technology may differ, influencing the resulting quality of PET flakes. The dividing line was formed by Poland, the Czech Republic, Slovakia, Hungary, and Slovenia, with these countries already considered as the Eastern part of the EU market. Both information, i.e., the type of collection system and the allocation to one of the markets, was determined by knowing where the producers' plants were located. However, the countries of operation were assessed as confidential information and thus will not be provided in this thesis.

Supplier Code	Number of batches	DRS/Mono-collection	EU market
A	7	Yes	E
B	3	No	W
C	13	Yes	E
D	5	Yes/No	E
D_CM	30	Yes/No	E
E	1	No	E
F	11	No	E
G	1	No	E
H_CM	2	Yes	W
I_CM	6	No	W
K_CM	10	Yes	W
L_CM	2	No	W
M_CM	5	Yes	W
N	6	Yes	E
O	48	No	E
P	2	Yes	E
Q	2	No	E
R	4	No	E

Figure 15: Information on suppliers. Suppliers D and D_CM are suppliers with two plants operating in different countries with different collection systems. Source: own source.

3.2 Methods

According to the standard EN 15348:2014 which describes how the characterization of rPET (flakes and pellets) should be done, there are seven mandatory and six optional procedures [133]. All mandatory analyses (except for the maximal size of the particles) and some optional ones (MFR, IV, colour) were used in this thesis. Some procedures were done according to no norm: a more elaborative description of such methods will be provided in the respective sections.

3.2.1 Entrance quality control analysis parameters

3.2.1.1 *Moisture content*

The determination of moisture content in flakes was performed in accordance with EN 15348:2014. In brief, the weight difference between raw and dried samples was used to determine the moisture content. 50 g of the sample was put into an oven preheated at 150 °C for 4 hours [133]. The analysis was done for four samples per batch, and an average was used as a final value [43].

3.2.1.2 *Contamination and other particles*

The determination of contamination according to the standard EN 15348:2014 was done as follows: 1000 g of a sample was heated for 1 hour at 220 °C. Contaminants were detected visually based on changes in appearance (e.g., darkening in PVC or melting in polyolefins) [133].

EN 15348:2014 is intended to evaluate PVC and polyolefins [133]; nevertheless, several other groups were identified on the same principle:

- PVC,
- PA,
- polyolefins together with polymers other than PVC, PET trays, and PA,
- PET trays,
- other non-polymeric contaminants (metals, wood, textile fibres, and so on) [43].

The standard was used also to evaluate other particles within a sample. Their amount was measured because they can influence some rPET properties significantly, especially a colour. The results are not in ppm but in percentage, as other particles were usually weightier. Among other substances, there were:

- flakes from barrier bottles,
- flakes with glue,
- coloured flakes (except for light blue flakes, light and dark colours are evaluated separately),
- opaque PET flakes,
- light blue flakes [43].

3.2.1.3 *Visual evaluation*

The visual evaluation aims to define the degree of degradation and quality of washing in flakes batches. Degradation was evaluated before and after the moisture content test, whereas the quality of washing was most apparent after heating. This evaluation was done visually; samples were compared to etalons to be matched to a corresponding grade. Degradation and washing were assessed individually, but a final grade is an average of both – value 1 being the best result and value 5 the worst [43].

3.2.1.4 *PET dust content*

Determination of PET dust content was done based on the norm EN 15348:2014. The dust content was the fraction of the particles in the flake batch, that could pass through 1 mm mesh, and the weight after sieving was used. Then, the dust fraction was determined in percentage [133]. Polymeric constitution was not determined further as these fine particles were hard to define visually or by FTIR-ATR instrumentation.

3.2.1.5 Bulk density

The bulk density was determined according to the standard EN ISO 60:1999. A standardized funnel and measuring cylinder were used. Flakes were slowly poured over the funnel into the cylinder up to the rim; the sample with the cylinder was then weighed [134]. To account for flake heterogeneity, the measurements were taken three times and the average of the three measurements was used as the result [43].

3.2.2 Spectrophotometry (L^* , a^* , b^*)

The reflectance analysis was performed with a spectrophotometer CM-5 by Konica Minolta according to the principles given by the standard EN ISO/CIE 11664-4:2019 [123]. The area used was 30 mm, the observer was set to 10° (Primary D65). During the measurements, the cuvette was filled with flakes up to a mark and measured [43]. The flake samples were often very heterogeneous – especially these with blue, degraded, or coloured flakes and opaques. Thus, the measurement was done three times per batch with a different flake fraction from the batch sample. The average of three measurements was used as final value for further data processing. This way, 145 samples representing individual batches were measured.

3.2.3 Determination of intrinsic viscosity

Measurement of viscosity was done according to EN ISO 1133-2:2011 [130]. During standard operation, this method is used to evaluate the final rPET pellets' quality, not for the flakes themselves [43]. Thus, a method needed to be developed that could be used to measure flake samples. As it was complicated to produce a cylinder-like sample from a flake batch, only the fragments of PET bottlenecks were used. 10 ml of bottleneck fragments were placed in an aluminium container for each measured batch. After drying, the content of the container was placed into the device and measured. Samples consisting of the bottlenecks were easier to handle, and although relating the final absolute IV value to the whole batch could be problematic, using the same measurement methodology for all samples at least provided data for comparison.

For the measurement, capillary plastometer Dynisco, model LMI 5000 was used. Device method A/B [135] and temperature 285 °C were selected. The sample was pre-heated for 300 s; the applied load weight was 2 160 g. Three clippings were produced – each after 10 s. The number of flags was set to 3; their length was 2 mm [43].

3.2.4 Identification of other polymers and other contaminants

After the entrance QC analysis, all contaminants separated from one batch of one producer were kept in a bag and archived. Within these samples, all particles representing contamination (i.e., PVC, PA, other polymers, blisters, and non-polymeric contaminants) were further separated into groups based on their visual properties. These groups were established in a previous thesis based on colour, opacity, and visual characteristics, and there were 59 groups containing plastic contaminants and 22 groups with non-plastic contaminants [104]. This division is now used as an aid in contaminant analysis for laboratory personnel [43] and was also used as a guide for this more detailed separation.

Evaluation of other polymeric and non-polymeric contaminants based on groups for one batch of PET flakes is shown in Figure 16. Each container represents one group. Each group was weighted, and individual group percentage was calculated from the total weight of contaminants in the related batch. Based on this information, it was further possible to inspect the frequency of occurrence for individual groups, batches with higher contaminant heterogeneity, and the relation of groups to the specific producers.

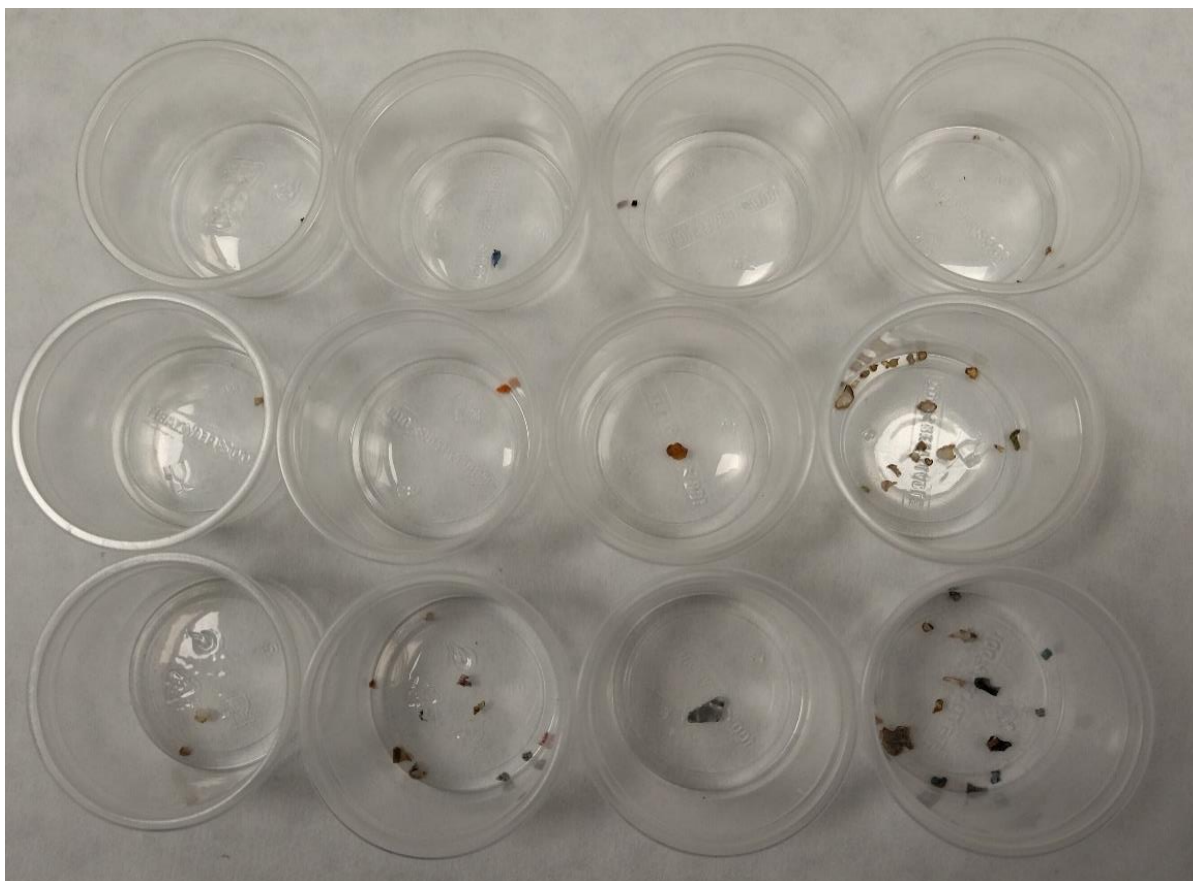


Figure 16: Evaluation of other polymeric contaminants based on groups for one batch of PET flakes. Source: own source.

3.2.4.1 FTIR-ATR measurement

Once a polymer was hard to determine and did not fit into the existing groups, the FTIR-ATR and subsequent spectral analysis were used to determine the polymeric type. FTIR spectrometer ALPHA II was used together with the ALPHA-P module for ATR measurements. ALPHA-P used Diamond 1; Refl #24E68AB82D as a crystal. Parameters of the measurements were set as follows: 32 as a number of scans and wavelength ranging from 400 to 4000 nm. The measurement and spectra evaluation were done in the software programme OPUS; evaluation was realised with the Spectrum Search function. For the comparison, spectra from purchased libraries (that are part of the OPUS software) were used. For new frequent contaminants that were not in the original grouping, new groups were established.

Some contaminants, that were assessed to some already-established group, were also analysed. The analysis aimed to verify the existing grouping of contaminants. As was mentioned, laboratory personnel use this partitioning as a guide when sorting the contaminating particles in incoming PET flakes [43], and it was also used for this analysis. Therefore, it was decided to verify this division and update the list for the laboratory staff in case of discrepancies. This analysis was performed on the FTIR-ATR instrument, and the contaminants were evaluated by comparing the purchased libraries in OPUS using the Spectrum Search function. Samples from the 18 most frequently occurring contaminant groups were examined; the total number of samples was 37. The results were compared to the original grouping.

3.3 Results and discussions

3.3.1 Entrance quality control analysis parameters

This section will contain the parameters measured when a new batch of PET flakes is received. The analysis included the analyses of PVC, other plastic contaminants, PA, PET trays, other contaminants (non-plastic materials such as wood, rubber, ...), moisture content, bulk density, PET dust, flakes with adhesives and barrier layers, differently coloured flakes (light and dark), light blue flakes, visual assessment, and opaque PET flakes [43].

3.3.1.1 Polyvinyl chloride (PVC)

Firstly, 63% of batches had PVC content smaller than 25 ppm; PVC ranged from 26 to 50 ppm in 21% of batches. Another 16% of batches were more problematic, containing more than 51 ppm of PVC. For more details, see Figure 17. Four batches exceeded 100 ppm, which is claimed to be a critical limit in scientific literature [29]. Still, most batches proved to contain only a small amount of PVC – less than 25 ppm. Thus, it is possible to assume that in most cases, producers managed to employ technologies to separate PVC effectively.

Producer	Average [ppm]	Min [ppm]	Max [ppm]	DRS/Mono-collection	EU market
A	28±16	8	58	Yes	E
B	53±45	6	96	No	W
C	30±25	2	75	Yes	E
D	3±3	0	7	Yes/No	E
D_CM	15±21	0	83	Yes/No	E
E	0±0	0	0	No	E
F	16±20	0	64	No	E
G	54±0	54	54	No	E
H_CM	93±24	76	110	Yes	W
I_CM	32±43	0	110	No	W
K_CM	43±22	6	82	Yes	W
L_CM	0±0	0	1	No	W
M_CM	69±39	25	104	Yes	W
N	8±13	0	30	Yes	E
O	19±13	1	58	No	E
P	16±3	14	18	Yes	E
Q	48±18	35	61	No	E
R	48±16	30	65	No	E

Figure 17: The results of PVC analysis. Source: own source.

The maximum permissible limit in our case is 50 ppm PVC, as stated in Figure 7. Producers D, N, and O were recommended for PET flakes supplying regarding low PVC content. These manufacturers were the most suitable because of their low PVC content on average and relatively stable results. Manufacturers D_CM, A, C, and F also had a low average PVC content, but there were frequent fluctuations. This was particularly noticeable in the case of D_CM, which can be explained by the existence of two individual plants. Low PVC content could, therefore, only be guaranteed for certain batches in D_CM. Producers E, L_CM, and P have supplied only a few batches, but further cooperation could be recommended due to the low PVC content.

By contrast, producers K_CM, R, Q, B, G, M_CM, and H_CM cannot be recommended due to the higher PVC content. In some batches from these producers, the 50-ppm limit was exceeded (or was exceeded also in the case of overall average value). For producers K_CM, B, R, and M_CM, the high

PVC content was (with a few exceptions) stable. It can therefore be assumed that these producers need to evaluate their existing separation technologies that can be used for PVC removal and modify them. For example, they may introduce or improve the manual or machine vision separation of bottles with shrink sleeves [42, 43] or install NIR equipment (or increase its sensitivity to PVC if this equipment was already installed) [48]. For producers Q, G, and H_CM, it was difficult to find a reason and predict batch trends due to the small number of batches; however, the supply to date has exceeded PVC limit, and therefore further supply from them was not recommended.

No difference was observed in PVC content in different collection schemes applied (see Chapter 2.2.1 for detail on different collection systems: DRS, mono-collection, and co-collection). This was also assumed a priori to the analysis, as in the case of this analysis, based on practice, and based on previous research [104], PVC comes mainly from bottle components⁴ and thus cannot be eliminated in mono-collection or DRS systems. The highest averages were observed mainly for the Western market. Based on the experience from practice, it was assumed that this can be attributed to that the Western market is generally characterized by preferring automatic sorting over manual sorting [43]. However, automatic sorting is not perfect once used alone [42, 44], and this was assumed to be a reason for a higher PVC content in the flakes from Western suppliers. There were also a few Eastern producers with high PVC content. It was concluded that these producers had inefficient or no manual sorting or inadequate or missing NIR equipment, as was discussed in the paragraph above. Considering the type of market, a complete lack of NIR equipment is the most plausible, given manual sorting is generally still intensive in the Eastern EU [43]. Concerning PET flake suppliers with low PVC content (D, O, D_CM, A, C), these are suppliers from the Eastern part of the EU. It was concluded that these suppliers were likely to operate both manual and NIR sorting efficiently, resulting in an efficient separation of PVC bottles. If the assumptions were true, these results show that it is important to incorporate not only NIR technologies but also manual separation or other technologies such as machine vision sorters.

3.3.1.2 Other polymers

In general, only the evaluation of polyolefins and PVC are embedded in EN standards [133]. Still, as the plastic types are truly diverse and can significantly influence technology devices and rPET quality [43], it was decided to monitor every plastic contamination other than PET; as described in Chapter 3.2.1.2. Only 17% of the batches contained below 34 ppm of other polymers, next 29% fell in the 35–74 ppm range, and 9% in the 75–99 ppm interval. Thus, 56% of the batches contained less than 100 ppm of other polymers. Another 25% of batches contained 100 to 199 ppm, 16% of batches were in the 200–499 ppm range, and 4% of batches exceeded the 500-ppm threshold (Figure 18). Given the results, the contamination with other polymers was significantly more pronounced than PVC contamination.

⁴ In our case, the origin of PVC from the bottle components is derived from a visual assessment of the PVC-contaminating particles in the batches. Based on the analysis in previous thesis [104] and the contaminant analysis of the samples used in this work (refer to Chapter 3.3.4.1 or Figure 35), the PVC contaminants were in the vast majority evaluated as PET bottle label pieces.

Producer	Average [ppm]	Min [ppm]	Max [ppm]	DRS/Mono-collection	EU market
A	142±65	36	227	Yes	E
B	702±650	100	1391	No	W
C	148±95	46	315	Yes	E
D	28±33	0	68	Yes/No	E
D_CM	78±91	2	392	Yes/No	E
E	2±0	2	2	No	E
F	100±33	57	161	No	E
G	138±0	138	138	No	E
H_CM	345±240	175	514	Yes	W
I_CM	252±180	55	544	No	W
K_CM	268±123	58	430	Yes	W
L_CM	345±21	330	360	No	W
M_CM	303±191	157	535	Yes	W
N	135±76	15	231	Yes	E
O	63±39	0	205	No	E
P	106±20	92	120	Yes	E
Q	27±38	0	54	No	E
R	306±89	192	392	No	E

Figure 18: The results of other polymer analysis. Source: own source.

To minimise the difficulties in reprocessing PET flakes, an internal limit of 75 ppm of other polymers is set [103]. Only two producers with an average of plastic contaminants lower than 75 ppm and stable quality could be recommended: D and O. Fluctuations were rare for these manufacturers. These were assumed to be mainly caused by a failure in some part of the technology, usually resulting in the penetration of voluminous contaminant that greatly impacted the result; or the entry of bottles with non-separable contamination (such as PETG labels). Manufacturers E and Q contained almost no polymeric contamination but have so far supplied few batches, and therefore their quality could not be assessed reliably. Producer D_CM also had an average value close to 75 ppm but had significant fluctuations. Again, this was probably due to the existence of two plants, one of which had effective separation technology, while the other did not.

I_CM, K_CM, M_CM, R, H_CM, L_CM, and B could not be recommended suppliers because of an average of 252 ppm and higher. Focusing on suppliers with multiple batches (two or more), only supplier R demonstrated a consistently high content of plastic contamination. The conjectures to explain the trends in other polymer contamination are based both on practical experience and information on various possibilities to remove other polymers: mainly NIR [42, 48, 110], manual [43], and machine vision sorting [42]. It was assumed that this supplier completely missed some parts of the technology such as NIR or manual sorting. The other producers showed fluctuations which could also be caused by the non-functionality or absence of some part of the separation technology. Consequently, fluctuations could arise from the heterogeneity of the PET bottles supplied for individual batches.

The type of market showed to be a crucial factor for the content of other polymers in provided PET flakes. The averages for the Eastern market were significantly lower than those for the Western market. For all suppliers from the Eastern market, the average of other polymers was below 148 ppm. All the Western-market-producers had higher other polymers content on average. An exception was Eastern producer R, with an average of 306 ppm. Again, it was concluded that this result could be attributed to the use of manual separation, which is not employed to the same extent in the Western as in the Eastern EU [43].

Surprisingly, less contaminated batches were seen mainly in the co-collection. The assumption was that the separate collection minimizes the input of other plastic products, i.e., other plastics. Thus, PET bottles themselves were assessed to be a significant source of contamination. The conjecture is that while producers processing material from co-mingled collection assumed a lot of heterogeneous plastics in the stream and had a technology to eliminate contamination from both faulty sorted objects and bottle components, producers processing PET bottles from mono-collection or DRS likely underestimated PET bottle components and different contamination (e.g., from devices and packaging) to some extent and thus their products contained higher polymer contamination. Additionally, as will be discussed in Chapter 3.3.4.1, PETG and PS occurred most frequently. These polymers can be used as bottle components [89, 101, 108] and in these cases cannot be completely avoided in mono-collection and DRS collection systems.

Still, it is important to consider that contamination by other plastics – except for PVC and polyolefins – is not described by the EN standards [133] (see Chapter 3.2.1.2) or EFSA (see Figure 7) and therefore, it can be assumed that some manufacturers had no incentive to invest in more efficient separation or to lose material due to higher instrument sensitivity unless forced to do so by its customers. Consequently, this can also be a reason for a higher content of other polymers in some producers. In this case, the processor should address with the problematic suppliers whether the contamination level can be reduced to a processable value determined by a processor.

The analysis of flakes from the German market also examined the content of polymer contaminants. PVC with PA were evaluated together with other polymers. Unfortunately, it is not possible to assess the content of PVC and PA. Overall, however, in contrast to our results, contamination with other polymers was much more severe in the co-collection system (flakes from DRS showed very low levels of other polymers). Since 2009, however, there was a noticeable decrease in the level of contamination of PET flakes from co-collection (probably attributable to the development of separation technologies). The smallest difference was observed in the last year of measurement, i.e., 2014 [4]. Thus, it is questionable whether measurements made at this time would show the same trends as the results of this thesis. If so, one could suggest that the efficiency of sorting systems may be more determinant today than the collection scheme concerning contamination by other polymers.

3.3.1.3 Polyamide (PA)

Concerning PA, 87% of the batches contained little or no PA (maximal content was 9 ppm), and the other 10% ranged from 10 to 39 ppm. Extremes, i.e., the presence of PA above 40 ppm, occurred in only 3% of batches. The results related to producers can be seen in Figure 19.

Producer	Average [ppm]	Min [ppm]	Max [ppm]	DRS/Mono-collection	EU market
A	14±21	0	47	Yes	E
B	0±1	0	1	No	W
C	2±5	0	16	Yes	E
D	2±4	0	9	Yes/No	E
D_CM	7±18	0	93	Yes/No	E
E	0±0	0	0	No	E
F	1±2	0	7	No	E
G	0±0	0	0	No	E
H_CM	6±9	0	13	Yes	W
I_CM	4±9	0	23	No	W
K_CM	8±9	0	25	Yes	W
L_CM	32±39	5	60	No	W
M_CM	7±10	0	24	Yes	W
N	2±5	0	12	Yes	E
O	1±2	0	11	No	E
P	2±2	0	3	Yes	E
Q	1±2	0	3	No	E
R	3±6	0	12	No	E

Figure 19: The results of PA analysis. Source: own source.

The resulting averages for most manufacturers complied with the requirements (see Figure 7), not exceeding 14 ppm. The only exception was producer L_CM, which provided PET flakes containing 32 ppm PA on average, but this should not be taken too much into account due to the small number of batches for this producer. Extremes occurred for producers L_CM, A, and D_CM. Still, the 50-ppm limit (see Figure 7) was exceeded only in two batches in total. Based on practical experience, these extremes could be related to the high content of barrier flakes (assuming PA and barrier flakes are correlated), perhaps also to processing technology leading to higher abrasion and hence peeling of the PA layer (likely caused by more aggressive washing). Penetration of the PA from non-bottle applications may also be the reason [43]. Despite this, these manufacturers provided satisfactory results in terms of PA limits; thus, these variations should not be a reason to discontinue sourcing from them.

PA averages were typically slightly higher for the Western market with mono-collection or DRS schemes. Since the differences in the averages are in the order of units of ppm, no conclusions were drawn from this trend. The assumption, however, is that these trends will be more pronounced for barrier flakes analysis.

3.3.1.4 Other contaminants

Based on practical experience, contamination by extraneous materials is random in the resulting flakes [43]. Erratic extremes are typical for this type of contamination but tend to be diluted in the final average due to the low abundance in other batches. This is highlighted by the fact that 84% of batches contained 9 ppm or less of other contamination. Then, 9% of batches were in the interval of 10–39 ppm, and extremes above 40 ppm occurred in 7% of batches. Results for individual suppliers are provided in Figure 20.

Producer	Average [ppm]	Min [ppm]	Max [ppm]	DRS/Mono-collection	EU market
A	0±1	0	3	Yes	E
B	57±93	0	164	No	W
C	22±44	0	134	Yes	E
D	0±0	0	1	Yes/No	E
D_CM	14±45	0	238	Yes/No	E
E	0±0	0	0	No	E
F	0±0	0	0	No	E
G	20±0	20	20	No	E
H_CM	60±75	7	113	Yes	W
I_CM	1±1	0	2	No	W
K_CM	6±12	0	36	Yes	W
L_CM	0±0	0	0	No	W
M_CM	12±22	0	51	Yes	W
N	1±3	0	8	Yes	E
O	9±21	0	85	No	E
P	2±3	0	4	Yes	E
Q	5±6	0	9	No	E
R	1±1	0	3	No	E

Figure 20: The results of other contaminants analysis. Source: own source.

In addition to the average content of other impurities for each manufacturer, it was also examined how many batches of a given manufacturer contained other contamination and how many did not. Once focusing on manufacturers with more than three batches, the most frequent contamination occurred in supplier C (62% of batches), K_CM (60% of batches), and O (50% of batches). Although K_CM and O manufacturers were likely to contain other contaminants in the batch, the overall average of other contaminants for these manufacturers was not high compared to the rest of the manufacturers. From this, it was concluded that manufacturers K_CM and O processed the input with frequent occurrence of extraneous materials but were able to remove the foreign material effectively with few exceptions. Producer C encountered other contaminants often as well but crossed 100 ppm twice in two batches. For this manufacturer (perhaps also for manufacturer B with a similar type of extremes), this raises the question of whether the level of other contamination was purely dependent on the input material. If in the remaining 9 batches, the material was free of contamination but in next 2 batches, the contamination in the input material was high, it indicates the manufacturer could not cope with other contamination once it appeared in the input material. This could point to missing or ineffective sorting technologies (see Chapter 2.4.3 for more details on separation of other contaminants). Even if the extreme content of other contamination was a matter of a small number of batches, this introduced a degree of uncertainty for the processor [43]. High variation in other contaminants content was also typical for producer D_CM, although two plants with differently efficient sorting technologies were suspected to be a source of fluctuation in this case.

The limits for other contaminants are listed individually by EFSA for metals (50 ppm) and paper with wood (also 50 ppm), while other particles such as stones, textiles, or rubber are not listed (see Figure 7). In practice, therefore, the internal limit for all these particles has been set at 50 ppm as a whole, regardless of the specific material [103]. As a result, H_CM, B, and C producers did not receive the recommendation due to higher averages and C due to fluctuation. It should be noted that there were only a few batches from producers H_CM and B, perhaps this was a random extreme occurrence. D_CM was also evaluated as an unrecommended producer due to the high fluctuation and therefore uncertainty for the processors. The rest of the producers were assessed as not problematic, i.e., except for individual

fluctuations, the content of other contamination was low to sporadic. There were even producers whose batches did not contain other contaminants (almost) at all – of note were producers N, R, I_CM, A, D, and F, which did not show extreme occurrences in any of the batches despite larger batch volumes and had an overall average of 0 to 1 ppm. An assumption was made that these favourable results could be due to a range of factors such as quality of an input material, appropriate bottle design, the minimal contamination from process equipment (such as rubber conveyor belts) and packaging (i.e., pallets) [43], and very efficient sorting.

Other contamination was expected to vary by type of collection scheme – in particular, to be higher for producers processing material from the co-collection. However, it turned out that the content of other contamination is completely independent of the type of collection, even when only the extremes were examined. Similarly, whether a producer operated in the Western or Eastern market did not affect the results. As a result, bottle design and separation technology were evaluated as the main causes of contamination by other materials. Moreover, based on practical knowledge, extraneous materials often occur as a huge single piece of a contaminant in the flakes samples. These pieces tend to be volumetrically significant (this is particularly true for stones or rubber, and metals) [43]. Control analysis thus lags in representing the other contamination in a total volume of delivered flakes, as the occurrence of such pieces of other contaminants in sampled material is more likely to be a matter of coincidence.

A similar analysis was carried out on PET flakes in Germany. Although the metals and other materials were analysed separately, both analyses showed a similar trend: with a few exceptions, the content of other contaminants was the highest in PET flakes from the co-mingled collection. However, only samples up to 2014 were analysed and a significant reduction of extraneous materials in the flakes from the co-mingled collection system was observed from 2012. The authors of the study attributed this trend to the increasing efficiency of sorting technologies [4]. Together with results with this thesis, it seems that the separation technology is becoming more important than the collection system. Still, more experiments would be needed to confirm this hypothesis.

3.3.1.5 Moisture content

Most batches (84%) had a moisture content in the range of 0.50–0.99% water content. Meanwhile, 7% of the batches had a lower water content (below 0.50%). On the contrary, 9% of the batches contained 1.00% moisture or more. Although lower moisture content is preferred, the moisture content was not a problem for 90% of batches. Detailed results are displayed in Figure 21.

Producer	Average [%]	Min [%]	Max [%]	DRS/Mono-collection	EU market
A	0.70±0.08	0.59	0.82	Yes	E
B	0.80±0.05	0.75	0.84	No	W
C	0.83±0.15	0.55	0.97	Yes	E
D	0.98±0.79	0.58	2.40	Yes/No	E
D_CM	0.59±0.14	0.34	0.92	Yes/No	E
E	0.57±0.00	0.57	0.57	No	E
F	0.82±0.09	0.67	0.93	No	E
G	0.88±0.00	0.88	0.88	No	E
H_CM	0.82±0.00	0.82	0.82	Yes	W
I_CM	0.64±0.07	0.56	0.77	No	W
K_CM	0.92±0.12	0.68	1.09	Yes	W
L_CM	0.81±0.06	0.77	0.85	No	W
M_CM	0.69±0.19	0.58	1.02	Yes	W
N	1.27±0.22	0.99	1.61	Yes	E
O	0.65±0.10	0.38	0.90	No	E
P	1.11±0.15	1.01	1.22	Yes	E
Q	0.71±0.04	0.68	0.74	No	E
R	1.02±0.07	0.92	1.09	No	E

Figure 21: The results of moisture content analysis. Source: own source.

The maximal limit reported to EFSA for the moisture content is 1.00% (see Figure 7). Some producers had an average moisture content above 1.00%: N, P, and R. Producer P exceeded the 1.00% limit for both batches supplied. Supplier K_CM also tended to have a higher moisture content but did not exceed 1.00% on average. As a result, N, P, and R suppliers could not be recommended because of the consistently high moisture content and possible technology [121] or storage problems [43].

In terms of low average moisture content and not exceeding the 1.00% moisture limit throughout the delivery period, the manufacturers I_CM, O, and D_CM were particularly recommended. However, the other manufacturers also did not have a problem to comply with the 1.00% moisture limit, except for a few batches with a slight exceedance of this limit.

The analysis showed one interesting outlier. One batch of producer D contained 2.40% moisture, although other batches from producer D_CM did not show moisture content problems. Based on experience from practice, this could be attributed either to problems with the drying [121], storage, or transport technology on the manufacturer's side [43]. The same issues could be the reason for extreme moisture content in other batches, mainly from suppliers N, P, and R.

Although the producers with the highest averages were located in the Eastern market and processed material from the mono-collection/DRS system, no clearer pattern in moisture content could be observed concerning the type of market or collection system.

3.3.1.6 PET dust

Majority, i.e., 80% of the batches contained a maximum of 0.19% dust. Contents in the range of 0.20%–0.39% were observed in 15% of the batches. The remaining 5% of the batches contained 0.40% PET dust or more. Results related to the individual batches are described in Figure 22.

Producer	Average [%]	Min [%]	Max [%]	DRS/Mono-collection	EU market
A	0.29±0.07	0.17	0.37	Yes	E
B	0.16±0.12	0.08	0.29	No	W
C	0.19±0.31	0.03	1.19	Yes	E
D	0.09±0.14	0.01	0.33	Yes/No	E
D_CM	0.02±0.01	0.00	0.05	Yes/No	E
E	0.19±0.00	0.19	0.19	No	E
F	0.20±0.22	0.03	0.84	No	E
G	0.13±0.00	0.13	0.13	No	E
H_CM	0.44±0.38	0.17	0.71	Yes	W
I_CM	0.26±0.40	0.01	1.00	No	W
K_CM	0.05±0.03	0.02	0.11	Yes	W
L_CM	0.01±0.01	0.00	0.02	No	W
M_CM	0.08±0.02	0.05	0.10	Yes	W
N	0.11±0.05	0.05	0.20	Yes	E
O	0.12±0.07	0.02	0.29	No	E
P	0.45±0.06	0.40	0.49	Yes	E
Q	0.33±0.16	0.22	0.44	No	E
R	0.14±0.08	0.08	0.25	No	E

Figure 22: The results of PET dust content analysis. Source: own source.

PET dust limit is not reported to EFSA, thus is based on the internal standard. In this case, a very optimal maximal value is 0.12% [103]. Flakes from suppliers P, H_CM, and Q had a high PET dust content (above 0.33%), but due to the small number of batches, it was not possible to conclude whether this was due to a temporary failure in the dust separation technology or whether the suppliers did not have an efficient dust separation [3]. Suppliers A, I_CM, and F supplied dustier batches on average. While dustiness was a stable issue for supplier A (it was assumed this was due to the technology), suppliers I_CM and F had higher averages due to significant variation in a single batch. Thus, this was probably a random problem in the dust removal technology [3].

The recommended producers were those with low dust content and the most stable results. These criteria were met by suppliers D_CM, K_CM, and M_CM. For D_CM, the stability was surprising given the two manufacturing plants; most probably these two plants used the same dust separation technology. The flakes from producer L_CM had complying level of dust content but only two batches were examined, and therefore no conclusions could be drawn. Producers O, N, and D also had low averages, but slight fluctuations had to be expected.

The content of PET dust showed no connection with the type of market or collection system. This conclusion was expected as this parameter was assumed to be dependent on the technology rather than the input material [3].

3.3.1.7 Bulk density

The market was relatively diverse regarding bulk density, with averages ranging from 268 kg·m⁻³ to 357 kg·m⁻³. The minimal measured value was 226 kg·m⁻³, and the maximum was 362 kg·m⁻³. For several producers, higher variability of results was observed. Overall, the market could be divided into three intervals. Only 6% of batches had a bulk density below 249 kg·m⁻³. In the range 250–299 kg·m⁻³, 68% of the batches were situated. The remaining 26% had a bulk density greater than 300 kg·m⁻³. Figure 23 displays more detailed results.

Producer	Average [$\text{kg}\cdot\text{m}^{-3}$]	Min [$\text{kg}\cdot\text{m}^{-3}$]	Max [$\text{kg}\cdot\text{m}^{-3}$]	DRS/Mono-collection	EU market
A	299±14	277	320	Yes	E
B	313±43	265	347	No	W
C	290±27	236	349	Yes	E
D	268±34	238	324	Yes/No	E
D_CM	270±22	226	326	Yes/No	E
E	301±0	301	301	No	E
F	280±9	263	296	No	E
G	299±0	299	299	No	E
H_CM	357±7	352	362	Yes	W
I_CM	302±10	290	316	No	W
K_CM	270±14	240	292	Yes	W
L_CM	285±0	285	285	No	W
M_CM	269±30	248	321	Yes	W
N	296±12	283	312	Yes	E
O	286±22	246	343	No	E
P	283±24	265	300	Yes	E
Q	305±21	290	320	No	E
R	321±8	311	328	No	E

Figure 23: The results of bulk density analysis. Source: own source.

It was not appropriate to divide suppliers into those which can be recommended and those which cannot be, because the requirements for processors are completely individual according to the technology. Even though optimal range is set by a producer [103], based on practical experience, the most important thing is that the manufacturer keeps the bulk density stable [43]. Regarding stability, suppliers A, F, I_CM, K_CM, N, and R could be recommended; but suppliers B, C, D, and M_CM could not (producers with 2 batches and less were not considered). In general, lower values were typical for manufacturers D, M_CM, and K_CM; high values for suppliers H_CM and R. B also showed a high average value but also variations from batch to batch.

The hypothesis is that bulk density and PET dust are correlated variables: if the bulk density is high, finer grinding – thus, a more intensive formation of PET dust – is likely [43]. PET dust may remain in the sample if the dust removal technology is not sufficient [3]. However, deviations from this assumption were observed during the analysis. Once the average bulk density was high, but the average PET dust percentage was low (N: 0.11% dust and bulk density 290 $\text{kg}\cdot\text{m}^{-3}$; G: 0.13% dust and bulk density 299 $\text{kg}\cdot\text{m}^{-3}$; R: 0.14% dust and bulk density 321 $\text{kg}\cdot\text{m}^{-3}$), some producers had relatively efficient PET dust separation. On the other hand, some producers had a high average PET dust but a low average bulk density (F: dust 0.20% and bulk density 280 $\text{kg}\cdot\text{m}^{-3}$ and P: dust 0.45% and bulk density 283 $\text{kg}\cdot\text{m}^{-3}$). The PET dust removal efficiency of these suppliers was assumed to be poor.

An interesting phenomenon was that while market type did not show an effect on bulk density, the flakes with the highest bulk densities were predominantly from the co-collection scheme, while the lower values were observed mainly in the case of producers processing waste from the mono-collection system/DRS. No explanation could be found for this phenomenon – it is likely coincidental. Comparable results were found in a study of PET flakes on the German market, where PET flakes from co-collection tend to have higher bulk density than PET flakes from mono-collection. Still, the difference was not very significant [4].

3.3.1.8 Flakes with adhesives

More than half of the batches (53%) contained only up to 0.09% flakes with adhesives. Then, 28% of the batches were in the range of 0.10–0.29%, and content between 0.30% and 0.49% was measured in 15% of the batches. More than 0.50% flakes with glue residue were found in only 4% of the batches. See Figure 24 for more detailed information.

Producer	Average [%]	Min [%]	Max [%]	DRS/Mono-collection	EU market
A	0.31±0.10	0.15	0.44	Yes	E
B	0.05±0.03	0.03	0.08	No	W
C	0.27±0.11	0.10	0.52	Yes	E
D	0.06±0.08	0.00	0.15	Yes/No	E
D_CM	0.13±0.16	0.01	0.83	Yes/No	E
E	0.07±0.00	0.07	0.07	No	E
F	0.44±0.10	0.28	0.57	No	E
G	0.02±0.00	0.02	0.02	No	E
H_CM	0.38±0.37	0.11	0.64	Yes	W
I_CM	0.03±0.01	0.00	0.04	No	W
K_CM	0.10±0.04	0.05	0.16	Yes	W
L_CM	0.07±0.02	0.05	0.09	No	W
M_CM	0.07±0.05	0.00	0.11	Yes	W
N	0.38±0.10	0.17	0.45	Yes	E
O	0.05±0.06	0.00	0.35	No	E
P	0.24±0.19	0.10	0.37	Yes	E
Q	0.08±0.01	0.07	0.08	No	E
R	0.30±0.09	0.25	0.43	No	E

Figure 24: The results of flakes with adhesives analysis. Source: own source.

According to the internal requirements, a PET flake batch can contain up to 1.00% of PET flakes with adhesive residuals [103]. Therefore, it was not possible to address some batches as not recommended because even the batch with the highest value of all batches (0.83%) was still processable. Still, lower content of flakes with adhesive and stability of this value are preferred over higher values and fluctuations [43]. These conditions were met by the producers I_CM, O, B, D, M_CM, Q, and K_CM (producers with two batches and fewer are not mentioned due to the impossibility of assessing the fluctuations between batches). In practice, low values could occur due to the good quality of the washing process [43], but also to the input material – some adhesives cannot be easily washed off [72]. The supplier D_CM also had a low average value, but a large variation between batches could be observed, which was attributed to the existence of two plants. The rest of the batches already had higher averages and sometimes even higher variations. This could (again) be attributed to the quality of washing and input material. Thus, it can be assumed that manufacturers with a higher average amount of flakes with adhesives either had a less efficient washing process or there was a higher number of bottles with harder-to-wash-off adhesives in the feedstock.

While there is no apparent association between the content of flakes with adhesives and market type, the co-collection scheme typically yielded a lower content of flakes with adhesives. Additionally, higher content of PET flakes with adhesive residuals was found in material for producers with mono-collection or DRS schemes.

3.3.1.9 Barrier flakes

A small proportion of flakes with the barrier, i.e., up to 0.29%, was found in 15% of the batches. Many flakes (68%) were in the interval 0.30% to 0.99%; when divided into two intervals 0.30–0.59% and

0.60–0.99%, the same percentage of batches (34%) were situated in them. Between 1.00% and 1.49% was measured in 7% of the batches; 10% of the batches contained 1.50% barrier flakes or more. Thus, 18% of the batches contained a considerable proportion of barrier flakes that could cause operational complications [43]. An overview of the results is displayed in Figure 25.

Producer	Average [%]	Min [%]	Max [%]	DRS/Mono-collection	EU market
A	0.23±0.10	0.12	0.42	Yes	E
B	0.77±0.72	0.23	1.59	No	W
C	0.70±0.37	0.17	1.62	Yes	E
D	0.70±0.38	0.15	1.16	Yes/No	E
D_CM	0.87±0.63	0.06	2.81	Yes/No	E
E	0.24±0.00	0.24	0.24	No	E
F	0.76±0.20	0.48	1.11	No	E
G	0.94±0.00	0.94	0.94	No	E
H_CM	0.88±0.13	0.79	0.98	Yes	W
I_CM	1.65±0.31	1.33	2.17	No	W
K_CM	0.97±0.45	0.52	1.91	Yes	W
L_CM	3.91±0.41	3.62	4.20	No	W
M_CM	0.90±0.53	0.22	1.58	Yes	W
N	0.73±0.14	0.50	0.87	Yes	E
O	0.50±0.20	0.18	1.27	No	E
P	0.35±0.07	0.31	0.40	Yes	E
Q	0.65±0.09	0.59	0.71	No	E
R	0.51±0.11	0.43	0.66	No	E

Figure 25: The results of barrier flakes analysis. Source: own source.

The highest content of barrier flakes was observed for the supplier L_CM (average 3.91%). This supplier is mentioned despite the small number of batches because such a high level of barrier flakes was not observed for any other producer. The assumption is that this manufacturer processed material with a high proportion of PET bottles with a barrier, that are difficult to be separated [94]. However, the supplier should consider whether to introduce manual or machine vision separation of bottles likely to contain a barrier and thus reduce the proportion of barrier flakes. For suppliers K_CM, M_CM, B, C, and D, such a measure could help to minimise batch-to-batch variations in this parameter. The fluctuations for supplier D_CM were most likely attributed to the different quality of the two plants and subsequently, of a supplied material.

According to the experience from practice, the additional problem concerning producer L_CM could be that the proportion of 3.91% is high, and a reduction would likely lead to significant material losses for the manufacturer. Additionally, separating PET bottles with a barrier layer before they enter the line would impact the overall PET recycling rate and recyclability of PET bottles in addition to material losses. Then, the question is whether the producer is forced to reduce the proportion of bottles with a barrier in the input material or whether some purchasers (PET flakes processors) exist that can process such flakes without problems. Producer I_CM with a relatively high average value should also consider the introduction of separation of PET bottles with a barrier. Another assumption based on practical experience is that producers L_CM and I_CM bought raw bottles of the lowest quality in order to reduce costs.

The internal limit for barrier flakes is 1.00% [103]. Additionally, the manufacturers to be recommended are required to keep the barrier flakes level as stable as possible [43]. These requirements were met in batches of producers A and R. O was also considered a suitable supplier, except for occasional

fluctuations. Few samples were available from suppliers E, P, and Q, and the low barrier flakes levels were a good indicator. These suppliers appeared to process PET bottles with lower barrier layer levels. Alternatively, a barrier bottle separation step could be implemented in these plants. For supplier O, a low average but occasional fluctuation could be observed, which may be due to the heterogeneity of PET bottles processed across batches.

There was a clear division between the Western and Eastern market in the content of the barrier flakes. Hypothetically, this observation could point to the difference in bottle design in Eastern and Western countries, e.g., the difference in consumption of beer [96], juices, fresh milk [5], and so on because Western producers supplied batches with a higher proportion of barrier flakes than those from the Eastern market. However, the assumed association with differential consumption of the mentioned beverages has not been further investigated and would need to be verified with real data. In contrast, no association with the type of collection was observed.

3.3.1.10 Differently coloured flakes

Due to the nature of the results of the differently coloured flakes, a figure providing the basic statistical parameters will not be available as in previous cases. This is because the content of the colour flakes was proven to be unproblematic (in terms of internal requirements) and, with exceptions, relatively stable across batches and manufacturers. The exceptions will be discussed in the following text.

Although present in 75% of batches, light-coloured flakes did not prove to be a problematic indicator. The average of most suppliers was 0.02%, which has a negligible effect on the quality of rPET (based on standard operating procedures (SOPs), an internal limit is 0.10% [43], which is significantly higher than the measured maximum). Batches from suppliers B, D_CM, K_CM, L_CM, and R had an average above 0.02%. Still, these averages were lower than the upper limit for light-coloured flakes. The supplier should only draw attention to producers with significant fluctuations, which was particularly the case for the supplier D_CM. The assumption is that one of the production plants of this supplier had colour separators (refer to Chapter 2.2.2.3 for additional information on colour separators) and was therefore characterised by low light flakes content. The other plant probably did not have such a separator, which resulted in significant fluctuations – probably due to the penetration of a differently coloured bottle. D_CM was also the only supplier who supplied two batches with light-coloured flake content above 0.10%. This value is above the internal limit and based on practical experience, may already represent a technological inconvenience [43]. The measured maximum was 0.17%. This supplier is the only one that could not be recommended due to these extreme fluctuations.

Dark-coloured flakes are more problematic: they can affect the colour of the recyclate much more than light-coloured flakes [43]. Thus, an internal limit of dark-coloured flakes is 0.02% [103]. Although dark-coloured flakes were present in only 53% of batches, some exceeded 0.02% limit. This happened once for supplier P, three times for supplier R (out of a total of four batches delivered), in most batches from suppliers M_CM, I_CM, and K_CM, ten times for supplier D_CM and once for supplier A. Particularly for suppliers I_CM, K_CM, and M_CM, there were high extremes and high averages. For these producers (except for supplier A, where accidental penetration or momentary malfunction of the technology was assumed to be the cause), the absence of colour separators could be assumed as well as in the case of light-coloured flakes. For D_CM, the absence of such technology was expected in one of the plants. Alternatively, suppliers may have colour separators employed only before entering the technology for whole bottles but no longer for the final flakes. Based on practical experience, a single penetrating bottle may be significantly reflected in the measured value [43]. However, the latter option is more likely to be assumed for D_CM and R producers, where the peaks were lower. In any case, these producers (except producers A and P) could not be recommended due to high and frequent dark-coloured flakes content.

It was not possible to observe trends in the content of the differently coloured flakes concerning the type of market and collection system. The highest average of dark-coloured flakes was observed in two producers in the Western market with a mono-collection system or DRS; however, this phenomenon is rather attributed to technology and not to the market or collection system.

3.3.1.11 Light blue flakes

The content of light blue flakes varied widely across manufacturers and batches: 40% of batches were found to have low blue flake content, i.e., up to 0.99%. Then, 22% of batches contained between 1.00% and 4.99% light blue flakes; between 5.00% and 9.99% were measured in 20% of batches. Batches containing 10.00–14.99% accounted for 9% of the batches, and a further 9% of the batches had a light blue flakes content of more than 15.00%. The results for individual suppliers are displayed in Figure 26.

Producer	Average [%]	Min [%]	Max [%]	DRS/Mono-collection	EU market
A	2.70±0.95	1.24	3.55	Yes	E
B	11.08±9.61	0.00	17.16	No	W
C	4.61±6.33	0.01	19.02	Yes	E
D	0.59±0.90	0.00	2.19	Yes/No	E
D_CM	3.49±4.13	0.07	14.65	Yes/No	E
E	0.00±0.00	0.00	0.00	No	E
F	0.79±0.88	0.14	3.21	No	E
G	0.00±0.00	0.00	0.00	No	E
H_CM	7.36±1.70	6.16	8.56	Yes	W
I_CM	22.85±7.22	13.86	33.12	No	W
K_CM	10.56±3.63	2.95	14.81	Yes	W
L_CM	11.98±2.01	10.56	13.40	No	W
M_CM	14.63±7.00	3.02	21.69	Yes	W
N	13.31±9.20	4.48	28.65	Yes	E
O	3.03±2.88	0.14	9.74	No	E
P	0.30±0.09	0.24	0.36	Yes	E
Q	0.30±0.42	0.01	0.60	No	E
R	0.44±0.09	0.34	0.55	No	E

Figure 26: The results of light blue flakes analysis. Source: own source.

Based on practical experience, light blue flakes are not entirely undesirable in recycled flakes. The light blue flakes can be deliberately added to the recyclate (or left unseparated) to optimise the colour of the clear PET flakes [136]. Traditionally, an additive with a blue dye is used for colour improvement because it can suppress unwanted rPET discoloration [98]. However, it can be assumed that if light blue flakes are added to PET flakes, they can theoretically replace some of the dye and thus reduce the additive dosage. It is assumed that the light blue flakes may (although imperfectly) partially compensate for the yellowish barrier flakes and greyish degraded flakes. If some manufacturers used this approach, there should be a correlation between light blue flakes and visual evaluation or barrier flakes content.

Therefore, dividing flakes based on light blue flakes into suitable and unsuitable ones is not straightforward. In practice, however, the content above 15% of light blue flakes is considered high according to the internal requirements on PET flakes [103]. This percentage is also used as an internal limit, and according to SOPs, flakes crossing this limit could be considered more as a mix of blue and transparent PET flakes [43]. Therefore, further supplies from I_CM and M_CM suppliers could not be recommended because in their case, the averages approached or significantly exceeded the 15% threshold. This limit was also exceeded by the maxima of suppliers N, B, and C.

Furthermore, stability is generally preferred. In practice, this is also true for light blue flakes content, which affects the dosage of the additive which is important for batch processing planning [43]. The suppliers I_CM, M_CM, N, B, C, D_CM, and (to a lesser extent) K_CM and O were unstable. On the other hand, suppliers A, F, D, and R showed high stability (producers with two or fewer batches cannot be assessed). However, lower averages are also typical for the more stable producers.

In practice, light blue flakes separation is not as crucial as the separation of above-mentioned contaminating particles [43]. An assumption regarding the sensitivity of the colour separators is that it could be relatively low, as light blue flakes do not pose such a threat to the quality [43]. This way, unnecessary material losses could be avoided. Nevertheless, if the higher content of light blue flakes was non-intended and due to insufficient sorting of coloured bottles, a higher content of differently coloured flakes should be observed in these batches.

While no association with the type of collection system was observed, there was a clear association with the type of market. The high content of light blue flakes was typical for the Western market (13.07% on average), while for the Eastern market, it was considerably lower (2.46% on average). However, no general approximation on the type of market could be made as in general, light blue content is influenced by three criteria: by customer (PET flakes processor), the sorting facilities, and the price [90, 136].

First, as was discussed above, the light blue flakes can partially replace the blue additive with their pigments to suppress greying due to degradation and yellowing due to barrier flakes [136]. Therefore, it was concluded that there may be flake processors requiring an addition of light blue flakes. Then, flakes suppliers may add light blue flakes to the batches by default also for other processors. In addition to replacing the additive, the reason for the addition of light blue flakes may be the price reduction. Because coloured flakes are economically less valuable than clear flakes [90], adding light blue flakes may have a positive effect on price. A final influence may be the sorting strategy in recycling facilities, as clear and light blue flakes can be considered one colour fraction [90].

3.3.1.12 Visual evaluation

Most batches fell in the interval between 1.00 and 1.99 (78 % of batches). This meant that 54 % of the batches could be considered well-washed, and not degraded (or only slightly degraded). The evaluation was slightly worse for 24% of the batches which had poorer washing process quality and/or more significant degree of degradation (grade 1.50–1.99). Finally, 22% of the batches were severely degraded and/or poorly washed. Therefore, they received a grade between 2.00 and 3.00. No grade worse than 3.00 was given. More information on grades for individual producers is provided in Figure 27.

Producer	Average [-]	Min [-]	Max [-]	DRS/Mono-collection	EU market
A	1.14±0.24	1.00	1.50	Yes	E
B	1.17±0.29	1.00	1.50	No	W
C	1.70±0.61	1.00	3.00	Yes	E
D	1.00±0.00	1.00	1.00	Yes/No	E
D_CM	1.40±0.48	1.00	3.00	Yes/No	E
E	1.00±0.00	1.00	1.00	No	E
F	1.24±0.21	1.00	1.50	No	E
G	1.50±0.00	1.50	1.50	No	E
H_CM	2.25±0.35	2.00	2.50	Yes	W
I_CM	1.50±0.00	1.50	1.50	No	W
K_CM	2.25±0.29	2.00	2.75	Yes	W
L_CM	3.00±0.00	3.00	3.00	No	W
M_CM	1.38±0.25	1.00	1.63	Yes	W
N	2.19±0.25	2.00	2.50	Yes	E
O	1.12±0.19	1.00	1.50	No	E
P	1.19±0.27	1.00	1.38	Yes	E
Q	1.50±0.00	1.50	1.50	No	E
R	1.88±0.25	1.50	2.00	No	E

Figure 27: Results of visual evaluation analysis. Source: own source.

A worse grade was not a reason to reject a batch – no internal or EFSA-reported (refer to Figure 7) limit exists [103]; visual evaluation is rather informative. But the lower the grade, the better for the processor [43]. The high grades were mainly reported in the suppliers L_CM, H_CM, K_CM, and N. Refer to Chapter 3.2.1.3 for information of visual sorting analysis: high grade was a sign of either high degradation, poor washing, or a combination of both. It was observed that a high grade of the above-mentioned manufacturers was mainly due to degradation caused by a higher number of recycling cycles. Based on practical experience, this problem is already inevitable in countries with bottle-to-bottle recycling implemented for longer period [43].

Another key criterion was, as with the other parameters, stability [43]. Stability was the worst for suppliers D_CM and C. For D_CM, the fluctuations were attributed to the operation of two production plants in two countries, which probably implied a difference in the quality of the input material as well as in the efficiency of the washing technology. Supplier C supplied more degraded batches overall, but it also happened that the washing process was not effective. In one of the batches, the quality of the washing was significantly worse compared to the other batches, hence the variation in the visual assessment. In practice, this could have been caused by a change in factors of the washing technology, such as a change in conditions, chemicals, or abrasives [43].

It was not assumed that the type of collection system affects the visual assessment of the sample. Although producers working with material from mono-collection/DRS systems had higher-grade averages, no conclusions were made concerning recycling scheme. However, in the case of the Western market and specific countries in the Eastern market, the visual assessment was impaired, the assumption being that bottle-to-bottle recycling is employed for longer in some Western countries and additionally, higher amount of rPET content is used in bottles, resulting in increased degradation in this region. The higher proportion of rPET in bottles in some Western EU countries was also derived from the fact that, for example, there are initiatives in Germany (Petcycle) that promote a higher proportion of recycled PET in bottles [137]. However, furthermore, the necessary market analysis has not been carried out in this respect and therefore these assumptions should only be regarded as a conjecture.

In Eastern countries, it was generally common that there was only a slight deterioration of the grade due to degradation and the poorer rating was more likely due to the quality of the washing. Besides the results of the analysis, these trends were observed in everyday practice [43].

For the visual assessment, it should be noted that it was evaluated by the laboratory technicians by visual comparison with etalons. The problem is that comparison to etalons could sometimes be subjective and biased, especially when there was a significant difference in the amount of blue or barrier flakes in the sample being evaluated compared to the etalon. This could tend to distort visual perception. Generally, this method is rather non-reproducible and tentative, and it would be advisable to optimise this method.

3.3.1.13 PET Trays

Although 57% of the batches contained a maximum of 49 ppm of PET tray fragments, and a further 19% ranged from 50 to 99 ppm, high occurrences were not uncommon. Between 100 and 199 ppm of PET trays were present in 9% of the batches, and contents of 200 to 499 ppm were measured in 6% of the batches. PET tray contents above 500 ppm were recorded in 9% of batches (5% contained more than 2000 ppm). Thus, the results showed a great diversity among manufacturers. In Figure 28, more detailed information is provided.

Producer	Average [%]	Min [%]	Max [%]	DRS/Mono-collection	EU market
A	12±15	0	36	Yes	E
B	335±345	0	689	No	W
C	26±32	0	119	Yes	E
D	38±29	14	88	Yes/No	E
D_CM	619±1638	0	7727	Yes/No	E
E	0±0	0	0	No	E
F	62±56	0	202	No	E
G	0±0	0	0	No	E
H_CM	1906±2681	10	3801	Yes	W
I_CM	2383±1394	0	3984	No	W
K_CM	2±5	0	15	Yes	W
L_CM	0±0	0	1	No	W
M_CM	123±118	0	281	Yes	W
N	512±330	80	1087	Yes	E
O	76±64	0	217	No	E
P	8±11	0	16	Yes	E
Q	2±3	0	5	No	E
R	62±49	19	116	No	E

Figure 28: The results of PET trays analysis. Source: own source.

The content of PET trays is not officially regulated (see Figure 7 and [103]). However, based on the practice and literature (recall Chapter 2.4.1.2), high PET tray content is not desirable for processors. Additionally, based on practical experience, it is problematic to increase IV to the level required for PET bottles production once PET tray content is higher. As a result, 100 ppm of PET trays is an internal limit, but it is rather orientational than decisive (respectively, the limit is only deciding once the extremes occur [43]). Therefore, I_CM, H_CM, D_CM, N, and B producers could not be recommended due to the relatively high average content of PET trays. The producers with high averages typically showed huge variations between batches; the most noticeable variations were observed for the supplier D_CM.

While several batches from D_CM contained few PET trays (below 100 ppm), several batches reached enormous extremes in the order of thousands of ppm. These fluctuations were already evident in the

previous indicators and, as with the previous ones, the existence of two plants was likely the cause. The plant with the typically lower volume of PET trays was processing material from a mono-collection/DRS system where the content of PET applications other than PET bottles should be minimised. It was likely that the plant had very high-quality separation technologies with either manual separation or machine vision sorters incorporated. These two quality steps are needed to remove PET trays, which are indistinguishable from PET bottles by NIR [42, 43]. In contrast, the second plant processed waste from the co-collection system, where logically a higher level of PET trays can be expected. Yet, this plant probably also missed manual or machine vision separation entirely (or at least has one but with insufficient efficiency). Thus, PET trays could go through the entire cycle to PET flakes without much difficulty. The significant fluctuations at the L_CM supplier were probably due to the same reasons. The material came from the co-collection and if there were more PET trays in the processed PET bottles and the manufacturer did not have the appropriate technology, most of the PET trays from the input infiltrated into the final batch.

The producers K_CM, P, A, D, C, R, and F showed very low and relatively stable levels. The low PET tray content was assumed to be likely due to a combination of two factors: the quality of the feedstock and the availability of sufficiently efficient separation technology for the other PET forms, which are manual separation and/or machine vision separators [42, 43].

It was assumed that the content of PET trays would not be related to the type of market; these assumptions were confirmed. However, a surprising thing was found concerning the collection scheme: high PET tray content was not expected for producers with mono-collection/DRS material; the opposite turned out to be true for H_CM, N, and partly M_CM. It was concluded PET trays may be collected with PET bottles; however, PET trays may also be added intentionally to increase the volume of recycled material. However, these assumptions were not compared to the reliable data or practice, thus are only a conjecture. Apart from these exceptions, the PET tray content of the rest producers with mono-collection/DRS schemes was low and stable. Excluding producers with less than two batches supplied; producers with a mono-collection scheme occupied the rungs with the lowest PET tray contamination. Co-collection (when not considering one or two-batch suppliers), on the other hand, tended to show higher occurrences of PET trays as well as occurrence of fluctuations, most likely depending on the composition of the waste delivered. Eastern producers deviating from this trend perhaps had effective separation technologies (co-collection suppliers with low averages and deviations). Concerning producers treating waste from a mono-collection scheme, which contained more PET trays with higher fluctuations in final PET flakes, it was deduced that they either treated waste from such a scheme where other PET applications were sorted together with PET bottles or PET trays were added to separately selected PET bottles. In any case, it can be expected that these producers did not have effective separation technologies that could reduce the PET tray content in the input material.

3.3.1.14 Opaque flake

The opaque flake content was almost the same for all manufacturers, not exceeding 0.04%. The only producer found to have batches with a higher opaque flake content (5 batches with 0.05 to 0.09% opaque flakes) was D_CM. It was considered that the higher content could be related to missing colour separators [51] in one of the production plants. However, based on practical experience, even this content is not significant [43]. Moreover, the opaque flake limit is neither an EFSA requirement (not mentioned in Figure 7) nor an internal requirement [103] – it is instead only an informative parameter.

3.3.1.15 Limitations

This analysis has yielded many interesting findings regarding the different qualities of each supplier, both regarding the market they operate in and the type of waste collection from which PET bottles were supplied to recyclers for reprocessing into flakes. However, it had its limitations, which should be mentioned.

Firstly, the QC analysis of input flakes from which these data were obtained was carried out on a sample of a negligible quantity compared to the total quantity of PET flakes in the batch [43]. Unfortunately, the flakes are very heterogeneous in general and, even if sampling was carried out at several heights of different big bags [43], the limited representativeness of this samples must be considered. For example, a sample may be taken with a large particle made of another polymer (e.g., PETG), causing a considerable increase in the content of other polymers, even though the volume of other polymers in the rest of the batch may be minimal. In this case, a control sample was taken [43], which provided some degree of correction by averaging with the previous results or, conversely, confirm the previous analyses. However, these remain indicative values. Still, this method is ideal from the point of view of the trade-off between obtaining information on the quality of the batch and time efficiency.

Secondly, the entrance QC analysis was performed by more than one lab technician. Although parameters such as bulk density or moisture are relatively well reproducible, the assessment of some characteristics may initially be biased by lack of experience (e.g., recognition of PVC and other polymers). Some characteristics, such as visual assessment, then remained partly subjective even for trained workers. Thus, the results for some batches may be biased by the subjective perception of the lab technician.

In terms of relating the characteristics to the type of market, it is also important to note that most Western market processors had the abbreviation CM added to the initial letter (e.g., M_CM). In other words, most batches from the Western producers were delivered as consumer material (see Chapter 3.1 for more details). It is possible that the manufacturer was handling lower-quality batches in this way, which could have biased the deductions regarding the market. Still, although these methods introduce some limitations and a degree of uncertainty, they have been able to provide a range of information on more than 150 batches.

3.3.1.16 Conclusion of entrance QC analysis

The most important findings concerning the initial QC analysis are the results of the PVC and other contaminants analyses. Although most suppliers had no problems with PVC content, 16% of the batches exceeded the 50-ppm limit [103]. Despite the problems associated with PVC in PET have long been discussed and well described [5, 6, 29, 72], there are still batches with higher levels in almost one-fifth of the cases in our analysis. Other polymers were the most meaningful result of the analysis. Regarding the optimum for preventing damage to the technology (the 75-ppm limit for other polymers [103]), only two suppliers met this limit on average. In particular, 54% of the batches contained more than 75 ppm of other polymers. As already mentioned, other polymers are not subject to mandatory analysis or EFSA limits – only PA, PVC, and polyolefins are examined (refer to Figure 7 and [133]). However, as confirmed by practice [43] and literature (see Chapters 2.4.1.4, 2.4.1.5, and 2.4.2), other polymers can pose an equally serious risk to product quality and machine functionality. And as could be seen, there is indeed a problem with the content of other polymers across manufacturers. Besides, based on market trends in the content of other polymers and PVC, it was deduced that it is necessary to engage both manual (or machine vision) and automatic contaminant separation for these types of contamination.

Other parameters, such as PA, other contaminants, etc., were not as important, did not show any problem, and mostly no essential conclusion was drawn from them for either the collection type or the market.

It is worth noting that there are suppliers who have a problem with the separation of dark-coloured flakes. Furthermore, high degradation was observed during the visual evaluation of Western suppliers. Regarding the PET trays, there was surprising content in some batches from suppliers who process material from DRS or mono-collection systems.

For clarity, a table (Figure 29) was created for each supplier showing whether the QC analysis parameters were met. Green ticks stand for the parameters where the supplier's average was below the limit. Red crosses mean the average had a value above the limit. A yellow dash indicates that the parameter has not been exceeded on average, but that there have been other discrepancies for which the parameter could not be considered as clearly fulfilled. Typically, these were fluctuations; in the case of visual assessment, there were signs of degradation or poor washing. As a result, a count was made for each of these marks, and from their frequencies (and parameter importance), suitable and unsuitable suppliers were determined.

The cooperation with supplier E is recommended if possible. In that case, all parameters were met. However, only one batch from supplier E was available, and these results would need to be verified by multiple deliveries. The stable supplier O was the best performer that (on average) did not exceed any of the limits. Supplier D, could also be recommended, but care should be taken with moisture content (typically higher) and bulk density, which both fluctuated. Suppliers A and F are also recommended, but in their case, higher contents of other polymers should be anticipated.

In contrast, the CM suppliers (except L_CM) were problematic and did not meet several parameters, confirming the hypothesis of poorer quality in this type of supply compared to other batches. Also, suppliers R, B, and C could not be recommended due to several non-compliant parameters.

A case worth noting is supplier D_CM. This supplier delivered PET flakes produced in two countries. As a result, two phenomena are seen in the batches supplied. While some batches did not cross the limits, others showed substandard quality and even extremes. It was assumed that the inconsistency in these supplies is due to the difference in the technology employed in two existing plants. Thus, the recommendation is not to discontinue buying from this supplier, but rather to request supplies only from the plant that does not have quality problems.

For suppliers with average quality who have not been recommended or not recommended above, the processor should be guided by which parameters the PET flakes producer did not meet. For example, if there was a non-conformity for parameters such as PET tray content, it is not nearly as serious as a non-conformity with PVC content.

Producer	PVC	OP	PA	OC	MC	PET Dust	BD	FwA	BF	DCF	LCF	LB	VE	PET trays	✓	✗	—
A	—	✗	✓	✓	✓	✗	✓	✓	✓	✓	✓	✓	✓	✓	11	2	1
B	✗	✗	✓	✗	✓	—	✗	✓	✗	✓	✓	✗	✓	✗	6	7	1
C	—	✗	✓	✗	✓	—	✗	✓	—	✓	✓	—	—	✓	6	3	5
D	✓	✓	✓	✓	✗	✓	✗	✓	—	✓	✓	✓	✓	✓	11	2	1
D_CM	✓	✗	✓	—	✓	✓	—	✓	—	—	✓	✓	—	✗	7	2	5
E	✓	✓	✓	✓	✓	—	✓	✓	✓	✓	✓	✓	✓	✓	13	0	1
F	✓	✗	✓	✓	✓	—	✓	✓	✓	✓	✓	✓	✓	—	11	1	2
G	✗	✗	✓	✓	✓	—	✓	✓	✓	✓	✓	✓	—	✓	10	2	2
H_CM	✗	✗	✓	✗	✓	✗	✓	✓	—	✓	✓	✓	—	✗	7	5	2
I_CM	✗	✗	✓	✓	✓	✗	✓	✓	✗	✗	✓	✗	—	✗	6	7	1
K_CM	✗	✗	✓	✓	—	✓	✓	✓	—	✗	✓	—	—	✓	7	3	4
L_CM	✓	✗	✗	✓	✓	✓	✓	✓	✗	✓	✓	—	—	✓	9	3	2
M_CM	✗	✗	✓	✓	✓	✓	✗	✓	—	✗	✓	✗	—	✗	6	6	2
N	✓	✗	✓	✓	✗	✓	✓	✓	✓	✓	✓	✗	—	✗	9	4	1
O	✓	✓	✓	✓	✓	✓	—	✓	✓	✓	✓	✓	✓	—	12	0	2
P	✓	✗	✓	✓	✗	✗	—	✓	✓	—	✓	✓	✓	✓	9	3	2
Q	✗	✓	✓	✓	✓	✗	—	✓	✓	✓	✓	✓	—	✓	10	2	2
R	✗	✗	✓	✓	✗	—	✓	✓	✓	✗	✓	✓	—	—	7	4	3

Figure 29: Suppliers evaluation. The abbreviations stand for: OP – other polymers, OC – other contaminants, MC – moisture content, BD – bulk density, FwA – flakes with adhesives, BF – barrier flakes, DCF – dark-coloured flakes, LCF – light-coloured flakes, LB – light blue flakes, VE – visual evaluation. Source: own source.

3.3.2 Spectrophotometry (L^* , a^* , b^*)

3.3.2.1 L^* parameter

The L^* value for flakes was 64.04 on average. 25% of batches had an L^* value lower than 62.99. A slightly higher number of batches (32%) ranged from 63.00 to 64.99. A value higher than 65.00 was measured in the case of 43% of samples. The lowest measured value was 52.71; the highest was 70.47. Thus, a wide range of results was observed. See Figure 30.

Producer	Average [-]	Min [-]	Max [-]	DRS/Mono-collection	EU market
A	66.77±0.93	65.73	67.90	Yes	E
B	67.05±3.01	63.75	69.63	No	W
C	64.61±1.39	61.22	66.31	Yes	E
D	66.85±2.18	65.02	70.00	Yes/No	E
D_CM	63.71±2.70	58.50	68.56	Yes/No	E
E	69.30±0.00	69.30	69.30	No	E
F	67.43±1.46	64.77	70.47	No	E
G	67.57±0.00	67.57	67.57	No	E
H_CM	61.27±1.92	59.91	62.63	Yes	W
I_CM	58.62±1.30	57.27	60.20	No	W
K_CM	53.90±1.11	52.80	55.44	Yes	W
L_CM	53.04±0.45	52.71	53.36	No	W
M_CM	64.86±1.10	62.93	65.54	Yes	W
N	57.74±1.42	55.55	59.12	Yes	E
O	64.38±1.08	61.34	66.82	No	E
P	67.89±2.67	66.00	69.78	Yes	E
Q	68.19±0.58	67.78	68.60	No	E
R	68.06±1.37	67.02	69.97	No	E

Figure 30: The results for L^* parameter. Source: own source.

In general, the higher the L^* value of the regranulate, the better [43]. Thus, it is advisable to process material that itself has the highest L^* parameter. The reason is that a decrease in L^* parameter means a shifting of colour towards grey, which is typical for degraded flakes [75]. A higher L^* parameter should ensure a lower degree of degradation and consequently, the nicer colour of final rPET pellets [43, 75]. No spectrophotometric limits are set by EFSA (control as per Figure 7) or internal limits [103]. Based on the practical observations, it was considered that flakes of above-average quality should achieve a value of at least 65.00. Results higher than this value were measured in 43% of batches.

As a result, suppliers with higher L^* averages and stable colour should be preferred, such as R, F, and A. Suppliers B and D also had higher average L^* but with more significant fluctuations. Suppliers E, Q, P, and G seemed promising in terms of colour, but there were not enough batches (i.e., less than three) to provide more reliable conclusions. Importantly, there were suppliers that never reached an L^* value lower than 65.00 (E, Q, R, G, P, A, and D). In contrast, there were suppliers with a low average L^* (L_CM, K_CM, I_CM, N, and H_CM) that never managed to reach the 65.00 threshold. These batches were likely very degraded and thus not much recommended for producing rPET pellets of brighter colour.

Flakes from the co-collection system proved to have a higher L^* value. Nevertheless, it is important that this conclusion could be biased due to four producers with less than two batches. Correlation with the market was more significant. Eastern producers tended to achieve higher L^* values, whereas lower values were typical for Western producers. Again, this is likely related to previously discussed

assumption concerning PET recycling history and differences in rPET content used in PET bottles in different regions (see Chapter 3.3.1.12). However, this explanation was not further verified by reliable data.

Theoretically, a variable related to the degree of degradation was found. This relation is displayed in Figure 31, where increasing degradation is observable together with decreasing L^* value. Neither a^* nor b^* seem to show such a relation to degradation. Thus, the L^* parameter could be considered an appropriate indicator of degradation; more accurate and reproducible than currently used visual evaluation. In addition, greying caused by degradation that translates in decreasing L^* value has been already observed in scientific literature [75]. Creation of a table with ranges of L^* values corresponding to the individual grades for degradation evaluation or to directly use measured L^* values to describe the degradation is therefore proposed. Correlation analysis will be performed in the following chapter to support or deny these recommendations.

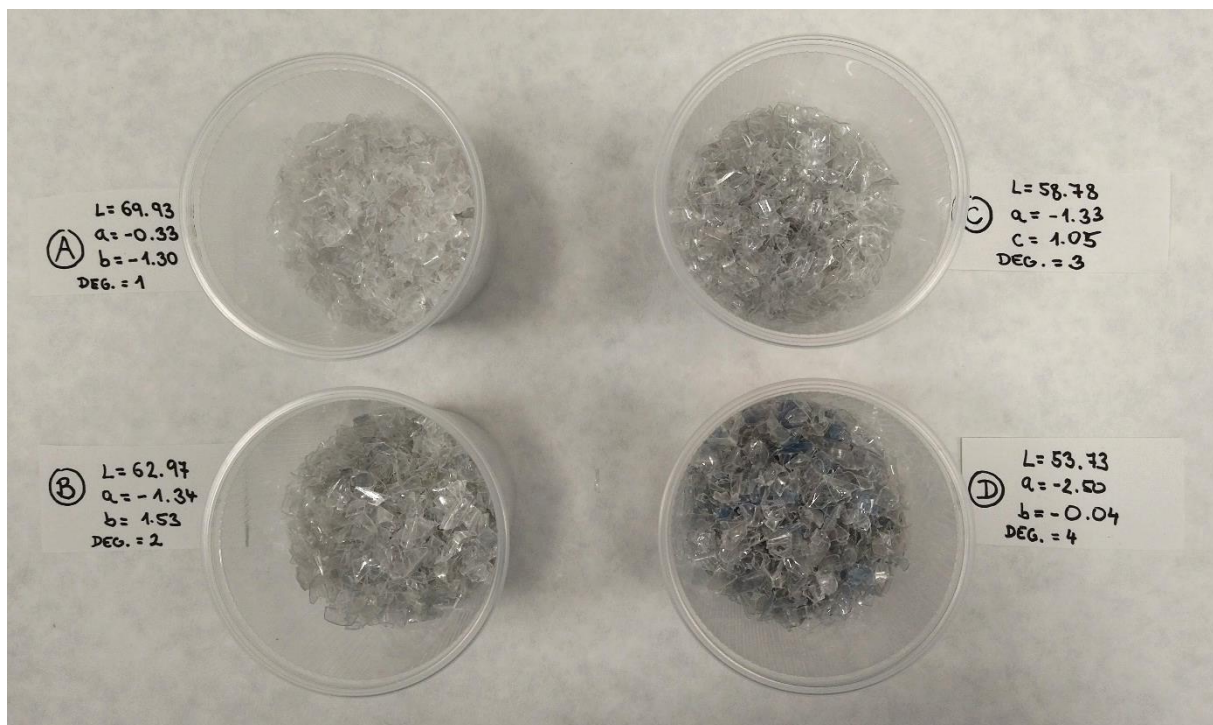


Figure 31: Colour parameters for differently degraded PET flakes. Source: own source.

3.3.2.2 a^* parameter

In 34% of batches, a^* value higher than -1.49 was observed. Additionally, 29% of batches had a a^* value between -1.50 and -1.99. Interval -2.00 to -2.99 contained 25% of batches. A value of a^* less than -2.99 was measured in 12% of batches. That means batches were rather green-coloured than red-coloured, but not significantly (refer to Figure 14 for graphical display of a^* axis). For more detailed information on results, see Figure 32.

Producer	Average [-]	Min [-]	Max [-]	DRS/Mono-collection	EU market
A	-1.78±0.32	-2.20	-1.33	Yes	E
B	-1.95±1.58	-3.50	-0.33	No	W
C	-1.80±0.79	-3.67	-1.19	Yes	E
D	-1.40±0.43	-1.71	-0.79	Yes/No	E
D_CM	-1.89±0.52	-3.11	-1.19	Yes/No	E
E	-0.56±0.00	-0.56	-0.56	No	E
F	-1.76±0.17	-2.05	-1.50	No	E
G	-1.16±0.00	-1.16	-1.16	No	E
H_CM	-2.07±0.04	-2.09	-2.04	Yes	W
I_CM	-4.09±0.13	-4.23	-3.89	No	W
K_CM	-2.67±0.30	-3.02	-2.36	Yes	W
L_CM	-3.18±0.30	-3.40	-2.97	No	W
M_CM	-3.60±0.76	-4.57	-2.63	Yes	W
N	-3.30±0.77	-4.79	-2.76	Yes	E
O	-1.64±0.38	-2.47	-1.15	No	E
P	-1.56±0.00	-1.56	-1.56	Yes	E
Q	-1.23±0.01	-1.24	-1.22	No	E
R	-1.62±0.15	-1.76	-1.50	No	E

Figure 32: The results for a* parameter. Source: own source.

No internal [103] or EFSA (see Figure 7) limit exists for a* parameter. Based on practice, flakes with a* value close to zero should be preferred [43]. The lowest values (thus those most distant from 0) were observed in suppliers I_CM, M_CM, N, L_CM, and K_CM. Still, this material is processable in terms of a*. Batches with high a* and low fluctuations were from producers D, R, O, F, A, and C. Additionally, E, G, Q, and P supplied batches with high a* value, but due to a small number of supplied batches, it is hard to predict the stability of this parameter.

As well as in the case of L*, no general trend could be applied in relation to the collection scheme, but there was a clear pattern in the type of market. Western producers tended to produce batches with low a*, thus more green material. Eastern producers, on the other hand, provided batches with higher a*, closer to zero. This can be related to the degradation; nevertheless, this pattern was also observed in light blue flakes content. This relation will be further elaborated in Chapter 3.3.5.

3.3.2.3 b* parameter

A wide range was measured for b*: results varied from around -5.00 to 3.00. The lowest values, i.e., values below -2.00; were measured in 7% of the batches. From -1.99 to -0.50 there were 14%; and from -0.49 to -0.01; 13% of the batches were measured. The range of 0.00 to 0.49 was measured in 21% of the batches. The interval of 0.50 to 1.99 was the largest one with 35% of batches. A b* value greater than 2.00 was found in 10% of the batches. Further details are displayed in Figure 33.

Producer	Average [-]	Min [-]	Max [-]	DRS/Mono-collection	EU market
A	1.25±0.34	0.84	1.88	Yes	E
B	-0.38±2.02	-1.79	1.94	No	W
C	1.99±1.45	-1.68	3.14	Yes	E
D	0.96±0.47	0.72	1.66	Yes/No	E
D_CM	0.64±1.26	-2.74	3.14	Yes/No	E
E	-0.66±0.00	-0.66	-0.66	No	E
F	1.60±0.75	0.24	3.27	No	E
G	1.74±0.00	1.74	1.74	No	E
H_CM	0.03±0.04	0.00	0.06	Yes	W
I_CM	-3.31±0.77	-4.00	-2.10	No	W
K_CM	0.19±0.34	-0.09	0.66	Yes	W
L_CM	-0.03±0.14	-0.12	0.07	No	W
M_CM	-2.89±1.85	-5.46	-0.80	Yes	W
N	-0.47±1.23	-2.67	1.10	Yes	E
O	0.04±0.51	-1.23	1.16	No	E
P	0.96±0.45	0.63	1.28	Yes	E
Q	2.90±0.12	2.81	2.98	No	E
R	1.48±0.12	1.31	1.59	No	E

Figure 33: The results for parameter b^* . Source: own source.

I_CM and L_CM producers had the lowest average b^* values. For the producers with low b^* , the shade of flakes should be rather blue. In contrast, the highest values were found by supplier Q (but only on average; individual batches for producers F, C, and D_CM exceeded this average). Flakes with a high b^* should be yellowish rather than bluish (refer to Figure 14 for graphical display of b^* axis).

No internal [103] or EFSA (see Figure 7) limit exists for the b^* parameter. Based on practice, a key factor is stability. It is preferable if the supplier's production is stable, also due to changes in additive dosage [43]. When not considering suppliers with two or fewer deliveries, R, A, K_CM, D, and O can be considered stable. Opposed to that, large fluctuations in the rest of producers were not the exception. The instability was typical for suppliers B, M_CM, C, D_CM, and N.

No association with collection systems was observed, but there was a dependence on the type of market. While the Eastern market had higher average of the parameter b^* , i.e., in the positive values (with exceptions for producers N and E), flakes from the Western market tended to be situated around zero to lower negative numbers. It is assumed that both degradation and light blue flake content are reflected in the b^* parameter, which was generally higher for more degraded flakes than for less degraded ones. This assumption will be tested by correlation analysis in Chapter 3.3.5.

3.3.2.4 Conclusion of spectrophotometric analyses

The most important parameter for spectrophotometric measurements was chosen to be L^* , which seemed to describe the degradation of the sample: the higher the L^* , the lower the degradation. This was in accordance with previous research [75] and additionally, supported the hypothesis of higher sample degradation for suppliers from the Western EU market. The L^* parameter has been proposed as a method for assessing degradation.

Parameters a^* and b^* did not show clear trends or problems with significant extremes in suppliers. However, they will be subjected to correlation analysis from which the possibility of using these parameters to improve existing methods will be assessed. Finally, although an effort was done to

evaluate suitable producers for each colour parameter, no supplier was unrecommended based on the spectrophotometric results.

3.3.3 Intrinsic viscosity

The first purpose of the IV analysis was to evaluate the EU PET flake market based on IV values. Only manufacturers with two or more batches will be included in the comparison⁵. According to the results, the IV averages ranged between 0.68 and 0.79. Of these, two producers were below 0.72; five producers showed values between 0.73 and 0.74. Seven producers fell in the interval of 0.75 to 0.77. Two producers reached an IV value of 0.78 or more. See Figure 34 for all averaged results.

Producer	IV [dl·g⁻¹]
A	0.77±0.01
B	0.72±0.00
C	0.76±0.01
D	0.75±0.01
D_CM	0.73±0.01
F	0.78±0.04
H_CM	0.75±0.00
I_CM	0.73±0.01
K_CM	0.73±0.01
L_CM	0.73±0.01
M_CM	0.73±0.01
N	0.75±0.02
O	0.75±0.01
P	0.77±0.00
Q	0.68±0.01
R	0.79±0.02

Figure 34: The results of IV analysis. Source: own source.

Producers R, F, and P supplied PET flakes with the highest IV. In contrast, the lowest IV was measured for K_CM, Q, and B. The lower the IV of the input material, the more challenging it is to raise it to the desired value [43]. IV-related limit is not given by EFSA (Figure 7) or internal limits [103]. Based on practical experience, the processor was advised to source flakes from manufacturers, ideally with an average IV of 0.75 and higher (even though some literature sources claim 0.70 to be the minimal limit [6]). IV value distinguishes between undegraded and degraded flakes. Generally, a lower IV value means higher degradation [3, 72, 74]. This causes problems from a certain level onward. The IV value of flakes is only affected by the input material, no other factor [43].

No correlation between collection systems was expected or observed. However, as in the case of colour analysis, there was a relatively clear distinction between the Western and Eastern market. The Eastern market with a higher average IV seems to process less degraded bottles. As was assumed in Chapter 3.3.1.12, this could support the conjecture of longer recycling history and higher shares of rPET in bottles in Western market. The only surprising result was the average IV for producer Q. This producer should have a lower degradation level and therefore higher IV on average because it was situated in the

⁵ The reasons for excluding manufacturers with one batch supplied are: (1) there was not enough material in one batch for repeated measurements and therefore a measurement error could not be captured, and (2) any IV variation between deliveries could not be inferred from one batch. In conclusion, these results were not provided because they could have biased the discussion of the results.

Eastern market. However, this supplier represents the producer with the lowest IV. This is most likely caused by measurement error.

The second purpose of the IV measurement was to develop a new method for measuring this parameter. Currently, a method often used to measure IV of polymers is dilute solution viscosity [138]. This method was considered relatively laborious, to use hazardous chemicals, and in the case of heterogeneous recycled PET flakes batches, also as highly unrepresentative [43]. It was hypothesized that the use of a plastometer could eliminate these negatives.

The very first challenge, however, was that only fragments of PET bottle-necks had to be collected for measurement in the plastometer (see Chapter 3.2.3). Thus, the method may partially lose its sensitivity and accuracy and serve more as comparative information. Moreover, picking these fragments is very time-consuming (up to half an hour of preparation per measurement). Secondly, after drying, the transfer of the sample to the plastometer needed to be as fast as possible. However, if an obstruction occurred (e.g., clogging of the funnel with bottle fragments), it delayed sample dispensing, and the PET began to incorporate moisture. As a result, the final IV value was biased (reduced). Again, this represented a big issue for the accuracy of the results, but also for the reproducibility.

Our results, although consistent with the trends observed for the rest of the measured parameters, cannot be considered with certainty to be accurate. Nor is it appropriate to choose this method in practice precisely because of the mentioned problems, such as measuring only PET bottleneck fragments, time demanding procedure, or difficulties with sample transfer into a measuring device. Nevertheless, this method would deserve further development because of the advantages it can bring (especially the elimination of chemicals). For example, it was proposed to compress the sample flakes into cylinders (as is mentioned by the standard [130]) that perhaps could be easier to handle and transfer to the chamber of the plastometer. Compressing the flakes into cylinders could also save time with the picking of PET bottleneck fragments. Compared to dilute solution viscosity, the change of the method could also save time and effort. Thus, this method deserves further research to develop an appropriate method for recycled PET flake IV measurements.

3.3.4 Other polymers and other contaminants

3.3.4.1 Trends found in plastic contamination

First, the most frequently encountered groups of contaminants were identified. The most common contaminants were transparent clear or slightly yellowish to orange PETG and PS or high impact polystyrene (HIPS) pieces. This type of contaminant was present in 131 batches out of 158, i.e., 83% of the batches. No other group of plastic contaminants was as prevalent. This finding is important. First, this group was more frequent than any group of PVC; thus, PETG and possibly PS or HIPS are the most frequent contaminants. If PS and HIPS fragments were prevalent in some samples, it is assumed that these contaminants could be possibly eliminated by increasing the sensitivity of the NIR separation equipment [110]. However, if it was predominantly PETG, this represents a problem as separation of PETG is difficult [44]. The second most frequent group of contaminants was a group containing darkened PVC labels. These were present in 91 batches, i.e., 58% of the batches. Thus, PVC contamination is still a problem.

No other group of plastic contaminants was as significant as the PETG/PS/HIPS or blackened PVC labels above. But several other frequently occurring groups were observed. These were the groups containing:

- another type of PVC labels with different discolouration (27% of batches),
- residuals of PP labels (25% of batches),
- polyolefin particles (23% of batches),
- PETG with incorporated inks and pigments (23% of batches),

- black contaminants with gloss, in particular PS and HIPS (19% of batches),
- PETG labels (20% of batches); see Figure 35.

It is important to note that five out of the seven most frequently occurring groups were thought to come from PET bottle components (blue bars in Figure 35). The other two groups (the most frequent consisting of PETG, PS, and HIPS, and the less frequent consisting of only PS and HIPS) may have come in part from poorly sorted material [100, 112] (hence shown as violet bars in Figure 35). However, they were also considered to be partly from bottle components – either the bottles themselves or the labels [89, 101, 108]. As a result, the most common contamination was evaluated to come predominantly from bottle components. Poorly sorted material was identified as a minor source compared to bottle components. This means that the introduction of a DRS or mono-collection system is not the way to reduce plastic contamination. In contrast, a change in bottle design is necessary.

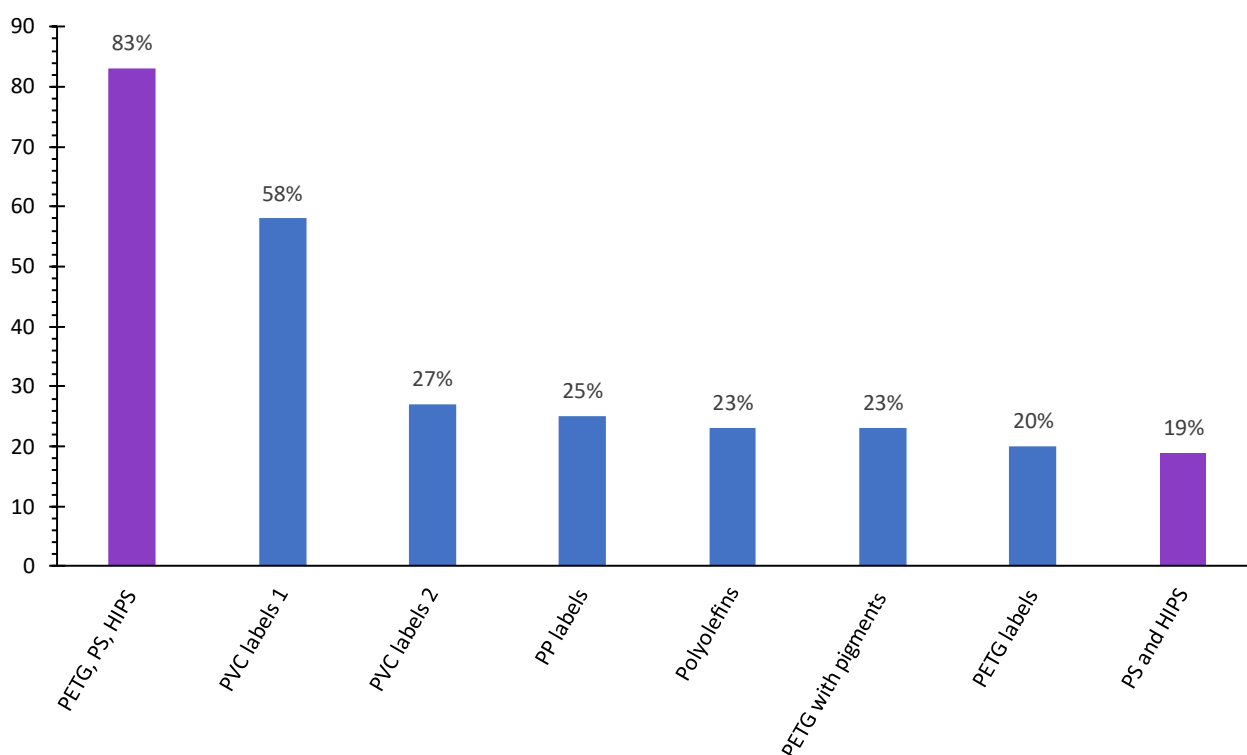


Figure 35: The most frequent groups of other contaminants (purple: origin in both bottle components and faulty-sorted material; blue: origin mainly or only in bottle components). Source: own source.

A result worth pointing out is that although polyolefins were present in a substantial part of the supply, they were not nearly as significant contaminants as, for example, PETG or PS, or HIPS. This is a substantial finding given that the standard EN 15348:2014 only describes the content of polyolefins, not the more frequent other plastic contaminants [133].

For non-plastic (other) contaminants, the frequency of occurrence per group was no higher than 9% of batches. Wood (9% of batches), rubber (8% of batches), and aluminium can residue (6% of batches) were the most frequently found extraneous contaminants.

Suppliers with the most heterogeneous batches in terms of groups (i.e., with the highest number of groups) were examined. Heterogeneity was observed in batches from supplier O, which contained up to 16 groups in one batch. This phenomenon was attributed to the co-collection system. It should be noted, however, that supplier O did not have problems complying with limits for other contaminants and other polymers, and thus probably contained several dissimilar contaminants with low mass. Other producers

were also heterogeneous concerning groups, including these processing materials from mono-collection or DRS collections. Additionally, supplier O contained some contaminants that have only been found in its batches. Of the plastics, it was group 58 with strange pieces resembling pebbles, see Figure 36. The polymer type was not identified, as it was not possible to examine this group on FTIR-ATR due to an irregular surface. Regarding other contaminants, various metal residues and stones were typical for producer O.



Figure 36: Contaminant group 58. Source: own source.

In terms of contamination by other plastics, there were generally groups that originated from bottles and were therefore expected in all types of collection systems (refer to Figure 35). Groups that could be attributed mainly to faulty sorting were logically expected to be more common in co-collection material. Contrary to expectations, however, the occurrence of such materials was also observed in flakes from the mono-collection/DRS system: not sporadically but even comparably to the flakes from the co-collection system. Considering how such extraneous and non-bottle-component plastics, especially in flakes from the DRS system, can occur at all is a topic for a broader discussion. Most importantly, our findings refute the argument that DRS/mono-collection solves the problem of contamination by other plastics. Contamination can also be introduced into the systems where PET bottles are collected separately, so based on these findings, preventing contamination was assessed a misguided argument in discussions about the introduction of mono-collection and DRS systems.

3.3.4.2 Evaluation of contaminant sorting QC analysis based on FTIR-ATR results

Apart from FTIR-ATR analysis of unidentified samples (see Chapter 3.3.4.3), few particles that were assessed to some existing groups were also subjected to measurements. The aim was to evaluate the current grouping. This grouping was used for analysis in this thesis and is used as a guide for contaminant recognition for lab technicians [43]. The 18 most frequent groups were evaluated in this analysis.

In 7 groups, there was no difference in the original composition of groups and measured results. In one group, a new type of plastic was determined but as this type of plastic was close in composition to the original polymer composition of a group (for example, SAN – styrene acrylonitrile in a group of styrenic plastics), this was not considered a discrepancy.

Nevertheless, in 10 inspected groups, the unexpected polymer was found such as PS in a group containing PETG or polybutylene terephthalate (PBT) in a group containing PS. Still, these findings do not represent a problem for regular laboratory operations since polymers discovered in 7 groups are still considered “other polymers” in the entrance QC analysis. In conventional analysis, polymers other than PVC and PA are not distinguished from each other, thus the results remain the same. The problem was found in 3 groups containing PVC and PA even though their presence was not expected in the groups. This could lead to mistaking one plastic for another and consequently, incorrect QC analysis results. The discrepancies in 10 groups also showed that the sorting of contaminants in this thesis can be partly biased. Still, the results of the sorting were used mainly to determine the general trends in contamination. Thus, no details were used for the analysis, and most likely, these observations were not burdened by mistaking the types of polymers.

An updated document with contaminant grouping was made to make the sorting more correct in the future. The reason the original contaminant grouping differed from the new one can be explained by the origin of the samples. Whereas in the previous paper, contaminant samples from only one producer were evaluated [104], this time, samples from different suppliers were identified.

An ideal option would be to conduct a detailed FTIR-ATR analysis of contamination in future research. Still, it is important to acknowledge that there will always be discrepancies in the results, not only due to the resemblance in some polymeric spectra and visual characteristics of polymer particles but also due to the diversity of plastic products and components that can infiltrate PET flakes. The question therefore remains whether it would be more appropriate to use research capacity in other areas of recycled PET flake quality.

3.3.4.3 New types of contaminants determined by FTIR-ATR

During sorting the contaminants into groups, several particles were not possible to assign to the existing groups. These were subjected to FTIR-ATR analysis, and they were identified based on the match with spectra from OPUS libraries.

Polypropylene and ethylene-propylene-diene monomer (PP+EPDM)

PP+EPDM material is a result of combining PP with ethylene-propylene-diene monomer (EPDM) rubber. The material can come with various properties due to the possibility of modifications in the manufacturing process. It is used in a variety of applications, such as automotive, toys, packaging, or household and construction products and materials [139]. This hybrid material (see Figure 37) was found in a batch that came from co-collection sorting, so the origin from faulty sorting was presumed. Based on information about NIR in Chapter 2.2.2.3, it was concluded that PP+EPDM should be easy to separate once producers can use NIR sorters and a device is prepared for the occurrence of PP+EPDM. Still, this rubber-thermoplastic composite was found only in one sample and a more significant occurrence was not considered to be likely.

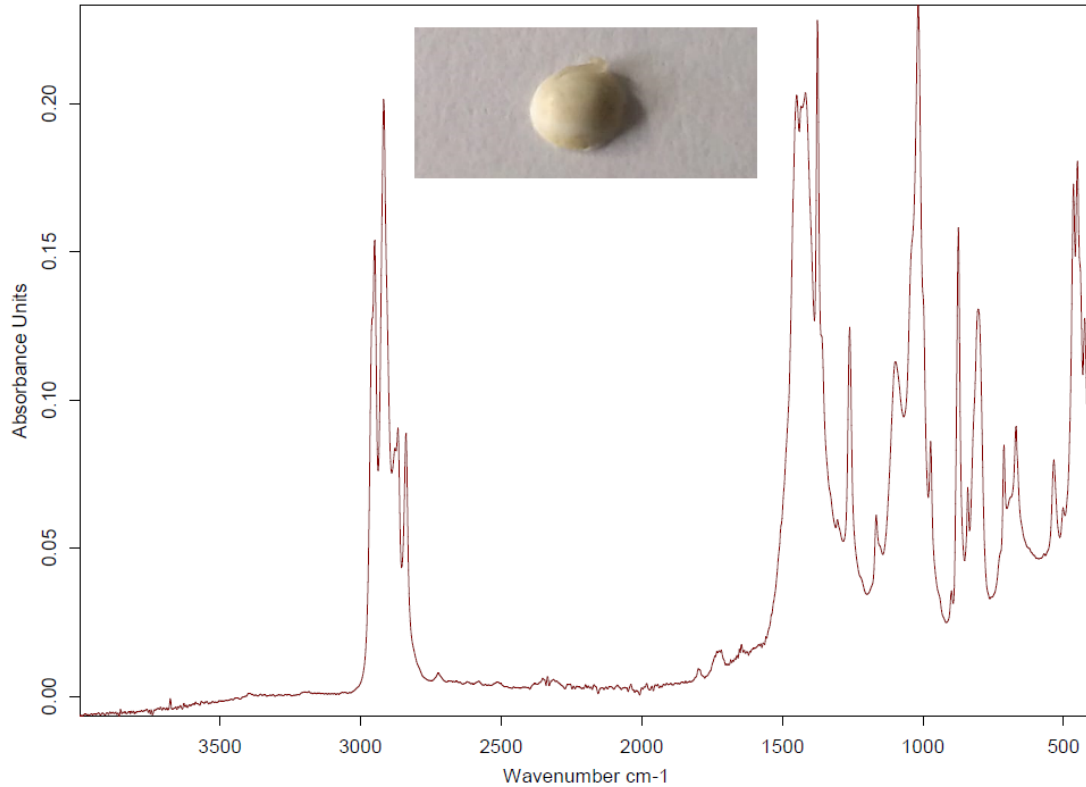


Figure 37: PP+EPDM contaminant particle and its spectra. Source: own source.

Poly (methyl methacrylate) (PMMA)

PMMA is a material known to be used in applications where inorganic glass needs to be substituted [140]. In the original grouping, this polymer was expected but it was not found among the analysed particles [104]. In this analysis, two particles were identified as PMMA (see Figure 38). Both particles were found in material from the co-mingled collection. The separation of PMMA should not be a problem with a properly adjusted NIR device [110].

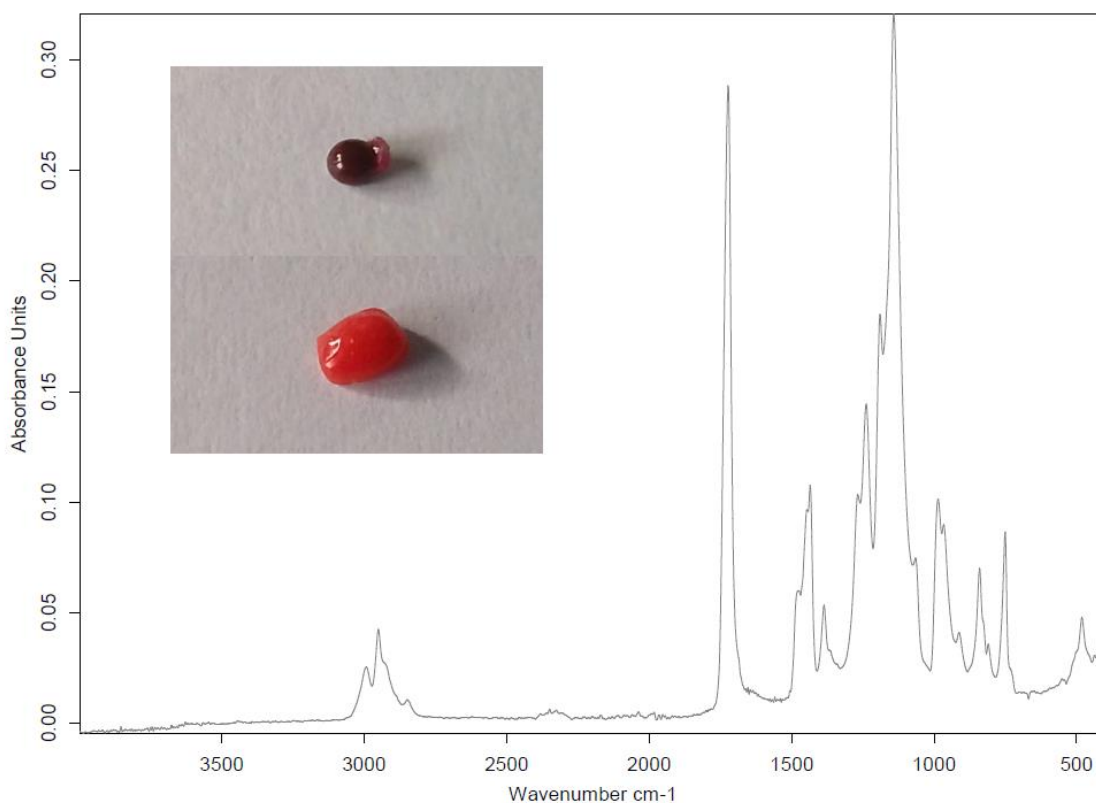


Figure 38: An example of PMMA spectra with particles identified as PMMA. Source: own source.

Polyurethane (PU)

PU is claimed to be a very universal material with a range of applications. For consumers, a known application is PU foam, but this plastic can be rigid and elastic as well. For PET bottles, the usage of PU as an adhesive is also important [141]. Two particles containing PU were found in the FTIR-ATR analysis. The first one was PET probably with a layer of PU adhesive, yielding combined polyester and PU spectrum. The second particle was a rubber-like piece of PU (see Figure 39). Based on information about NIR in Chapter 2.2.2.3, it was assumed that sorting of particles made only of PU should be performed effectively with NIR systems. Nevertheless, the problem could arise once PU is in a form of a layer on PET bottle flake. The problem is quite the same as for PET flakes with the PA layer [94]. Both particles containing PU were found in batches produced from co-mingled sorted material.

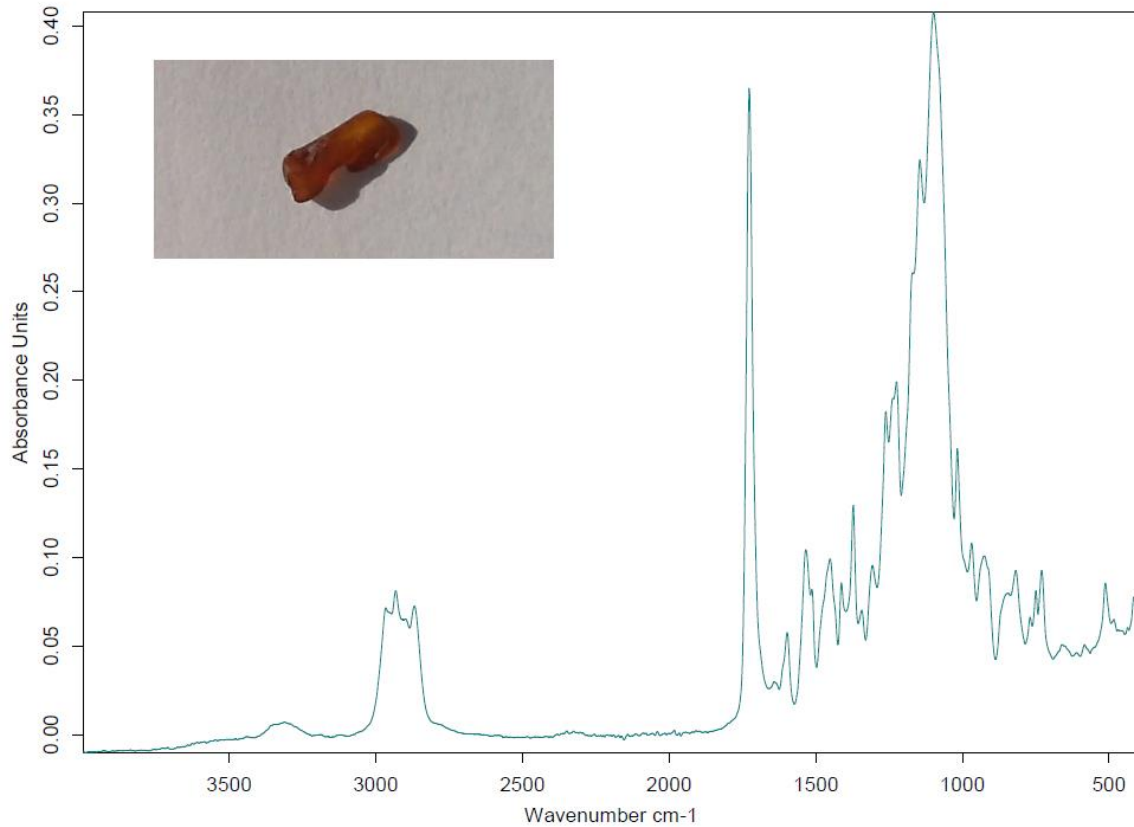


Figure 39: PU particle and its spectrum. Source: own source.

Polydimethylsiloxane (PDMS)

PDMS or silicone is a rather inert, non-toxic material and as such, it is used in special applications like coatings, medicine, or automotive [142]. It can be also used as an additive, especially as a release agent [143]. The measured particle was matt, opaque, and very brittle (see Figure 40). PDMS occurrence in PET flakes was not expected; moreover, it was found in a batch that produced bottles from DRS/mono-collection system. Based on information about NIR in Chapter 2.2.2.3, it is expected that PDMS separation based on NIR spectra should not be problematic, except for black particles such as the one found in this analysis. It would be useful to examine the topic further to determine the source of PDMS contamination.

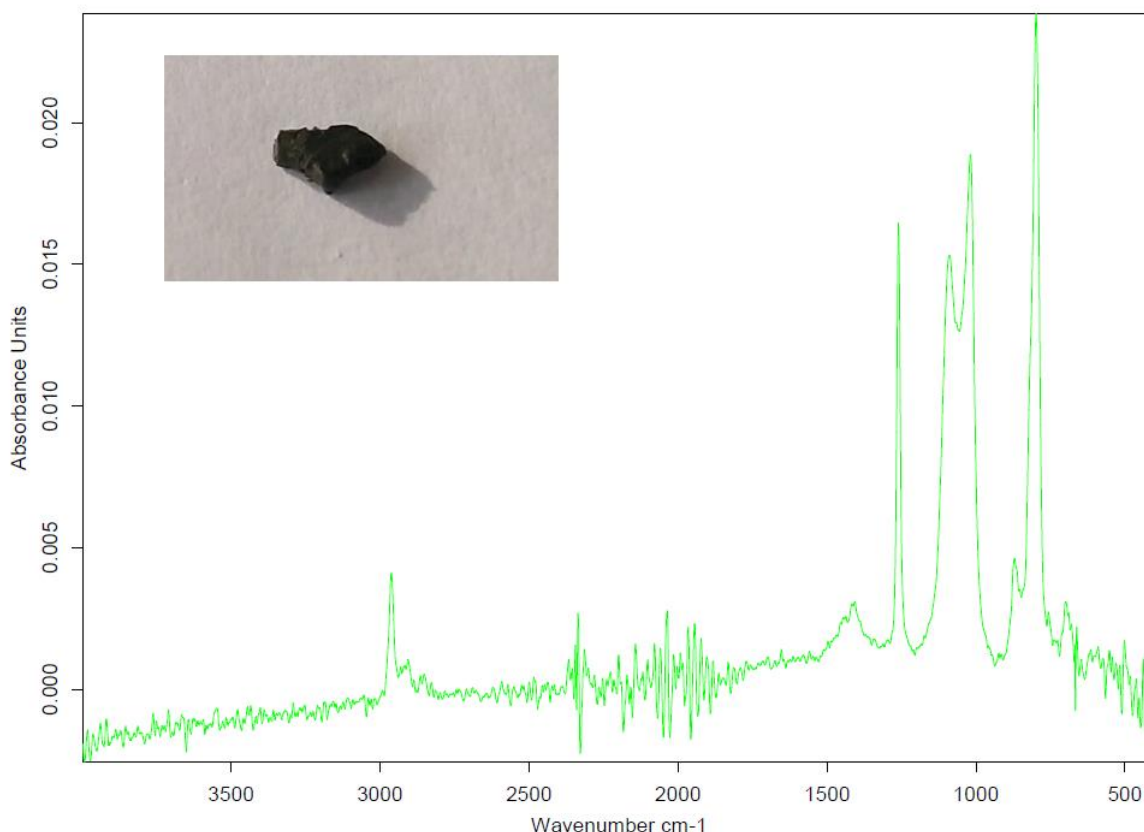


Figure 40: PDMS particle and its spectrum. Source: own source.

3.3.5 Correlation analysis

During the discussion so far, several hypotheses about the relationship between the parameters have emerged. A large dataset was used to analyse the correlation between the pairs of variables. This analysis was performed between all possible pairs of variables measured in the input QC analysis. Subsequently, these parameters were also correlated with the L^* , a^* , and b^* parameters, where only those batches that could be spectrophotometrically measured were included. Still, only the ones where the correlation was hypothesised will be discussed. No data cleansing was performed before the correlation analysis.

3.3.5.1 Contaminant correlation

For contaminants, the link to the sorting technology of the manufacturer was often mentioned. Hypothetically, this dependence should be reflected by a correlation between PVC, other polymer contaminants, other contaminants, PA, and PET trays. Correlation with PET trays could be expected, especially with other polymers and other contaminants where the employment of manual separation is considered to play a significant role. However, PA is specific – in fact, the PA layer must be peeled off from the barrier flakes, which will possibly affect the PA content [43]. This variable should probably not be correlated with others. The results are shown in Figure 41.

	PVC	Other polymers	Other contaminants	PA	PET trays
PVC	x	0.524	0.301	-0.019	0.163
Other polymers	0.524	x	0.198	0.140	0.247
Other contaminants	0.301	0.198	x	0.070	0.172
PA	-0.019	0.140	0.070	x	0.218
PET trays	0.163	0.247	0.172	0.218	x

Figure 41: Pearson coefficients obtained in contamination correlation analysis. Source: own source.

Based on the results, a correlation between PVC, other polymers, and other contaminants was found. PVC and other polymers yielded the highest correlation coefficient. For other contaminants, the origin and separation may be more complex (e.g., completely different separators for aluminium and metals [54, 55]). This fact could explain the lower correlation. Low Pearson coefficient values for PA were not surprising. A higher correlation with residual contaminant particles was expected for PET trays, but it is evident that the content of PET trays is also influenced by other factors.

3.3.5.2 Light blue flakes

The light blue flakes were assumed to be related to the visual evaluation, where light blue flakes could be added intentionally to reduce the need for additives. It was also believed that light blue flakes were related to other coloured flakes. Additionally, there could be a link between the light blue flakes and the barrier flakes, possibly to suppress the yellowing of the recyclate due to the thermal degradation of the barrier compounds (for origin of these hypotheses, refer to Chapter 3.3.1.11). See the results of the correlation in Figure 42.

	Visual evaluation	Light flakes	Dark flakes	Barrier flakes
Light blue flakes	0.380	0.316	0.001	0.406

Figure 42: Light blue flakes correlated to selected variables. Source: own source.

An intended addition of light blue flakes due to the visual evaluation seemed to be possible, at least in some batches. The Pearson coefficient between light blue and visual evaluation could be skewed as the quality of washing is also evaluated in visual evaluation but should not be a reason for higher light blue flakes content. Also, there was a high correlation with barrier flakes.

Correlation analysis between light blue flakes, light-coloured flakes, and dark-coloured flakes yielded different results. Light blue and light flakes showed a certain degree of correlation; probably due to turquoise flakes that may penetrate together with light blue flakes but cannot be considered as light blue [43]. The same phenomenon for dark blue flakes was expected, but it seems that dark blue flakes are separated with a high degree of precaution as they have a much greater impact on the final colour [43].

3.3.5.3 Barrier flakes

First, barrier flakes were expected to correlate with PA based on the assumption that the more flakes with a barrier, the greater the chance of PA layer detachment. The next aim was to determine if the content of barrier flakes affects the visual evaluation performed during QC analysis. The washing quality that is evaluated in the visual inspection is only detectable after the flakes are heated and it is characterized by the yellowing of the flakes [43]. However, this is a yellowing of poorly washed chemicals which has nothing to do with the barrier layer on the barrier flakes. Nevertheless, there is a risk of distortion of visual perception, especially in batches with elevated levels of barrier flakes.

The Pearson coefficient between barrier flakes and PA was 0.238. Although this value is not negligible, a significantly higher value of the correlation coefficient was expected. It seems that the direct correlation between the quantity of barrier bottles and the PA content is not always relevant – mainly because the peeling off the barrier layer is probably further influenced by the intensity of abrasion in the washing process [43], but also perhaps by the design and manufacturing technology of the barrier bottles themselves. In addition, residual PA from other applications is evaluated as the PA content but is not related to the barrier flakes.

The Pearson coefficient between barrier flakes and visual evaluation was 0.447. This coefficient is considerable. Even though hypothetically, higher content of barrier flakes could be typical for degraded and poorly washed flakes, the possibility of biasing visual evaluation is also possible. This could pose a problem in the visual evaluation. Therefore, the evaluation of the washing quality by the laboratory

staff should be revised and if an irregularity is found, the method should be modified to avoid bias due to barrier flakes.

3.3.5.4 Colour parameters and visual evaluation

Value L* showed to yield the highest Pearson coefficient in the correlation analysis (see Figure 43). This supports our proposal to change the method of degradation evaluation for PET flakes, described above and in Chapter 3.2.1.3. The finding is also consistent with previous research [75]. It is important to consider, that visual evaluation is an average of degradation and quality of washing. Once only degradation would be considered, it is possible to believe that the correlation would be much higher.

	L*	a*	b*
Visual evaluation	-0.544	-0.408	-0.066

Figure 43: Pearson coefficient values between colour parameters and visual evaluation. Source: own source.

3.3.5.5 Colour parameters and barrier flakes

b* parameter was presumed to yield the highest Pearson coefficient and as such should be the most related to barrier flakes. This hypothesis was already examined and generally confirmed by research, that determined the presence of a barrier layer in a bottle based on b* [5]. However, the a* and L* parameters had higher Pearson coefficients (see Figure 44). The analysis of the mentioned study took place on individual bottles; in the case of this thesis, a heterogeneous sample of flakes from many bottles was measured, possibly containing different coloured flakes, contaminants, and light blue flakes that can largely influence the b* parameter [5].

	L*	a*	b*
Barrier flakes	-0.496	-0.506	-0.224

Figure 44: Pearson coefficients between barrier flakes and colour parameters. Source: own source.

L* and a* approximation to barrier flakes content should be done with caution. The existing method of evaluation of the barrier flakes was evaluated to be sufficient, and the result of the spectrophotometric analysis to be rather indicative information.

3.3.5.6 Colour parameters and light blue flakes

It was hypothesized that light blue flakes would be most highly correlated with the parameter b*, as this corresponds to the axis between blue and yellow (see Figure 14). Although these two variables showed a correlation, the a* parameter yielded a significantly higher correlation with light blue flakes (see Figure 45).

	L*	a*	b*
Light blue flakes	-0.505	-0.87	-0.742

Figure 45: Correlation between light blue flakes and colour parameters. Source: own source.

This difference was surprising and therefore it was decided to inspect the results visually by comparing the batches with different light blue flakes content. Indeed, the a* parameter appeared to decrease with increasing light blue flake content, while the b* parameter showed no clear pattern (see Figure 46). This may be due to the influence of barrier flakes on the b* parameter [5].

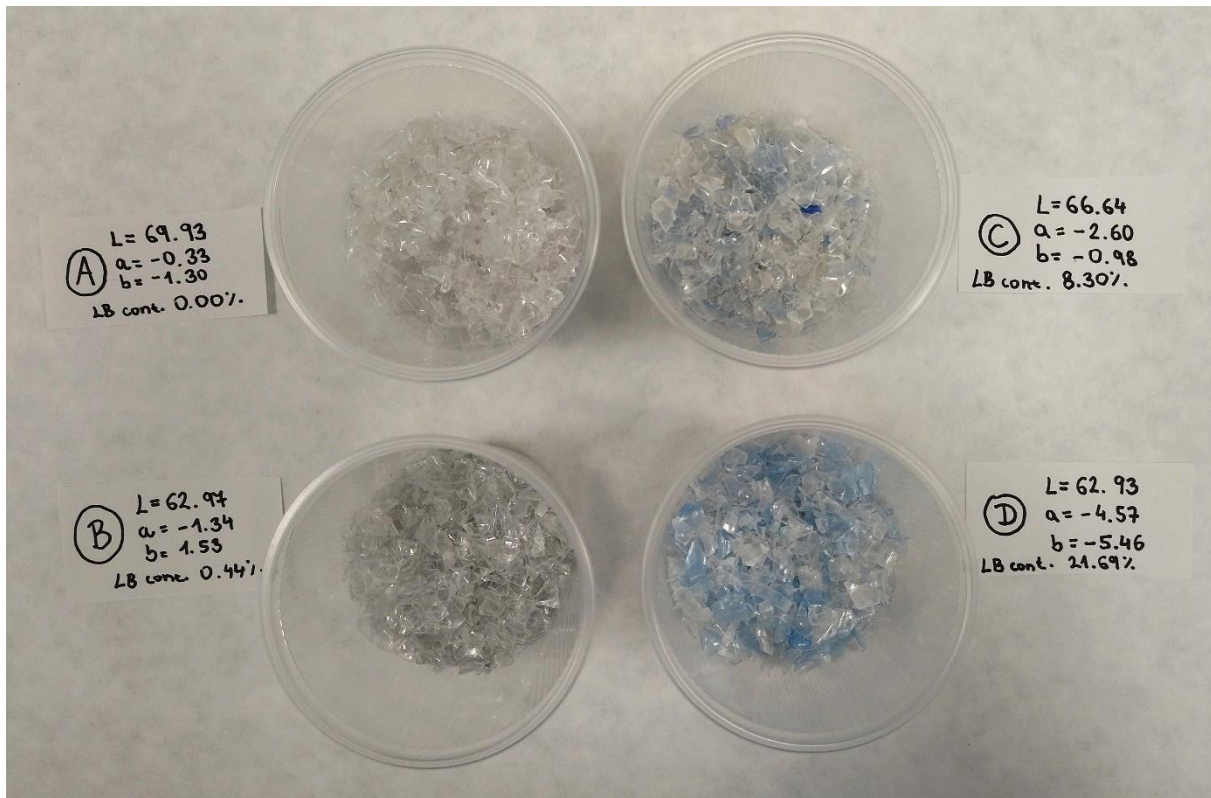


Figure 46: Relation between light blue flakes and colour parameters. Source: own source.

The use of the a^* parameter instead of the current method of light blue flake analysis would have to be subjected to further tests to reflect on whether this method can provide reliable information.

3.3.5.7 Bulk density and PET dust

It was assumed that the finer the PET flakes (i.e., the higher the bulk density), the higher the PET dust content. This topic was already inspected in Chapter 3.3.1.7. The Pearson coefficient obtained in correlation analysis was 0.334. Thus, PET dust was evaluated as partly dependent on bulk density (i.e., fineness of grinding) – nevertheless, it is important to consider that PET dust is also dependent on PET dust removal technologies [3]. Batches with low PET dust removal efficiencies yet low bulk density and at the same time, batches with higher bulk density but highly effective dust removal were observed in QC analysis. Such cases undermine the apparent notion of a clear positive correlation between these two variables.

4 CONCLUSION

This work aimed to provide a detailed qualitative analysis of the EU PET flake market. Also, an effort was made to find quality trends and their causes, propose new methods in the field of PET flakes quality measurement, make recommendations connected to the individual suppliers, as well as to map current challenges in food-grade rPET recycling. In addition, a comprehensive literature review focusing on recycling technologies and quality parameters was carried out in the theoretical part.

The first large group of parameters that were measured were the entrance QC quality parameters. The contamination of PET flakes with other plastics was the most important result. The limit of other polymers optimal for the proper functioning of the technology and the quality of rPET was observed in only 46% of the batches, and on average in only 4 suppliers out of 18. Although contamination by other polymers can have profound consequences (and according to the results of the analyses, it is a frequently occurring contamination), this parameter is not determined by EFSA. Furthermore, many manufacturers had problems complying with the limit for PVC content, even though PVC is a long and intensively discussed and monitored contaminant.

Furthermore, spectrophotometric analyses were performed. The parameter L^* was determined as an important indicator of the degradation of PET flakes. This parameter was thus proposed to develop a new method to determine the degradation of PET flakes to replace the existing visual method. This proposal was encouraged by correlation analysis between L^* and visual evaluation.

Concerning IV analysis, the aim was to propose a new method for IV measurement of PET flakes with a plastometer. Yet, the method was not reliable and accurate and thus no conclusions were drawn, although the results were consistent with the assumptions. Still, with further development of this method, it is believed that the method of measuring IV with a plastometer would be applicable in practice.

The analysis of polymer contaminants showed that the most frequent and abundant contaminants are PETG, PS, polyolefins, HIPS, and PVC. Firstly, the EFSA requirements do not take the occurrence of PETG, PS, and HIPS into account despite their frequent occurrence. Furthermore, the most frequent polymer contaminants originate wholly or at least partially in the components of PET bottles, making PET bottles themselves the most critical source of contamination in the EU.

For a general assessment of the EU market, this market has been divided into Western and Eastern. The Western market was found to be characterised by a higher degradation of PET flakes but also by a higher content of PVC and other polymers. Additionally, all the recommended suppliers operated in the Eastern market, while most of the non-recommended suppliers operated in the Western market. This may be due to the absence of manual sorting in Western countries, but also due to the multiple recycling cycles of PET bottles in the Western EU.

As far as collection systems are concerned, it was found that since the polymeric contamination mainly comes from the components of PET bottles, the collection system does not influence the contamination content. Moreover, batches from DRS and mono-collection also contained faulty-sorted objects. Thus, it needs to be acknowledged that the implementation of DRS for PET bottles is not a guarantee of lower input contamination and therefore may not be of significant help to recyclers.

While this work will yield important insights, optimising PET bottle recycling is still a long way off. To start with, given the results of the contamination origin analysis, the need for appropriate eco-design of PET bottles to better manage polymer contamination should be emphasised, either at the initiative of manufacturers or by directly embedding it in legislation. Furthermore, modifying the EFSA requirements to include a limit on other polymers was recommended. In the area of quality, it would also be appropriate to investigate other parameters of PET flakes such as volatile content, metal content, pH, etc., to a similar extent.

BIBLIOGRAPHY

1. WELLE, Frank. Twenty years of PET bottle to bottle recycling—An overview. *Resources, Conservation and Recycling*. 1 September 2011. Vol. 55, no. 11, p. 865–875. [Accessed 10 January 2023]. DOI 10.1016/J.RESCONREC.2011.04.009. Available from: <https://www.sciencedirect.com/science/article/abs/pii/S0921344911000656>
2. EFSA Panel on food contact materials, enzymes, flavourings and processing aids (CEF). Scientific Opinion on the criteria to be used for safety evaluation of a mechanical recycling process to produce recycled PET intended to be used for manufacture of materials and articles in contact with food. *EFSA Journal* [online]. July 2011. Vol. 9, no. 7. [Accessed 20 March 2023]. DOI 10.2903/j.efsa.2011.2184. Available from: <http://doi.wiley.com/10.2903/j.efsa.2011.2184>
3. PAWLAK, Andrzej, PLUTA, Mirosław, MORAWIEC, Jerzy, GALESKI, Andrzej and PRACELLA, Mariano. Characterization of scrap poly(ethylene terephthalate). *European Polymer Journal* [online]. September 2000. Vol. 36, no. 9, p. 1875–1884. [Accessed 6 March 2023]. DOI 10.1016/S0014-3057(99)00261-X. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S001430579900261X>
4. SNELL, H, NASSOUR, A and NELLES, M. Qualitative comparison of polyethylene terephthalate flakes from various collection systems in Germany. *Waste Management & Research: The Journal for a Sustainable Circular Economy* [online]. 17 January 2017. Vol. 35, no. 2, p. 163–171. [Accessed 10 March 2023]. DOI 10.1177/0734242X16686413. Available from: <http://journals.sagepub.com/doi/10.1177/0734242X16686413>
5. THODEN VAN VELZEN, E. U., BROUWER, M. T. and MOLENVELD, K. *Technical quality of rPET* [online]. Wageningen : Wageningen UR Food & Biobased Research, 2016. [Accessed 28 April 2023]. ISBN 978-94-6257-723-7. Available from: <https://edepot.wur.nl/392306>
6. AWAJA, Firas and PAVEL, Dumitru. Recycling of PET. *European Polymer Journal* [online]. July 2005. Vol. 41, no. 7, p. 1453–1477. [Accessed 8 April 2023]. DOI 10.1016/j.eurpolymj.2005.02.005. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0014305705000728>
7. FORREST, Martin J. 1. Introduction to Polyethylene Terephthalate Recycling. In: *Recycling of Polyethylene Terephthalate* [online]. 2nd ed. Berlin/Boston: De Gruyter, 2019. p. 1-13. [Accessed 3 January 2023]. ISBN 978-3-11-064030-4. Available from: <https://www.degruyter.com/document/doi/10.1515/9783110640304-001/html?lang=en>
8. PLASTICS EUROPE. Plastics - the Facts 2021: An analysis of European plastics production, demand and waste data. In: *Plastics Europe* [online]. c2021. [Accessed 4 March 2023]. Available from: <https://plasticseurope.org/wp-content/uploads/2021/12/Plastics-the-Facts-2021-web-final.pdf>
9. TANIGUCHI, Ikuo, YOSHIDA, Shosuke, HIRAGA, Kazumi, MIYAMOTO, Kenji, KIMURA, Yoshiharu and ODA, Kohei. Biodegradation of PET: Current Status and Application Aspects. *ACS Catalysis* [online]. 8 April 2019. Vol. 9, no. 5, p. 4089–4105. [Accessed 1 March 2023]. DOI 10.1021/acscatal.8b05171. Available from: <https://pubs.acs.org/doi/10.1021/acscatal.8b05171>
10. DEVASAHAYAM, Sheila, BHASKAR RAJU, Guntamadugu and MUSTANSAR HUSSAIN, Chaudhery. Utilization and recycling of end of life plastics for sustainable and clean industrial

- processes including the iron and steel industry. *Materials Science for Energy Technologies* [online]. December 2019. Vol. 2, no. 3, p. 634–646. [Accessed 9 March 2023]. DOI 10.1016/j.mset.2019.08.002. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S2589299119300618>
11. FORREST, Martin J. 5. Separation and sorting technologies. In: *Recycling of Polyethylene Terephthalate* [online]. 2nd ed. Berlin/Boston: De Gruyter, 2019. p. 75-99. [Accessed 3 January 2023]. ISBN 978-3-11-064030-4. Available from: <https://www.degruyter.com/document/doi/10.1515/9783110640304-005/html>
 12. MUÑOZ MENESES, Rodrigo A., CABRERA-PAPAMIJA, Gerardo, MACHUCA-MARTÍNEZ, Fiderman, RODRÍGUEZ, Luis A., DIOSA, Jesús E. and MOSQUERA-VARGAS, Edgar. Plastic recycling and their use as raw material for the synthesis of carbonaceous materials. *Heliyon* [online]. March 2022. Vol. 8, no. 3, p. e09028. [Accessed 27 February 2023]. DOI 10.1016/j.heliyon.2022.e09028. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S2405844022003164>
 13. BENYATHIAR, Patnarin, KUMAR, Pankaj, CARPENTER, Gregory, BRACE, John and MISHRA, Dharmendra K. Polyethylene Terephthalate (PET) Bottle-to-Bottle Recycling for the Beverage Industry: A Review. *Polymers* [online]. 11 June 2022. Vol. 14, no. 12, p. 2366. [Accessed 27 February 2023]. DOI 10.3390/polym14122366. Available from: <https://www.mdpi.com/2073-4360/14/12/2366>
 14. AL-SABAGH, A.M., YEHIA, F.Z., ESHAQ, Gh., RABIE, A.M. and ELMETWALLY, A.E. Greener routes for recycling of polyethylene terephthalate. *Egyptian Journal of Petroleum* [online]. March 2016. Vol. 25, no. 1, p. 53–64. [Accessed 12 April 2023]. DOI 10.1016/j.ejpe.2015.03.001. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S1110062115000148>
 15. UEKERT, Taylor, SINGH, Avantika, DESVEAUX, Jason S., GHOSH, Tapajyoti, BHATT, Arpit, YADAV, Geetanjali, AFZAL, Shaik, WALZBERG, Julien, KNAUER, Katrina M., NICHOLSON, Scott R., BECKHAM, Gregg T. and CARPENTER, Alberta C. Technical, Economic, and Environmental Comparison of Closed-Loop Recycling Technologies for Common Plastics. *ACS Sustainable Chemistry & Engineering* [online]. 12 January 2023. Vol. 11, no. 3, p. 965–978. [Accessed 18 April 2023]. DOI 10.1021/acssuschemeng.2c05497. Available from: <https://pubs.acs.org/doi/10.1021/acssuschemeng.2c05497>
 16. CHIRAYIL, Cintil Jose, MISHRA, Raghvendra Kumar and THOMAS, Sabu. Materials Recovery, Direct Reuse and Incineration of PET Bottles. In : THOMAS, Sabu, RANE, Ajay, KANNY, Krishnan, V.K., Abitha and THOMAS, Martin George, *Recycling of Polyethylene Terephthalate Bottles* [online]. Elsevier, 2019. p. 37–60. [Accessed 20 January 2023]. ISBN: 978-0-12-811361-5. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9780128113615000031>
 17. JANKAUSKAITE, Virginija, MACIJAUSKAS, Gintaras and LYGAITIS, Ramūnas. Polyethylene Terephthalate Waste Recycling and Application Possibilities: a Review. *MATERIALS SCIENCE (MEDŽIAGOTYRA)*. 2008. Vol. 14, no. 2, p. 119–127. [Accessed 13 March 2023]. Available from: <https://matsc.ktu.lt/index.php/MatSc/article/view/26243>
 18. DIRECTIVE (EU) 2019/904 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 5 June 2019 on the reduction of the impact of certain plastic products on the environment (Text with EEA relevance). In: *Official Journal of the European Union* [online].

- 12 June 2019. [Accessed 2 February 2023]. Available from: <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32019L0904&from=EN>
19. LUBONGO, Cesar and ALEXANDRIDIS, Paschalis. Assessment of Performance and Challenges in Use of Commercial Automated Sorting Technology for Plastic Waste. *Recycling* [online]. 23 February 2022. Vol. 7, no. 2, p. 11. [Accessed 13 March 2023]. DOI 10.3390/recycling7020011. Available from: <https://www.mdpi.com/2313-4321/7/2/11>
 20. DUROJAYE, Omoniyi, LASEINDE, Timothy and OLUWAFEMI, Ifetayo. A Descriptive Review of Carbon Footprint. In : AHRAM, T., KARWOWSKI, W., PICKL, S. and TAIAR, R., *Human Systems Engineering and Design II* [online]. Cham : Springer International Publishing, 2019. p. 960-968. [Accessed 4 April 2023]. Advances in Intelligent Systems and Computing. ISBN 978-3-030-27927-1. Available from: http://link.springer.com/10.1007/978-3-030-27928-8_144
 21. VISCOTEC. Carbon Footprint: Plastic producers to be forced to measure the carbon footprint of their products?. In: *Viscotec* [online]. November 2017. [Accessed 4 February 2023]. Available from: <https://www.viscotec.at/visconews-successful-partnerships-and-more/details/carbon-footprint/>
 22. WONG, Chee. A Study of Plastic Recycling Supply Chain. In: *CILT UK* [online]. The Chartered Institute of Logistics and Transport UK, 2010. [Accessed 27 March 2023]. ISBN 1-904564-36-4. Available from: <https://www.ciltuk.org.uk/portals/0/documents/pd/seedcornwong.pdf>
 23. ROY, Poritosh, NEI, Daisuke, ORIKASA, Takahiro, XU, Qingyi, OKADOME, Hiroshi, NAKAMURA, Nobutaka and SHIINA, Takeo. A review of life cycle assessment (LCA) on some food products. *Journal of Food Engineering* [online]. January 2009. Vol. 90, no. 1, p. 1–10. [Accessed 4 April 2023]. DOI 10.1016/j.jfoodeng.2008.06.016. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0260877408002793>
 24. BROCK, Alice and WILLIAMS, Ian D. Life Cycle Assessment of Beverage Packaging. *Detritus* [online]. December 2020. Vol. 13, p. 47–61. [Accessed 12 April 2023]. DOI 10.31025/2611-4135/2020.14025. Available from: <https://digital.detritusjournal.com/doi/14025-368>
 25. FERRARA, Carmen, DE FEO, Giovanni and PICONE, Vincenza. LCA of Glass Versus PET Mineral Water Bottles: An Italian Case Study. *Recycling* [online]. 15 July 2021. Vol. 6, no. 3, p. 50. [Accessed 29 April 2023]. DOI 10.3390/recycling6030050. Available from: <https://www.mdpi.com/2313-4321/6/3/50>
 26. ALPLA. New Study Quantifies the Environmental Impacts of Packaging. *ALPLA* [online]. 10 April 2019. [Accessed 4 February 2023]. Available from: <https://blog.alpla.com/en/press-release/newsroom/new-study-quantifies-environmental-impacts-packaging/04-19>
 27. FRANKLIN ASSOCIATES. Life Cycle Impacts for Postconsumer Recycled Resins: PET, HDPE, and PP. *The Association of Plastic Recyclers* [online]. December 2018. [Accessed 10 February 2023]. Available from: <https://plasticsrecycling.org/images/library/2018-APR-LCI-report.pdf>
 28. BARTOLOME, Leian, IMRAN, Muhammad, GYOO, Bong, A., Waheed and HYUN, Do. Recent Developments in the Chemical Recycling of PET. In: *Material Recycling - Trends and Perspectives* [online]. InTech, 2012. [Accessed 6 March 2023]. ISBN 978-953-51-0327-1.

Available from: <http://www.intechopen.com/books/material-recycling-trends-and-perspectives/recent-developments-in-the-chemical-recycling-of-pet>

29. PACI, M and LA MANTIA, F.P. Influence of small amounts of polyvinylchloride on the recycling of polyethyleneterephthalate. *Polymer Degradation and Stability* [online]. January 1999. Vol. 63, no. 1, p. 11–14. [Accessed 24 February 2023]. DOI 10.1016/S0141-3910(98)00053-6. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0141391098000536>
30. QIN, Dan, WANG, Chaosheng, WANG, Huaping, CHEN, Ye, JI, Peng and XI, Zhenhao. Modeling and Optimizing of Producing Recycled PET from Fabrics Waste via Falling Film-Rotating Disk Combined Reactor. *International Journal of Polymer Science* [online]. 2017. Vol. 2017, p. 1–13. [Accessed 29 April 2023]. DOI 10.1155/2017/1062493. Available from: <https://www.hindawi.com/journals/ijps/2017/1062493/>
31. BROUWER, Marieke T., ALVARADO CHACON, Fresia and THODEN VAN VELZEN, Eggo Ulphard. Effect of recycled content and rPET quality on the properties of PET bottles, part III: Modelling of repetitive recycling. *Packaging Technology and Science* [online]. 3 August 2020. Vol. 33, no. 9, p. 373–383. [Accessed 20 February 2023]. DOI 10.1002/pts.2489. Available from: <https://onlinelibrary.wiley.com/doi/10.1002/pts.2489>
32. CALABRESE, A., COSTA, R., LEVIALDI GHIRON, N., MENICHINI, T., MISCOLI, V. and TIBURZI, L. Operating modes and cost burdens for the European deposit-refund systems: A systematic approach for their analysis and design. *Journal of Cleaner Production* [online]. March 2021. Vol. 288, p. 125600. [Accessed 29 March 2023]. DOI 10.1016/j.jclepro.2020.125600. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0959652620356468>
33. UNESDA SOFT DRINKS EUROPE. State of implementation of Deposit Refund Systems (DRS) across Europe. *UNESDA* [online]. c2022. [Accessed 24 February 2023]. Available from: <https://www.unesda.eu/deposit-refund-systems/>
34. SEYRING, Nicole, DOLLHOFER, Marie, WEISSENBACHER, Jakob, BAKAS, Ioannis and MCKINNON, David. Assessment of collection schemes for packaging and other recyclable waste in European Union-28 Member States and capital cities. *Waste Management & Research: The Journal for a Sustainable Circular Economy* [online]. 11 July 2016. Vol. 34, no. 9, p. 947–956. [Accessed 1 March 2023]. DOI 10.1177/0734242X16650516. Available from: <http://journals.sagepub.com/doi/10.1177/0734242X16650516>
35. SAMOSEBOU.CZ. A Guide to Sorting and Recycling of Waste in the Czech Republic. *Samosebou.cz* [online]. 8 April 2022. [Accessed 8 April 2023]. Available from: <https://www.samosebou.cz/2022/04/08/a-guide-to-sorting-and-recycling-of-waste-in-the-czech-republic/>
36. COELHO, T.M., CASTRO, R. and GOBBO, J.A. PET containers in Brazil: Opportunities and challenges of a logistics model for post-consumer waste recycling. *Resources, Conservation and Recycling* [online]. January 2011. Vol. 55, no. 3, p. 291–299. [Accessed 4 February 2023]. DOI 10.1016/j.resconrec.2010.10.010. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0921344910002387>
37. GHESLA, Pamela, GOMES, Luciana, CAETANO, Marcelo, MIRANDA, Luis and DAI-PRÁ, Léa. Municipal Solid Waste Management from the Experience of São Leopoldo/Brazil and Zurich/Switzerland. *Sustainability* [online]. 16 October 2018. Vol. 10, no. 10, p. 3716.

- [Accessed 4 February 2023]. DOI 10.3390/su10103716. Available from: <http://www.mdpi.com/2071-1050/10/10/3716>
38. LAL PATEL, Munna, JAIN, Rajnikant and SAXENA, Alok. Assessment of the Municipal Solid Waste & Status of Implementation of Municipal Solid Waste (Management & Handling), Rules, 2000 in the State of Madhya Pradesh, 2008 - A case study. *Waste Management & Research: The Journal for a Sustainable Circular Economy* [online]. 17 June 2010. Vol. 29, no. 5, p. 558–562. [Accessed 4 February 2023]. DOI 10.1177/0734242X10372662. Available from: <http://journals.sagepub.com/doi/10.1177/0734242X10372662>
 39. WAHAB, D. A., HUSSAIN, A., SCAVINO, E., MUSTAFA, M.M. and BASRI, H. Development of a Prototype Automated Sorting System for Plastic Recycling. *American Journal of Applied Sciences* [online]. July 2006. Vol. 3, no. 7, p. 1924–1928. [Accessed 4 April 2023]. DOI 10.3844/ajassp.2006.1924.1928. Available from: <http://www.thescipub.com/abstract/?doi=ajassp.2006.1924.1928>
 40. BRUNO, Edward A. Automated Sorting of Plastics for Recycling. In: *Pollution Prevention Infohouse* [online]. [2000]. [Accessed 6 March 2023]. Available from: <https://p2infohouse.org/ref/09/08620.pdf>
 41. DA SILVA, Daniel José and WIEBECK, Hélio. Current options for characterizing, sorting, and recycling polymeric waste. *Progress in Rubber, Plastics and Recycling Technology* [online]. 2020. Vol. 36, no. 4, p. 284-303. [Accessed 1 May 2023]. DOI 10.1177/1477760620918603. Available from: <http://journals.sagepub.com/doi/10.1177/1477760620918603>
 42. JI, Tianchen, FANG, Huaiying, ZHANG, Rencheng, YANG, Jianhong, FAN, Lulu and LI, Jiantao. Automatic sorting of low-value recyclable waste: a comparative experimental study. *Clean Technologies and Environmental Policy* [online]. 10 October 2022. Vol. 25, p. 949-961. [Accessed 1 March 2023]. DOI 10.1007/s10098-022-02418-7. Available from: <https://link.springer.com/10.1007/s10098-022-02418-7>
 43. RPET INWASTE. *Standard Operating Procedures (SOPs) of rPET InWaste s.r.o.* Rosice U Brna, 2022.
 44. EPBP. European PET Bottle Platform Technical Opinion: Glycol-modified polyethylene terephthalate (PETG). In: *EPBP* [online]. Brussels, 2020. [Accessed 8 April 2023]. Available from: <https://www.epbp.org/download/333/non-endorsement-petg-components>
 45. BAUER, Markus, LEHNER, Markus, SCHWABL, Daniel, FLACHBERGER, Helmut, KRANZINGER, Lukas, POMBERGER, Roland and HOFER, Wolfgang. Sink–float density separation of post-consumer plastics for feedstock recycling. *Journal of Material Cycles and Waste Management* [online]. 24 May 2018. Vol. 20, no. 3, p. 1781–1791. [Accessed 4 April 2023]. DOI 10.1007/s10163-018-0748-z. Available from: <http://link.springer.com/10.1007/s10163-018-0748-z>
 46. MENESES QUELAL, Washington Orlando, VELÁZQUEZ-MARTÍ, Borja and FERRER GISBERT, Andrés. Separation of virgin plastic polymers and post-consumer mixed plastic waste by sinking-flotation technique. *Environmental Science and Pollution Research* [online]. 2022. Vol. 29, no. 1, p. 1364–1374. [Accessed 4 April 2023]. DOI 10.1007/s11356-021-15611-w. Available from: <https://link.springer.com/10.1007/s11356-021-15611-w>

47. CAMPANALE, Claudia, SAVINO, Ilaria, POJAR, Iulian, MASSARELLI, Carmine and URICCHIO, Vito Felice. A Practical Overview of Methodologies for Sampling and Analysis of Microplastics in Riverine Environments. *Sustainability* [online]. 20 August 2020. Vol. 12, no. 17, p. 6755. [Accessed 4 April 2023]. DOI 10.3390/su12176755. Available from: <https://www.mdpi.com/2071-1050/12/17/6755>
48. WU, Xiaoyu, LI, Jia, YAO, Linpeng and XU, Zhenming. Auto-sorting commonly recovered plastics from waste household appliances and electronics using near-infrared spectroscopy. *Journal of Cleaner Production* [online]. February 2020. Vol. 246, p. 118732. [Accessed 16 March 2023]. DOI 10.1016/j.jclepro.2019.118732. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0959652619336029>
49. BECKER, Wolfgang, SACHSENHEIMER, Kerstin and KLEMENZ, Melanie. Detection of Black Plastics in the Middle Infrared Spectrum (MIR) Using Photon Up-Conversion Technique for Polymer Recycling Purposes. *Polymers* [online]. 2017. Vol. 9, no. 9. [Accessed 1 May 2023]. DOI 10.3390/polym9090435. Available from: <https://www.mdpi.com/2073-4360/9/9/435>
50. LUIJSTERBURG, Benny and GOOSSENS, Han. Assessment of plastic packaging waste: Material origin, methods, properties. *Resources, Conservation and Recycling* [online]. April 2014. Vol. 85, p. 88–97. [Accessed 6 March 2023]. DOI 10.1016/j.resconrec.2013.10.010. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0921344913002176>
51. WANG, Zhaokun, PENG, Binbin, HUANG, Yanjun and SUN, Guanqun. Classification for plastic bottles recycling based on image recognition. *Waste Management* [online]. April 2019. Vol. 88, p. 170–181. [Accessed 6 March 2023]. DOI 10.1016/j.wasman.2019.03.032. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0956053X19301643>
52. LU, Weisheng and CHEN, Junjie. Computer vision for solid waste sorting: A critical review of academic research. *Waste Management* [online]. April 2022. Vol. 142, p. 29–43. [Accessed 6 March 2023]. DOI 10.1016/j.wasman.2022.02.009. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0956053X22000678>
53. ELAMRI, Adel, ZDIRI, Khmais, HARZALLAH, Omar and LALLAM, Abdelaziz. Progress in Polyethylene Terephthalate Recycling. In : *Polyethylene Terephthalate: Uses, Properties and Degradation* [online]. Nova Science Publishers, 2017. [Accessed 9 April 2023]. ISBN 978-1-53611-991-6. Available from: <https://hal.science/hal-02953197/document>
54. SERRANTI, Silvia and BONIFAZI, Giuseppe. Techniques for separation of plastic wastes. In: PACHECO-TORGAL, Fernando, KHATIB, Jamal, COLANGELO, Francesco and TULADHAR, Rabin, *Use of Recycled Plastics in Eco-efficient Concrete* [online]. Elsevier, 2019. p. 9-37. [Accessed 14 March 2023]. ISBN 9780081026762. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9780081026762000025>
55. DALMIJN, W. L. and DE JONG, T. P. R. The development of vehicle recycling in Europe: Sorting, shredding, and separation. *JOM* [online]. 6 November 2007. Vol. 59, no. 11, p. 52–56. [Accessed 15 March 2023]. DOI 10.1007/s11837-007-0141-1. Available from: <http://link.springer.com/10.1007/s11837-007-0141-1>
56. SESOTEC. FLAKE PURIFIER+. In: *Sesotec* [online]. c2008-2023. [Accessed 4 March 2023]. Available from: <https://www.sesotec.com/emea/en/lp/flake-purifier-plus>
57. SESOTEC. VARISORT+. In: *Sesotec* [online]. c2008-2023. [Accessed 4 March 2023]. Available from: <https://www.sesotec.com/emea/en/lp/varisort-plus>

58. SESOTEC. VARISORT Multi-Sensor Sorting Systems for the Recycling Industry. In: *Sesotec* [online]. c2008-2023. [Accessed 4 March 2023]. Available from: https://www.sesotec.com/sites/593fc2aac25e5b0640a20ff8/content_entry59e75c78ffe9ecdb05f0bdf9/5a97c50bffe9ec732a731491/files/varisort-pr-en.pdf?1652433049
59. ALAERTS, Luc, AUGUSTINUS, Michael and VAN ACKER, Karel. Impact of Bio-Based Plastics on Current Recycling of Plastics. *Sustainability* [online]. 9 May 2018. Vol. 10, no. 5, p. 1487. [Accessed 11 February 2023]. DOI 10.3390/su10051487. Available from: <http://www.mdpi.com/2071-1050/10/5/1487>
60. WELLE, Frank. Simulation of the Decontamination Efficiency of PET Recycling Processes based on Solid-state Polycondensation. *Packaging Technology and Science* [online]. 2014. Vol. 27, no. 2, p. 141–148. [Accessed 1 March 2023]. DOI 10.1002/pts.2013. Available from: <https://onlinelibrary.wiley.com/doi/10.1002/pts.2013>
61. COMMISSION REGULATION (EC) No 282/2008 of 27 March 2008 on recycled plastic materials and articles intended to come into contact with foods and amending Regulation (EC) No 2023/2006 (Text with EEA relevance). In: *Official Journal of the European Union* [online]. 28 March 2008. [Accessed 10 March 2023]. Available from: <https://eur-lex.europa.eu/legal-content/EN/TXT/HTML/?uri=CELEX:32008R0282>
62. COMMISSION REGULATION (EU) 2022/1616 of 15 September 2022 on recycled plastic materials and articles intended to come into contact with foods, and repealing Regulation (EC) No 282/2008 (Text with EEA relevance). In: *Official Journal of the European Union* [online]. 20 September 2022. [Accessed 10 March 2023]. Available from: <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32022R1616>
63. SILANO, Vittorio, BARAT BAVIERA, José Manuel, BOLOGNESI, Claudia, CHESSON, Andrew, COCCONCELLI, Pier Sandro, CREBELLI, Riccardo, GOTT, David Michael, GROB, Konrad, MORTENSEN, Alicja, RIVIÈRE, Gilles, STEFFENSEN, Inger-Lise, TLUSTOS, Christina, VAN LOVEREN, Henk, VERNIS, Laurence, ZORN, Holger, DUDLER, Vincent, MILANA, Maria Rosaria, PAPASPYRIDES, Constantine, TAVARES POÇAS, Maria de Fátima, LIOUPIS, Alexandros, VOLK, Katharina and LAMPI, Evgenia. Safety assessment of the process RE-PET, based on EREMA Basic technology, used to recycle post-consumer PET into food contact materials. *EFSA Journal* [online]. March 2020. Vol. 18, no. 3. [Accessed 20 February 2023]. DOI 10.2903/j.efsa.2020.6049. Available from: <http://doi.wiley.com/10.2903/j.efsa.2020.6049>
64. LAMBRÉ, Claude, BARAT BAVIERA, José Manuel, BOLOGNESI, Claudia, CHESSON, Andrew, COCCONCELLI, Pier Sandro, CREBELLI, Riccardo, GOTT, David Michael, GROB, Konrad, MENGELERS, Marcel, MORTENSEN, Alicja, RIVIÈRE, Gilles, STEFFENSEN, Inger-Lise, TLUSTOS, Christina, VAN LOVEREN, Henk, VERNIS, Laurence, ZORN, Holger, DUDLER, Vincent, MILANA, Maria Rosaria, PAPASPYRIDES, Constantine, TAVARES POÇAS, Maria de Fátima, LIOUPIS, Alexandros and LAMPI, Evgenia. Safety assessment of the process Omorika Recycling, based on PET direct iV+ technology, used to recycle post-consumer PET into food contact materials. *EFSA Journal* [online]. October 2021. Vol. 19, no. 10. [Accessed 20 February 2023]. DOI 10.2903/j.efsa.2021.6872. Available from: <http://doi.wiley.com/10.2903/j.efsa.2021.6872>
65. LAMBRÉ, Claude, BARAT BAVIERA, José Manuel, BOLOGNESI, Claudia, CHESSON, Andrew, COCCONCELLI, Pier Sandro, CREBELLI, Riccardo, GOTT, David Michael, GROB, Konrad, MENGELERS, Marcel, MORTENSEN, Alicja, RIVIÈRE, Gilles, STEFFENSEN, Inger-Lise, TLUSTOS, Christina, VAN LOVEREN, Henk, VERNIS,

- Laurence, ZORN, Holger, DUDLER, Vincent, MILANA, Maria Rosaria, PAPASPYRIDES, Constantine, TAVARES POÇAS, Maria de Fátima, LIOUPIS, Alexandros and LAMPI, Evgenia. Safety assessment of the process Greenpet, based on the Starlinger iV+ technology, used to recycle post-consumer PET into food contact materials. *EFSA Journal* [online]. July 2022. Vol. 20, no. 7. [Accessed 20 February 2023]. DOI 10.2903/j.efsa.2022.7387. Available from: <http://doi.wiley.com/10.2903/j.efsa.2022.7387>
66. LAMBRÉ, Claude, BARAT BAVIERA, José Manuel, BOLOGNESI, Claudia, CHESSON, Andrew, COCCONCELLI, Pier Sandro, CREBELLI, Riccardo, GOTT, David Michael, GROB, Konrad, MENGELERS, Marcel, MORTENSEN, Alicja, RIVIÈRE, Gilles, STEFFENSEN, Inger-Lise, TLUSTOS, Christina, VAN LOVEREN, Henk, VERNIS, Laurence, ZORN, Holger, DUDLER, Vincent, MILANA, Maria Rosaria, PAPASPYRIDES, Constantine, TAVARES POÇAS, Maria de Fátima, LIOUPIS, Alexandros and LAMPI, Evgenia. Safety assessment of the process rPET InWaste, based on the NGR technology, used to recycle post-consumer PET into food contact materials. *EFSA Journal* [online]. May 2022. Vol. 20, no. 5. [Accessed 20 February 2023]. DOI 10.2903/j.efsa.2022.7273. Available from: <http://doi.wiley.com/10.2903/j.efsa.2022.7273>
67. HORODYTSKA, O., CABANES, A. and FULLANA, A. Non-intentionally added substances (NIAS) in recycled plastics. *Chemosphere* [online]. 2020. Vol. 251. [Accessed 9 March 2023]. DOI 10.1016/j.chemosphere.2020.126373. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S004565352030566X>
68. THODEN VAN VELZEN, Eggo Ulphard, BROUWER, Marieke T., STÄRKER, Carina and WELLE, Frank. Effect of recycled content and rPET quality on the properties of PET bottles, part II: Migration. *Packaging Technology and Science* [online]. 2020. Vol. 33, no. 9, p. 359–371. [Accessed 7 March 2023]. DOI 10.1002/pts.2528. Available from: <https://onlinelibrary.wiley.com/doi/10.1002/pts.2528>
69. FRANZ, Roland and WELLE, Frank. Contamination Levels in Recollected PET Bottles from Non-Food Applications and their Impact on the Safety of Recycled PET for Food Contact. *Molecules* [online]. 28 October 2020. Vol. 25, no. 21, p. 4998. [Accessed 9 March 2023]. DOI 10.3390/molecules25214998. Available from: <https://www.mdpi.com/1420-3049/25/21/4998>
70. WELLE, Frank. Food Law Compliance of Poly(ethylene Terephthalate) (PET) Food Packaging Materials. In: KOMOLPRASERT, Vane and TUROWSKI, Petra, *Food Additives and Packaging* [online]. Washington, DC : American Chemical Society, 2014. p. 167-195. ACS Symposium Series. [Accessed 14 February 2023]. ISBN 9780841230248. Available from: <https://pubs.acs.org/doi/abs/10.1021/bk-2014-1162.ch016>
71. ERIKSEN, M.K., PIVNENKO, K., OLSSON, M.E. and ASTRUP, T.F. Contamination in plastic recycling: Influence of metals on the quality of reprocessed plastic. *Waste Management* [online]. September 2018. Vol. 79, p. 595–606. [Accessed 28 January 2023]. DOI 10.1016/j.wasman.2018.08.007. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0956053X1830494X>
72. SCHEIRS, John. Recycling of PET. In: *Polymer Recycling: Science, Technology and Applications*. Chichester: John Wiley & Sons, 1998. p. 119–183. ISBN 0-471-97054-9.
73. TORRES, N., ROBIN, J.J. and BOUTEVIN, B. Study of thermal and mechanical properties of virgin and recycled poly(ethylene terephthalate) before and after injection molding. *European Polymer Journal* [online]. October 2000. Vol. 36, no. 10, p. 2075–2080. [Accessed 2 February

- 2023]. DOI 10.1016/S0014-3057(99)00301-8. Available from:
<https://linkinghub.elsevier.com/retrieve/pii/S0014305799003018>
74. MILGROM, J. Polyethylene Terephthalate (PET). In: EHRIG, R. J. (ed.), *Plastics Recycling: Products and Processes*. Hanser, 1992. p. 45–73. ISBN 3-446-15882-0.
 75. ALVARADO CHACON, Fresia, BROUWER, Marieke T. and THODEN VAN VELZEN, Eggo Ulphard. Effect of recycled content and rPET quality on the properties of PET bottles, part I: Optical and mechanical properties. *Packaging Technology and Science* [online]. 2020. Vol. 33, no. 9, p. 347–357. [Accessed 11 February 2023]. DOI 10.1002/pts.2490. Available from: <https://onlinelibrary.wiley.com/doi/10.1002/pts.2490>
 76. SCHEIRS, John. Melt Filtration of Contamination in Recycled Polymers. In : *Polymer Recycling: Science, Technology and Applications*. Chichester: John Wiley & Sons, 1998. p. 101–119. ISBN 0-471-97054-9.
 77. FORREST, Martin J. 6. Recycling Technologies for Polyethylene Terephthalate. In: *Recycling of Polyethylene Terephthalate* [online]. 2nd ed. Berlin/Boston: De Gruyter, 2019. p. 101-119. [Accessed 3 January 2023]. ISBN 978-3-11-064030-4. Available from: <https://www.degruyter.com/document/doi/10.1515/9783110640304-006/html?lang=en>
 78. WRÓBEL, G. and BAGSIK, R. The importance of PET filtration for the possibility of material recycling. *Journal of Achievements in Materials and Manufacturing Engineering* [online]. November 2010. Vol. 43, no. 1., p 178-191. [Accessed 3 February 2023]. Available from: http://jamme.acmsse.h2.pl/papers_vol43_1/43117.pdf
 79. OBERMAYR, Robert. Finding the Right Melt Filtration System for Post-Consumer Recycling. In: *Plastics Technology* [online]. 21 February 2020. [Accessed 1 May 2023]. Available from: <https://www.ptonline.com/blog/post/finding-the-right-melt-filtration-system-for-post-consumer-recycling>
 80. NGR. P:REACT. In: *NGR* [online]. c2023 [Accessed 2 March 2023]. Available from: https://www.ngr-world.com/wp-content/uploads/2018/09/NGR_PREACT_191010_EN.pdf.
 81. GAO, Qiu, NAN-XUN, Huang, ZHI-LIAN, Tang and GERKING, L. Modelling of solid state polycondensation of poly(ethylene terephthalate). *Chemical Engineering Science* [online]. February 1997. Vol. 52, no. 3, p. 371–376. [Accessed 2 March 2023]. DOI 10.1016/S0009-2509(96)00414-9. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0009250996004149>
 82. CRUZ, S. A. and ZANIN, M. PET recycling: Evaluation of the solid state polymerization process. *Journal of Applied Polymer Science* [online]. 2005. Vol. 99, no. 5, p. 2117–2123. [Accessed 2 March 2023]. DOI 10.1002/app.22526. Available from: <https://onlinelibrary.wiley.com/doi/10.1002/app.22526>
 83. SCHIAVONE, Robert J. Solid state polymerization (SSP) of low molecular weight poly(ethylene terephthalate) (PET) copolyesters compared to conventional SSP of PET. *Journal of Applied Polymer Science* [online]. 2002. Vol. 86, no. 1, p. 230–238. [Accessed 2 March 2023]. DOI 10.1002/app.10952. Available from: <https://onlinelibrary.wiley.com/doi/10.1002/app.10952>
 84. COOPER, T.A. Developments in plastic materials and recycling systems for packaging food, beverages and other fast-moving consumer goods. In: FARMER, Neil, *Trends in Packaging of Food, Beverages and Other Fast-Moving Consumer Goods (FMCG)* [online]. Woodhead

- Publishing, 2013. p. 58-107. [Accessed 17 March 2023]. ISBN 978-0-85709-503-9. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9780857095039500047>
85. KOO, Hyun Jin, CHANG, Gap Shik, KIM, Seong Hun, HAHM, Wan Gyu and PARK, Seong Yoon. Effects of recycling processes on physical, mechanical and degradation properties of PET yarns. *Fibers and Polymers* [online]. 2013. Vol. 14, p. 2083–2087. [Accessed 10 February 2023]. DOI 10.1007/s12221-013-2083-2. Available from: <http://link.springer.com/10.1007/s12221-013-2083-2>
 86. GEYER, B., LORENZ, G. and KANDELBAUER, A. Recycling of poly(ethylene terephthalate) – A review focusing on chemical methods. *Express Polymer Letters* [online]. 2016. Vol. 10, no. 7, p. 559–586. [Accessed 11 February 2023]. DOI 10.3144/expresspolymlett.2016.53. Available from: <http://www.expresspolymlett.com/letolt.php?file=EPL-0006898&mi=c>
 87. MASMOUDI, Fatma, FENOUILLOT, Françoise, MEHRI, Afef, JAZIRI, Mohamed and AMMAR, Emna. Characterization and quality assessment of recycled post-consumption poly(ethylene terephthalate) (PET). *Environmental Science and Pollution Research* [online]. 2018. Vol. 25, p. 23307–23314. [Accessed 21 March 2023]. DOI 10.1007/s11356-018-2390-7. Available from: <http://link.springer.com/10.1007/s11356-018-2390-7>
 88. KANEKA BELGIUM NV. Modifiers spur growth in PET recycling. *Plastics, Additives and Compounding* [online]. July-August 2005. Vol. 7, no. 4, p. 32–33. [Accessed 5 March 2023]. DOI 10.1016/S1464-391X(05)70430-X. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S1464391X0570430X>
 89. NIAOUNAKIS, Michael. Types, Forms, and Uses of Flexible Plastic Packaging. In : *Recycling of Flexible Plastic Packaging* [online]. Elsevier, 2020. p. 97-137. [Accessed 15 February 2023]. ISBN 978-0-12-816335-1. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9780128163351000049>
 90. VAN DONGEN, Cees, DVORAK, Robert and KOSIOR, Ed. *Design Guide for PET Bottle Recyclability*. UNESDA & EFBW, [2011].
 91. LEÓN ALBITER, Noel, SANTANA PÉREZ, Orlando, KLOTZ, Magali, GANESAN, Kishore, CARRASCO, Félix, DAGRÉOU, Sylvie, MASPOCH, Maria Lluïsa and VALDERRAMA, César. Implications of the Circular Economy in the Context of Plastic Recycling: The Case Study of Opaque PET. *Polymers* [online]. 31 October 2022. Vol. 14, no. 21, p. 4639. [Accessed 11 February 2023]. DOI 10.3390/polym14214639. Available from: <https://www.mdpi.com/2073-4360/14/21/4639>
 92. ROOSEN, Martijn, MYS, Nicolas, KLEINHANS, Kerstin, LASE, Irdanto Saputra, HUYSVELD, Sophie, BROUWER, Marieke, THODEN VAN VELZEN, Eggo U., VAN GEEM, Kevin M., DEWULF, Jo, RAGAERT, Kim, DUMOULIN, Ann and DE MEESTER, Steven. Expanding the collection portfolio of plastic packaging: Impact on quantity and quality of sorted plastic waste fractions. *Resources, Conservation and Recycling* [online]. March 2022. Vol. 178, p. 106025. [Accessed 12 February 2023]. DOI 10.1016/j.resconrec.2021.106025. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0921344921006339>
 93. ROOSEN, Martijn, MYS, Nicolas, KUSENBERG, Marvin, BILLEN, Pieter, DUMOULIN, Ann, DEWULF, Jo, VAN GEEM, Kevin M., RAGAERT, Kim and DE MEESTER, Steven. Detailed Analysis of the Composition of Selected Plastic Packaging Waste Products and Its

- Implications for Mechanical and Thermochemical Recycling. *Environmental Science & Technology* [online]. 2020. Vol. 54, no. 20, p. 13282–13293. [Accessed 13 February 2023]. DOI 10.1021/acs.est.0c03371. Available from: <https://pubs.acs.org/doi/10.1021/acs.est.0c03371>
94. MORRIS, Barry A. Designing flexible packaging for sustainability. In: *The Science and Technology of Flexible Packaging* [online]. 2nd ed. Elsevier, 2022. p. 709–761. [Accessed 12 February 2023]. ISBN 978-0-323-85435-1. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9780323854351000181>
 95. BARREDO, Asier, ASUETA, Asier, AMUNDARAIN, Izotz, LEIVAR, Jon, MIGUEL-FERNÁNDEZ, Rafael, ARNAIZ, Sixto, EPELDE, Eva, LÓPEZ-FONSECA, Rubén and GUTIÉRREZ-ORTIZ, José Ignacio. Chemical recycling of monolayer PET tray waste by alkaline hydrolysis. *Journal of Environmental Chemical Engineering* [online]. 2023. Vol. 11, no. 3, p. 109823. [Accessed 1 May 2023]. DOI 10.1016/j.jece.2023.109823. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S2213343723005626>
 96. NAKAYA, Masaki, UEDONO, Akira and HOTTA, Atsushi. Recent Progress in Gas Barrier Thin Film Coatings on PET Bottles in Food and Beverage Applications. *Coatings* [online]. 8 December 2015. Vol. 5, no. 4, p. 987–1001. [Accessed 21 February 2023]. DOI 10.3390/coatings5040987. Available from: <https://www.mdpi.com/2079-6412/5/4/987>
 97. BERG, Dennis, SCHAEFER, Karola, KOERNER, Andrea, KAUFMANN, Robert, TILLMANN, Walter and MOELLER, Martin. Reasons for the Discoloration of Postconsumer Poly(ethylene terephthalate) during Reprocessing. *Macromolecular Materials and Engineering* [online]. 2016. Vol. 301, no. 12, p. 1454–1467. [Accessed 21 February 2023]. DOI 10.1002/mame.201600313. Available from: <https://onlinelibrary.wiley.com/doi/10.1002/mame.201600313>
 98. PINTER, Elisabeth, WELLE, Frank, MAYRHOFER, Elisa, PECHHACKER, Andreas, MOTLOCH, Lukas, LAHME, Vera, GRANT, Andy and TACKER, Manfred. Circularity Study on PET Bottle-To-Bottle Recycling. *Sustainability* [online]. 1 July 2021. Vol. 13, no. 13, p. 7370. [Accessed 15 February 2023]. DOI 10.3390/su13137370. Available from: <https://www.mdpi.com/2071-1050/13/13/7370>
 99. CHEN, Tingting, JIANG, Guodong, LI, Guoyu, WU, Zhipeng and ZHANG, Jun. Poly(ethylene glycol-co-1,4-cyclohexanedimethanol terephthalate) random copolymers: effect of copolymer composition and microstructure on the thermal properties and crystallization behavior. *RSC Advances* [online]. 2015. Vol. 5, no. 74, p. 60570–60580. [Accessed 25 February 2023]. DOI 10.1039/C5RA09252C. Available from: <http://xlink.rsc.org/?DOI=C5RA09252C>
 100. SEPAHI, Mohammad Taregh, ABUSALMA, Hisham, JOVANOVIĆ, Vukica and EISAZADEH, Hamid. Mechanical Properties of 3D-Printed Parts Made of Polyethylene Terephthalate Glycol. *Journal of Materials Engineering and Performance* [online]. 2021. Vol. 30, no. 13, p. 6851–6861. [Accessed 4 February 2023]. DOI 10.1007/s11665-021-06032-4. Available from: <https://link.springer.com/10.1007/s11665-021-06032-4>
 101. FERREIRA, Isaac, VALE, Diogo, MACHADO, Margarida and LINO, Jorge. Additive manufacturing of polyethylene terephthalate glycol /carbon fiber composites: An experimental study from filament to printed parts. *Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications* [online]. 2018. Vol. 233, no. 9, p.

- 1866–1878. [Accessed 4 February 2023]. DOI 10.1177/1464420718795197. Available from: <http://journals.sagepub.com/doi/10.1177/1464420718795197>
102. CHEN, Tingting, ZHANG, Weikai and ZHANG, Jun. Alkali resistance of poly(ethylene terephthalate) (PET) and poly(ethylene glycol-co-1,4-cyclohexanedimethanol terephthalate) (PETG) copolyesters: The role of composition. *Polymer Degradation and Stability*. October 2015. Vol. 120, p. 232–243. [Accessed 4 February 2023] DOI 10.1016/J.POLYMDEGRADSTAB.2015.07.008. Available from: <https://www.sciencedirect.com/science/article/pii/S0141391015300392>
103. RPET INWASTE. Requirements for purchased product and quality system. In: *rPET InWaste* [online]. c2023. [Accessed 15 April 2023]. Available from: https://rpet-inwaste.com/data/blocks_files/63.pdf
104. SNOPOKOVÁ, Martina. *Identifikace zdrojů kontaminace při recyklaci PET* [online]. Brno, 2021 [Accessed 23 February 2023]. Available from: <https://www.vutbr.cz/studenti/zav-prace/detail/131501>. Bachelor thesis. Brno University of Technology, Faculty of Chemistry, Institute of Chemistry and Technology of Environmental Protection. Supervisor Jiří Kučerík.
105. LEWANDOWSKI, Krzysztof and SKÓRCZEWSKA, Katarzyna. A Brief Review of Poly(Vinyl Chloride) (PVC) Recycling. *Polymers* [online]. 27 July 2022. Vol. 14, no. 15, p. 3035. [Accessed 7 February 2023]. DOI 10.3390/polym14153035. Available from: <https://www.mdpi.com/2073-4360/14/15/3035>
106. YU, Jie, SUN, Lushi, MA, Chuan, QIAO, Yu and YAO, Hong. Thermal degradation of PVC: A review. *Waste Management* [online]. 2016. Vol. 48, p. 300-314. [Accessed 23 April 2023]. DOI 10.1016/j.wasman.2015.11.041. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0956053X15302233>
107. KANG, Wook Geon and KIM, Jongkyoung. A Survey on Consumer Perception on Removability of PET Bottle Labels. *KOREAN JOURNAL OF PACKAGING SCIENCE AND TECHNOLOGY* [online]. 2021. Vol. 27, no. 2, p. 63–70. [Accessed 20 March 2023]. DOI 10.20909/kopast.2021.27.2.63. Available from: <http://koreascience.or.kr/article/JAKO202129557735273.pdf>
108. KÜPPERS, Bastian, CHEN, Xiaozheng, SEIDLER, Irina, FRIEDRICH, Karl, RAULF, Karoline, PRETZ, Thomas, FEIL, Alexander, POMBERGER, Roland and VOLLPRECHT, Daniel. Influences and Consequences of Mechanical Delabelling on PET Recycling. *Detritus* [online]. 2019. Vol. 06, p. 39-46 [Accessed 1 March 2023]. DOI 10.31025/2611-4135/2019.13816. Available from: <https://digital.detritusjournal.com/doi/13816-220>
109. UPASANI, Prasad S., JAIN, Ashwin K., SAVE, Ninad, AGARWAL, Uday S. and KELKAR, Anil K. Chemical recycling of PET flakes into yarn. *Journal of Applied Polymer Science* [online]. July 2011. Vol. 123, no. 1, p. 520–525. [Accessed 15 February 2023]. DOI 10.1002/app.34503. Available from: <https://onlinelibrary.wiley.com/doi/10.1002/app.34503>
110. ZHU, Shichao, CHEN, Honghui, WANG, Mengmeng, GUO, Xuemei, LEI, Yu and JIN, Gang. Plastic solid waste identification system based on near infrared spectroscopy in combination with support vector machine. *Advanced Industrial and Engineering Polymer Research* [online]. April 2019. Vol. 2, no. 2, p. 77–81. [Accessed 18 March 2023]. DOI 10.1016/j.aiepr.2019.04.001. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S2542504818300113>

111. ITIM, Bachir and PHILIP, Mathew. Effect of multiple extrusions and influence of PP contamination on the thermal characteristics of bottle grade recycled PET. *Polymer Degradation and Stability* [online]. July 2015. Vol. 117, p. 84–89. [Accessed 16 February 2023]. DOI 10.1016/j.polymdegradstab.2015.04.004. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0141391015001275>
112. SASTRI, Vinny R. Commodity Thermoplastics: Polyvinyl Chloride, Polyolefins, and Polystyrene. In: *Plastics in Medical Devices* [online]. Elsevier, 2010. p. 73-119. [Accessed 5 March 2023]. ISBN 978-0-8155-2027-6. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9780815520276100066>
113. MIRÓN, V., FERRÁNDIZ, S., JUÁREZ, D. and MENGUAL, A. Manufacturing and characterization of 3D printer filament using tailoring materials. *Procedia Manufacturing*. 2017. Vol. 13, p. 888–894. [Accessed 12 March 2023]. DOI 10.1016/J.PROMFG.2017.09.151. Available from: <https://www.sciencedirect.com/science/article/pii/S2351978917307886>
114. AURAS, Rafael, HARTE, Bruce and SELKE, Susan. An Overview of Polylactides as Packaging Materials. *Macromolecular Bioscience* [online]. 2004. Vol. 4, no. 9, p. 835–864. [Accessed 19 February 2023]. DOI 10.1002/mabi.200400043. Available from: <https://onlinelibrary.wiley.com/doi/10.1002/mabi.200400043>
115. NAGARAJAN, Vidhya, MOHANTY, Amar K. and MISRA, Manjusri. Perspective on Polylactic Acid (PLA) based Sustainable Materials for Durable Applications: Focus on Toughness and Heat Resistance. *ACS Sustainable Chemistry & Engineering* [online]. 2016. Vol. 4, no. 6, p. 2899–2916. [Accessed 19 February 2023]. DOI 10.1021/acssuschemeng.6b00321. Available from: <https://pubs.acs.org/doi/10.1021/acssuschemeng.6b00321>
116. CASTRO-AGUIRRE, E., AURAS, R., SELKE, S., RUBINO, M. and MARSH, T. Enhancing the biodegradation rate of poly(lactic acid) films and PLA bio-nanocomposites in simulated composting through bioaugmentation. *Polymer Degradation and Stability*. August 2018. Vol. 154, p. 46–54. [Accessed 19 February 2023]. DOI 10.1016/J.POLYMDEGRADSTAB.2018.05.017. Available from: <https://www.sciencedirect.com/science/article/pii/S0141391018301708>
117. ZHAO, Xiaoying, CORNISH, Katrina and VODOVOTZ, Yael. Narrowing the Gap for Bioplastic Use in Food Packaging: An Update. *Environmental Science & Technology* [online]. 21 April 2020. Vol. 54, no. 8, p. 4712–4732. [Accessed 19 February 2023]. DOI 10.1021/acs.est.9b03755. Available from: <https://pubs.acs.org/doi/10.1021/acs.est.9b03755>
118. RAMESHKUMAR, Saranya, SHAIJU, P., O’CONNOR, Kevin E. and P, Ramesh Babu. Bio-based and biodegradable polymers - State-of-the-art, challenges and emerging trends. *Current Opinion in Green and Sustainable Chemistry* [online]. February 2020. Vol. 21, p. 75–81. [Accessed 19 February 2023]. DOI 10.1016/j.cogsc.2019.12.005. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S2452223619300665>
119. BRIASSOULIS, Demetres, PIKASI, Anastasia and HISKAKIS, Miltiadis. End-of-waste life: Inventory of alternative end-of-use recirculation routes of bio-based plastics in the European Union context. *Critical Reviews in Environmental Science and Technology* [online]. 2019. Vol. 49, no. 20, p. 1835–1892. [Accessed 19 February 2023]. DOI 10.1080/10643389.2019.1591867. Available from: <https://www.tandfonline.com/doi/full/10.1080/10643389.2019.1591867>

120. GERASSIMIDOU, Spyridoula, LANSKA, Paulina, HAHLADAKIS, John N., LOVAT, Elena, VANZETTO, Silvia, GEUEKE, Birgit, GROH, Ksenia J., MUNCKE, Jane, MAFFINI, Maricel, MARTIN, Olwenn V. and IACOVIDOU, Eleni. Unpacking the complexity of the PET drink bottles value chain: A chemicals perspective. *Journal of Hazardous Materials* [online]. May 2022. Vol. 430, p. 128410. [Accessed 24 February 2023]. DOI 10.1016/j.jhazmat.2022.128410. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0304389422001984>
121. KUMARTASLI, Sedat and AVINC, Ozan. Important Step in Sustainability: Polyethylene Terephthalate Recycling and the Recent Developments. In: MUTHU, Subramanian Senthilkannan and GARDETTI, Miguel Angel, *Sustainability in the Textile and Apparel Industries* [online]. Cham: Springer International Publishing, 2020. p. 1-19. [Accessed 25 March 2023]. ISBN 978-3-030-38012-0. Available from: https://link.springer.com/10.1007/978-3-030-38013-7_1
122. KONICA MINOLTA. Spectrophotometer CM-5: Stand alone Top-Port Spectrophotometer with maximum Flexibility and Versatility CM-5. In: *Konica Minolta* [online]. c2019. [Accessed 2 March 2023]. Available from: https://d1c96hlcey6qkb.cloudfront.net/2c71c08c-36a1-4a84-8732-2918e00c00bb/d7b973df1a144d898cda4ee946939712?response-content-disposition=inline%3B%20filename%2A%3DUTF-8%27%27CM-5_ENG.pdf.pdf&response-content-type=application%2Fpdf&Expires=1683331200&Signature=c~ivQM9bqNumdRsLs1BSMRqw~j7PSxByW7PJ-U38O9VJwabwM-k45ope4U~pXbbTWEo2GH6CTqXas9YlbpNqVISJBqXxdMFXfyU5dYUTjZwtiqbHitbmJOnu3m4str02ntoOvgs2OFtE12d-rwAQn5kydZqqai8PAP1SJhOJb~9YUGKOS1hEgTOzQqAJptKtI6VEZFWYXrSSOqv~hhrzCE5MZzBujHOCPKlvFWpeEUPyt3EYy--fBJEp-euR1fvwHPStOZCB22TKvZXCMPaY4N5CeWZaNb~GckoqLkLGrkPe-iCPXFBmCfCx-V2P0fU2ELEPSHv4~-IKhOQU67Osw__&Key-Pair-Id=APKAI33AGAEAYCXFBDA
123. EN ISO/CIE 11664-4:2019. 2019. *Colorimetry - Part 4: CIE 1976 L*a*b* colour space*. City of Brussels: European Committee for Standardization.
124. KORIFI, Rabia, LE DRÉAU, Yveline, ANTINELLI, Jean-François, VALLS, Robert and DUPUY, Nathalie. CIEL*a*b* color space predictive models for colorimetry devices – Analysis of perfume quality. *Talanta* [online]. January 2013. Vol. 104, p. 58–66. [Accessed 18 March 2023]. DOI 10.1016/j.talanta.2012.11.026. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0039914012009587>
125. DELAZIO, Alexandra, ISRAR, Ali and KLATZKY, Roberta L. Cross-modal correspondence between vibrations and colors. In: *2017 IEEE World Haptics Conference (WHC)* [online]. IEEE, June 2017. p. 219–224. [Accessed 12 April 2023]. ISBN 978-1-5090-1425-5. Available from: <http://ieeexplore.ieee.org/document/7989904/>
126. ABBAS, Ouissam, DARDENNE, Pierre and BAETEN, Vincent. Near-Infrared, Mid-Infrared, and Raman Spectroscopy. In: PICÓ, Yolanda, *Chemical Analysis of Food: Techniques and Applications* [online]. Elsevier, 2012. p. 59–89. Available from: [Accessed 1 May 2023]. ISBN 9780123848628. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9780123848628000030>
127. RATNER, Buddy D. and CASTNER, David G. Surface Properties and Surface Characterization of Biomaterials. In: *Biomaterials Science* [online]. 4th ed. Elsevier, 2020. p.

- 53–75. [Accessed 1 May 2023]. ISBN 9780128161371. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9780128161371000064>
128. BRADLEY, Mike. Is ATR an Effective Sampling Technique for Carbon Black Rubber Analysis? In: *ThermoFisher Scientific* [online]. 11 October 2015. [Accessed 18 December 2022]. Available from: <https://www.thermofisher.com/blog/materials/is-atr-an-effective-sampling-technique-for-carbon-black-analysis/>
 129. MALET-MARTINO, Myriam, MARTINO, Robert and BONNET, Pierre-Antoine. Nutritional Supplements. In: PICÓ, Yolanda, *Chemical Analysis of Food: Techniques and Applications* [online]. Elsevier, 2012. p. 539–573. [Accessed 1 May 2023]. ISBN 9780123848628. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9780123848628000170>
 130. EN ISO 1133-2:2011. 2011. *Plastics — Determination of the melt mass-flow rate (MFR) and melt volume-flow rate (MVR) of thermoplastics — Part 2: Method for materials sensitive to time-temperature history and/or moisture*. City of Brussels: European Committee for Standardization.
 131. HOLTZ, Yan. Correlation matrix with ggally. In: *the R Graph Gallery* [online]. c2018. [Accessed 10 April 2023]. Available from: <https://r-graph-gallery.com/199-correlation-matrix-with-ggally.html>
 132. VILLALOBOS ALVA, Jalil. Statistical Data Analysis. In: *Beginning Mathematica and Wolfram for Data Science* [online]. Berkeley, CA: Apress, 2021. p. 209–242. [Accessed 10 April 2023]. ISBN 978-1-4842-6593-2. Available from: http://link.springer.com/10.1007/978-1-4842-6594-9_6
 133. EN 15348:2014. 2014. *Plastics – Recycled plastics – Characterisation of poly(ethylene terephthalate) (PET) recyclates*. City of Brussels: European Committee for Standardization.
 134. EN ISO 60:1999. 1999. *Plastics - Determination of apparent density of material that can be poured from a specified funnel*. City of Brussels: European Committee for Standardization.
 135. DYNISCO. Dynisco LMI5000 Series Melt Indexer: Operating Manual. *Azurr-tech* [online]. c2023. [Accessed 10 April 2023]. Available from: https://www.azurr-tech.cz/en/file/1d2095ce444b18e2183d15ee8a31730c/21466/LMI5000_manual_ENG.pdf
 136. SALMINEN, Pontus. *Using recycled polyethylene terephthalate (PET) in the production of bottle trays* [online]. Degree Thesis. 2013. [Accessed 29 April 2023]. Available from: https://www.theseus.fi/bitstream/handle/10024/57923/Salminen_Pontus.pdf?sequence=1&isAllowed=y
 137. GRANT, Andy, LAHME, Vera, CONNOCK, Toby and LUGAL, Leyla. How Circular is PET? In: *Zero waste Europe* [online]. Zero Waste Europe, February 2022. [Accessed 1 May 2023]. Available from: https://zerowasteurope.eu/wp-content/uploads/2022/02/HCIP_V13-1.pdf
 138. POLYMER SOLUTIONS. Dilute Solution Viscosity. In: *Polymer Solutions* [online]. 6 June 2019. [Accessed 26 March 2023]. Available from: <https://www.polymersolutions.com/blog/dilute-solution-viscosity/>
 139. BIRON, Michel. Detailed Accounts of Thermoplastic Resins. In: *Thermoplastics and Thermoplastic Composites* [online]. 3rd ed. Elsevier, 2018. p. 203–766. [Accessed 20 April 2023]. ISBN 9780081025017. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9780081025017000047>

140. ALI, Umar, KARIM, Khairil Juhanni Bt. Abd and BUANG, Nor Aziah. A Review of the Properties and Applications of Poly (Methyl Methacrylate) (PMMA). *Polymer Reviews* [online]. 2015. Vol. 55, no. 4, p. 678–705. [Accessed 20 April 2023]. DOI 10.1080/15583724.2015.1031377. Available from: <http://www.tandfonline.com/doi/full/10.1080/15583724.2015.1031377>
141. AKINDOYO, John O., BEG, M. D. H., GHAZALI, Suriati, ISLAM, M. R., JEYARATNAM, Nitthiyah and YUVARAJ, A. R. Polyurethane types, synthesis and applications – a review. *RSC Advances* [online]. 2016. Vol. 6, no. 115, p. 114453–114482. [Accessed 20 April 2023]. DOI 10.1039/C6RA14525F. Available from: <https://pubs.rsc.org/en/content/articlelanding/2016/ra/c6ra14525f>
142. KAUSAR, Ayesha. Polydimethylsiloxane-based nanocomposite: present research scenario and emergent future trends. *Polymer-Plastics Technology and Materials* [online]. 2020. Vol. 59, no. 11, p. 1148–1166. [Accessed 20 April 2023]. DOI 10.1080/25740881.2020.1719149. Available from: <https://www.tandfonline.com/doi/full/10.1080/25740881.2020.1719149>
143. WYPYCH, George. GENERIC TYPES. In: *Handbook of Antiblocking, Release, and Slip Additives* [online]. 3rd ed. Toronto: ChemTec Publishing, 2014. p. 9-36. [Accessed 25 April 2023]. ISBN 9781895198836. Available from: <https://linkinghub.elsevier.com/retrieve/pii/B9781895198836500040>

LIST OF ABBREVIATIONS

AI	artificial intelligence
ATR	attenuated total reflectance
CM	customer material
C_{res}	residual contamination
C_{mod}	model concentration
DRS	deposit-refund system
EC	European Commission
EFSA	European Food Safety Agency
EPDM	ethylene-propylene-diene monomer
EU	European Union
EVOH	ethylene vinyl alcohol
FTIR	Fourier transform infrared (spectroscopy)
FTIR-ATR	Fourier transform infrared (spectroscopy) with attenuated total reflectance
IR	infrared
IV	intrinsic viscosity
LCA	life cycle assessment
LSP	liquid-state polycondensation
MIR	mid-infrared
NIAS	non-intentionally added substances
NIR	near-infrared
PA	polyamide
PBT	polybutylene terephthalate
PDMS	polydimethylsiloxane
PE	polyethylene
PET	polyethylene terephthalate
PETG	polyethylene terephthalate glycol
PLA	polylactic acid
PMMA	poly (methyl methacrylate)
PP	polypropylene
PP+EPDM	polypropylene and ethylene-propylene-diene monomer
ppm	parts per million
PS	polystyrene
PU	Polyurethane
PVC	polyvinylchloride
QC	quality control
RGB	red, green, blue
rPET	recycled PET
SAN	styrene acrylonitrile
SOPs	standard operating procedures
SSP	solid-state polycondensation