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Technical quality of rPET

Technical quality of rPET that can be obtained from Dutch PET bottles that have been collected, sorted and mechanically recycled in different manners

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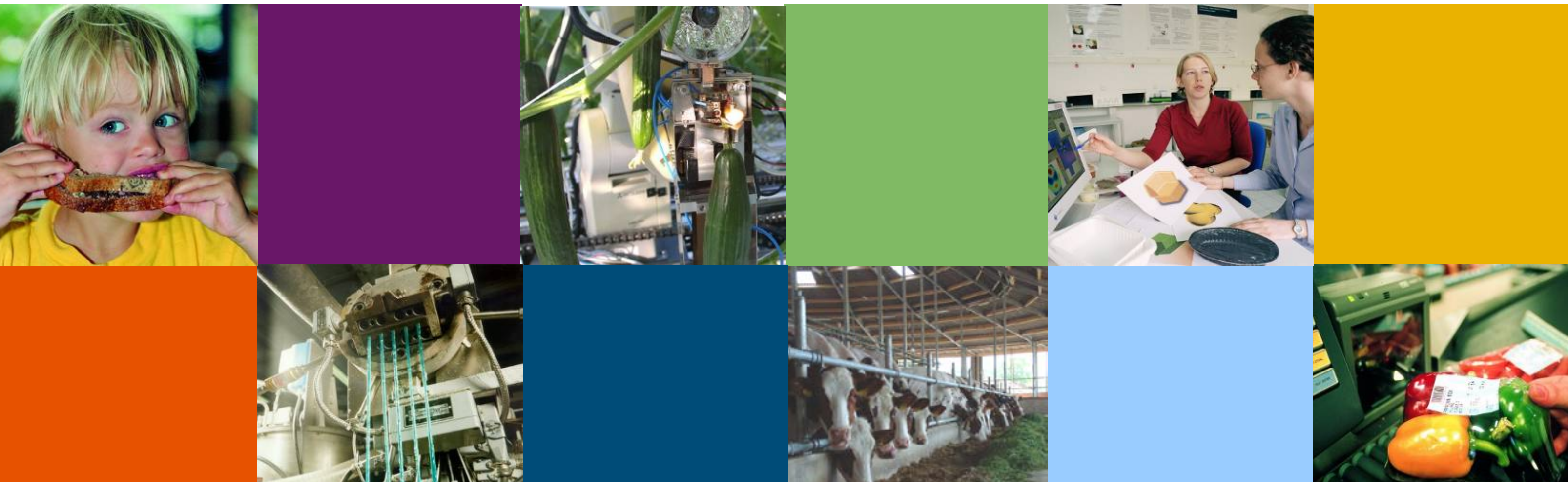

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Management Summary in Dutch - Uitgebreide samenvatting

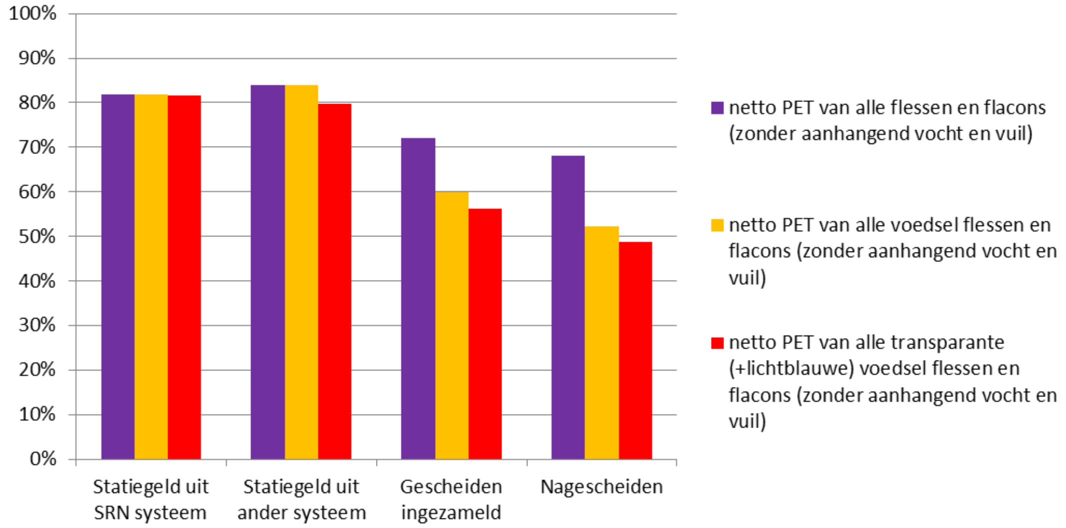
In 2015 is er een wetenschappelijk onderzoeksproject uitgevoerd met als doel de mechanische recyclingprocessen van gesorteerde PET flessen in rPET producten beter te begrijpen. Drie verschillende aspecten werden onderzocht. Allereerst werd de samenstelling van de aanwezige PET-fles-grondstoffen uitgebreid onderzocht. Dit zijn de getelde of gesorteerde PET-flessen die in Nederland beschikbaar zijn als grondstof voor recycling. Ten tweede werd er systematisch onderzoek gedaan naar de invloed van verontreinigingen op de eigenschappen van gerecycleerd PET (rPET). Ten derde werden PET-flessen uit verschillende inzamelsystemen, hierna genoemd PET-fles-grondstoffen, met verschillende mechanische recyclingprocessen omgezet in rPET, waarvan de eigenschappen op een systematische wijze werden geanalyseerd met als doel om relaties te kunnen vinden tussen de grondstoffen, de processen en de verkregen kwaliteiten rPET.

Samenstelling van PET-fles-grondstoffen

In Nederland leveren drie inzamelsystemen gesorteerde PET flessen als grondstof voor mechanische recycling. Deze drie grondstoffen zijn flessen uit statiegeldsystemen, flessen uit gescheiden inzameling en flessen uit nascheiding. Deze PET-fles-grondstoffen verschillen in samenstelling. De samenstelling van de PET-flessen-grondstoffen werd onderzocht door deze grondstoffen te sorteren in categorieën van soorten flessen, flacons, andersoortige verpakkingen en andere materialen en vervolgens voor elke categorie van PET flessen en flacons de gemiddelde materiaalsamenstelling te bepalen.

Hiervoor was het nodig om de zogenoemde PET barrièreflessen te kunnen herkennen, zodat deze in aparte categorieën gesorteerd kunnen worden. PET-barrièreflessen zijn PET-flessen waaraan een additionele laag, coating of additief is toegevoegd zodat de instroom van zuurstofgas door de fles wordt verlaagd en de houdbaarheid van de gebottelde drank of verpakt product wordt verlengd. Er bestaan barrièreflessen met Nylon, EVOH, zuurstof-absorbers, koolstof-coatings en zo meer. Aangezien dergelijke barrièreflessen het gerecycleerde PET onevenredig meer geel of bruin laten verkleuren, is het belangrijk te weten hoeveel van deze flessen er in de PET-fles-grondstoffen bevinden. Hiertoe werd een bestaande detectiemethode voor PET maalgoed aangepast voor stukken PET-fles, door stukken PET-fles tussen twee metalen plaatjes te klemmen en in de oven te verhitten. De test toont alleen de onder hitte (220 °C) verkleurende barrières aan, en zal de niet-verkleurende barrière-flessen niet aantonen. Barrière-flessen die niet onder hitte verkleuren werden niet in deze analyse gedetecteerd. Op deze manier werden 175 PET flessen en flacons geanalyseerd op de aanwezigheid van een barrière. Uiteindelijk bleken er slechts een beperkt aantal flessen en flacons op de Nederlandse markt een barrière te bevatten die onder hitte verkleuren. Deze werden op een aparte sorteerlijst geplaatst, waarmee de PET-fles-grondstoffen werden gesorteerd.

De gemiddelde materiaalsamenstelling werd voor elke verpakkingscategorie bepaald door de flessen te ontleden en elk verpakkingsonderdeel te analyseren op materiaal met nabij-infrarood spectroscopie (NIR) en zo nodig ook met infrarood spectroscopie (IR). Dit werd gedaan voor tenminste 25 (zover als mogelijk) verschillende flessen / flacons per categorie. Hieruit werd de gemiddelde materiaalsamenstelling per categorie berekend. Door de samenstelling van de grondstoffen in termen van soorten flessen te combineren met de gemiddelde samenstelling per categorie fles werd inzicht verkregen in de materiaalsamenstelling van de PET-fles-grondstoffen, zie Figuur a. De figuur geeft inzicht in het netto-percentage PET (afkomstig van de flessen-body's) van alle PET flessen en flacons (paars), van alleen de PET voedsel flessen en flacons (oranje) en van alleen de PET voedsel flessen en flacons die transparant waren (rood).



Figuur a: Netto PET gehalte van alle flessen en flacons (dus minus aanhangend vocht en vuil), ten opzichte van het totaal gewicht van de gesorteerde PET producten (bruto). De statiegeld grondstof is hierin gesplitst tussen statiegeld uit het SRN systeem en uit een ander systeem.

Hieruit bleek dat de PET-fles-grondstoffen uit statiegeldsystemen bijna uitsluitend PET flessen bevatten die voldoen aan de ontwerprichtlijnen van de European PET Bottle Platform (EPBP). In één van de twee PET-fles-grondstoffen afkomstig van statiegeldsystemen werden geen barrièreflessen aangetroffen en 0,2% gekleurde flessen. In de PET-flessen-grondstof van het andere statiegeldsysteem was dat 0,6% barrièreflessen en 4,9% gekleurde flessen. Dit contrasteert met de samenstelling van de PET-fles-grondstoffen afkomstig uit bron- en nascheiding. Ten eerste bevatten deze grondstoffen forse hoeveelheden (17-24%) aan non-food flacons, 1 tot 2% barrière PET flessen, minder dan 1% opake PET flessen, 4-10% niet-flessen PET objecten en ongeveer 2% in totaal aan foutief gesorteerde objecten van PE, PP, PS, PVC en zelfs andere materialen. Van een veel groter deel van de PET flessen aanwezig in deze grondstoffen is het ontwerp niet in overeenstemming met de EPBP ontwerprichtlijnen, door de aanwezigheid van barrières, aluminium sluitcaps, PS krimp-labels, glazen balletjes, metalen balletjes, metalen veren, POM, ABS en siliconen ventielen. Deze ongewenste materialen en componenten werden vooral aangetroffen in de sorteercategorieën PET food flacons en PET non-food flacons. Bij de non-food flacons werden ze vooral aangetroffen in de mechanieken voor handpompen en spray-guns.

Invloed van verontreinigingen op de rPET kwaliteit

De invloed van 10 verschillende verontreinigingen op de eigenschappen van rPET werd systematisch onderzocht. Hiertoe werd in samenspraak met de 'industrial board' (zie inleiding, paragraaf 1.3.2) eerst vastgesteld welke 10 verontreinigingen belangrijk zijn en in welke concentratie deze verwacht worden in PET-flessen-grondstoffen. Vervolgens werd PET maalgoed gemaakt van schone PET fleslichamen (zonder label, dop, sluiting, etc.). De verontreinigen werden droog gemengd met het PET maalgoed, geëxtrudeerd tot regranulaat en nagecondenseerd (SSP-behandeling). Het PET regranulaat werd geanalyseerd met kleurenmeting, DSC, GPC, IV en GC. Daarnaast werd een deel van het granulaat spuitgegoten tot test-staafjes. Deze testplaatjes werden met DMTA en haze (mate van troebelheid) onderzocht. Bijna alle verontreinigingen bleken de eigenschappen van het rPET negatief te beïnvloeden. De meest voorkomende invloeden waren toename van troebelheid (haze) en verandering van de temperatuur waarbij kristallisatie begint (de zogenoemde onset). Tijdens het afkoelen vindt de kristallisatie dan al bij hogere temperaturen plaats in de aanwezigheid van PVC en EVOH. Tijdens opwarmen vindt de kristallisatie dan bij lagere temperaturen plaats in aanwezigheid van PLA en PE. In het algemeen leiden polyolefine verontreinigingen zoals PE, PP en PS afkomstig van zowel foutief gesorteerde objecten als van fles-eigen componenten tot meer deeltjesverontreiniging en dus een troebeler PET. Bovendien versnellen PE en PS het kristallisatiegedrag van PET zonder dat de kristallisatiegraad verhoogd wordt. Hotmelt-resten en barrière-additief Amosorb leiden tot een iets gelere kleur rPET.

Barrièremateriaal EVOH reageert met PET tot vernette structuren, wat kan worden afgeleid van de toename in de gemiddelde molecuulmassa (Mz). PVC dat afkomstig is van sorteerfouten kleurt het PET geler en beïnvloed het kristallisatiegedrag. Een verandert kristallisatiegedrag kan onder andere invloed hebben op het blaasproces van PET flessen en de gasdoorlaatbaarheid van het materiaal (houdbaarheid). Het verlaagt het gemiddelde molecuulgewicht en de intrinsieke viscositeit van rPET en na SSP behandeling zijn afbraakproducten waarneembaar met gaschromatografie. Dit laatste waarschijnlijk doordat het PVC gedurende verhitting zoutzuur elimineert, wat de afbraak van PET katalyseert. De belangrijkste invloeden van de verontreinigingen op de kwaliteiten rPET zijn samengevat weergegeven in Tabel a.

Tabel a: Belangrijkste invloed van verontreinigingen op rPET kwaliteit.

Verontreiniging	**Belangrijkste invloed op rPET kwaliteit**
Inkt	Reductie van de intrinsieke viscositeit (IV) en molecuulgewichten voor nacondensatie
Amosorb	Vergeling
PS	Vergeling, Haze (toegenomen troebelheid), ΔIV laag; vertraagt re-condensatie
PP	Haze en deeltjesverontreiniging ΔIV laag; vertraagt re-condensatie
PLA	Eerdere kristallisatie bij lagere temperaturen tijdens opwarmen ΔIV laag; vertraagt re-condensatie
EVOH	Vernet met PET gedurende de nacondensatie Eerdere kristallisatie bij hogere temperaturen tijdens afkoelen ΔIV laag; vertraagt re-condensatie
LDPE	Haze, deeltjesverontreiniging Eerdere kristallisatie bij lagere temperaturen tijdens opwarmen
Nylon (MXD6)	
Hot-melt	Vergeling Eerdere kristallisatie bij hogere temperaturen tijdens afkoelen Zwarte stippen na extrusie
PVC	Vergeling en wordt roder Vorming vluchtige afbraakproducten Reductie van IV en molecuulgewichten voor nacondensatie ΔIV laag; vertraagt re-condensatie Eerdere kristallisatie bij hogere temperaturen tijdens afkoelen

Kwaliteit rPET die kan worden gemaakt uit verschillen PET-fles-grondstoffen

Tenslotte werden verschillende PET-fles-grondstoffen mechanisch gerecycleerd in rPET granulaat met twee verschillende processen met een standaard laboratoriumopstelling. Hierdoor konden de meetresultaten goed met elkaar worden vergeleken. Het standaard mechanische recyclingproces bootst de huidige verwerking van de Nederlandse statiegeldflessen na. Het geavanceerde recyclingproces start met het standaardproces en is vervolgens uitgebreid met additionele scheidingsstappen, welke bedoeld zijn om contaminanten af te scheiden, zoals het fijn zeven van het ongewassen PET maalgoed met een 2 mm zeefplaat, machinale NIR-sortering van het gewassen maalgoed en kleur-sortering hiervan. PET–flessen-grondstoffen van twee verschillende statiegeldsystemen werden gerecycleerd volgens het standaard proces en de PET-flessen-grondstoffen van bron- en nascheiding werden gerecycleerd volgens beide recyclingprocessen. Aan de PET-flessen-grondstoffen van bron en nascheiding werden PET flessen uit het statiegeldsysteem toegevoegd om een mogelijk toekomstig scenario na te bootsen waarbij de plicht van het voeren van een statiegeldsysteem zou zijn afgeschaft. Deze samengestelde stromen werden gerecycleerd met het geavanceerde proces. Tenslotte werden beide samengestelde flessenstromen nogmaals gerecycleerd met het geavanceerde proces waarbij de grondstof eerst werd voorgesorteerd om te voldoen aan de strengere sorteerspecificatie DKR 325 (alleen flessen en flacons).

Allereerst moet worden opgemerkt dat geen van de in het laboratorium geproduceerde rPET granulaten voldeden aan gangbare inkoopvoorwaarden van frisdrankproducenten ten aanzien van kleurwaarden en haze-waarden. Dit contrasteert met de industriële praktijk. Blijkbaar leidt de laboratoriumaanpak tot een systematisch lagere kwaliteit rPET dan wat industrieel gebruikelijk is. Dientengevolge mogen de resultaten van dit onderzoek niet absoluut maar wel vergelijkenderwijs worden gebruikt.

Redelijk goede kwaliteiten rPET konden worden verkregen uit PET-fles-grondstoffen afkomstig van statiegeldsystemen die met het standaard proces werden gerecycleerd, ten aanzien van kleur, haze (mate van troebelheid), intrinsieke viscositeit en kristallisatiegedrag. Wel werden er kleine verschillen waargenomen tussen de rPET producten gemaakt van flessen van beide verschillende statiegeldsystemen. Het rPET van flessen van het SRN-systeem was wat donkerder, terwijl het rPET van flessen van het andere statiegeldsysteem lichter maar wel iets geler was. Dit kan waarschijnlijk worden verklaard doordat een frisdrankproducent van wie de flessen in het SRN worden verwerkt reheat-additief (carbon black) toepast, terwijl dit niet in flessen uit het andere systeem wordt toegepast. Daar staat tegenover dat het andere statiegeldsysteem ongeveer 0,6% barrièreflessen bevat, wat de lichte vergeling verklaart.

Wanneer echter PET-fles-grondstoffen afkomstig uit bron- en nascheiding worden gerecycleerd met het standaard proces worden er minderwaardige kwaliteiten rPET verkregen ten aanzien van kleur, haze, deeltjesverontreiniging, intrinsieke viscositeit, molecuulgewichten en vluchtige afbraakproducten. De combinatie van relatief hoge concentraties contaminanten in deze flessen-grondstoffen en het onvermogen van het standaardproces om deze af te scheiden veroorzaakt waarschijnlijk deze mindere rPET kwaliteiten.

Wanneer daarentegen deze PET-fles-grondstoffen uit bron- en nascheiding worden gerecycleerd met het geavanceerde recyclingproces worden er veel betere rPET producten verkregen in termen van kleur, haze, deeltjesverontreiniging en de sterke reductie in het vluchtige afbraakproducten. In veel maar niet alle kwaliteitsaspecten zijn deze rPET producten vergelijkbaar met de rPET producten gemaakt van statiegeldflessen met het standaardproces. Een belangrijk verschil tussen beide processen is de opbrengst, zowel de netto massarendementen als de netto PET-opbrengsten zijn 5 à 10% lager voor het geavanceerde proces in vergelijking met het standaardproces. Dit grotere verlies wordt vooral veroorzaakt door de additionele machinale NIR-sortering van het gewassen maalgoed, waardoor extra materiaal uit de grondstofstroom verdwijnt.

Door de toevoeging van grote PET flessen afkomstig uit het statiegeldsysteem aan de PET-flessen-grondstoffen van bron- en nascheiding worden de kwaliteiten rPET verkregen met het geavanceerde proces weinig beïnvloed, alhoewel de rendementen licht verbeteren. Deze toevoeging zorgt er voor dat de contaminanten als fles-eigen componenten en sorteerfouten worden verdund. Het concentratieniveau blijft echter voldoende hoog om de rPET kwaliteit licht negatief te beïnvloeden. Ook de invloed van een additionele sorteerstap van DKR 328-1 naar DKR 325 heeft relatief weinig invloed op de kwaliteit van het geproduceerde rPET. Deze additionele sorteerstap elimineert contaminanten die afkomstig zijn van sorteerfouten. Echter er blijven nog voldoende contaminanten over die afkomstig zijn van de PET flessen en flacons zelf (vreemd kunststof, glas, metaal) die blijven drukken op de rPET kwaliteit.

Aanbevelingen voor de betrokken in de rPET keten, met het oog op zo veel mogelijk hergebruik van PET met een zo hoog mogelijke kwaliteit, zijn:

1. Kies en implementeer een tracer techniek voor het snel, mechanisch sorteren van non-food flacons uit de PET producten.
2. Gebruik tegelijkertijd deze tracer technologie om andere ongewenste PET flessen (barrière-flessen, opake PET verpakkingen en PET trays) te markeren en makkelijk automatisch uit te sorteren.
3. De gehele keten[1] actief stimuleren om de design richtlijnen van EPBP na te leven.
4. Sorteerbedrijven blijven aanmoedigen om PET grondstoffen te produceren met een minimale hoeveelheid sorteerfouten en restafvalcomponenten en nieuwe striktere specificaties ontwikkelen die passen bij de nieuwe tracer technologie.
5. Het bestuderen en herontwerpen van PET non-food flacons met handpomp of spray-guns zodat er weinig of geen glas, metaal, POM, silicone, ABS of andere plastics met een dichtheid hoger dan 1 g/ml worden geïntroduceerd in het te recyclen materiaal.

Om een zo goed mogelijke eindkwaliteit rPET te verwezenlijken zullen de inspanningen van alle betrokkenen in de keten op elkaar moeten worden afgestemd. Wanneer er op één plek in de keten een verandering plaatsvindt zal dat gevolgen hebben op de andere ketenonderdelen. Welk inzamel- en hergebruiksysteem er in de toekomst ook gevoerd gaat worden, de individuele verantwoordelijkheden zullen moeten worden afgestemd op systeemniveau.

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[1] *Met de hele keten wordt geduid op alle bedrijven die invloed kunnen uitoefenen op het ontwerp van de PET verpakkingen, zoals bedrijven die PET verpakkingen ontwerpen, produceren, inkopen, op de markt zetten en verkopen.*

Abstract

In 2015 a research project has been conducted in the Netherlands aimed to improve the scientific understanding of mechanical recycling processes from sorted PET bottle feedstocks into rPET products. In the Netherlands three different types of PET bottle feedstocks are produced, originating from deposit refund, separate collection and mechanical recovery systems. The PET bottle feedstocks originating from these three different collection systems have different compositions. First the composition of these feedstocks in terms of packaging and material categories was determined by object-wise sorting. In order to be able to identify barrier bottles, first a new barrier bottle detection method was developed. This yielded a list of barrier PET bottles and flasks present on the Dutch market, which in turn was used to correctly sort the PET bottles and flasks in the different categories. Secondly, the average material composition per packaging category was determined by disassembling packages into components, weighing the components and analysing the components by near infrared (NIR) and in case necessary infrared (IR). This comprehensive analysis of the PET bottle feedstocks revealed that in bottle feedstocks from deposit refund systems almost exclusively PET-bottles are present that were designed according to the European PET Bottle Platform (EPBP) design guidelines. In one of the two studied deposit refund PET bottle feedstock no barrier bottles and 0.2% coloured PET bottles were present. And in the other deposit refund PET bottle feedstock 0.6% barrier bottles and 4.9% coloured bottles were present. In contrast, PET bottle feedstocks originating from separate collection systems and mechanical recovery systems contained substantial amounts of non-food PET flasks (17-24%), 1-2% barrier PET bottles, <1% opaque PET bottles, 4-10% non-bottle PET objects and about 2% in total of faulty sorted objects from PE, PP, PS, PVC and even some residual materials. A larger portion of the PET bottles present in these feedstocks were not compliant with the EPBP design guidelines, due to the presence of barriers, metal caps, PS shrink labels, glass balls, metal springs, metal balls, POM, ABS and Silicone valves. These undesired components were especially found in the packaging categories PET food flasks and PET non-food flasks .

The impact of ten impurities on the properties of rPET were systematically studied. First of all, PET flakes were made from clean soft drink bottles. The contaminants were dry mixed in the PET bottle flakes, extruded into granulates and subjected to SSP treatment. The PET granulates were analyses with colour measurements, DSC, GPC, IV, GC and injection moulded test specimen were tested with Haze measurements and DMTA. Almost all impurities were found to impact the quality of the rPET in a negative manner. The most common type of impact by the impurities was a change in the onset of crystallisation. During cooling the crystallisation occurred at higher temperatures with PVC and EVOH as contaminants and during heating it occurred at lower temperatures with PE and PLA as contaminants. Two impurities were found to have markedly different behaviour. EVOH most probably cross-links with PET as can be deduced from the rise in Mz molecular weights. PVC causes PET chains to degrade faster, as can be

discerned from the yellowing, the low molecular weights prior to SSP, the relatively low gain in IV during SSP and the emission of degradation products.
Finally, various PET bottle feedstocks were mechanically recycled into rPET granulates with two different processes with a standard laboratory set-up for a comparative analysis. A straight-forward standard recycling process which mimics the current recycling process for bottles from the deposit-refund system and an advanced mechanical recycling process with additional separation equipment (sieving with a 2 mm screen, flake-sorting and colour-sorting). Two different PET bottle feedstocks from deposit refund systems were subjected to the standard process. PET bottle feedstocks from separate collection and mechanical recovery were subjected to both the standard and the advanced recycling process. To both PET bottle feedstocks large PET bottles from the deposit refund system were added to mimic a potential future scenario in which the deposit refund systems would be abolished and the recycling with advanced recycling process was repeated. And finally both composed PET bottle feedstocks were first subjected to an additional pre-sorting step to let them comply with sorting specification DKR 325 (bottles and flasks only) and then subjected to the advanced mechanical recycling process.

None of rPET granulates produced was found to qualify with the purchase specifications of major beverage manufacturers. The rPET produced was too dark and too hazy. Apparently the laboratory execution of the recycling processes yields rPET with less good properties in comparison to the industrial practise. Therefore, the results of this study cannot be used in absolute terms, but should only be used in a comparative manner.

Fairly good qualities of rPET could be obtained from PET bottles feedstocks from deposit refund systems with the standard mechanical recycling process, in terms of colour, haze, IV and crystallisation behaviour. The same standard process with PET bottle feedstocks originating from separate collections and mechanical recovery, however, yielded inferior qualities of PET recyclates, in terms of colour, haze, particle contamination, molecular weights and the emission of degradation products. The presence of relatively high concentrations of contaminants in these feedstocks, combined with the inability of the standard process to remove these, is the likely cause of these poor rPET properties.

When, however, the same feedstocks were mechanical recycled with the advanced recycling process the resulting rPET products were much better, in terms of colour, haze, particle contamination and the largely reduced emission of degradation products. In many (but not all) quality aspects these rPET products have comparable properties as those made from deposit refund bottles and the standard process. A major difference between both recycling processes are the yields; both the net recovered masses as the net PET yields are lower by 5 to 10% for the advanced process in comparison to the standard process. This loss in yield can mostly be attributed to the additional flake sorting step.

With the addition of the large PET bottles from the deposit refund system to the PET bottles of the separate collection system and the mechanical recovery system the obtained rPET qualities are hardly affected, although the process yields do slightly improve. This addition dilutes the contaminants originating from bottle-components and faulty sorted objects, but the level of contaminants remains relatively high. Hence the properties of the final products are hardly affected by this dilution. Also the additional pre-sorting step from DKR 328-1 to 325 hardly affects the quality of the rPET produced. Most likely the level of contaminants that originate from bottle-components is relative high and hence the drop in contaminants from faulty sorting objects is hardly noticed in the final product.

In the end, the quality of rPET is influenced by multiple stakeholders with strong interactions between the efforts done by the various stakeholders. No matter what type of collection & recycling system will be developed in the future, the individual responsibilities in relation to the total system achievement (rPET quality) will need to be managed to be able to achieve high qualities of rPET.

Content

1 Introduction

This research project deals with the technical quality of rPET that can be obtained from Dutch PET bottles that have been collected, sorted and mechanically recycled in different manners. This introduction describes the context, project organisation, objectives and the structure of this research project. In this report some scientific terms are used to indicate recycling systems, these terms are explained in Appendix A.

1.1 Context

1.1.1 *Political and societal*

For several years Dutch politicians are debating a possible abolishment of the obligation to operate a deposit refund system for large (>0.5 litre) PET bottles for water and soda's and the potential amalgamation of these bottles to the collection and recycling systems for the other plastic packages. An important issue in this debate is the technical quality of rPET that can be attained via the two collection routes[2]. Many stakeholders care about the closed loop recycling of large PET bottles via the deposit refund system, since this is considered *the* example of closed loop recycling of plastic packages. This raises the question which control measures have to be taken in order to attain a sufficient high quality of rPET for closed loop recycling in sufficient high volumes for the Dutch market, in case of system amalgamation. These control measures can relate to the beverage producers (design guidelines), collecting services (instructions for the households), sorting facilities (specifications) and recycling plants (implementation of technology). All these stakeholders have a partial influence on the technical quality of the recycled PET that can be attained, see Figure 1.

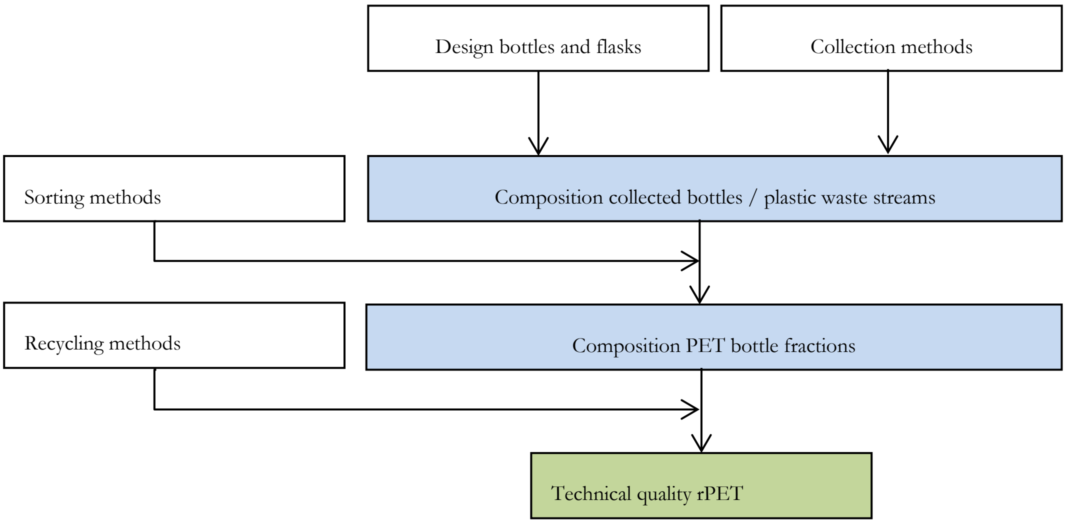

Figure 1: The technical quality of the retrieved rPET is determined by a complex interaction between the bottle design, collection methods, sorting methods and recycling methods.

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[2] *In the Dutch context this means 1) deposit refund systems and 2) the combination of the separate collection system and mechanical recovery system for plastic packaging waste.*

It is of importance that the results of this study are scientifically peer reviewed and supported by stakeholders, so they can be used in the societal debate. Therefore it was decided to establish a scientific review board and an industrial review group, see paragraph 3.1.

1.1.2 *Technical*

In the past decades several recycling systems have been developed globally which mechanically recycle PET bottles into bottles, trays, fibre fill, strapping, etc. In the Netherlands PET bottles from the deposit refund system, separate collection system and the mechanical recovery system are all used to make new bottles, trays and fibre fill. The PET bottles from the deposit refund system are recycled into food-grade rPET for bottles and trays without the need for a preceding additional sorting step. The PET bottles from the separate collection and mechanical recovery systems are not directly recycled into food grade bottles and trays, they require an additional preceding sorting step in order to sufficiently remove non-food flasks and impurities. The latter is required to comply with the EFSA condition that the input material should not contain more than 5% of non-food flasks (EFSA 2011), although from a FDA (US Food and Drug Administration) point of view a higher amount of non-food flasks might be possible (FDA 2006). EFSA introduced this 5% limit as a precautionary measure to limit the possible contamination of rPET from non-food flasks. However, evidence is building both in the USA and in the scientific community (Welle 2013) that this limit doesn't address the food safety issues for rPET correctly. Nevertheless, this is the legal limit in Europe at the moment, which also Dutch PET recycling companies have to comply to.[3]

In countries like Belgium[4], France[5] and Switzerland[6] separately collected PET bottles are mechanically recycled in fibres, trays and bottles. In Spain[7] two PET bottle feedstocks (separate collected and mechanically recovered PET bottles) are mechanically recycled into similar products. Whereas in countries such as Germany[8], Denmark[9] and Sweden[10] the predominantly present PET bottle feedstocks originate from deposit refund systems, which are also used to make similar products. This simple comparison between countries reveals that closed loop recycling is possible via various different collection routes. The specific manner in which the mechanical recycling is conducted, the yields and the attained qualities will differ from each other, but these have not been documented in a comparative manner.

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[3] *Also see review remark no 1 of Mr. Frank Welle in the Appendix N.*

[4] *https://www.fostplus.be/nl/sorteren-recycleren/alles-over-recyclage/pmd-recycleren*

[5] *http://www.ecoemballages.fr/recyclagebouteillesplastique/*

[6] *http://www.petrecycling.ch/de/*

[7] *https://www.ecoembes.com/en/citizens/packages-and-recycling-process/pick-up-sorting-and-recycling-process*

[8] *http://www.dpg-pfandsystem.de/index.php/de/die-pfandpflicht-fuer-einweggetraenkeverpackungen/ruecknahmepflicht-und-pfanderstattung.html*

[9] *http://www.dansk-retursystem.dk/*

[10] *http://pantamera.nu/pantsystem/statistik/pantstatistik/*

Recent Dutch research in laboratories of both Cumapol[11] and Food & Biobased Research have shown that mechanically recovered and sorted PET bottles (complying with DKR specification 328-1) can be recycled into a high quality rPET, of which the technical quality is comparable to rPET's made from PET bottles originating form separate collection and deposit refund systems. The European associations of beverage producers (UNESDA and EFBW) have published a design guide in 2011, which qualitatively describes the impact of the most relevant contaminants on the rPET quality. They summarised this in a qualitative overview of which bottle-materials are easy to handle for PET bottle recycling facilities, which bottle-materials can conditionally be dealt with in recycling facilities and which bottle-materials are problematic (van Dongen et al.). This is valuable qualitative information on the European level. According to our industrial board most PET bottles in the Dutch deposit refund system are well-suited for recycling. These PET bottles have been designed for recycling. Only a minority of the bottles present in the deposit refund systems contain a colour or a barrier and are slightly less suited for recycling. However, as long as these coloured bottles can be sorted out and the concentration of these barrier bottles remains low, the total mixture can still be recycled to food grade rPET. In the separate collection and mechanical recovery system, many more PET bottles and flasks are expected to be present that can only be recycled after applying additional sorting and/or recycling steps.

The high quality rPET that is obtained from deposit refund bottles can not only be related to the selective collection method but also stems from the favourable bottle design, which excludes the need for an additional sorting step and allows for a relative simple recycling process. From the comparison with other countries it is apparent that also a high quality rPET can be obtained from PET bottles from separate collection and mechanical recovery systems. Since the design of the bottles and flasks present in these fractions is less well suited for recycling, additional sorting and recycling steps are required to obtain rPET material with a similar high quality as the material obtained from deposit refund systems.

This yields the overall impression that the effort that needs to be done at the end of the recycling chain (sorting and recycling) is strongly dependent on the effort that has been done at the start of the recycling chain (collection method and design of the bottles). Although this hypothesis appears to be plausible, it will have major ramifications for the stakeholders and it is therefore of great importance that it is scientifically verified.

1.1.3 *Scientifically*

Various scientific research groups have contributed in the past years to enlarge the knowledge of PET bottle recycling. Two scientific review papers on PET bottle recycling have appeared. The review paper of Awaja from 2005 (Awaja and Pavel 2005) gives a firm basis regarding the polymer and its use, but lacks information on the modern super-clean technologies, of which the

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[11] *http://www.cumapol.nl/*

large scale adaptation would follow a few years later. The review paper of Welle from 2011 (Welle 2011) discusses these super-clean technologies in detail.

A large part of the most recent literature deals with the solid state condensation process (SSP). PET only absorbs contaminants with low molecular masses (<300 g/mole), like limonene, toluene, acetaldehyde, etc. (Welle 2013) and these are sufficiently volatile to remove with SSP within 5 hours (Welle 2014). The volatile contaminants are being removed during the whole recycling process, roughly 90% is removed during extrusion and the remaining 10% is removed during SSP (Oliveira et al. 2014). A downside of SSP is the yellowing of the PET product. Rieckmann investigated this yellowing process extensively and found that it relates to the temperature (above 190ºC), the oxygen concentration in the SSP reactor (less oxygen means less yellow discoloration) and of the diethylene glycol (DEG) co-monomer content (more DEG means more yellowing) (Rieckmann et al. 2013). Krehula investigated the washing process with warm alkaline solutions. The optimum was found to be 15 minutes at 75ºC with 0.5M sodium hydroxide between the effective removal of hot melt residues and PET chain scission (as derived from the concentration of dissolved PET oligomers in washing water) (Krehula et al. 2012) Finally, Badia has found that the DMTA technology yields valuable insights in the morphology of recycled PET. Multiple recycling of PET results in reduced lamellar thicknesses (Badía et al. 2009).

Besides these scientific publications, WRAP has also produced an extensive applied research report on the issues with closed loop (bottle to bottle) recycling of PET in the United Kingdom (Dvorak et al. 2013). British PET bottle recyclers regarded 'black specs' as the largest quality issues they have to deal with, which they attributed to the presence of PVC. Laboratory research confirmed that although PVC is a likely cause, it is not the only cause, since charred paper fibres were observed in some of these black specs. Other quality issues that the PET recyclers reported were the presence of coloured plastic objects, metal, other heavy plastics (PS, ABS, silicone valves, POM), films, other materials (glass, paper) and fines (sand, dust, etc.). Especially metal springs used in the pump mechanisms of flasks with a spray gun or hand pump were reported to be difficult to separate.

Applied research confirmed that the removal of fines by sieving (< 2 mm) improves the quality of rPET greatly, probably because PVC and other contaminants are predominantly present in the fine fraction. Additionally wind sifting of crudely milled PET material helps to reduce the contamination with blisters, films, labels and nylon barrier layers (Dvorak et al. 2013) [12].

In conclusion, much is already known about PET bottle recycling processes. However, to the best of our knowledge, there are no publications available that relate the technical quality of the

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[12] *The quality of British PET bottle fractions is regarded by Dutch stakeholders as more polluted than those from the Netherlands, Belgium or Germany. Hence these observations could also be true for Dutch material, but this is not sure.*

recycled rPET to the collection system with which it was retrieved from the consumers and hence to the composition of this PET bottle fraction, including the contaminants present.

1.1.4 *Level of the available knowledge*

From the sectorial sustainability action plan of the Dutch beverage industry (FWS) and the discussions within the project group "PET bottle recycling", it is apparent that there are still many unanswered questions regarding PET bottle recycling. Within the industry there are some clues for the causes of quality deterioration of a number of relevant quality aspects of rPET. However, none of them are systematically studied and documented. Hence no relations can be drawn between the quantities of contaminants present within the PET bottle products, the sorting and recycling methods and the technical quality of the obtained rPET. Thus it is still impossible to recommend improvement measures that are evidence based. This relates to the following list of quality aspects of rPET:

- Darkening of rPET which is related to the thermal degradation of PET additives, contaminants, virgin PET itself, barrier bottles and possibly ink residues.
- Yellowing of rPET which is related to the degradation of PVC, EVOH, barrier bottles[13], additives like colorants, and is also related to the SSP conditions and the type of virgin PET (especially the DEG co-monomer content).
- Reduction of the optical clarity (haze) which is related to particle contamination.
- Presence of black specs and other large, clearly visible imperfections that are related to PVC, paper fibres and non-meltable contaminants. These imperfections may result in the loss of mechanical properties, especially in blow outs (rupture and failure during the bottle blowing process due to the presence of non-meltable contaminants in the preform.
- Molecular weight distribution of rPET and PET morphology. Although it is known that SSP is able to restore the average molecular weight of PET to levels similar of virgin PET, evidence suggests that recycling does alter the morphology, chain structure and present terminal groups. Furthermore, the molecular weight distribution is broadened by SSP (the polydispersity is enlarged). This indirectly results in changes of processability and raise the variance in bottle wall thickness.

All these quality attributes limit the applicability of rPET in bottles. An improved understanding of the relations between contaminants present, their concentrations and the impact on the quality attributes of rPET will enable stakeholders to jointly propose on evidence-based measures to improve the quality of rPET.

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[13] *Barrier bottles is a generic term for all type of PET based bottles to which something (multilayer, additives, absorbers, coating) is added to reduce the gas permeability.*

1.2 Objectives and project structure

This research project aims to determine the technical quality of mechanically recycled PET that can be attained with PET bottle products from different Dutch collection systems, which have been subjected to different sorting methods and recycling processes. Moreover, this project also aims to unravel the relationships between the design parameters of PET bottles, the contaminants present, the presence of faulty sorted objects and the technical quality of mechanically recycled PET.

These evidence-based relationships can be used to propose recommendations regarding bottle design, sorting methods and recycling methods to improve the quality of rPET.
The added-value of this research project is that relationships are clarified between bottle design, collection methods, sorting methods and recycling methods on one side and the technical quality of the obtained rPET on the other side. Additionally, the impact of merging the PET bottles from the deposit refund system to the recycling process of the separately collected and mechanically recovered PET bottle products will be investigated and clarified. This project aims to contribute to achieve technical-scientific consensus between the main industrial stakeholders on the consequences of a potential future system integration.

1.2.1 *Main objectives*

1. To determine the influence of bottle design, collection method, sorting method and recycling method on the technical quality of mechanically recycled PET.
2. To improve the understanding of the mechanisms of quality decay of rPET.
3. To propose evidence-based recommendations to improve the technical quality of rPET.
4. To achieve consensus on these recommendations between the most important industrial stakeholders.

Objectives 1 and 2 relate to the scientists and this report. The results of this study will be used by the industrial board to achieve objectives 3 and 4. The latter objectives therefore lie outside the scope of the scientists and this report. Nevertheless, the quality of the scientific output should be sufficient for the industrial stakeholders to define their recommendations.

1.2.2 *Scope and constraints*

This research project studies the Dutch situation. Therefore, PET bottle products will be investigated that are currently present in the Netherlands or that will likely be present in the future from deposit refund systems, separate collection systems and mechanical recovery systems.

This study attempts to mimic the actual Dutch situation as accurately as possible, on a laboratory scale. It is not an industrial research, yet findings from the laboratory scale research may be interpreted for industrial purposes. Industrial research is not suitable for systematic comparative scientific research, since every recycling facility has different equipment, machine settings and feedstocks.

This project will not deal with food safety and migration issues. This project is limited to the technical quality. The approval of food safety for recycling processes is a long dedicated process (see EU 2008/282) that is not part of this project for reasons of content, time and budget.

1.2.3 *Work packages*

Three work packages are derived from the main objectives, these are:

- Work package 1: To determine the composition of PET bottle fractions that are collected and sorted in different manners, containing counted bottles from the deposit refund system, sorted PET bottles from separate collection that comply to DKR 328-1 and sorted PET bottles from mechanical recovery that comply to DKR 328-1. These compositions will be determined in detail and will contain: materials that are integral parts of PET bottles, faulty sorted objects, residual waste and contaminants.
- Work package 2: To comprehend the relation between the PET feedstock and the quality of the retrieved rPET, in such a way that the influence of bottle-materials, faulty sorted objects and contaminants on the rPET quality is clarified and the underlying mechanisms of quality decay are revealed.
- Work package 3: To determine the influence of the composition of PET bottle products (as they are collected and sorted) in combination with the sorting and recycling methods on the technical quality of the retrieved mechanically recycled rPET. Additionally also future scenarios are investigated in case the PET bottles from the deposit refund system are collected and recovered together with the rest of the plastic packaging waste and subsequently sorted.

Work package 1 will render objective information on the composition of PET bottle fractions from separate collection, mechanical recovery and deposit refund systems in terms of materials that are an integral part of PET bottles, faulty sorted objects and contaminants. This data will be used to draw relations between the composition of the PET fractions and the technical quality of the rPET. Moreover a detailed understanding of the composition of the fractions is necessary to propose meaningful recommendations.

Work package 2 will provide a scientific basis for understanding the underlying mechanisms of quality decay in rPET. This will help to find the causes of quality decay, which can be traced back to bottle design, collection methods and sorting methods. This is an important step in achieving consensus among industrial stakeholders.

Work package 3 will generate objective information on the relation between the composition of the PET bottle products, the recycling method and the technical quality of the retrieved rPET. Besides the current PET bottle products (from mechanical recovery, separate collection and deposit refund) also potential future PET bottle products will be studied which will exist after the

PET bottles from the deposit refund system have been merged with the two other existing collection systems.

1.3 Project organisation

The overall impression of the project organisation is given in Figure 2. The project organisation encompasses a client, an industrial board with an industrial reviewer, a project team with a project manager and a scientific review board. The tasks and responsibilities of all involved parties are described separately in the next sections.

1.3.1 *Client*

This project is financed by Stichting DDL and Stichting Afvalfonds. Mr. Cees de Mol van Otterloo of Stichting Afvalfonds is the formal client. He is also the chairman of the industrial board "project group PET bottle recycling".

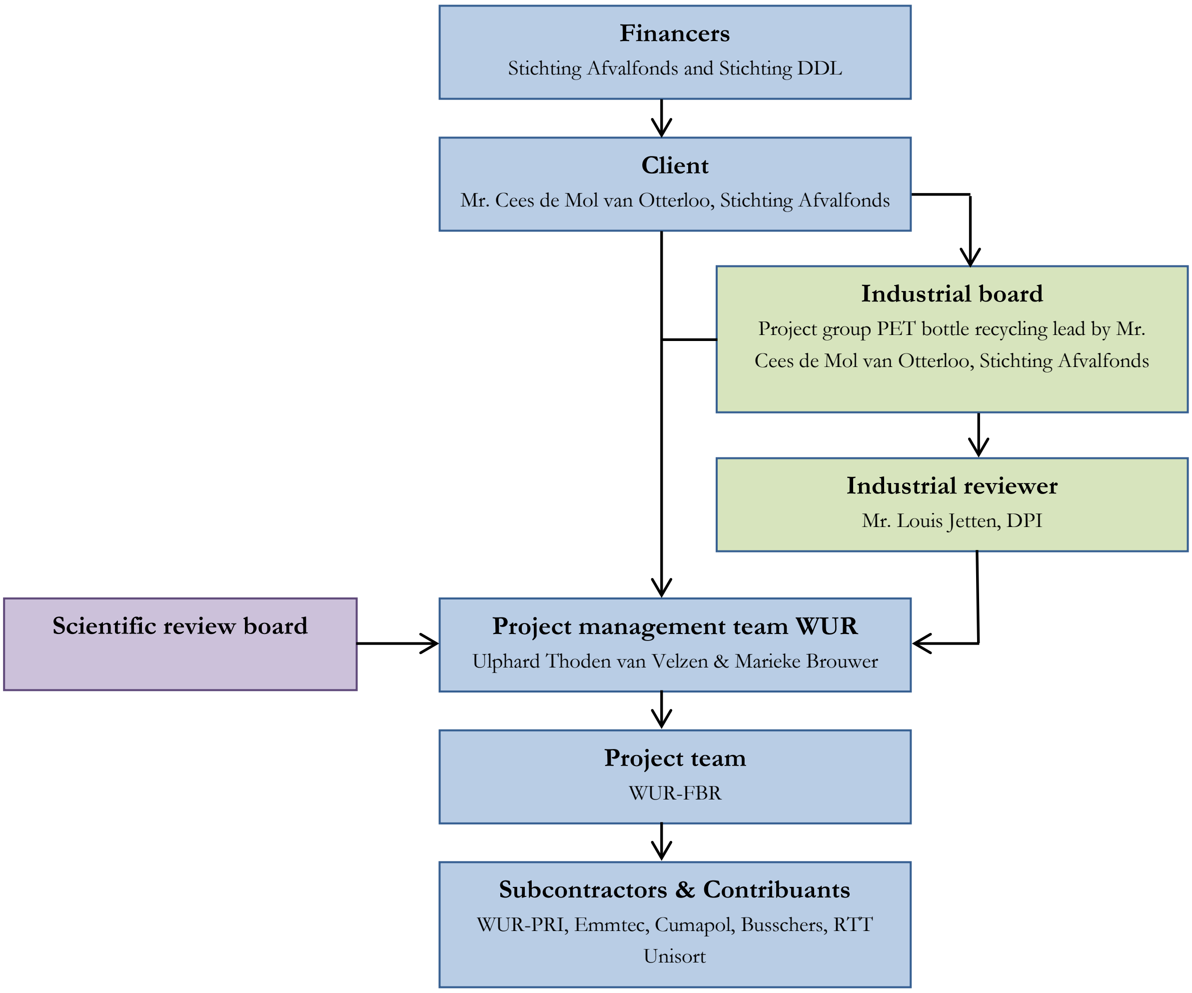

Figure 2: Project organisation.

1.3.2 *Industrial board*

The industrial board of this project is named “Project group PET bottle recycling”. This board is chaired by Cees de Mol van Otterloo of Stichting Afvalfonds. This board has the following members:

- Cees de Mol van Otterloo / Stichting Afvalfonds (chairman)
- Aafko Schanssema / NRK Verpakkingen (secretary)
- Herman Snellink / Suez
- Roger Beuting / Van Scherpenzeel
- Stefan Morssinkhof / Morssinkhof Plastics
- Hans Kuipers / 4PET Holding
- Jan Burger / Coca-Cola
- Louis Jetten / DPI Value Centre

Mr. Louis Jetten of DPI Value Centre has an additional role within this project, namely as industrial reviewer. This role implies that he supervises the choices that are made within the project and coordinates the interests of the industrial partners in that regard. This role should guarantee the applicability of the research results for the Dutch industrial practise of PET bottle recycling.

The industrial board has an advisory role within the project. The board advises the client and the industrial reviewer regarding important decisions within the project. This board is comprised of the most relevant stakeholders of the Dutch PET bottle recycling network. The commitment of these industrial partners to this project will add to the understanding of this research project and guarantees the applicability of the results for the involved Dutch industrial parties.

The subcontractors were WUR-PRI for haze measurements, Busschers for wind sifting experiments and Emmtec for Partisol measurements. Two partners contributed free of charge, Cumapol for the laboratory scale SSP treatment and the DSC and IV measurements and RTT Unisort for flake sorting.

1.3.3 *Scientific review board*

The quality of research plan, results and report is guaranteed by a scientific review board. This board will advise the researchers on methodological aspects. Furthermore, the board will verify that the results are interpreted in the correct manner. The opinions of the scientific review board will be documented. The board will be composed of three members from other EU countries with proven experience in PET bottle recycling. The members of the scientific review board are: Ms. Sigbritt Karlsson of Högskolan in Skövde in Skövde (Sweden), Mr. Frank Welle of Fraunhofer Institute for Process Engineering and Packaging (IVV) in Freising (Germany) and Mr. Edward Kosior of Nextek in London (United Kingdom).

2 Methods

The main objectives have been translated into three sub-objectives and concomitant work packages. The research methods applied in these three separate work packages are explained in the separate paragraphs below.

2.1 Work package 1: Methods to determine the composition of PET bottle products

Work package 1 aims at determining the composition of various PET bottle products that are relevant for the Netherlands; meaning counted PET bottles products from deposit refund systems, sorted fractions from separate collection systems and mechanical recovery systems. A detailed understanding of the composition of PET bottle products, including the contaminants present, is important to be able to relate the quality of the rPET made from these products to their composition. Only by knowing the origin of problematic contaminants, dedicated recommendations can be made for effective control measures. In order to understand the quality of rPET three types of contaminants are relevant; 1) non-PET materials that are an integral part of PET bottles and flasks, 2) faulty sorted objects and 3) enclosed product residues and attached moisture and dirt.

2.1.1 *Design of PET bottles and flasks: categorisation and composition*

To study the material composition of PET bottles and flasks present on the Dutch market, were collected directly as post-consumer waste from households of colleagues of the FBR institute. Packages that were missing in the sampling were purchased in supermarkets (Aldi, Lidl, Albert Heijn, Jumbo and Spar in Wageningen). This rendered a good cross-section of PET packages on the Dutch market. The collected PET bottles and flasks were categorised according to the categories below. The sorting decision tree for the categorisation of these PET bottles and flasks is clarified in Appendix B.

- PET bottles > 1 litre, clear transparent (incl. light blue), divided in barrier and non-barrier
- PET bottles > 1 litre, coloured, divided in barrier and non-barrier
- PET bottles ≤ 1 litre, clear transparent (incl. light blue), divided in barrier and non-barrier
- PET bottles ≤ 1 litre, coloured, divided in barrier and non-barrier
- Opaque PET bottles, divided in barrier and non-barrier
- PET food flasks, divided in barrier and non-barrier
- PET non-food flasks, divided in barrier and non-barrier

Since, in every category barrier bottles and flasks can potentially be present, an analysis method was necessary to identify barrier bottles and flasks. This method can be used to screen all suspected bottles and flasks and identify those that contain barrier functions. The positive barrier bottles and flasks are then listed in a sorting list and all PET bottle categories can be split into two (normal and barrier). However, such an analysis method was absent and hence has been developed for this project. The European PET bottle platform already had developed an analytical test for the presence of flakes of barrier bottles in PET milled goods, the so-called oven provocation test (EPBP 2010).[EPBP 2010] This test was modified to function for approximately 5 x 5 cm large PET bottle wall cuttings, as it was needed to check every bottle separately. Since, these cuttings tend to shrink and curl up after exposure to 220°C for one hour, adaptations were deemed necessary. This modified analysis protocol is a result of this research project and hence its results are discussed in paragraph 3.1.2. For the sake of readability of the report, the modified analysis method is described here.

The bottles and flasks that were suspected to contain a barrier function, and were hence tested, encompassed: turbid transparent PET bottles, thin PET bottled for fruit juices and nectar juices, PET flasks for sauces, sterilised dairy based beverages and some other product types of which we did not suspect a barrier are checked as well, such as ice tea, vitamin waters, cola, etc. to verify the method. Based on the value of the difference in normalised b* colour value it was decided whether or not the bottle was a barrier bottle. In a few cases delamination could even be observed, with a barrier layer that was more shrunken than the exterior PET layers, see separate text below. The identified barrier bottles and flasks were listed on a sorting list for barrier bottles.

For each PET bottle and flask category the material composition was determined by measuring the material composition of the individual dry and clean PET bottles and flasks. The bottles were disassembled and all the constituents were weighted separately. The plastics components were identified with a NIR analyser [IOSYS Siro], components that were not detectable by NIR analyser, such as very small objects were analysed by FT-IR [Varian Scimitar 1000 MIR equipped with a Pike MIRacle™ ATR (Diamond w/ZnSe lens single reflection ATR plate)] and based on the spectra the material type was determined. The material composition was expressed as mass percentages of all constituents that were an integral part of the bottle and it involves various types of plastics (PET, PE, PP, PS, PVC, PA, POM, ABS, Silicone-rubber), glass, metal and paper. Per category a weighted average and standard deviation was calculated of the material composition of the category. During disassembling special attention was given to details that are relevant for mechanical recycling, such as the ease at which labels can be removed, the type of batch number prints and or best-before-dates (laser-engraved or inkjet-printed) and their location (on the bottle neck, cap or label) Also the colour of the bottle & product volume were noted per packaging. It was intended to study at least 25 typical examples of PET bottles / flasks per sorting category and to calculate the weight-averaged mass percentages over these 25 individual specimen[14]. In a few cases much more than 25 specimen per category were studied, but for a few categories (opaque bottles, coloured PET bottles) there were only a few bottles on the market and the amount of specimen was much lower than 25. This approach of averaging the

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[14] *Individual specimen = no double measurements for the same product/packaging combination. In some bases a similar bottles with different product (e.g. flavours) are included in the list of specimen.*

composition of 25 typical bottles and flasks per sorting category had previously proven to be effective in the analysis of beverage cartons (Thoden van Velzen et al. 2013).[Thoden van Velzen 2014]

Barrier bottle analysis method

Pieces of about 5 x 5 cm of PET material are cut from the bottle wall in such a way that these pieces are as flat as possible and fold lines and wrinkles are avoided. These PET cuttings were cleaned (with water and soap) to make sure that no product residues, hot-melt-residues or other dirt was present on its surfaces. The initial bottle wall thickness is determined with a mechanical thickness measurement tool by measuring at least five different locations and averaging the values. This yields the average initial thickness ($\bar{x}_{initial}$) and the standard deviation in the initial thickness ($\sigma_{x-initial}$). Then the initial b*-colour value is determined with a Minolta Chromometer. These values are also determined on five different locations on the cutting and averaged. This yields an average initial b*-value ($\bar{b}^*_{initial}$) and a standard deviation ($\sigma_{b*-initial}$). This PET-cutting is clamped in between two thin sheets of aluminium of 5 x 5 x 0.05 cm and placed in an preheated GC-oven at 220°C for one hour exactly. (The GC-oven was chosen for its precise temperature control.) After 1 hour the clamped piece of PET was quenched to room temperature by throwing it in a bucket of water for 1 minute, removing the clamp and drying the PET piece. The piece of PET is now shrunken, thicker and darker, but still flat. The final thickness and b*-colour values are determined by measuring at five different locations and averaging, yielding the average final thickness ($\bar{x}_{final}$), the standard deviation in the final thickness ($\sigma_{x-final}$), the average final b*-value ($\bar{b}^*_{final}$) and the standard deviation ($\sigma_{b*-final}$). These parameters are filled into the equation below to obtain the normalised difference in b* colour value.

Equation 1: The difference in thickness normalised b* colour value.

$$\Delta\bar{b}^*_{norm.} = \bar{b}^*_{final} \times \frac{100\ \mu m}{\bar{x}_{final}} - \bar{b}^*_{initial} \times \frac{100\ \mu m}{\bar{x}_{initial}}$$

Parameter	Meaning	Unit
$\Delta\bar{b}^*_{norm.}$	Difference in thickness normalised b* colour value	[-]
$\bar{b}^*_{final}$	Average final b* colour value	[-]
$\bar{x}_{final}$	Average final thickness	[µm]
$\bar{b}^*_{initial}$	Average initial b* colour value	[-]
$\bar{x}_{initial}$	Average initial thickness	[µm]

Equation 2: The standard deviation in the difference in thickness normalised colour values.

$$\sigma_{\Delta b*norm} = \sqrt{\left(\sigma^2_{b*norm.final} + \sigma^2_{b*norm.initial}\right)}$$

Equation 3: The standard deviation in the final thickness normalise colour value.

$$\sigma_{b*norm_{final}} = 100 * \sqrt{\left[\left(\frac{1}{\bar{x}_{final}{}^{2}}\right) * \left(\sigma_{b*final}{}^{2} + \left(\frac{\bar{b}^{*}_{final}}{\bar{x}_{final}{}^{2}}\right)^{2} * \sigma_{\bar{x}_{final}}{}^{2}\right)\right]}$$

Parameter	Meaning	Unit
$\sigma_{b*norm_{final}}$	Standard deviation in the thickness normalised final b* colour value	[-]
$\bar{b}^{*}_{final}$	Average final b* colour value	[-]
$\bar{x}_{final}$	Average final thickness	[µm]
$\sigma_{b*final}$	Standard deviation in the final b* colour value	[-]
$\sigma_{\bar{x}_{final}}$	Standard deviation in the final thickness	[µm]

The standard deviation in the thickness normalised b* colour value was derived from the square root of the sum of the squares of the final and initial normalised standard deviations, see Equation 2. The latter are derived from applying the error propagation laws to Equation 1 1, which yields Equation 3, which was used to calculate both $\sigma_{b*norm.final}$ and $\sigma_{b*norm.initial}$.

In case the difference in thickness normalised b* colour values was larger than the threshold value of 1.6, the bottle was identified as a barrier bottle. This threshold value was derived from two measurements with PET cuttings from PET bottles that were known to not contain any barrier layer or additive to two of those from PET bottles that were known to contain these barrier layers and/or additives, according to industrial representatives.

This barrier bottle measurement method will identify which bottles contain barrier function that cause excessive yellowing or browning of the PET resin during thermal treatment. This is the case for most barrier functions based on EVOH layers, PA layers, oxygen scavengers such as Amosorb, etc. However, a few barrier layers such as SiOx layers will not cause yellowing according to An Vossen of Plarebel[15] and hence bottles with such a coating will not be identified by this method. This is, however, not regarded as a problem by the PET recycling industry, since only bottles that cause excessing yellowing / browning should be identified.

2.1.2 *Composition of collected and sorted PET bottle products*

Sorted PET bottle products were obtained from the operators of deposit-refund systems and the operators of sorting facilities. The composition of these collected and sorted PET bottle products were determined by NIR-assisted manual object-wise sorting, using the general sorting protocol for plastic packaging waste (Thoden van Velzen 2013) with a few additions in PET bottle and flask categories. In this sorting process the PET bottles and flasks were sorted into the

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[15] *Meeting with An Vossen, 5th of March 2015, Wageningen*

relevant categories (see paragraph 2.1.1 and Appendix B). Additionally, the faulty sorted objects (such as PS, PVC, PP, PE, PC, PLA) were resorted and categorised. Finally, objects made from non-plastic materials were also sorted into a few residual material categories (paper & board, textiles, metals, glass, organic & indefinable residue).

Table 1 shows the list of the samples of PET bottle products taken and their weight. Although more samples would have helped to understand the variance in composition, the amount of samples was limited to those shown in the table as this represents the work-load that could be performed with the framework of this contract. The aim was to sample at least one bale of each origin for PET bottle products and to take a few additional smaller samples (bigbag size) from PET bottle products made from the separate collection and the mechanical recovery systems.

Table 1: Overview of the PET bottle products sampled for compositional analysis.

PET bottle product	Origin	Weight, [kg gross]	Sample day
SRN counted transparent	Deposit refund	217.35	14-5-‘15
Other system	Deposit refund via Cumapol	223.11	27-2-‘15
Suez 1, DKR 328-1	Separate collection	327.48	21-4-‘15
Suez 2, DKR 328-1	Separate collection	39.62	21-5-‘15
Schönmackers, DKR 328-1	Separate collection	15.16	29-4-‘15
Attero Wijster, DKR 328-1	Mechanical recovery	86.11	30-1-‘15
Omrin & Augustin, DKR 328-1	Mechanical recovery	269.14	24-3-‘15

The coloured PET bottle product from the SRN deposit refund system was not analysed, since this product was (and is) not used to recycle into new bottles. The results of the compositional analysis was expressed in weighted averages with standard deviations.

For all sorting categories of PET bottles and flasks, the percentages of attached moisture and dirt were determined from each sample by taking 10 representative bottles or flasks from each category, weighing post-consumer bottles, cleaning and drying them and weighing again. This yielded the percentages of attached moisture and dirt (PAMD in Equation 4) and these values were averaged per sorting category.

Equation 4: Percentage of attached moisture and dirt.

$$PAMD = 100\% - \frac{Net\ weight\ of\ dried\ and\ cleaned\ PET\ bottles}{Gross\ weight\ of\ PET\ bottles\ as\ received}$$

This collected data was used to calculate for each of three main origins of the PET bottle products (deposit refund, separate collection and mechanical recovery) weight-averaged values for all the PET bottles and flasks.

Additionally, the net PET material content was calculated for each PET bottle product from the average composition in terms of packaging categories, the average material composition of each packaging category and the average percentage of attached moisture and dirt. This net PET material content was calculated on three levels:

- For all PET bottles and flasks,
- For all transparent PET bottles and flasks and
- For all transparent food PET bottles and flasks.

2.2 Work package 2: Methods to study the impact of contaminants on the quality of retrieved rPET using model studies

The impact of the presence of low concentrations of contaminants on the quality of rPET was studied by adding relevant contaminants at realistic concentrations to clean rPET milled goods in a systematic way. These contaminated materials were extruded, treated by SSP and injection moulded into test specimen. The impact of the presence of the contaminant on the rPET was determined with the following analytical tools, of which a few were conducted on the extruded granulate prior to SSP treatment, a few were conducted on the granulate after SSP treatment and a few on the injected moulded test specimen.

2.2.1 *Origin and preparation of the rPET*

Coca Cola Western Europe kindly supplied us 150 kg empty PET bottles. About 90% large, empty 1.5 litre PET bottles that have not been used and about 10% small, empty PET bottles. The large bottles had rPET content of 50% and the small of 25%, so this mixture roughly had rPET content of 47.5 mass%. Additionally, it is this company's policy to use reheat additive in their bottles. There were no ink-jet prints on these bottles with batch-numbers or best-before dates. Therefore, the quality rPET with which we started was relatively rich in rPET and reheat additive.

Every single bottle was visually inspected. In case labels, hot-melt-residue or other contaminants were observed, these materials were completely cleaned. Labels, caps etc. were manually removed. Hot-melt residues were removed with white spirit and a hot wash with dish washing liquid, rinsed and dried in the full summer sun for several hours. In case a bottle was too contaminated it was rejected.

Roughly 140 kg of the cleaned PET bottles were milled with a WEIMA WLK04 shredder and a 2 cm sieve plate, yielding milled goods of 2-5 cm size. These milled goods proved too large for direct extrusion and hence this milled goods were additionally milled with a Wanner C17.26 shredder and a 0.8 cm sieve plate to sub-centimetre size milled goods, which could be extruded.

2.2.2 *Selection of the contaminants and concentrations*

Ten contaminants for the model studies were chosen by the industrial board, based on their industrial experience. The concentrations of the contaminants were chosen to reflect realistic levels. These were proposed by the researchers and subsequently amended by the industrial review board. The chosen contaminants and their concentrations are listed in Table 2. The justification of the chosen concentrations is written below.

Table 2: Type of contaminant chosen and their concentrations.

Contaminant	Origin	Concentration
PVC	Sorting fault	0.1%
Ink	Neck prints with batch numbers	300 prints cut from other bottles
Hot-melt residues	Integral part of PET bottles	0.33%
PE	Sorting fault	0.7%
PP	Labels as integral part of PET bottles	1.9%
EVOH	Barrier resin	0.1%
Nylon MXD-6	Barrier resin	0.1%
Amosorb	Oxygen scavenger	30 Amosorb containing bottles on 10 kg
PS	Shrink label as integral part of PET bottles	0.76%
PLA	Sorting fault	0.3%

PVC

The PVC concentration was set to be 0.1%, just like the concentration limit in specification DKR 328-1[16]. Pure granular PVC powder was chosen, hence, without additives. Type Kingfa ND95 NC005 was used.

Ink

The concentration for ink was set at 300 prints cut of other bottles and added to the 10 kg rPET to reflect the situation that all 300 bottles would have an inkjet print on their neck.

Hot-melt residue

The concentration was set at 0.33% since 10 kg bottles roughly contain 333 bottles with 0.1 gram of hot-melt per bottle. Type Paramelt Latyl E395 was used.

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[16] *http://www.gruener-punkt.de/en/download.html#c3353*

PE

The concentration was set at 0.7% since the DKR 328-1 specification allows for maximally 10% PET-trays which are maximally composed off 93% PET and 7% PE. A normal commercial grade LDPE granulates was used Sabic 2404NO.

PP

The PP concentration was set at 1.9%, roughly reflecting the bottle to label weight ratio of 50:1. Predominantly black Coca Cola Zero labels present from the original bottles, were cleaned and used.

EVOH

The concentration was set to 0.1% to reflect a sample with 10% barrier bottles containing 1% EVOH. Type EVAL F171B was used.

Nylon

Similarly to EVOH the concentration was set to 0.1%. MXD-6 Nylon of Mitsubishi was used.

Amosorb

The concentration was set to 30 bottles of which it is known that they contain Amosorb in 10 kg rPET.

PS

The concentration was set to 0.76 reflecting the bottle to shrink label ratio of 13:1 found on the Dutch market. Friesland-Campina was kind to supply us with 76 gram of blue-red coloured shrink label.

PLA

The concentration was set to 0.3%, reflecting the contamination of 10 kg of rPET with one tray of 30 grams of PLA. Type Ingeo NTR 2003D was used.

2.2.3 *Processing*

The rPET flakes were mixed with contaminants and extruded in granulates. These granulates were subjected to SSP treatment. These SSP-treated granulates were injection moulded into test specimen. Analysis was performed on the granulates and the test specimen.

Sample preparation

PET was cut to <8mm pieces using a Wanner C17.26 cutting mill provided with a 8mm sieve to allow homogeneous feeding and transportation, see paragraph 2.2.1. Contaminants as listed in Table 2 were cut to similar sizes as the recycled PET granulate and were added using dry blending.

Extrusion

Compounding extrusion was performed using a Berstorff ZE 25 (25 mm, 40D) co-rotating twin screw extruder with degassing unit melt filter and strain pelletizing. Processing conditions were discussed with Stefan Morssinkhof of Morssinkhof Plastics[17] in order to mimic current Dutch recycling processes. It was the mutual consensus that for this study the washed milled goods would not be dried before processing. Morssinkhof explained that they do not dry their flakes before processing, because they want to have a longer SSP process for the decontamination of the granulates. Other recycling processes dry the flakes before extruding in a single screw extruder. Still, vacuum degassing was used to prevent excessive degradation. The level of vacuum applied in the extruder was tuned in such a way that the intrinsic viscosity of the granulate was 0.64, as this was approximately the target level (0.65) recommended by Stefan Morssinkhof. To obtain short residence times a screw setup with 1 mixing zone (melt-lock) to allow vacuum degassing was used. A description of the screw and barrel configurations are given in Table 3 and shown in Figure 3. The extruded filament was water-cooled in a trough and pelletized by a rotating knife.

Processing characteristics were:

- Throughput: 10 kg/hr
- Speed: 300 rpm
- Pressure (measured): ~ 25 bar
- Vacuum: 0.2 bar
- Melt temperature (measured) : 265-267 °C.
- Melt filter: 50 micron

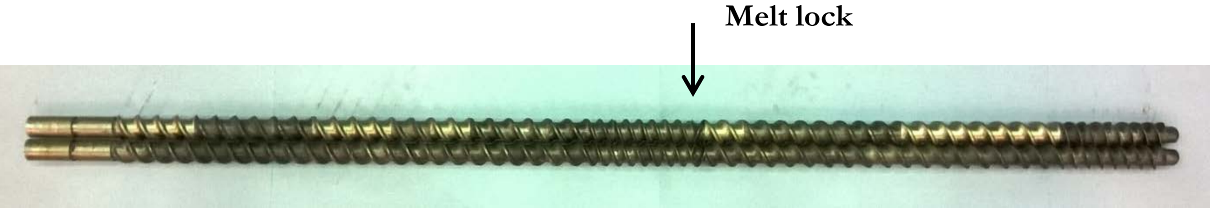

Figure 3: Picture of the screw layout used.

Table 3: Extruder configuration settings.

Screw configuration	Barrel configuration
10x transport large	Zone 1 & 2 Open atmospheric degassing
4x transport small	Zone 3 t/m 6 Closed
1x Melt lock	Zone 7 Vacuum degassing
10x transport large	Zone 8 t/m 10 closed
6x transport small	Zone 11 & 12 Melt filter

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[17] *Email-conversation with Stefan Morssinkhof, Wednesday 17 June 2015*

SSP treatment

2 kg batches of the eleven rPET samples (one reference rPET and 10 contaminated rPET samples) were subjected to solid state condensation treatment at Cumapol in Zevenaar. The process time was 8 hours, the pressure was 47.5 mbar and the temperature was 217°C. During this SSP treatment the rPET granulates were flushed with 0.04 ml nitrogen gas per minute.

Injection moulding

Roughly 1 kg of SSP-treated rPET granulates were injection moulded into different types of test specimen. One type of test specimen (bone shaped rods/tensile bars) was intended for DMTA and 50 x 50 x 2 mm plates were intended for Haze measurements. These test specimen were manufactured using a DEMAG D25NC IV injection moulding machine. Materials were processed at a cylinder temperature of 275°C and the mould temperature was set at 70°C. Prior to injection moulding materials were dried at 120°C for 4 hours using a desiccant dryer, to prevent hydrolysis.

2.2.4 *Analysis*

Various measurements were performed on the rPET granulates, the SSP-treated rPET granulates and the injection moulded test specimen. An overview is given in Table 4.

Table 4: Overview of the analysis performed on the intentionally contaminated rPET samples.

Substrates	Measurements
rPET granulate prior to SSP treatment	Colour (amorphous and crystallised)
	GPC
	IV
	DSC
	GC
rPET granulate after SSP treatment	Colour (crystallised)
	GPC
	IV
	GC
	Partisol
Test specimen	Haze
	DMTA

Colour

The colour measurements were performed with a Minolta Chromometer on a tray filled with loose rPET granulates. Measurements were performed in fivefold and averaged, yielding average values for L*, a* and b* and their standard deviations.

GPC

GPC measurements are performed to determine the molar mass and distribution using a Viscotech VE 2001 GPC max provided with a TDA305 Triple Detector Array (RALLS + LALLS, RI Detector and Viscometer). Columns used are a PSS PFG analytical linear M and guard column, molecular range ~ 250 – 2.5·10e^{6} D (PMMA in HFIP). Solvent was HFIP with 0.02M KTFA. All measurements were performed in duplicate.

IV

The intrinsic viscosity was determined at Cumapol with Dynisco IV meter named Kayeness LMI 4003. The granulate was dried at 160°C and about 5 mbar vacuum prior to the measurement. The IV was determined with 2.16 kg weight at 285°C.

GC

GC measurements were performed according to ASTM F2013-10 using a Agilent 3800 Varian GC provided with a Restek Rxi-5ms, 30m x 0.25mm x 0.25µm column. PET samples (finely milled PET pellets) were weighed into a 20-mL head-space vials, sealed, and then heated at 150°C for 60 min. After heating, the gas above the sealed sample of PET polymer was injected onto a capillary GC column. The acetaldehyde, benzene and limonene peaks were separated on the columns, and the gas concentration of these components was calculated in ppm [mg/kg]. Standards were used to determine the retention times and the response factors. The standards were measured prior to analyses in the same measurement line-up. In Figure 4 a typical gas chromatogram is shown.

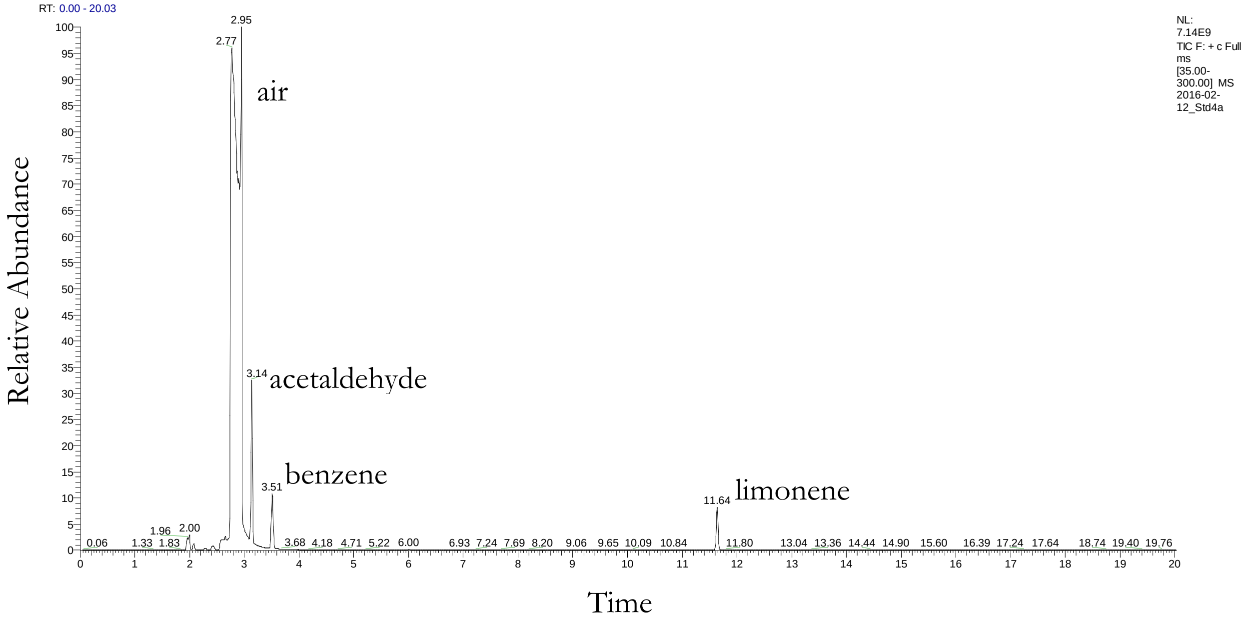

Figure 4: Volatile components as measured using head space gas chromatography. (Acetaldehyde at 3.14 min, benzene at 3.51 min and limonene at 11.64 min)

An additional check was performed with SIM mass spectroscopy to verify the identity of the peaks. Indeed the peak at r.t. 2.03 min. was identified as acetaldehyde with a mass of 44 g/mole and a straight-forward fragmentation pattern. The peak at r.t. 3.49 min. with a mass of 78 g/mole was identified as benzene with slightly more complex fragmentation pattern. The peak at r.t. 11.62 min. with a mass of 136 g/mole with a more elaborate fragmentation pattern was identified as limonene. The combined GC-SIM measurements increased the sensitivity and selectivity enormously.

DSC

The DSC analysis was performed at Dufor with a Netzsch DSC 200F3 240-20-0725-L under nitrogen atmosphere with about 5 mg PET granulate in Aluminium pans. The temperature program was composed of the first heating run from 40 to 280°C, a cooling run to 40°C and a second heating run to 280°C, all at 10°C/minute.

$$40^{\circ}C \xrightarrow[\text{First heating}]{10^{\circ}C/\text{min}} 280^{\circ}C \xrightarrow[\text{Cooling}]{10^{\circ}C/\text{min}} 40^{\circ}C \xrightarrow[\text{Second heating}]{10^{\circ}C/\text{min}} 280\,^{\circ}\mathrm{C}$$

Partisol

The Partisol measurements were performed by Emmtec. PET granulates were washed to remove attached dust, thermally treated to return them to the amorphous state and dissolved in hexafluoro-isopropanol at a concentration of 10 g/100 ml. These solutions were passed through an automatic particle counting cell. For every solution 10000 images were made and the amount of particles were counted. The threshold value for this analyses was set at 38. Additionally, these particles were classified in 5 size groups in µm; 1-3, 3-5, 5-10, 10-25 and 25-100. These figures can roughly be converted to amount of particles per gram by multiplying with a factor 250. However, this introduces a fairly large systematic error due to variations in density and viscosity, which is approximately 5 million particles per gram.

Haze

The Haze measurements were performed by PRI with an Hazeguard on the 5x5 cm large test specimen. This machine measures the amount of light that is scattered more than 2.5° from the incoming light-beam, according to ASTM D1003. Also dedicated Haze measurements were conducted with different light frequencies. The reported unit is the percentage of light that is scattered more than 2.5°.

DMTA

DMTA measurements were performed using Rheometrics RSAII solids analyser at a frequency of 1 Hz and a heating rate of 5°C/min. Measurements were performed in the tensile mode using samples of 30*5*1mm. E', E" and tan(δ) were plotted against the temperature to study thermal transitions.

2.3 Work package 3: To investigate the relation between the composition of PET bottle products, standardised mechanical recycling methods and the quality of rPET

Four different PET bottle products were subjected to two different sorting and mechanical recycling processes. Additionally, two PET bottle products were composed to reflect a hypothetical future situation in which the deposit refund system for PET would be abolished and the bottles would be recycled via separate collection and mechanical recovery. The latter PET bottle products were subjected to two different sorting and mechanical recycling processes. An overview of the PET bottle products and the three sorting and recycling processes to which they are subjected is given in Table 5.

The PET bottle products that are obtained from deposit refund systems (SRN, other) were only subjected to the standard mechanical recycling process, because these relatively clean bottle products don't require more elaborate sorting and recycling processes. Moreover, this process has been developed for this feedstock; PET bottles from deposit refund systems. This standard mechanical recycling process is described in paragraph 2.3.2 Standard process.

The PET bottle products originating from the current separate collection and mechanical recovery systems were subjected to both the standard and the advanced mechanical recycling process. This advanced recycling process had several additional process steps, such as NIR-pre-sorting, sieving with a 2 mm screen, flake sorting and colour sorting, see paragraph 2.3.3. This second, more advanced, laboratory process was configured to convert PET bottle fractions of separate collection and mechanical recovery origins into food grade bottle rPET as a mimic of industrial recycling processes in other countries like France.

For the hypothetical scenario, PET bottle products that would originate from separate collection and mechanical recovery systems with the PET bottles from the deposit refund system added have been subjected to the advanced recycling process and to a process in which the PET bottle products have been made to comply to DKR 325 (bottles only) prior to this advanced process.

The precise conditions and configuration was advised by the industrial reviewer L. Jetten and checked to current industrial practice with industrial review board members S. Morssinkhof and H. Kuipers. The conditions of the standard and advanced process are described in the following paragraphs, the exact conditions of washing per batch are shown in Appendix F.

Table 5: Matrix of measurements that will be performed on rPET products made from various input PET bottle fractions with three different mechanical recycling processes. The numbers are the codes used.

	Sorting and recycling process		
PET-bottle products	**Standard***	**Advanced***	**Advanced + DKR 325**
Deposit refund SRN, clear & light blue	1		
Deposit refund, other via Cumapol	2		
Separate collected and sorted fraction	3	4	
Mechanical recovered and sorted fraction	5	6	
Separate collected and sorted fraction when deposit refund bottles are added to the collection system for plastic packages		7	8
Mechanical recovered and sorted fraction when deposit refund bottles are added to the collection system for plastic packages		9	10

*The standard and advanced recycling process are explained in paragraph 2.3.3 and 2.3.4.

2.3.1 *Calculation of the composition of PET bottle products for future scenarios of collection system integration*

The composition of the PET bottle products after system amalgamation were predictively calculated. This calculation was based on the composition of the analysed PET bottle fractions and weighing factors that represent the amounts of PET bottles that the different systems are expected to contribute to the new merged collection system. These weighing factors, which are shown in Table 6, represent the weight of PET bottles that originate from the contributing systems, see Equation 5.

For the PET bottles and flasks that originate from the separate collection system the weighing factor equalled the amount of PET-bales produced at sorting facilities that comply to DKR 328-1. This amount was derived from the total amount of sorted products registered to originate from separate collection in 2014 (102 kton according to Stichting Afvalfonds) multiplied by the sorting division for PET (9.2% PET sorting yield based on outgoing weights)(Thoden van Velzen 2014).

For the PET bottles that originate from the deposit refund system SRN, the weighing factor was derived from the total amount of counted PET bottles derived from deposit refund systems in the Netherlands in 2014 (21 kton according to Stichting Afvalfonds) multiplied by the market share of SRN compared to other deposit refund systems (80% was chosen based on a two year old report (Warringa et al. 2014), a new report estimates this as 75% (Ansems et al. 2015)), multiplied by the fraction of Dutch households that is connected to a separate collection system for plastic packaging waste (roughly 90% in comparison to mechanical recovery), multiplied by the collection percentage for large PET bottles. The latter was crudely estimated to be 70%, based on the collection results of Fost-Plus in Belgium (FostPlus 2014), Redilo in Switzerland (2008) and own calculations on the current separate collection system (Thoden van Velzen 2015). Furthermore, it is in good agreement with one of the two assumptions used in the last environmental analysis of the framework treaty (Ansems et al. 2015)

Likewise the weighing factor for the bottles originating from the other deposit refund systems were calculated as 21 ktons times 20% times 90% times 70%. The applied weighing factors are listed in Table 6.

Table 6: Applied weighing factors to calculate compositions of sorted products after the deposit refund bottles have been added to the separate collection system and the mechanical recovery system.

	Weighing factors for bottles originating from, [kton]			
	Separate collection	**Mechanical recovery**	**SRN**	**Other deposit refund systems**
Merged separate collection system	9.38	-	10.60	2.63
Merged mechanical recovery system	-	1.74	1.51	0.38

The weighing factors for calculating the composition after adding the bottles from the deposit refund system to the mechanical recovery system are calculated in a similar manner, but with different parameters, see Table 6.

For the PET bottles and flasks that originate from the mechanical recovery system the weighing factor equalled the amount of PET-bales produced at sorting facilities that comply to DKR 328-1. This amount was derived from the total amount of sorted products registered to originate from mechanical recovery in 2014 (28 kton according to Stichting Afvalfonds) multiplied by the sorting division for PET (6.2% PET sorting yield based on outgoing weights)[18]. For the bottles that originate from the deposit refund systems and are added to the mechanical recovery system, the calculations are similar, but the percentage of Dutch households contributing to mechanical

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[18] *Bergsma, A. 2015. Personal communication*

recovery was set to 10% and the expected recovery yields for the large PET bottles were set to 90% (Thoden van Velzen et al. 2013).

Equation 5: concentration of a packaging category in a merged system is calculated from the concentrations in the contributing systems and weighing factors.

$$c_{merged\ system}^{small\ PET\ bottles} = \frac{W_{sep.coll.} \times c_{sep.coll.}^{small\ PET\ bottles} + W_{DRS1} \times c_{DRS1}^{small\ PET\ bottles} + W_{DRS2} \times c_{DRS2}^{small\ PET\ bottles}}{W_{sep.coll.} + W_{DRS1} + W_{DRS2}}$$

Parameter	Meaning	Unit
$c_{merged\ system}^{small\ PET\ bottles}$	Concentration of the category "small PET bottles" in a PET bottle product from a merged system.	[%]
$W_{sep.coll.}$	Weighing factor for separate collection; the amount of PET bottle product the original separate collection system contributes to the new merged system.	[kton]
$c_{sep.coll.}^{small\ PET\ bottles}$	Concentration of the category "small PET bottles" in the original separate collection system.	[%]

2.3.2 *Composition of the PET bottle feedstocks; mimicking pre-sorting*

The composition of PET bottle products from both deposit refund systems equalled the composition of the feedstocks 1 and 2 in the standard recycling process. For all other combinations of feedstocks and processes (3 - 10), different pre-sorting steps were required. The applied pre-sorting efficiencies are listed in Table 7. These efficiencies were mutually chosen to reflect the common experience of the industrial board. The numbers were verified and approved by the individual industrial partners. Only for the required pre-sorting of non-food flasks not a standard efficiency was chosen. It was the shared opinion of the industrial review group that although this sorting task is difficult, since it cannot be performed automatically and hence has currently to be done manually, the efficiency will be adjusted to achieve 5% of non-food flasks in the input feedstock.

Table 7: Applied pre-sorting efficiencies.

Type of sorting	Applied efficiency
Manual sorting of residual waste and other types of packaging	75%
Manual sorting of coloured bottles & opaque bottles	99%
Manual sorting of non-food flasks	Adjusted to fit < 5% in feedstock
De-metallisation with magnets and Eddy-current separators	99%
NIR machine sorting to remove non-PET objects	90%

For combination 3 (separately collected PET bottle product that is recycled with the standard process), a manual pre-sorting step was conducted to remove non-PET bottle objects with an estimated efficiency of 75%, metals were removed with a efficiency of 99% and coloured PET bottles, opaque PET bottles were removed with an estimated efficiency of 99%. The efficiency of manual pre-sorting of non-food PET flasks was set to 78%, since this efficiency would be necessary to achieve a feedstock with less than 5% of non-food flasks.

For combination 4 (separately collected PET bottle product that is recycled with an advanced process), a NIR pre-sorting step with 90% efficiency will remove most of the non-PET material, for non-bottle PET objects the manual pre-sorting efficiency remains 75% and all the other separation efficiencies are also equal to combination 3.

For combination 5 (mechanically recovered PET bottle product that is recycled with the standard process), almost all the pre-sorting steps are executed just like for combination 3. Only the efficiency in removing non-food-flasks had to be raised to 85.4%, since the concentration in the input is much larger.

For combination 6 (mechanically recovered PET bottle product that is recycled with an advanced process), a NIR pre-sorting step with 90% efficiency will remove most of the non-PET material, for non-bottle PET objects the manual pre-sorting efficiency remains 75% and all the other separation efficiencies are also equal to combination 5.

Combinations 7-10 dealt with PET bottle products from merged collection systems. The composition of PET bottle products made from merged collection systems were estimated with Equation 5 for each packaging category. The concentrations of each category were corrected to let the sum of the concentrations equal 100%. This calculation was executed for merged systems of separate collection and deposit refund systems and of mechanical recovery and deposit refund systems. These compositions of both PET bottle products of future collection systems after a potential merger were the starting point for the further calculations.
For feedstock 7, a similar pre-sorting was mimicked as for feedstock 4, only the required sorting efficiency for non-food-PET flasks was lowered to 35%, as the concentration of these flasks is reduced due to the merger with the large PET bottles from the deposit refund systems.
For feedstock 9, a similar pre-sorting was mimicked as for feedstock 6, only the required sorting efficiency for non-food-PET flasks was lowered to 57%, as the concentration of these flasks is reduced due to the merger with the large PET bottles from the deposit refund systems.

For feedstock 8 and 10, an additional sorting step was required to upgrade the PET bottle products from DKR 328-1 to 325 (bottles only). The composition of the feedstock material was calculated with equation 5 for merged systems (similar to feedstock 7 and 9). An additional manual sorting step (with 75% efficiency) was mimicked. The subsequent pre-sorting steps were similar to those of feedstock 7 and 9 with the exceptions that the efficiencies for the sorting of non-food PET flasks had to be 39% and 60% for feedstock 8 and 10, respectively.

2.3.3 *Standard process*

This process is meant to mimic the current industrial recycling practices in the Netherlands in which PET bottles from a deposit refund system are recycled to food grade bottle rPET. This process is adapted to laboratory conditions. Photos of the used lab-equipment can be found in Appendix C.

Step 1 Input quality check

This step is modelled by calculation, as described in paragraph 2.3.1 and 2.3.2. The average composition of the relevant PET bottle feedstocks were taken (result of task 1). The concentration of the two metal packaging categories were lowered in accordance with the previously agreed upon removal efficiency for metals (99% in Table 7). Also the concentration of the four types of residual waste (paper & board, organics & indefinable, glass, textiles) and all non-PET-bottle packages were lowered with previously agreed removal efficiencies (75% in Table 7). Finally, the concentrations were corrected by re-indexation (to let the sum equal 100%). This was the composition of the samples after the input quality check. In order to reconstitute these feedstock samples for the recycling process, bottles and objects that were previously sorted in task 1 in the categories were weighed to obtain in total 10 kg of feedstock. Obviously, the bottles and objects from the correct collection system were used for this feedstock reconstitution. The accuracy level was 1 gram for all categories contributing more than 5 gram and 10 mg for all categories contributing less than 5 gram.

Step 2 Rinsing/pre-wash bottles and flasks with water

The bottles and flasks are pre-washed with water of 50°C for 5 minutes in order to wash off the attached moisture and dirt. The 10 kg samples are washed in 2 runs of 5kg, as this is the capacity of the laboratory washing mill.

Step 3 Milling to 1-3 cm flakes

After pre-washing the bottles and flask, the wet bottles and flasks were milled with a WEIMA WLK04 mill with 2 cm screen. The bottles and flasks were pre-washed and milled wet to prevent dirt from being pushed into the PET material during the milling action. The milled flakes were about 1-3 cm.

Step 4 Centrifuging

The wet flakes were dried in a centrifuge in order to make the samples suitable for wind sifting. The flakes were dried as good as possible, until no water came out of the centrifuge.

Step 5 Wind sifting

The samples were wind sifted at the test set-up Busschers Haaksbergen. It is expected that the light fraction will predominantly contain labels, nylon layers, loose foils, paper, etc., and that the PET will be concentrated in the heavy fraction.

Step 6 Hot wash

The flakes were washed in a hot wash of 75°C, for 15 minutes with a alkaline solution of 0.1 M, in order to wash off product residues, hot melt and other attached dirt. The aim was to have the water above 75°C during the whole washing procedure, therefore the process was started with hot water of 80°C, which during the process cooled down with max. 5°C . The washing procedure was done in 2 runs of 4 kg, with 150 L water and a rotating stirrer. The temperature and conductivity of the water were monitored in the process. The floating plastics are scooped off the water after washing, as a first floatation separation.

Step 7 Rinsing with cold water

After washing the flakes were rinsed very well with cold water to rinse off all the alkaline solution from the flakes.

Step 8 Floatation separation

Floatation separation was used to remove polyolefins, such as caps and closures, and to concentrate the PET.. The flakes were divided in several buckets filled with water and stirred. The floating flakes were removed from the water.

Step 9 Centrifuging

The wet flakes were dried in a centrifuge.

Step 10 Thermal drying

After centrifuging the samples were dried in an oven overnight, at 60 °C.

Step 11 Fine milling

The dry flakes were milled fine in pieces of around 0.5 cm, with a Wanner mill equipped with a sieve plate of 0.8 cm.

Step 12 Extrusion, SSP and Injection moulding

See method work package 2, the method for extrusion, SSP and injection moulding is the same.

2.3.4 *Advanced process*

This process is meant to mimic the industrial recycling process in France in which PET bottles from separate collection systems are recycled to food grade bottle rPET.

Step 1 Input quality check

This step is similar to step 1 of the standard process, only the faulty sorted objects are removed by NIR machines with an estimated 90% efficiency and clearly visible contaminants by manual quality control. This step is modelled by calculation , as described in paragraph 2.3.1 and 2.3.2. The composition of the samples is done in the same way as the standard samples.

For the samples with modelled DKR 325 composition (third recycling process), this is modelled within the calculation of input material. These samples are further processed with the same advanced process.

Step 2 – 11 Standard process

These steps are similar to the standard process.

Step 12 Vibrating screen

The fine flakes are sieved by a vibrating screen of 2mm to remove the fines and to concentrate the PET (Dvorak et al. 2013).

Step 13 Flake sorting (material and colour)

Flake sorting (based on material) was conducted with a RTT Unisort NIR Flake sorter in the test laboratory of RTT in Zittau (D). The chosen sorting program was to allow only PET and to blow out all particles that are identified with NIR as different plastics and the non-identified particles. The 6 kg samples were sorted within 3-4 minutes. The weights of the NIR sorted product and the reject were determined. The qualities of both products were tested with a NIR flake analyser to render the concentration of PET particles.

Unfortunately, the flake colour sorter was not available at RTT Unisort during the visit. According to the director Bert Handschick, it could be mimicked fairly well with manual sorting, because the efficiencies are very high and the losses are low. Hence, the NIR-flake sorted samples were subjected to a manual colour sorting, to obtain a product of only transparent and light-blue PET particles.

Step 14 – 16 Extrusion, SSP, Injection moulding

These steps are similar to the standard process step 12-14.

2.3.5 *Technical measurements*

In order to assess the technical quality of the rPET produced with the 10 different combinations of feedstocks and recycling processes, a list of measurements were conducted on the different products made, see Table 8.

Table 8: Technical measurements conducted on the rPET products.

Substrates	**Measurements**
Washed PET flakes	Hexane extraction
rPET granulate prior to SSP treatment	Colour (amorphous and crystallised)
	GPC
	IV
	DSC
	GC
rPET granulate after SSP treatment	Colour (crystallised)
	GPC
	IV
	GC
	Partisol
Test specimen	Haze

These technical measurements were conducted in the same manner as in task 2, section 2.2 in this report. Although DMTA analysis was intended in the research plan, this analysis was not performed in task 3, since the results from task 2 proved that it did not have added-value. The only new measurement was the hexane extraction. This was performed with a soxhlet extraction set-up (capacity of about 150 gram PET flakes, 500 ml of pre-distilled n-hexane, overnight extraction, 20 h), with the intention to evaluated the amount of hot-melt residues that the washing procedure did not remove.

2.3.6 *Cross-calculation of material content of samples*

The material composition per packaging type for the PET bottles was determined in work package 1, which form the main part of the feedstock samples for task 3. The composition of the other packaging types (trays etc.) was estimated, or determined in previous research projects. The applied material composition per packaging type is listed in Appendix G.

The composition of the feedstock samples of task 3, before the modelled pre-sorting were a direct result of task 1 and were hence known (Appendix G). These compositions were described in terms of gross weights, including moisture and dirt. Based on the average moisture and dirt content of the sample's input, the net material weights were calculated.

The net material composition per feedstock sample were obtained by performing a matrix cross-calculation of the feedstock sample composition in terms of packaging types with the average material compositions per packaging type. These net material compositions of the 10 feedstock samples in task 3 were also used to determine the origin of the impurities. Two types of origin for impurities were discerned:1) impurities from components of the PET bottle or flasks and that are hence integral parts of the bottle / flask design and 2) impurities from sorting faults. So for each impurity the net composition in the feedstock was calculated and this was expressed as percentages. Moreover for each impurity also a distribution of the origin (sorting faults, design component) was calculated and this was also expressed in terms of percentages.

2.3.7 *Calculation of the mechanical recycling yields*

The yields per process step were determined by dividing the material weight after the process step with the material weight before the process step. The complete chain yield was determined by multiplying these yields, see Equation 6.

The net recovery of mass (R_m) was obtained from the total net material weights after and before the process as shown in equation 6. The net PET yield (η^{PET}_{net}) was obtained similarly using the net PET weights in this equation.

Equation 6: Calculation method of mechanical recycling yields

$$Chain\ Yield = \frac{W_{After\ process\ 1}}{W_{Before\ process\ 1}} * \frac{W_{After\ process\ 2}}{W_{Before\ process\ 2}} * \ldots * \frac{W_{After\ process\ n}}{W_{Before\ process\ n}}$$

Parameter	**Meaning**	**Unit**
$W_{After\ process\ n}$	Weight after process step, expressed in net material (based on all material in sample), and expressed in net PET (based on PET content in sample)	[g]
$W_{Before\ process\ n}$	Weight before process step, expressed in net material (based on all material in sample), and expressed in net PET (based on PET content in sample)	[g]

For each process step the weights required to calculate the process yield were obtained in a slightly different manner. This is explained in the text below.

The mechanical recycling process was divided in several process steps of which the independent yields were determined separately:

- Pre-sorting
- Pre-wash and wet-milling
- Wind sifting
- Hot wash & floatation separation

- Sieving with a 2 mm vibration screen
- Flake sorting - material
- Flake sorting - colour

Both the recovery of mass and the net PET yields were determined based on net material weights. In the industrial practice material yields are mostly determined by dividing the output weight by the gross input weight. In the laboratory situation the samples were relatively small and hence small losses have much more impact than in the industrial situation. Therefore the industrial yields are likely to be larger than those measured in the laboratory. It is important to regard the results of these tests as indications and use the results in a comparative manner. The results cannot be compared to the industrial practice in quantitative terms.

For each process step three parameters were determined:

- Net material yield (R_m, net recovery of mass)
- PET concentration in the sample
- Net PET yield

Pre-sorting

The pre-sorting yields were determined from on the material compositions of the samples before and after pre-sorting (calculated as described in paragraph 2.3.2). Based on the material compositions the net material weight of both before and after pre-sorting was determined, as well as the net PET concentration and the net PET weight.

Pre-wash and wet-milling

The net material weight before processing was determined by Equation 7:

Equation 7: Net material before pre-wash

$$W_{Before\ pre-wash} = W_{input\ sample\ (gross)} * (100\% - \text{Average moisture and dirt content of sample origin})$$

The net material weight after wet-milling was determined by subtracting the added moisture (added weight) and the weight of the removed dirt of this process step. The weight of the dirt was determined by sampling some of the output flakes and measuring the dirt content.

The net PET weight before processing was determined by multiplying the net material weight with the PET concentration in the sample. The PET concentration was previously established in the cross-calculation explained in paragraph 2.3.6. After processing, the net PET weight was also determined by multiplying the net material weight with the PET concentration. The new PET concentration, after the pre-wash and wet milling, was determined by dividing the net PET weight before processing (no PET losses, so the same as after processing) by the net material weight after processing. The net material weight after processing was the net material weight minus the sludge that was removed during this washing step.

After wet-milling the material was mechanically dried with a basket centrifuge. No material was lost in this process, only water is separated from the samples. Thus no net material yields had to be calculated.

Wind sifting

After wind sifting the PET concentrations in the heavy and light fraction were determined, as well as the gross weights of the fractions produced. The gross material weight of the input material was obtained by adding up the gross material weights of the light and heavy fractions. The net recovery of mass was estimated by dividing the gross weight of the heavy fraction by the gross input weight. As the moisture and dirt content in these samples is unknown this will give the best estimate of the net material yield, assuming the percentages of attached moisture and dirt are similar for both products.

Hot wash & floatation separation

This process step was conducted in two batches per sample. A sample of the input material was taken to determine the moisture and dirt content of the input material. The input material was weighed before washing. Based on this information the net material weight was determined in the same way as described in Equation 7. Also the net PET weight could be determined, using the PET concentration determined for the heavy fraction in the previous processing step.

After floatation separation the samples were dried in an oven. The weights of the sinking fraction and the floating fraction were determined. As the washing process was very extensive, the assumption was made that these weights represent the net material weights, and thus no attached moisture and dirt is present on the samples. The PET concentration of the floating fraction was measured, by sorting analyses. The PET weight of the sinking and floating fraction together is determined by Equation 8. The net material weight was multiplied by the PET concentration of the input sample, which provided the net PET weight of the input. The weight of the coarse sludge (which mainly consists of fine PET flakes) was subtracted. The net PET weight in the sinking fraction was determined by subtracting the measured weight in the floating fraction from the net PET weight of the combined fractions.

Equation 8: Weight of net PET material in sinking + floating fraction

$$W_{net\ PET\ (Sinking+Floating)} = (W_{Floating} + W_{Sinking}) * C_{PET,\ after\ windsifting} - W_{coarse\ sludge}$$

After this process step the material is milled fine. There is no material lost in this process, thus this yield is not taken into account.

Vibrating screens

The input weight was measured, and this represents the net material weight. With the PET concentration of the previous process step, the net PET weight was determined. After sieving the material, the weights of the fine and the coarse fractions were determined. The PET concentration in the input material of the next process step is known (described in the next paragraph) and used to calculate the net PET weight in the fine fraction.

Flake sorting – material

After flake sorting the weight of the product and rejects are weighted. Together these weights represent the input weight. The PET concentration in the product and the reject fractions were determined by automated flake sorting with the Unisort NIR-flakesorter. This analysis yields a detailed composition for all plastics. However, a small percentage of flakes reflect the light too strong and are registered as unknown flakes, while they are just PET flakes, so the determined PET concentrations are slightly underestimated. The PET concentration in the input sample was derived from the PET concentrations in the product and the reject and their relative weights.

Flake sorting – colour

This sorting step was mimicked by manual sorting, and was done very precisely. The light blue PET flakes were allocated to the transparent fraction, and thus not sorted out. No losses occurred, only the coloured flakes were removed from of the sample. The weights of the product and reject were measured. The assumption was made that the PET concentration does not alter due to this sorting step, which was likely since this material was already sorted with a NIR flake sorter and should hence only contain PET flakes. Therefore, it was assumed that the PET concentration after the colour sorting process equalled the PET concentration before the colour sorting process and was hence equal to the PET concentration determined after the NIR flake sorting step. With these PET concentrations, the net PET weights were determined. In practice this yield will be lower, since machine sorting will always create some losses.

3 Results

The results of the three work packages are presented in the three following paragraphs.

3.1 Results of work-package 1

3.1.1 *Composition of PET packages per category*

The material composition of seven different PET bottle and flask categories were studied by disassembly and NIR and IR identification of the individual components. These compositions were averaged per category. For each category it was attempted to collect 25 individual different bottles. Unfortunately, this was not possible for the opaque and coloured bottles, since these are hardly present on the Dutch market. The amount of analysed bottles is listed in Figure 5. These averaged material compositions are shown in Figure 6. In Figure 7 some typical PET packages are shown to give an impression of the material composition of the packages. In Appendix D the detailed composition per category including standard deviations is given.

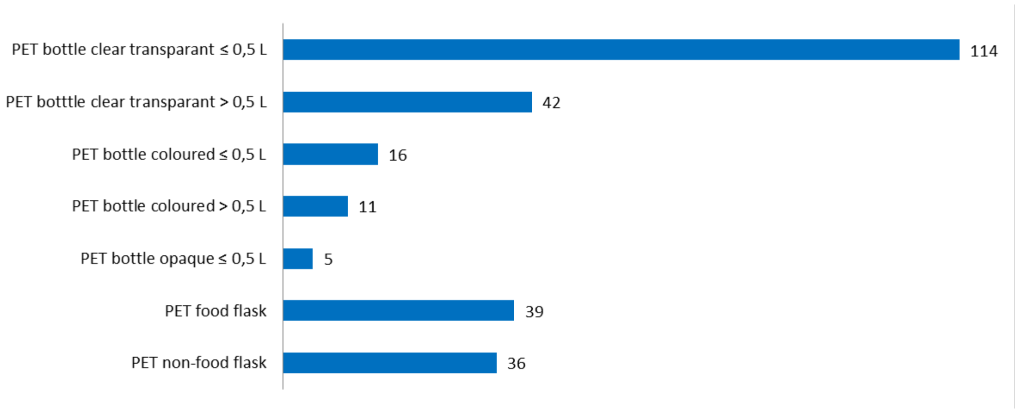


Figure 5: Amount of bottles and flasks studied per category.

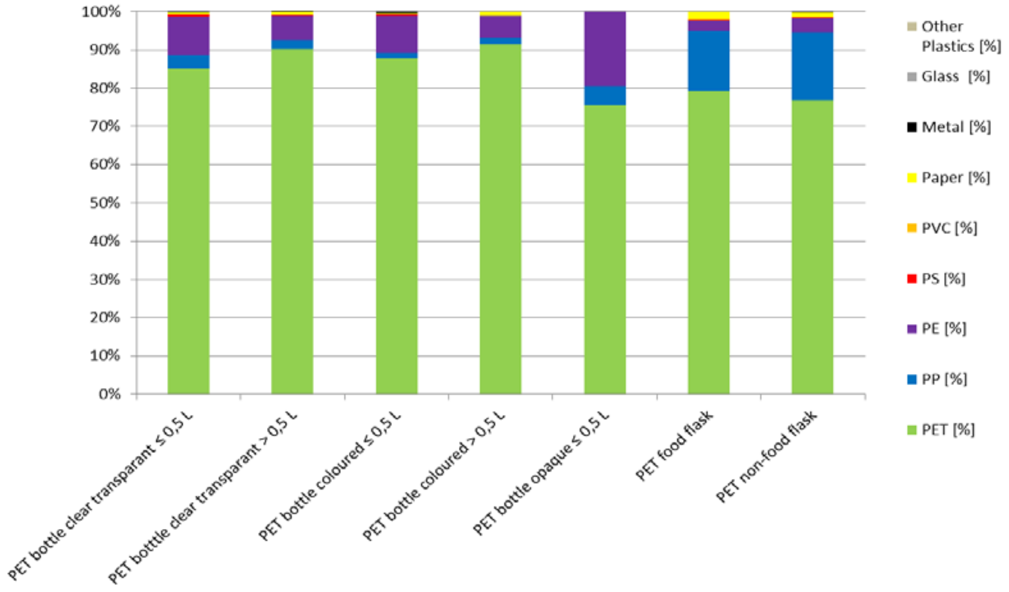
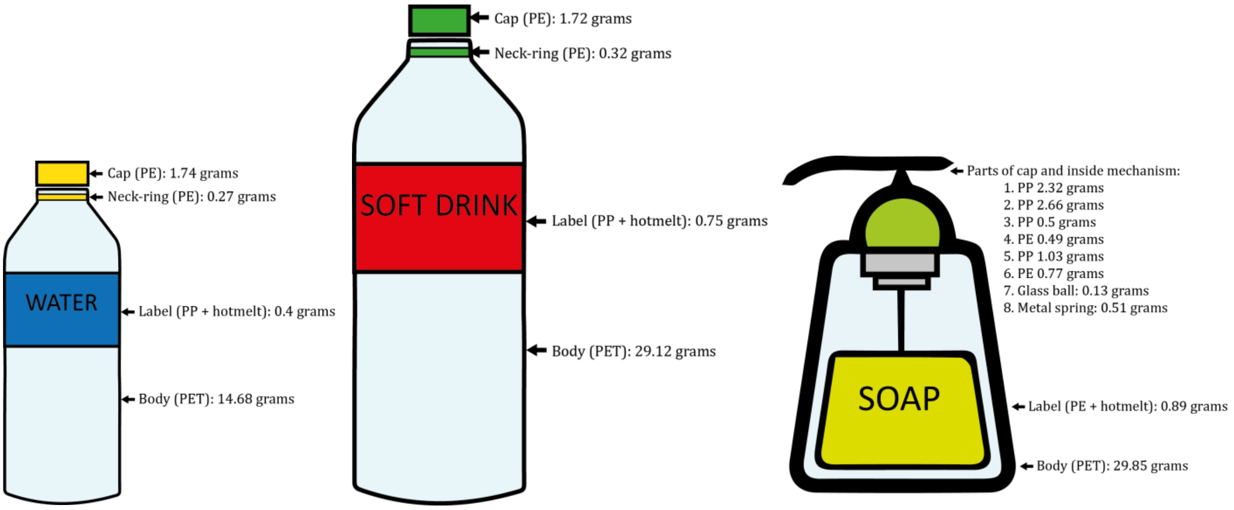


Figure 6: Average material composition of the PET bottle and flask categories.



a.) **b.)** **c.)**

Figure 7: The material composition of some typical PET bottles and flask. a.) typical transparent PET bottle 0.5 L. b.) typical transparent PET bottle 1.5 L c.) typical non-food soap flask with pump mechanism

Most PET bottles and flasks on the Dutch market are composed of PET bodies, PP and paper based labels, PP and PE based caps and closures. No PVC components were found. PS was found on a limited amount of small bottles as labels and in one non-food-flask for shower gel as a decorative over-lid.

Specialty plastics with a density larger than 1 g/ml (PA, ABS, POM, Silicone rubber) were mostly found in flasks. 8 out of 31 Food flasks were found to contain silicone rubber valves. And 2 out of 36 non-food flasks were found to contain POM based plungers in spray guns and one flask contained a silicone rubber valve. Only one small PET bottle with PA label was found.
Only a small number of bottles were found to contain metal and glass parts. 4 out of 114 small transparent PET bottles had aluminium caps under the lid (for juices) and aluminium screw caps (small PET wine bottles). In the category non-food flasks 6 out of 36 flasks had metal parts. These were spray guns and hand pumps with small metal balls and metal springs. In the same category 5 out of 36 flasks were found to contain small glass balls, also in spray guns and hand pumps. The details of the materials found in the 7 PET bottle categories are listed in Appendix E.

3.1.2 *Identification of barrier bottles and flasks present on the Dutch market*

The barrier bottle identification test was evaluated with several bottles of which the producers or other stakeholders had stated that they did contain a barrier function and several bottles of which the producers had stated that they did not contain any barrier, see Table 9.

Table 9: The results of the barrier bottle identification test for two PET bottles known to be free of barrier functions and two bottles / flasks that do contain a barrier function.

PET bottle / flasks	$\overline{x}_{initial}$	$\overline{b}^{*}_{initial}$	$\overline{x}_{final}$	$\overline{b}^{*}_{final}$	$\Delta\overline{b}^{*}_{norm.}$
Bottles that do NOT contain any barrier function					
Coca Cola light 500 ml	254 ± 4	2.0 ± 0.1	510 ± 4	7.4 ± 0.1	0.65 ± 0.05
Coca Cola Zero 1.5 ltr	288 ± 2	2.0 ± 0.3	609 ± 12	6.9 ± 0.6	0.44 ± 0.14
Bottles known to contain a barrier function					
Apple juice 250 ml	269 ± 36	2.2 ± 0.3	491 ± 9	32 ± 1	5.6 ± 0.3
Tomato ketchup 220 ml	471 ± 29	2.6 ± 0.2	598 ± 13	25 ± 3	3.6 ± 0.5

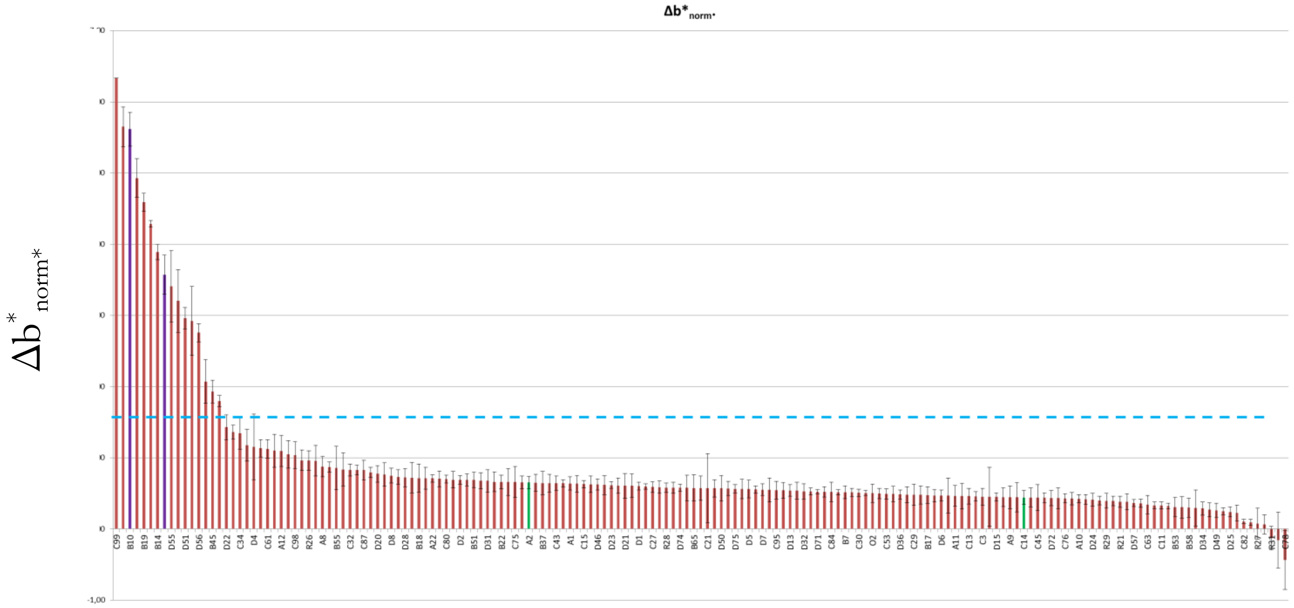


Figure 8: The results of the barrier bottle identification test for 175 different bottles of the Dutch market. Displayed are the normalised differences in b* colour value. The purple bars are for known barrier bottles, the green bars for known non-barrier bottles. The blue dashed line is the threshold value of 1.6.

In total 175 different PET bottles and flasks were tested. The result is shown in Figure 8. Based on the data of the reference bottles (Table 9), visual inspection of the heat treated PET cuttings (what is clearly more brown to the normal eye), visual delamination and packaging technological knowledge (barrier functions are potentially useful for beer, wine, sauces, juices), the threshold value was set to 1.6. The value of 1.6 was chosen as there was a clear gap between the barrier bottles and the non-barrier bottles. The lowest value found for a barrier bottle was 1.8 (orange juice), the highest value found for a non-barrier bottle was 1.4 (ice tea). Hence in case the normalised difference in b* colour value exceeded 1.6, the bottle was identified as barrier bottle. In some cases (as with PET wine bottles) the PET cuttings did not only discolour, but they also clearly delaminated, which is a sign that the packaging has a barrier. The PET bottles identified as barrier bottles present on the Dutch market are listed below in Table 10.

Table 10: the anonymised list of barrier PET bottles present on the Dutch market.

All PET wine & beer bottles
Brand X, 330 ml, Multivitamin juice
Brand X, all types of sauces and ketchups, in contrast to all the other brands
Brand X, 1.5 ltr, Cranberry juice
Brand X, 330 ml, Apple juice
Brand X, 330 ml, Orange juice
Brand X, 400 ml, four different types of juice

The barrier bottle test does not allow to test for barrier functionalities in coloured PET bottles, since the colorant itself discolours as well during the heat treatment. Only in case of delamination (as happened with green PET wine bottles and brown beer bottles) a barrier bottle can be identified with this test. The PET bottles and flasks used to determine the material composition (paragraph 3.1.1) were mostly also tested for barrier functionality, see Figure 9.

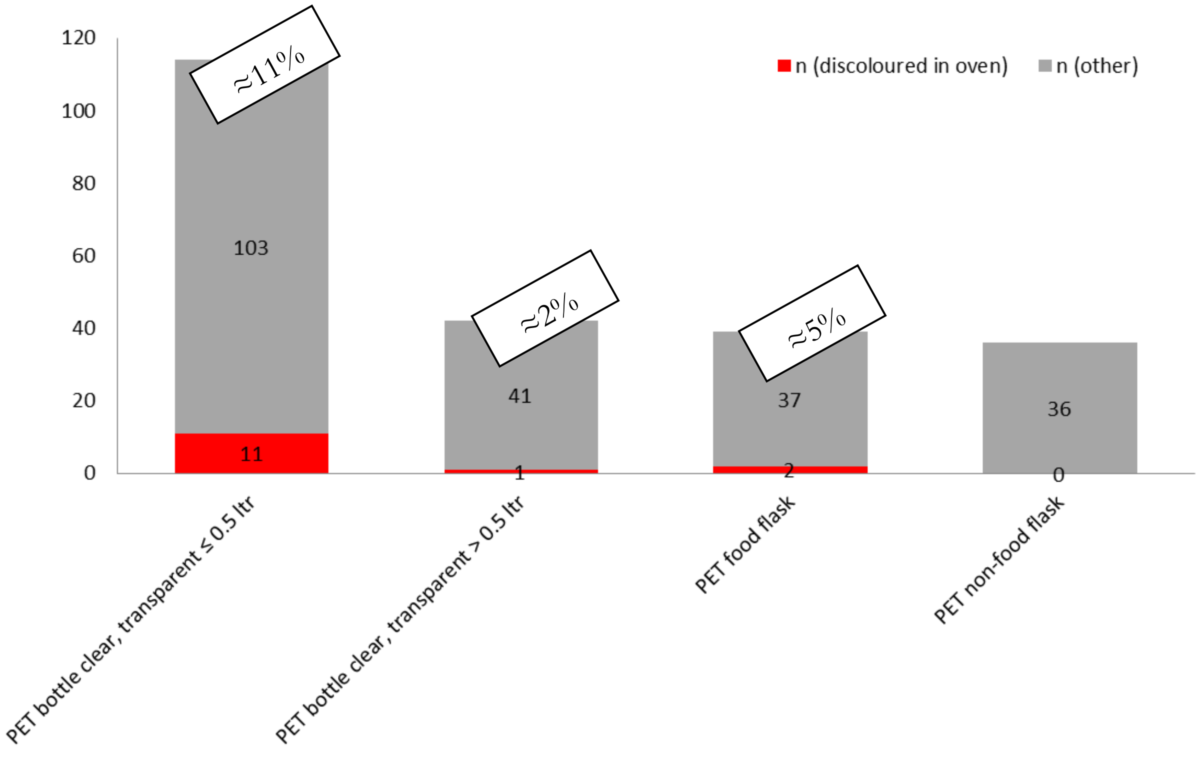


Figure 9: The division of barrier PET bottles over the PET bottle categories. n(other) indicating both the amount of tested bottles that were found not to contain a barrier and non-tested bottles, that could not have a barrier based on the type product they contained (for instance water). Percentages refer to total amount of barrier bottles in the categories, expressed in percentage.

Most of the barrier PET bottles were found in the category of small transparent PET bottles, only one was found in the large transparent PET bottle category and 2 were found in the PET food flask category. This list of barrier bottles present on the Dutch market was used by the sorting assistants to establish the composition of the sorted PET products. During the sorting of the PET bottle products a few green and brown PET bottles and green and transparent wine bottles were found. Tests revealed that these were also barrier bottles and hence they were also classified in the barrier bottle category.

3.1.3 *Composition of sorted PET products*

The material composition of the sorted PET products is shown in Figure 10 with respect to the main sorting categories.

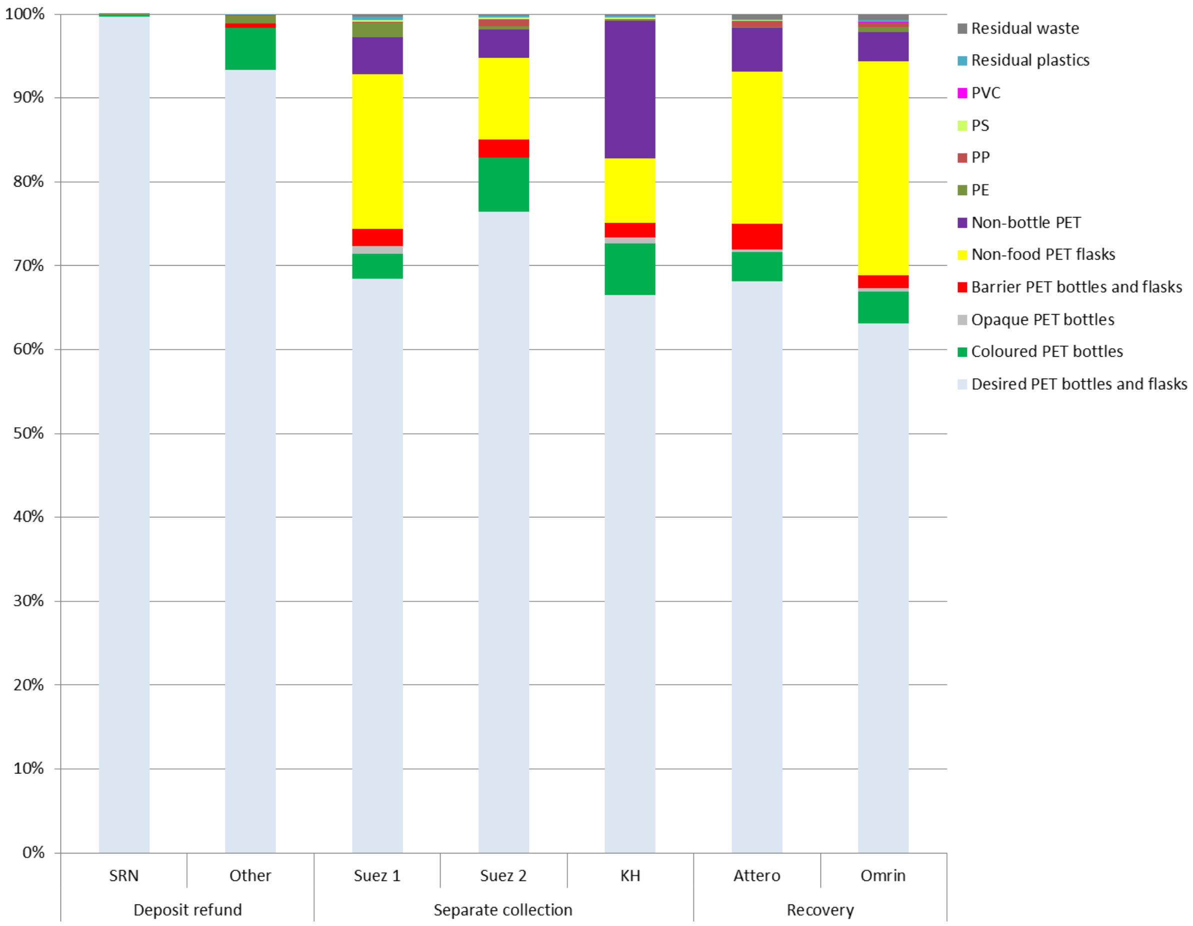


Figure 10: The material composition of the sorted PET bottle products.

The material compositions were weight-averaged for the three main origins of sorted PET bottle products (deposit refund systems, separate collection and mechanical recovery), these averages are listed in Table 11.

Table 11: Weight-averaged compositions of sorted PET bottle products different origins.

Category	Deposit-refund	Separate collection	Mechanical recovery
Desired PET bottles and flasks	96.5 ± 4.5%	69 ± 5%	64 ± 4%
Coloured non-barrier PET bottles	3 ± 3%	4 ± 3%	3.7 ± 0.3%
Opaque bottles	0 ± 0%	0.9 ± 0.1%	0.5 ± 0.9%
Barrier PET bottles and flasks	0.3 ± 0.4%	2.1 ± 0.3%	1.9 ± 1.2%
Non-food PET flasks	0 ± 0%	17 ± 8%	24 ± 6%
Non-bottle PET	0 ± 0%	5 ± 8%	3.9 ± 1.4%
PE	0.5 ± 0.6%	1.5 ± 1.3%	0.5 ± 0.4%
PP	0.1 ± 0.1%	0.2 ± 0.4%	0.6 ± 0.1%
PS	0 ± 0%	0.19 ± 0.02%	0.04 ± 0.06%
PVC	0 ± 0%	0.02 ± 0.01%	0.01 ± 0.01%
Residual plastics	0 ± 0%	0.4 ± 0.1%	0.2 ± 0.1%
Residual waste	0 ± 0%	0.3 ± 0.1%	0.7 ± 0.1%

The levels of attached moisture and dirt were determined for the PET bottle categories of the seven samples. These parameters were weight-averaged with respect to the material composition of all the samples and subsequently weight-averaged to overall parameters for PET bottles and flasks from the three main types of origin, see Table 12.

Table 12: The percentage of attached moisture and dirt (PAMD) for each sample and the main types of origin.

PET bottle feedstock	PAMD per sorted product	PAMD per main type of origin
SRN	9.3 ± 0.2%	Deposit refund 7 ± 2%
Other	5.8 ± 0.3%	
Suez 1	6.3 ± 0.4%	Separate collection 7 ± 3%
Suez 2	11.8 ± 0.3%	
KH	9.8 ± 0.4%	
Omrin 1	13.7 ± 0.3%	Mechanical recovery 13 ± 4%
Omrin 2	10.8 ± 0.3%	
Attero	18.0 ± 0.3%	

3.1.4 *Net PET composition of sorted PET products*

The net PET material content was calculated from the average composition for PET bottle products from different origin in terms of packaging categories (Figure 10), the average material composition per packaging category (Figure 6 and Appendix D) and the average percentages of attached moisture and dirt (Table 12). This matrix calculation was carried out for three groups of PET bottle categories;

1. All the PET bottles and flasks
2. All the PET food bottles and flasks
3. All the transparent and light-blue PET food bottles and flasks.

The results of these matrix calculations are shown in Figure 11.

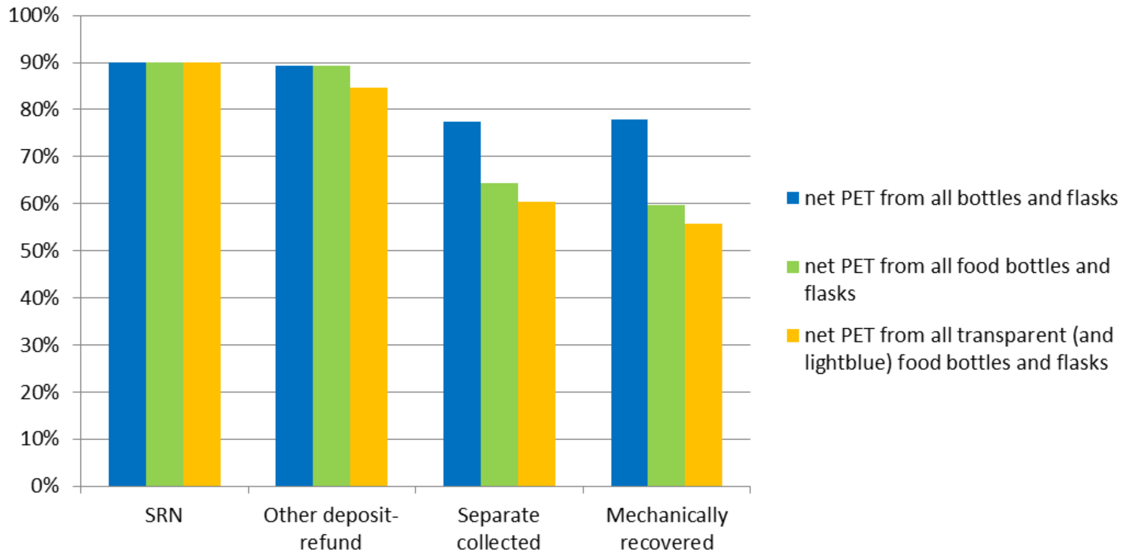


Figure 11: The net PET material content of sorted PET products originating from three different collection systems in [% w dm/w dm].

Hence, the net PET material content of PET bottle products from deposit refund systems is roughly 90%. For sorted PET bottle products originating from separate collection this net PET material content drops to 60-65% depending on what cross section of bottles is relevant. For sorted PET bottle products originating from mechanical recovery this net PET material content is even lower 55-60%. The main reasons for the lower net PET material content of PET bottle products from the latter two origins compared to those of deposit refund systems is the relative high concentration of non-food flasks.

The industry is used to net contents that are calculated by comparing the net PET product weight divided by the gross feedstock weight. To provide a net PET/gross feedstock weight material content, the net PET content from Figure 11 is corrected with 100% minus the percentage of attached moisture and dirt and are shown in Figure 12. The net/net percentages are used in the remainder of the report.

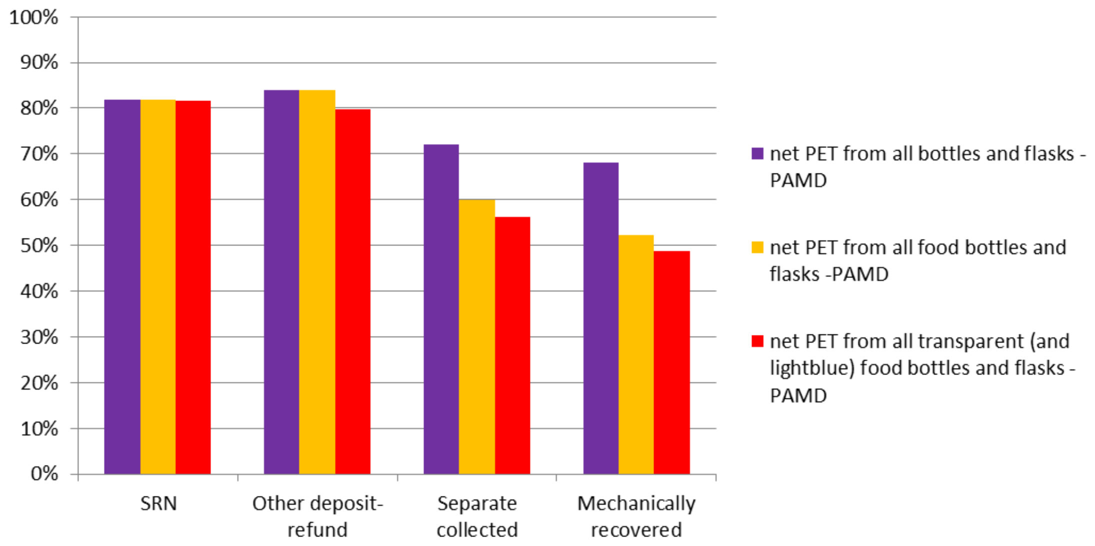


Figure 12: The net PET material content of sorted PET products originating from three different collection systems in [% w dm/w gross].

3.1.5 *Conclusions work package 1*

The material composition of Dutch PET bottle products have been determined in detail. PET bottle products originating from deposit refund systems almost exclusively contain desired PET bottles that comply to the EPBP design guidelines. The net PET content of these products is roughly 80% (see Figure 12). The other 20% are almost completely identified as bottle components like labels, caps, closures and attached moisture and dirt. The two analysed PET bottle products are slightly different. The SRN product contained no barrier bottles and 0.2% coloured bottles. Whereas in the PET bottle product from the other deposit refund system about 0.6% barrier bottles and 4.9% coloured bottles were found.

The PET bottle products that originate from the separate collection and mechanical recovery systems are much more complex. These products contain 17 to 24% PET non-food flasks, which is a serious obstacle for food-grade bottle-to-bottle recycling. Furthermore these products also contain less than 10% non-bottle PET products, mainly PET trays. These objects also need to be removed to allow for a sufficient high quality of rPET that could be used in bottle applications for food. But on top of those large contaminants these PET bottle products also contain a large amount of contaminants with smaller concentrations, such as barrier bottles (~2%), opaque bottles (~1%) and packaging objects from different types of plastics. Additionally, the material compositional analysis of small PET bottles and PET food flasks, revealed the presence of undesired materials (glass, metals) and plastics (POM, silicone rubber, ABS, PA) that can infringe recycling. Most undesired components were found to be present in the category non-food flasks and especially in the spray gun and hand pump parts.

3.2 Results of work-package 2

3.2.1 *Introduction*

Table 13 gives an overview of the deliberately contaminated rPET samples made with the type and concentration of contaminants that were added (see Section 2.2).

Table 13: Overview of the samples made in task 2.

Sample	Contaminant	Concentration	Application form/source
0, rPET	None		Reference rPET
1, ink	Ink labels	300 prints	Cuttings from the neck of PET bottles
2, Amosorb	Amosorb	30 bottles per 10 kg	Barrier bottles
3, PS	PS labels	0.76%	Printed PS labels, Friesland-Campina
4, PP	PP labels	1.9%	Printed PP labels, Coca Cola
5, PLA	PLA	0.3%	PLA tray, Ingeo NTR 2003D
6, EVOH	EVOH	0.1%	Virgin granulate, Eval F171B
7, PE	LDPE	0.7%	Virgin LDPE granulate, Sabic 2404NO
8, Nylon	Nylon (MXD6)	0.1%	Virgin granulate, Mitsubishi
9, Hotmelt	Hot-melt	0.33%	Virgin hot-melt, Paramelt Latyl E395
10, PVC	PVC	0.1%	Virgin PVC granulate, Kingfa ND95 NC005

3.2.2 *Extrusion*

All material combinations of rPET with contaminants could be processed into granulates. These granulates were used for analyses (GC, partisol, colour), SSP and subsequent injection moulding. The recycled PET has a greyish colour and some of the contaminants cause a further colour change in the granulate. Addition of the hotmelt and PVC leads to yellowing, addition of LDPE labels results in hazy granulates and addition of Amosorb and PP results in greenish and purple granulates. PP and LDPE gave some pressure fluctuations during extrusion and addition of PVC resulted in an increase in pressure to 100 bars, see Figure 13. In Figure 14 a photo of the granulates is included.

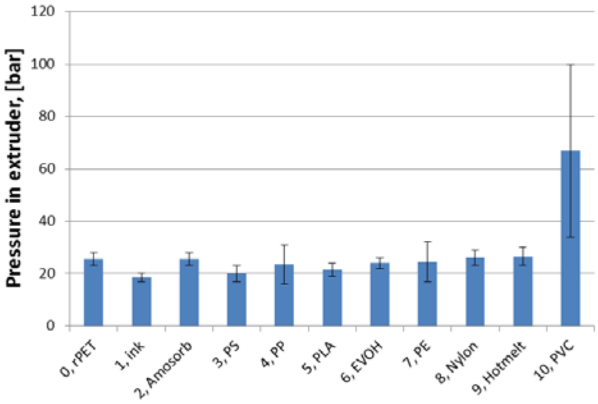


Figure 13: The pressure in the extruder during processing the samples for task 2 [bar].

Figure 14: Samples of the recycled PET granulates. Variations in the greyish colour caused by contaminants. From left to right: 0 rPET, 1 ink, 2 Amosorb, 3 PS, 4 PP, 5 PLA, 6 EVOH, 7 PE, 8 Nylon, 9 Hotmelt, 10 PVC.

3.2.3 *Partisol*

The total amount of particles counted per 10,000 images and the estimated amount of particles per gram are listed in Table 14. The conversion from amount of particles into particles per gram involves several assumptions regarding viscosity and density that creates an estimated standard error of 5 million particles per gram.

Table 14: The main results of the Partisol measurements on the samples made in task 2.

Code	Nature of the solution	Total particle count per 10000 images	Estimated amount of particles per gram, [millions per gram]
0, rPET	Clear	65,225	16 ± 5
1, ink	Clear	40,373	10 ± 5
2, Amosorb	Clear	27,722	7 ± 5
3, PS	White	39,880	10 ± 5
4, PP	Dark grey – white	772,117	193 ± 5
5, PLA	Clear	27,182	7 ± 5
6, EVOH	Hazy	32,986	8 ± 5
7, PE	White	193,094	48 ± 5
8, Nylon	Clear	34,187	9 ± 5
9, Hotmelt	Hazy	54,435	14 ± 5
10, PVC	Clear	78,606	20 ± 5

These results show that the amount of particles present in the reference rPET sample is already fairly high. According to Emmtec normal levels for Partisol counts of rPET are approximately 15,000 particles per 10,000 images and a virgin PET has a count of about 370 particles/ 10,000 images. This high particle count for the rPET reference (code 0) probably relates to the origin of this reference sample, which was made from clean bottles with about 47.5% rPET and reheat additive. Most of the rPET samples to which contaminants were added have similar levels of particle contamination, due to the fact the concentration of these contaminations is relatively low. Furthermore, in some cases the contaminants will dissolve in the solvent and hence will not be visible as particles (ink, Amosorb, nylon, EVOH, PLA). Additionally the carbon black reheat additive present in the rPET could possibly coagulate with impurities in recycled plastics, so that the total count doesn't increase (Luijsterburg 2015). The contaminants that show high levels of particle contamination are the samples to which PE and PP have been added. These contaminants do not dissolve in the solvent and are hence visible as contaminants. Moreover, their concentration was chosen relatively high.

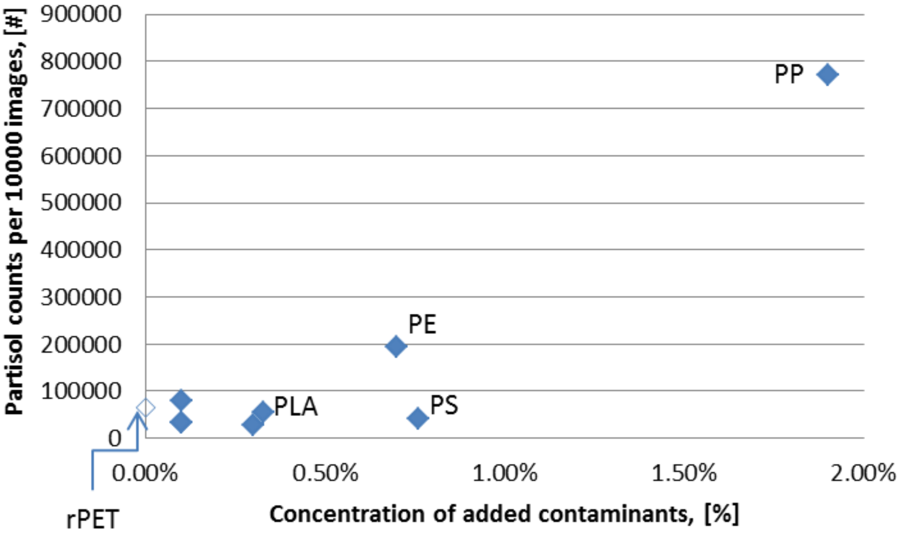


Figure 15: Relationship between the concentration of added contaminants and Particle count.

Figure 15 shows the relationship between the concentration of the deliberately added contaminants and the amount of particles counted in 10000 images, for the eight contaminants to which a concentration could be assigned. Hence only for PP and PE a clear increase in the particle contamination could be observed.

Figure 16 shows the normalised particle size distributions of the deliberately added contaminants. For normal rPET most contaminants present (~70%) are smaller than 3 µm, 20% smaller than 3 -5 µm, and only a small fraction is larger. This only significantly changes for PE and PP. The addition of these contaminants results in even more small particles. Both contaminants also produced most pressure fluctuations during extrusion, besides PVC. Which is likely caused by the passing of this molten dispersion through the melt filter. The high degree of small particles for PE and PP is caused by the fact that these contaminants produce a blend of small particles and that the concentration of these contaminants is the highest of all.

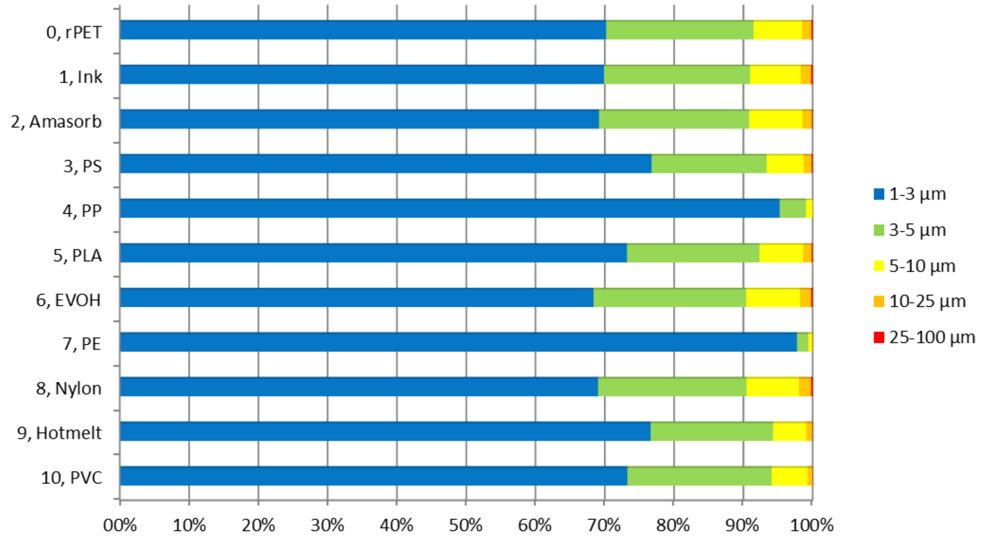


Figure 16: Normalised particle size distributions.

3.2.4 *Colour of the granulates*

Table 15: The CIELAB colour values of the rPET granulates with deliberately added contaminants in the amorphous and crystallised state prior to SSP and crystallised after SSP. L* indicating the lightness (black / white), a* indicating colouring along the red / green axis, b* indicating colouring along the yellow / blue axis. Clearly deviating results are coloured.

Code	Amorphous prior to SSP			Crystallised prior to SSP			Crystallised after SSP		
	L*	a*	b*	L*	a*	b*	L*	a*	b*
0, rPET	40.4±0.5	0.88±0.03	2.33±0.03	49.9 ± 0.8	-0.02±0.04	-0.4±0.4	63.1±0.2	-0.63±0.01	1.17±0.03
1, ink	41.0±0.3	0.57±0.02	1.71±0.07	50.2±0.6	-0.43±0.02	-1.3±0.2	62.2±0.7	-1.28±0.04	0.45±0.16
2, Amosorb	41.3±0.9	0.31±0.02	2.82±0.13	52.5±0.5	-0.82±0.01	0.34±0.14	63.3±0.4	-1.11±0.04	2.47±0.02
3, PS	43.7±0.1	-0.71±0.01	1.69±0.07	51.3±0.2	-1.40±0.01	0.0±0.2	62.1±1.0	-2.74±0.08	2.21±0.03
4, PP	37.7±0.4	0.1±1.0	0.18±0.8	39.6±0.6	0.85±0.02	-0.23±0.06	50.3±0.3	-0.91±0.01	0.59±0.06
5, PLA	44.9±0.6	-0.1±0.1	1.3±0.1	53.7±0.9	-0.92±0.04	-1.3±0.2	63.5±0.1	-1.89±0.01	0.66±0.07
6, EVOH	40.3±0.1	0.52±0.01	1.54±0.04	50.6±0.4	-0.28±0.01	-1.3±0.1	63.5±0.1	-0.97±0.01	-0.34±0.08
7, PE	43.9±0.9	0.08±0.01	1.48±0.06	53.9±0.5	-0.79±0.04	-0.96±0.13	65.1±0.3	-1.17±0.01	1.37±0.04
8, Nylon	41.0±0.1	0.55±0.02	2.27±0.01	50.3±0.1	-0.32±0.01	-0.53±0.08	64.1±0.4	-1.14±0.06	1.28±0.01
9, Hotmelt	41.1±0.1	0.51±0.01	3.62±0.03	51.4±0.3	-0.57±0.02	1.78±0.08	62.6±0.1	-0.09±0.01	4.73±0.06
10, PVC	40.7±0.1	1.18±0.05	3.14±0.04	50.9±0.3	0.29±0.03	-1.3±0.1	61.8±0.1	0.46±0.01	1.28±0.01

The CIELAB colour values of the rPET granulates with the deliberately added contaminants are listed in Table 15. The L*-values of all the rPET granulates are all relatively low, indicating that the rPET granulates are relatively dark. Compared to for instance the purchase specifications of Coca-Cola (L* >67 for crystallised pellets), all rPET-granulates are too dark. This is most probably caused by the fact that the feedstock were PET-bottles with 47.5% rPET and the presence of reheat additive and the samples were produced under laboratory conditions. The results can therefore only be compared between samples.

The darkest rPET is the rPET to which PP labels were added. Since most of these labels were black zero Cola labels, it is well understandable that this rPET is the darkest.
The a*-value is for most crystallised rPET granulates negative, indicating that on the green to red colour scale, most rPET granulates tend slightly to green. The only exception is number 10, this the rPET to which PVC is added which has a positive a* value meaning that it is slightly red.
The b*-value is for most crystallised rPET granulates after SSP treatment positive, indicating that on the blue to yellow colour scale, most rPET granulates tend to yellow. The only exception is number 6, the rPET to which EVOH is added, which has a negative b* value indicating that it is slightly blue. The rPET granulates that have most strongly coloured yellow are the rPET granulates to which hotmelt, Amosorb oxygen scavenger and PS have been added. Only the b* value of the rPET granulate to which hotmelt has been added is unacceptable in comparison with the purchase specifications of Coca Cola (b* < 3).

3.2.5 *Haze measurements on test specimen*

The results of the haze measurements are shown in Table 16. The reference rPET has a haze-value of 24% and is too hazy in comparison to purchase specifications that demand that the haze should be less than 13%. The test specimen made from rPET to which PE, PP and PS contaminants are added become extremely hazy. Whereas test specimen made from rPET to which Amosorb, PLA, EVOH, Nylon and PVC are added become less hazy in comparison to the reference rPET. With the exception of the specimen with PVC, this can be explained by the drop in particle contamination as determined with the Partisol measurement.

Table 16: Haze measurements on 5x5 cm test specimen made from SSP-treated rPET granulates to which contaminants were deliberately added.

Code	Hazeguard, [%]	Extended Haze measurement, [%]	Transmittance, [%]
0, rPET	24 ± 2	26	55.1 ± 0.3
1, ink	27 ± 1	29	52.6 ± 0.7
2, Amosorb	17 ± 1	12	60.5 ± 0.1
3, PS	88.3 ± 0.2	86	21.5 ± 0.3
4, PP	100 ± 3	100	0.1 ± 0.01
5, PLA	20 ± 2	13	62.3 ± 0.4
6, EVOH	20 ± 1	23	55.8 ± 0.6
7, PE	73.9 ± 0.7	71	40.0 ± 0.2
8, Nylon	19 ± 1	13	58.8 ± 0.4
9, Hotmelt	28.1 ± 0.5	29	49.1 ± 0.2
10, PVC	21.8 ± 0.3	25	55.8 ± 0.2

3.2.6 *IV and GPC*

The intrinsic viscosities and the molar masses determined with GPC for the rPET granulates to which contaminants were deliberately added are shown in Figure 17, Figure 18 and Figure 19.

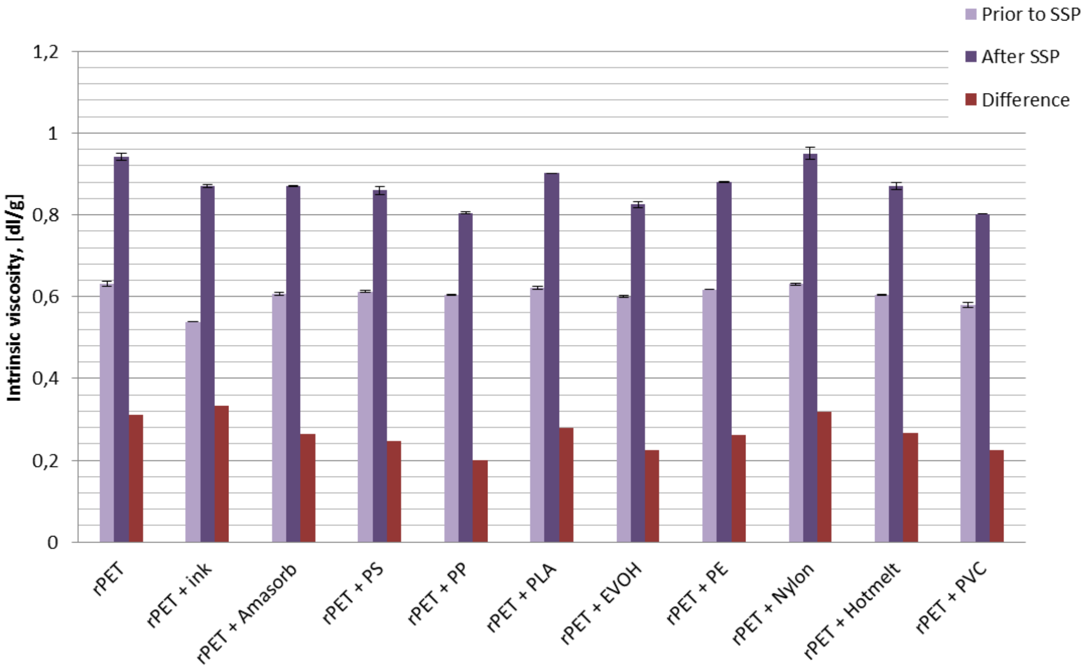


Figure 17: The Intrinsic viscosities of the rPET granulates prior and after SSP [dl/g].

All rPET granulates have too low intrinsic viscosities prior to SSP treatment (desired is 0,65 dl/g according to Stefan Morssinkhof of Morssinkhof Plastics) , this is probably caused by the processing in laboratory conditions and with flakes that were not dried prior to processing. The SSP treatment allows the intrinsic viscosity to reach sufficiently high values for reuse in new bottles (IV > 0.78 or 0.80 dl/g). The rPET sample to which PVC was deliberately added just reached the threshold value, indicating that the presence of PVC has partially counteracted the SSP treatment in restoring the PET chains. This is apparent from the lower difference in IV values as compared with this value for reference rPET. Most contaminants show a reduced difference in IV between prior and after the SSP treatment. With the exception of the contaminants Nylon and ink, all other contaminants hampered the recovery of the IV during the SSP process. Since these measurements were single measurements, with unknown standard deviations, the results should be treated with care and the conclusion should be that most contaminants appear to hamper the recovery of IV during SSP treatment. The IV prior to SSP was remarkably low for PVC and ink contaminants. Apparently both contaminants enhance depolymerisation. This can be understood for PVC which forms hydrochloric acid during heating, which in turn can split the ester bonds in PET.

Gel partitioning chromatography (GPC) measurements are used to study the influence of contaminants on the molecular mass (distribution) of PET. In industry IV measurements are performed as a measure of molecular mass. However, IV measurements do not give any information on the molecular mass distribution and is a rather insensitive method. GPC results before and after SSP are presented in Figure 18 and Figure 19. The numbers are shown in Appendix I. The molecular mass (M_w) is most comparable to the IV (M_v) and ranges from 39,000 to 47,000 g/mole before SSP. After SSP the Mw increase to values around 70,000 g/mole. After SSP most samples are very similar with respect to the molar mass distribution apart from the sample containing EVOH, see Figure 20. In this case a very clear increase in the M_z (z-average) is observed indicating a larger amount of molecules with very high molecular mass. This can be explained by a reaction between PET and EVOH during SSP. Eventually this reaction could lead to crosslinking and the presence of gel in PET (material that does not properly melt). The polydispersity (the ratio between M_n and M_w) is clearly enlarged by the SSP treatment which implies that the molecular weight distribution broadens.

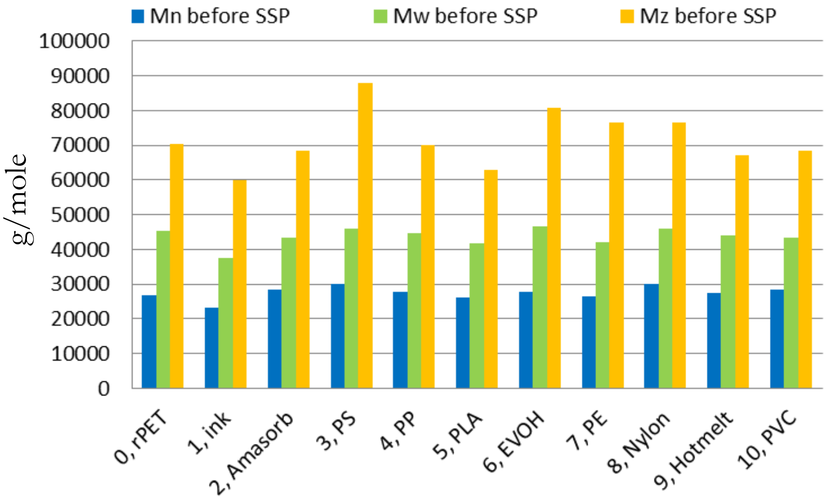


Figure 18: The three types of average molecular weights for the PET samples prior to SSP treatment.

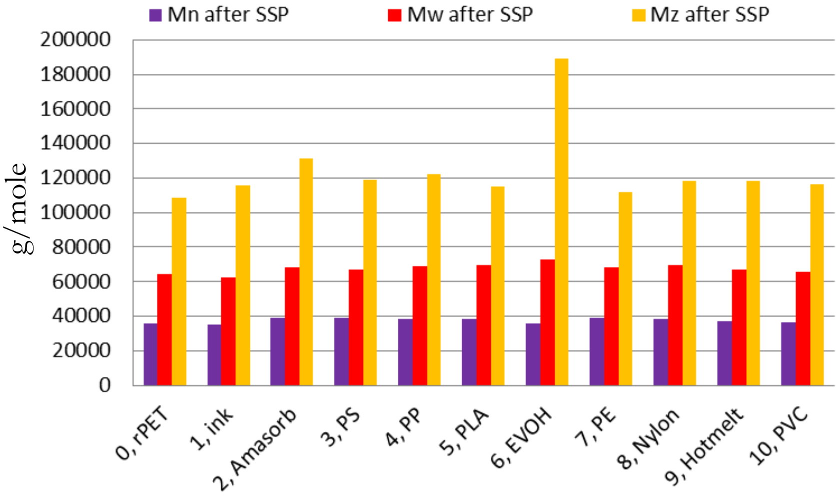


Figure 19: The three types of average molecular weights for the PET samples after SSP treatment.

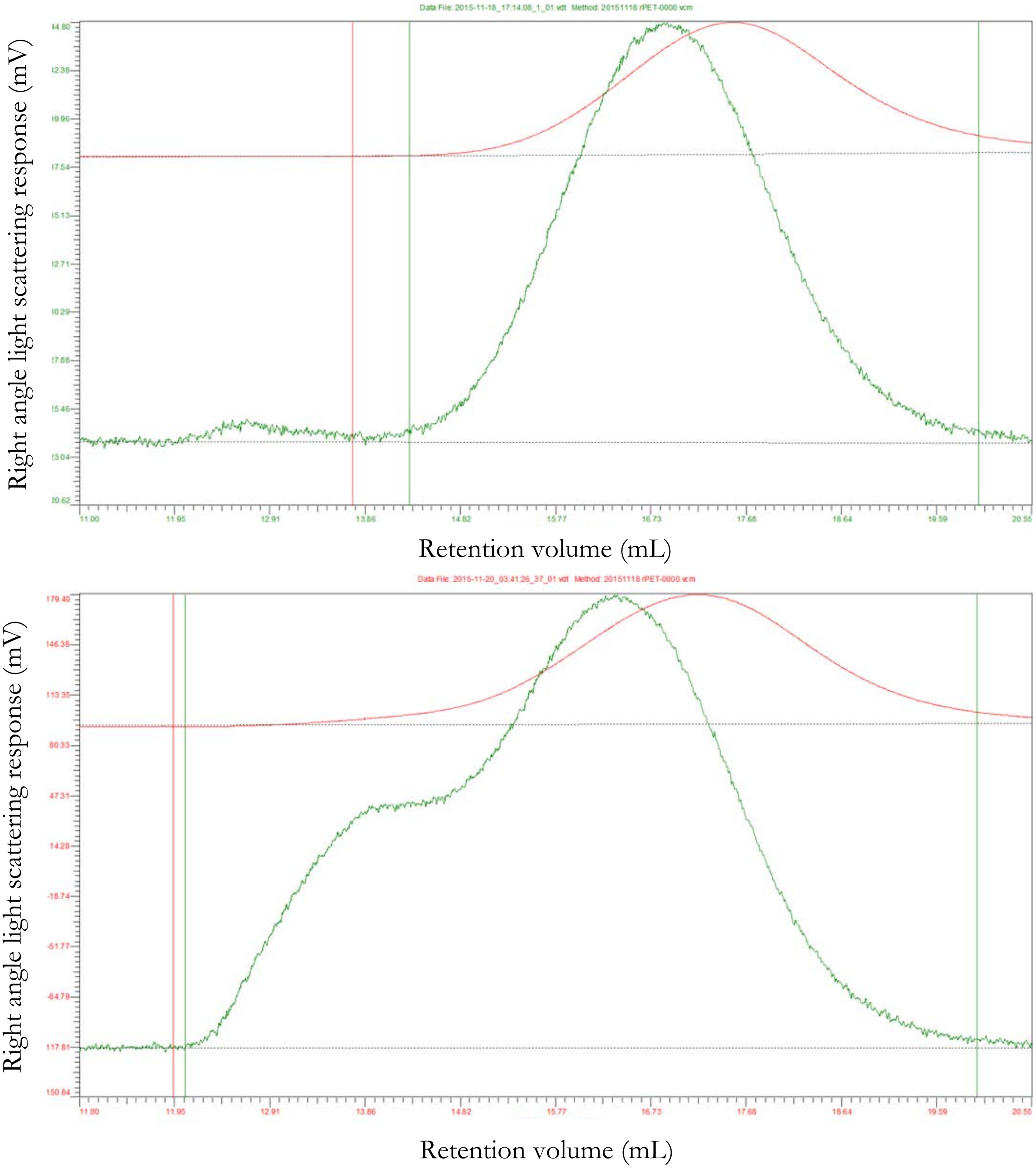


Figure 20: Gel-partitioning chromatogram for normal rPET (above) and rPET to which EVOH is added (below). In GPC's the largest masses run first of the column and the smallest masses as last. Hence the broad shoulder on the left side indicates the presence of molecules with a high molar mass. Red line = refractive index response, [mV] Green line = Right angle light scattering response, [mV]

3.2.7 *DSC*

Three DSC thermograms were recorded per sample (see Figure 21), a first heating scan, a cooling scan and a second heating scan. The heating scans show a glass transition at 74-76°C, a crystallisation peak at about 120°C and a melting peak at around 250°C.

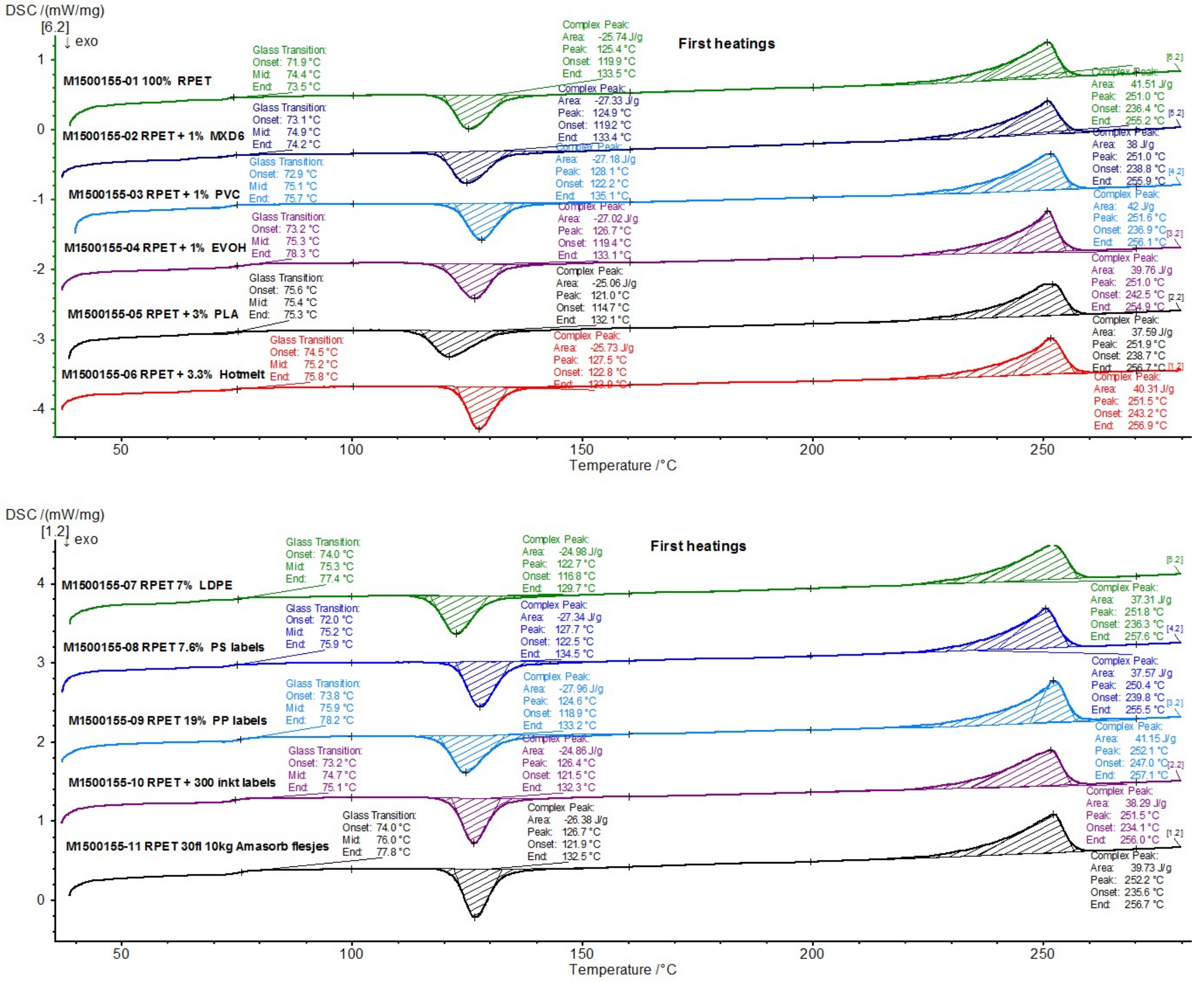


Figure 21: Thermograms showing the first heating scans of the rPET and 10 deliberately contaminated rPET samples. DSC thermograms show the enthalpy that is released or absorbed during the heating scans.

The degrees of crystallinity were derived from the peak area of the melting peak near 250°C from the second heating run. These peak areas (recorded enthalpies) were converted into crystallisation degrees, assuming the theoretical melt enthalpy of a PET crystal to be 140 J/g, see Table 17. These degrees of crystallisation were averaged. The melting peaks in the first heating runs and the crystallisation peak near 190°C in the first cooling run were found to deviate strong from the second heating run, yielding degrees of crystallisation that were 4% higher. Apparently the thermal history has a large impact on these deliberately contaminated rPET samples.

Also shown in Table 17 are the onsets of the crystallisation peaks of the first cooling runs. The higher the temperature of crystallisation, the more nucleating contaminants are present in the PET resin.

Table 17: The most important data from the DSC analysis. Clearly deviating numbers are presented in bold.

Code	Estimated degree of crystallinity, [%]	Onset of crystallisation peak from first heating run, [°C]	Onset of crystallisation peak from first cooling run, [°C]
0, rPET	24 ± 2%	119.9	199.8
1, ink	24 ± 2%	121.5	200.3
2, Amosorb	24 ± 2%	121.9	200.8
3, PS	24 ± 2%	122.5	199.7
4, PP	23 ± 2%	118.9	200.0
5, PLA	25 ± 2%	**114.7**	200.3
6, EVOH	24 ± 2%	119.4	**203.2**
7, PE	23 ± 2%	**116.8**	201.4
8, Nylon	24 ± 2%	119.2	199.6
9, Hotmelt	23 ± 2%	122.8	201.7
10, PVC	25 ± 2%	122.2	**202.0**

The DSC measurements show that the presence of the contaminants has a limited impact on the degree of crystallinity. It drops with less than 10% and this drop is not significant. Furthermore, the high content of reheat additive in this rPET is likely to either mask or interfere with any impacts of the studied contaminants on the degree of crystallinity. Contaminants such ink, PLA, PE and MXD-6 were found to lower the degree of crystallinity the most. This small reduction in degree of crystallinity could potentially negatively impact PET-material properties such as the gas permeability values and could result in shorter shelf lives for carbonated soft drinks. This is, however, not very likely, since the stretch blow moulding process of the bottles tends to increase the degree of crystallinity and is hence likely to compensate for these contaminant-effects.

The onset temperature of crystallisation was determined for the first heating run near 120°C and for the cooling run near 200°C, see Table 17. The onset temperature of crystallisation during heating is relevant for the bottle blowing process and should ideally be as high as possible (as close to 120°C as possible). Two contaminants PE and PLA were found to reduce this onset temperature. This could potentially result in crystallisation too early in the blowing process and is therefore undesired. This lower onset temperature appears to contradict with the lower estimated degrees of crystallinity. Since these measurements are single measurements, the conclusion is that there appears to be a tendency to lower the onset temperature.

The onset temperature of crystallisation during cooling is relevant for the preform injection moulding process as a too quick crystallising PET could give rise to partially crystallised preforms. Especially EVOH and PVC were found to raise this onset temperature.

3.2.8 *DMTA*

To study the influence of contaminants on the crystal structure of recycled PET, DMTA (dynamic mechanical thermal analysis) measurements were performed using injection moulded tensile bars. Badia et al reported severe changes in the microstructure and properties of PET due to recycling (Badía et al. 2009). The changes were monitored via shifts in the loss modulus (E”) around the glass transition temperature of about 75°C as measured via DMTA. Since after injection moulding all samples are amorphous, samples were first annealed at 120°C for 2 hours to induce crystallisation.

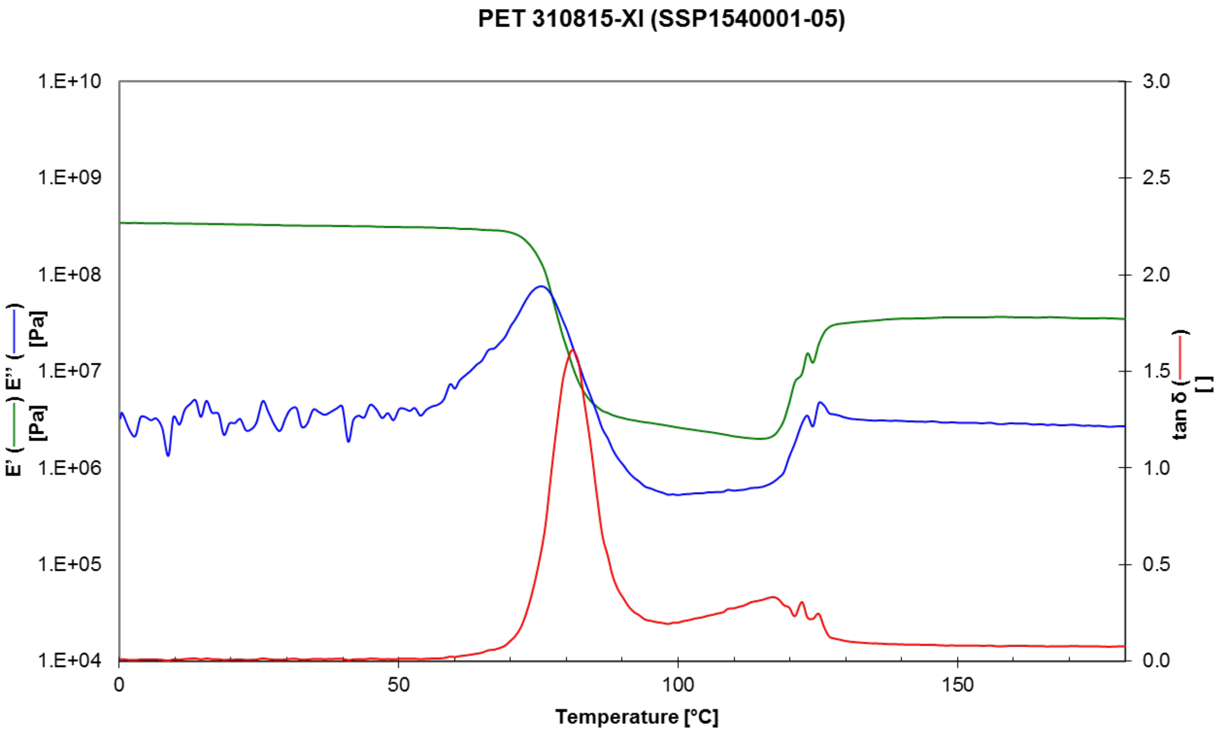


Figure 22: Typical DMTA thermograms of amorphous PET around the glass transition temperature.[19]

Figure 22 shows a typical DMTA graph of amorphous PET. In this graph a decrease of the stiffness (E’) is observed around the glass transition temperature near 75°C. At about 120°C the sample starts to crystallize and the E’ increases.

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[19] *DMTA technology is explained in* [*https://en.wikipedia.org/wiki/Dynamic_mechanical_analysis*](https://en.wikipedia.org/wiki/Dynamic_mechanical_analysis)

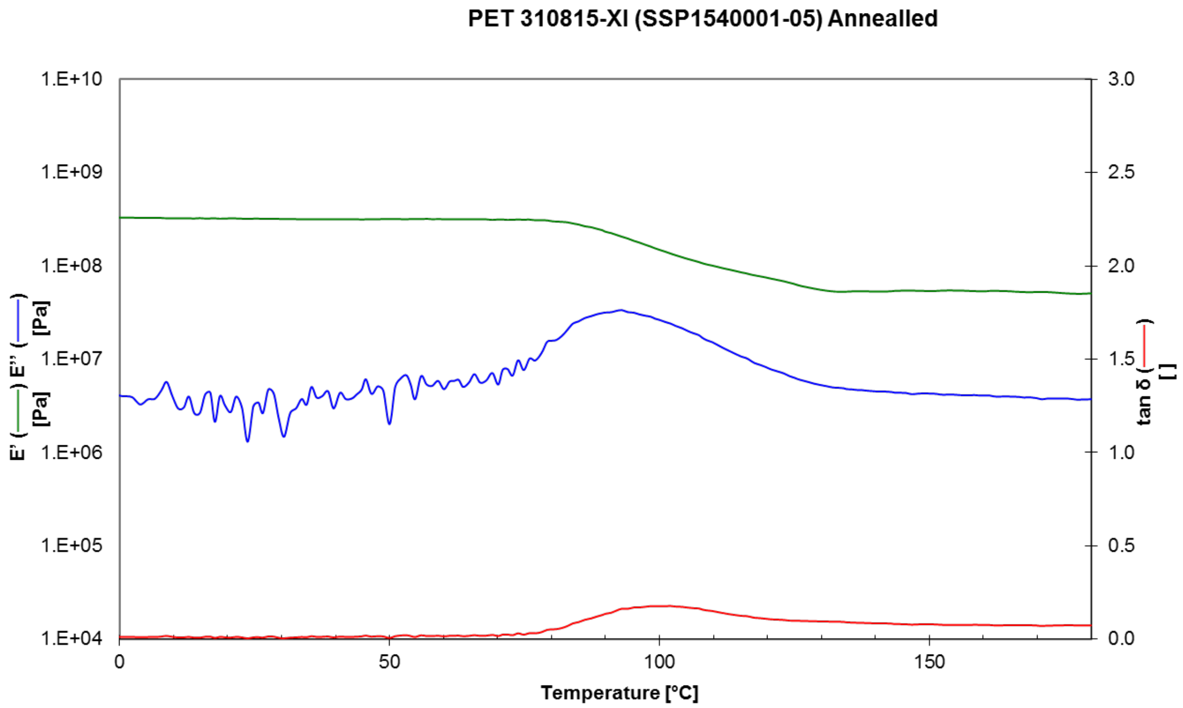

Figure 23: Typical DMTA thermogram of semi-crystalline PET.

Figure 23 shows a typical DMTA graph of a semi-crystalline PET sample. Again a drop in stiffness (E') is observed near the glass transition temperature. This drop is, however, much smaller than for amorphous PET since a large part of the PET material is crystalline.

Figure 24 shows in more detail the measured loss moduli (E'') around the glass transition temperature. As can be seen there is no difference between the reference rPET samples and the various PET samples to which contaminants were deliberately added. In other words the presence of the contaminants were not found to alter the DMTA results and hence also not change the lamellar crystalline structure of the rPET. The main difference with the results of Badia is that she studied rPET which was recycled multiple times, without SSP treatment and we studied rPET that was subjected to SSP and extruded once. Apparently the SSP treatment sufficiently repairs the PET chains so that after SSP treatment no difference in DMTA is observable. Hence DMTA is not a good analysis method to study the impact of contaminants on the morphology of rPET after SSP treatment, since the differences are too small to be observed in the DMTA thermograms.

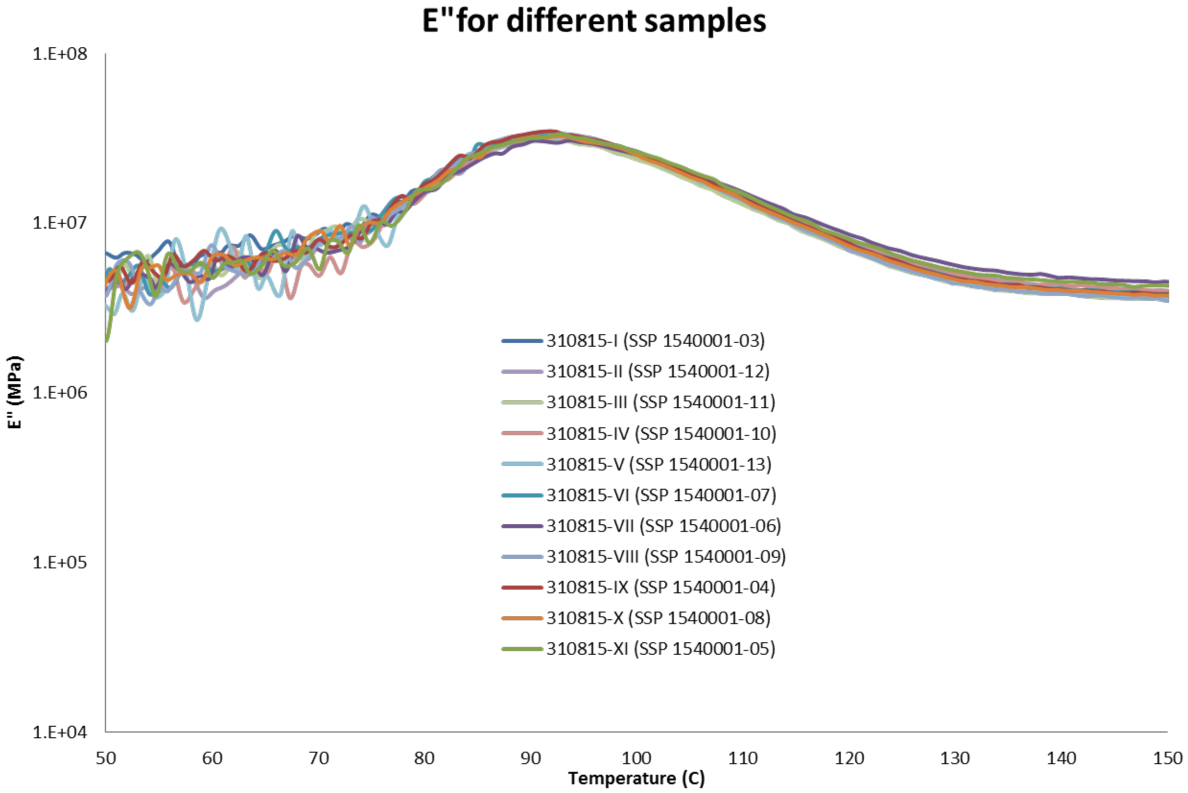

Figure 24: DMT-analysis of the loss moduli (E") around the glass transition temperature of PET.

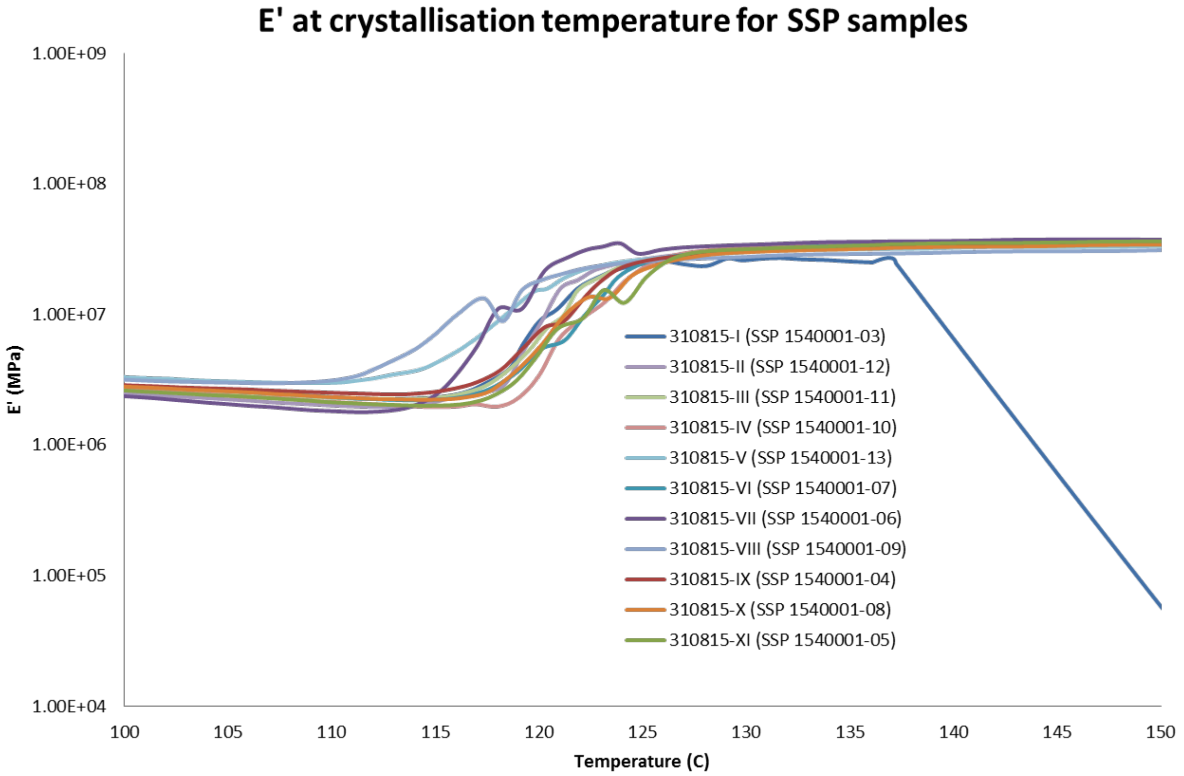

Figure 25: The onset of crystallisation studied with the stiffness (E').

The only measureable difference with DMTA is the onset of crystallisation, see Figure 25. Three samples with PP (310815-V), PE (310815-VIII) and EVOH (310815-VII) as contaminants clearly show a reduced onset temperature of crystallisation. This has already been observed with DSC for PE but not for PP and EVOH (Table 17). Moreover, DMTA does not give evidence of initiation of crystallisation by PLA. Control DSC measurements by Wageningen UR (Figure 25) indicate clear correspondence with DMTA results and in this case same contaminants also show a reduction in onset temperature of crystallisation in the DSC measurements. In Figure 26 a superposition of the first heating run with DSC for all the samples is shown. Some contaminants clearly influence the crystallisation behaviour and act as a nucleating agent. According these measurements PE, PP and EVOH can induce early crystallisation.

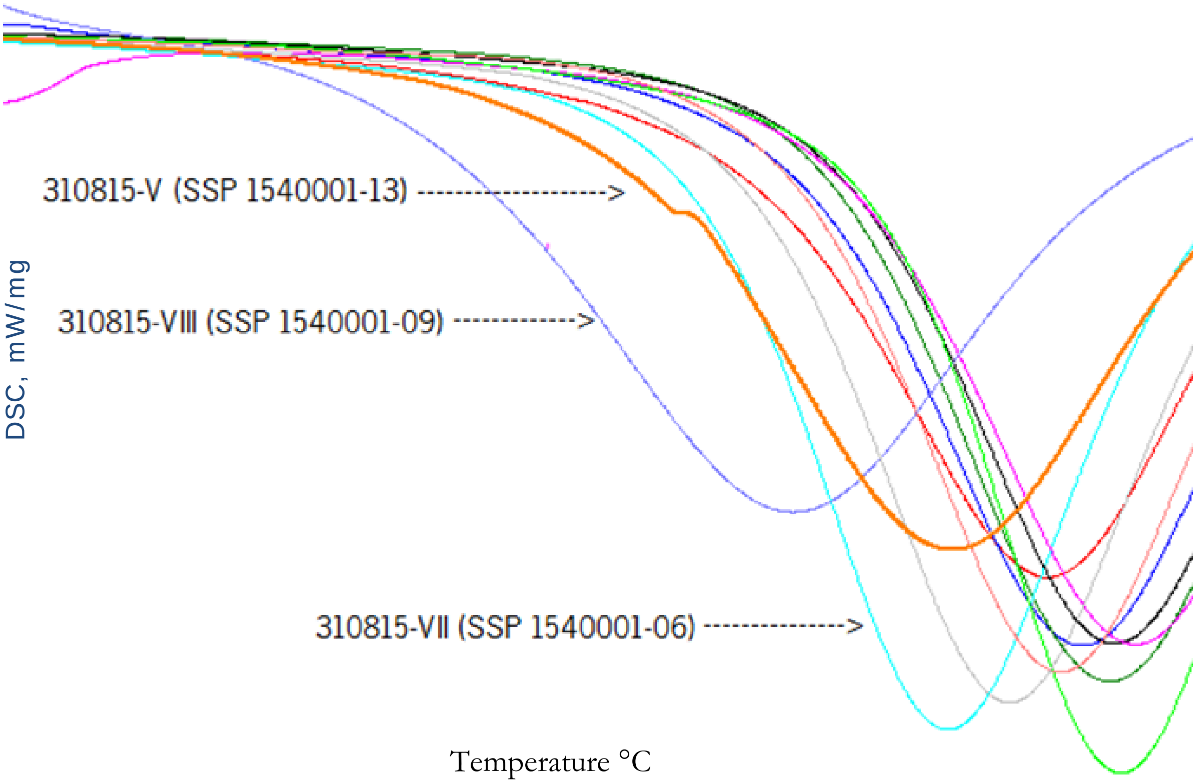

Figure 26: Superposition of the first heating run with DSC for all the samples in task 2 in the temperature range 100-130°C.

3.2.9 *Gas chromatography on the granulates*

The results of the headspace gas chromatography analysis are shown in Table 18. The rPET granulates contain substantial amounts of acetaldehyde and small amounts of benzene. The SSP treatment can reduce both. Acetaldehyde remains present but in lower concentrations and the benzene concentration is reduced to very low levels. The rPET granulate to which PVC was deliberately added had a much larger content of benzene, which was barely detectable after SSP treatment. The likely explanation is that PVC eliminates hydrochloric acid during heating and this acid is known to catalyse the degradation of PET. One of the known acid-catalysed degradation products of PET is benzene (Dzięcioł and Trzeszczyński 2000; Kumagai et al. 2014) [. Limonene was not detected, but was also not expected, because the rPET was made from unused bottles and no beverage flavours could have absorbed in the rPET.

Table 18: Results of the headspace GC analysis of the rPET granulates. The samples prior to SSP treated were measured twice and the results were averaged.

Code	**GC prior to SSP, [µg/g or ppm]**			**GC after SSP, [µg/g or ppm]**		
	Acetaldehyde	**Benzene**	**Limonene**	**Acetaldehyde**	**Benzene**	**Limonene**
0, rPET	13.5 ±1.2	0.005±0.002	0	3.6	0.0000	0
1, ink	9.3 ± 0.4	0.004±0.002	0	2.6	0.0000	0
2, Amosorb	11.1 ± 0.7	0.008±0.003	0	2.8	0.0000	0
3, PS	10.6 ± 1.3	0.009±0.004	0	2.9	0.0000	0
4, PP	8.7 ± 1.9	0.008±0.004	0	3.2	0.0000	0
5, PLA	9.8 ± 1.7	0.006±0.003	0	3.2	0.0000	0
6, EVOH	12.0. ± 2.7	0.014±0.003	0	2.7	0.0000	0
7, PE	7.9 ± 0.7	0.006±0.002	0	2.8	0.0000	0
8, Nylon	9.5 ±0.1	0.004±0.002	0	3.2	0.0000	0
9, Hotmelt	9.3±0.9	0.006±0.002	0	3.4	0.0000	0
10, PVC	10.3±0.2	**1.46 ± 0.14**	0	2.8	**0.0002**	0

In Appendix J, results from tests on samples before SSP treatment performed by Emmtech, requested by Stefan Morssinkhof, are shown. These results were send to us by Stefan Morssinkhof. The test method, equipment and settings of Emmtech are slightly different from the research method of the tests in this project. Therefore, the results of the test are slightly different. These additional test results are presented, in order to provide a complete overview of all available results. Both groups of results are roughly speaking in agreement with each other.

3.2.10 *Conclusions work package 2*

Contaminants deliberately added to rPET alter the properties of the rPET. The impacts of these contaminants on the rPET properties are listed in Table 19.

Table 19: The impacts of the contaminants on the properties of rPET.

Code	Contaminant	Main impact
0, rPET	None	
1, ink	Ink	Reduction of IV and molecular weights prior to SSP
2, Amosorb	Amosorb	Yellowing
3, PS	PS	Yellowing, Haze, ΔIV low; retards re-condensation during SSP process
4, PP	PP	Haze, particle contamination ΔIV low; retards re-condensation during SSP process
5, PLA	PLA	Faster crystallisation ΔIV low; retards re-condensation during SSP process
6, EVOH	EVOH	Cross-linking with PET during SSP Faster crystallisation ΔIV low; retards re-condensation during SSP process
7, PE	LDPE	Haze, particle contamination Faster crystallisation
8, Nylon	Nylon (MXD6)	
9, Hotmelt	Hot-melt	Yellowing, Faster crystallisation, Black specs.
10, PVC	PVC	Yellowing and Redding Benzene formation Reduction of IV and molecular weights prior to SSP ΔIV low; retards re-condensation during SSP process Faster crystallisation

Polyolefinic contaminants (PE, PP, PS) can originate from insufficiently removed caps, closures and labels or faulty sorted objects including PET-PE trays. They cause a fine particle contamination which turns the rPET hazy. Additionally PS and PE can also influence the act as nucleators and raise the speed of crystallisation of rPET.

Hotmelt doesn’t enhance the particle contamination. Most likely it coagulates with the reheat additive present. But during processing some large black specs were formed, which were so large that they were removed. It does make the rPET more yellow and it does increase the speed of crystallisation.

PVC makes the rPET more yellow and more red, increases the speed of crystallisation. It degrades PET prior to SSP which is apparent from the low IV values prior to SSP and the formation of the degradation product benzene.

EVOH crosslinks with rPET during SSP treatment and raises the speed of crystallisation. These cross-linked structures could result in the formation of undesired gel structures within the rPET.

The oxygen absorbing additive Amosorb makes the rPET slightly more yellow.

Nylon was found to have very limited effects on rPET in comparison to the other two barrier materials (EVOH and Amosorb).

PLA was found to lower the onset of crystallisation at heating and retard the re-condensation of PET during SSP treatment. A possible interpretation is that this contaminant which is also a condensation polymer combines with PET to form a co-polymer with different properties.

3.3 Results of work-package 3

3.3.1 *Introduction sample codes*

PET bottle products originating from different collection systems are mechanically recycled with a standard process and an advanced process. Besides the currently existing PET bottle products (1-6) also combinations of separately collected and deposit refund PET bottles and mechanically recovered and deposit refund bottles are tested (7-10). These PET bottle products are sorted to the current specification (DKR 328-1) and to a potential future specification (DKR 325). The matrix of combination and the codes used in this chapter are explained in Table 20.

Table 20: Codes of the samples, their origin and the mechanical recycling process that they are subjected to.

Code	Origin	Deposit refund bottles added	Sorting specification	Mechanical recycling process
1	SRN Deposit refund	N/A	N/A	Standard
2	Other Deposit refund	N/A	N/A	Standard
3	Separate collection	No	DKR 328-1	Standard
4	Separate collection	No	DKR 328-1	Advanced
5	MBT	No	DKR 328-1	Standard
6	MBT	No	DKR 328-1	Advanced
7	Separate collection	Yes	DKR 328-1	Advanced
8	Separate collection	Yes	DKR 325	Advanced
9	MBT	Yes	DKR 328-1	Advanced
10	MBT	Yes	DKR 325	Advanced

3.3.2 *Material composition of samples*

The material content after pre-processing, as input material for the laboratory mechanical recycling process is shown in Table 21. The material composition numbers shown in this table are calculated values, based on two different measurements (compositional analysis of PET bottle products in terms of packaging categories after pre-sorting, see Appendix H and the average material compositions per packaging category, see Appendix G). Both measurements are prone to variations and therefore these numbers will contain errors that cannot precisely be determined and should be used indicatively. Nevertheless, they render a good insight in the type of contaminants that are present and their expected concentrations. In a similar manner the amount of barrier bottles, and bottles with ink on the body is determined, the result is shown in Table 22.

Table 21: Material content of the samples after pre-processing, as input for laboratory mechanical recycling process

	1	2	3	4	5	6	7	8	9	10
PET	90.1%	89.9%	84.3%	84.7%	83.9%	84.2%	87.8%	87.9%	87.4%	87.5%
PP	2.4%	2.4%	6.2%	6.2%	7.3%	7.2%	4.2%	4.2%	4.8%	4.7%
PE	6.5%	6.7%	8.0%	7.7%	7.1%	7.0%	6.8%	6.8%	6.6%	6.6%
PS	0.2%	0.2%	0.4%	0.3%	0.4%	0.3%	0.2%	0.2%	0.2%	0.2%
PVC	-	-	0.005%	0.002%	0.004%	0.002%	0.001%	0.000%	0.001%	0.0001%
Paper	0.85%	0.84%	0.93%	0.92%	1.21%	1.11%	0.88%	0.88%	0.96%	0.95%
Metal	-	-	0.04%	0.04%	0.04%	0.04%	0.02%	0.02%	0.03%	0.02%
Glass	-	-	0.007%	0.004%	0.002%	0.002%	0.003%	0.002%	0.002%	0.002%
PA/POM/ABS, black, etc.	-	0.003%*	0.06%	0.06%	0.09%	0.09%	0.02%	0.02%	0.04%	0.02%
Silicon	-	-	0.03%	0.03%	0.04%	0.04%	0.01%	0.01%	0.01%	0.01%
Undefined/rest, incl. textiles	-	-	0.058%	0.023%	0.076%	0.031%	0.008%	0.002%	0.012%	0.003%

* some not NIR sortable (black) material is present in this sample, which is in this case most likely PE or PP.

Table 22: Amount of barrier bottles and bottles with ink on body in feedstock samples, prior to recycling.

	1	2	3	4	5	6	7	8	9	10
Share of barrier-bottles (PA, EVOH, Amosorb) in comparison to total amount of PET bottles, mass based.	-	0.6%	2.5%	2.6%	2.3%	2.2%	0.9%	0.9%	0.9%	0.9%
Share of bottles with ink on body in comparison to total amount of PET bottles, mass based.	32%	32%	35%	35%	35%	35%	33%	33%	33%	33%

The two pre-sorted PET bottle feedstocks that originated from the two deposit refund systems were free of undesired impurities such as PVC, glass, metal, silicones and undefined materials, but one of the two samples (2) did contain 0.6% barrier bottles. In all the other pre-sorted PET bottle feedstocks these contaminants were present, although in low concentrations. The mechanical recycling processes intend to reduce the concentration of these impurities further. Table 23 shows which process steps are expected to reduce these contaminants.

This analysis reconfirms that the impurities present in the bottle products originating from deposit refund systems can be removed with a standard process, whereas for the other PET bottle products the advanced process is required to lower the concentrations of these contaminants.

Table 23: Expected removal of impurities and contaminants in the standard and advanced recycling processes

	Standard				Advanced		
Contaminant/impurity	**Pre-wash & wet milling**	**Wind sifting**	**Hot wash**	**Floatation separation**	**Vibrating screens**	**Flake sorting (material/ colour)**	**Conclusion**
Ink printing							**No**
Amosorb		If loose				If loose	Some
PS		**Yes**		**Yes**	**Yes**	**Yes**	**Yes**
PP/PE		**Yes**		**Yes**		**Yes**	**Yes**
PLA		**Yes**		**Yes**		**Yes**	**Yes**
EVOH		If loose				If loose	Some
LDPE		**Yes**		**Yes**		**Yes**	**Yes**
Nylon (MXD6)		If loose	Some			If loose	Some
Hot-melt			**Yes**				**Yes**
PVC					**Yes**	**Yes**	**Only advanced**
Glass					**Yes**	**Yes**	**Only advanced**
Paper		**Yes**	**Yes**				**Yes**
Metal					**Yes, fine parts**	**Yes**	**Only advanced**
PA/POM/ABS, etc						**Yes**	**Only advanced**
Silicone				If floating		**Yes**	Some, **only advanced**

3.3.3 *Origin of impurities*

The origin of the impurities in the feedstock samples was derived these material compositions and is shown in Appendix K. Again, these numbers should be used as indicative numbers and were based on sets of measurements and estimations. However, it clarifies whether or not a contaminant primarily originates from the bottle design of PET bottles and flasks or from sorting faults. This analysis revealed that:

- PP, PE, PS, paper, metal and silicones are mainly originating from the design of the PET bottles and flasks.
- PVC originates only from faulty sorted objects (mainly PVC from non-packaging objects), as PVC is no part of the design of PET bottles and flasks.
- Glass and other heavy plastics originate both from the design of the flasks and from the faulty sorted objects.

3.3.4 *Net recycling chain yields*

The recovered masses, net PET material concentrations and net PET material yields are listed in the tables below for all the process steps. For most process steps these parameters could be calculated from the analysed compositions and determined product weights. Nevertheless, in a few instances not all required data was available and assumptions were required, this is explained in the method chapter.

Table 24: The net recovery of mass per chain step (which are executed subsequently), expressed in net masses [% w net/ w net].

Code	pre-sorting	pre-wash and wet-milling	wind sifting	hot wash & floatation separation	vibrating screens	flake sorting, material	flake sorting, colour
1	100%	99%	92%	92%			
2	94%	97%	91%	91%			
3	77%	93%	90%	85%			
4	76%	91%	90%	87%	99%	92%	99%
5	71%	98%	95%	89%			
6	71%	100%	93%	87%	99%	92%	98%
7	92%	95%	90%	90%	99%	93%	99%
8	92%	94%	89%	89%	99%	91%	99%
9	88%	99%	90%	88%	99%	90%	99%
10	92%	98%	89%	87%	99%	91%	99%

Table 25: The PET concentration at the end of each chain step (which are executed subsequently),. [% w net/ w net].

Code	pre-sorting	pre-wash and wet-milling	wind sifting	hot wash & floatation separation	vibrating screens	flake sorting, material	flake sorting, colour
1	90%	90%	93%	96%			
2	89%	90%	95%	98%			
3	82%	84%	90%	96%			
4	82%	85%	89%	95%	98%	99%	99%
5	82%	84%	89%	95%			
6	82%	84%	91%	95%	98%	100%	100%
7	87%	88%	94%	97%	99%	99%	99%
8	87%	88%	92%	96%	98%	99%	99%
9	86%	87%	90%	92%	98%	99%	99%
10	86%	87%	92%	95%	98%	98%	98%

Table 26: The net PET material yield per chain step, expressed in net weights, [% w net/w net].

Code	pre-sorting	pre-wash and wet-milling	wind sifting	hot wash & floatation separation	vibrating screens	flake sorting, material	flake sorting, colour
1	100%	99%	93%	95%			
2	95%	97%	92%	94%			
3	78%	94%	92%	90%			
4	78%	91%	92%	93%	98%	94%	99%
5	73%	99%	96%	95%			
6	73%	100%	94%	91%	97%	93%	98%
7	93%	95%	91%	94%	100%	93%	99%
8	93%	94%	89%	92%	99%	92%	99%
9	90%	99%	92%	90%	95%	90%	99%
10	93%	98%	90%	89%	98%	91%	99%

Table 27: Net chain yields for the complete recycling processes.

	Recovery of Mass (dry matter) [%]	Δ chain PET concentration (net) [%]	Net chain yield (net PET) [%]
1. SRN deposit, standard	84%	6%	88%
2. Other deposit, standard	75%	8%	79%
3. Separate collected, standard	55%	13%	61%
4. Separate collected, advanced	49%	17%	56%
5. Recovered, standard	59%	13%	66%
6. Recovered, advanced	51%	18%	56%
7. Separate collected + deposit, advanced	64%	12%	70%
8. Separate collected + deposit, DKR325, advanced	61%	12%	66%
9. Recovered + deposit, advanced	61%	13%	63%
10. Recovered + deposit, DKR325, advanced	61%	12%	65%

These values are indicative and will vary with varying feedstocks, as explained in the methods chapter. Due to this variance in the composition of the samples, one calculated yield exceeded 100%, and was set back to 100%. Additionally, the chain yields for colour sorting will be lower in the industrial reality.

In the industrial practice material yields are mostly determined by dividing the output weight by the gross input weight. In the laboratory situation the samples were relatively small. In practice, smaller batches result in higher losses. Therefore the industrial yields could deviate from the laboratory yields. It is important to see the results of these tests as an indication and use the results to compare between different samples. The results cannot be compared to practice directly.

The determined recoveries of mass (Table 27) are slightly smaller (about 5-10%) than the net PET material content as is shown in Figure 11. These differences are the process losses that can be attributed to the laboratory execution of the mechanical recycling in comparison to the conventional industrial execution. The difference in recovered mass between the standard and the advanced process is striking, this additional loss is predominantly caused by the additional flake sorting step. Addition of the deposit refund bottles to the separately collected and mechanically recovered PET bottles causes a slight increase (10%) in the net recovered mass and the net PET chain yields.
The difference in PET concentration before and after the mechanical recycling process gives some insight in the relative effort that is required to create rPET products from various PET bottle feedstocks. As expected, this effort is the smallest for deposit-refund bottles and the largest for separately collected an mechanically recovered PET bottles subjected to the standard process.

3.3.5 *Waste water analysis*

In Table 28 the waste water analysis results are shown of the waste water discarded after the washing step. Both the Kjeldahl number (total-N) and the chemical oxygen demand are reported.

Table 28: Results of the waste water analysis of the discarded washing water.

	total-N WI 4.25-115 [mg/kg]	**COD WI 4.25-138 [mg/kg]**
1. SRN deposit, standard	2	80
2. Other deposit, standard	1	240
3. Source separated, standard	3	710
4. Source separated, advanced	2	500
5. Recovered, standard	5	980
6. Recovered, advanced	3	730
7. Source separated + deposit, advanced	2	180
8. Source separated + deposit, DKR325, advanced	2	250
9. Recovered + deposit, advanced	4	290
10. Recovered + deposit, DKR325, advanced	3	370

Apparently the removed dirt from the PET bottles hardly contains any proteins, since the Kjeldahl number is low. The chemical oxygen demand does appear to vary with the origin of the PET bottles. The least polluted water is formed from washing deposit refund PET bottles in the standard process. Slightly more polluted is the washing water from the separate collected and mechanical recovered PET bottles. When comparing the standard with the advanced process (code 3 to 4 and code 5 to 6) a small difference becomes apparent in the waste water. The waste water of the advanced process is slightly less polluted than the waste water of the standard process. This can be attributed to the mimicked pre-sorting step in the advanced process in which a few undesired objects are already removed, such as the residual waste and other non-bottle packages. After adding the PET bottles from the deposit refund systems to the separately collected and mechanically recovered PET bottles the wastewater is slightly less polluted (comparing code 4 to 7 and 6 to 9).

3.3.6 *Extrusion*

Batches recycled PET from different sources and with different levels of sorting and washing (see Table 20) were processed into granules for analyses, SSP and injection moulding. The processing conditions during extrusion are listed in the methods and are identical to the processing conditions used in task 2. During extrusion, differences in colour were observed. The standard SRN rPET (1) has a grey colour like the reference material from task 3. The rPET granulate originating from the other deposit refund system (2) and from the separate collection system (3) have yellow to green colours. PET originating from separate collection system and from mechanical recovery that have been recycled with the standard recycling process (3 and 5) are difficult to process in an extruder. The extrusion process for sample 3 is instable as a result of large differences in the melt viscosity. Sample 5 also showed a significant increase in pressure during extrusion caused by non-melting materials obstructing the melt filter.

3.3.7 *Colour*

The determined colour values for PET granulates are listed in Table 29 and for the crystallised granulates after SSP treatment shown in Figure 27.

Table 29: The CIELAB colour values of the rPET granulates produced in task 3 in the amorphous and crystallised state prior to SSP and crystallised after SSP. L* indicating the lightness (black/white), a* indicating colouring along the red/green axis, b* indicating colouring along the yellow/blue axis. Clearly deviating numbers are coloured.

Code	Amorphous prior to SSP			Crystallised prior to SSP			Crystallised after SSP		
	L*	a*	b*	L*	a*	b*	L*	a*	b*
1	33.7±1.4	-0.8±0.1	1.5±0.2	44.4±1.2	-1.2±0.1	-2.4±0.5	54±3	-1.9±0.1	1.2±0.5
2	39.6±1.8	-1.5±0.1	6.7±0.3	49.3±2.2	-1.9±0.1	3.1±0.8	58.1±1.1	-2.1±0.1	5.4±0.6
3	33.4±1.8	-3.0±0.1	2.5±0.4	40.9±1.0	-3.4±0.1	-0.4±0.2	51.5±1.5	-2.9±0.1	3.5±0.2
4	33±3	-2.2±0.2	3.9±0.4	46.0±1.1	-3.0±0.1	1.3±0.4	55.3±0.5	-3.2±0.2	3.9±0.3
5	31.5±1.0	-2.8±0.1	7.4±0.3	43.0±0.2	-4.3±0.3	7.4±0.3	49.4±0.6	-2.9±0.1	8.1±0.4
6	31.4±1.3	-1.6±0.1	1.8±0.1	46±3	-2.6±0.2	-2.2±0.7	54.7±1.5	-3.2±0.2	3.5±0.5
7	34.1±0.9	-1.4±0.1	3.1±0.2	47.5±1.6	-2.3±0.1	-0.3±0.4	54.8±0.7	-2.9±0.1	2.9±0.6
8	33.9±0.9	-1.5±0.1	2.1±0.2	44.3±0.8	-2.2±0.1	-2.7±0.4	56±2	-3.1±0.1	2.1±0.5
9	33.3±1.1	-1.5±0.1	2.2±0.3	44.8±0.9	-2.3±0.1	-2.5±0.5	56.3±1.2	-3.0±0.1	1.3±0.5
10	33.3±2.2	-1.4±0.1	1.8±0.3	44±3	-2.1±0.1	-2.8±0.7	55.9±0.8	-2.9±0.1	1.9±0.4

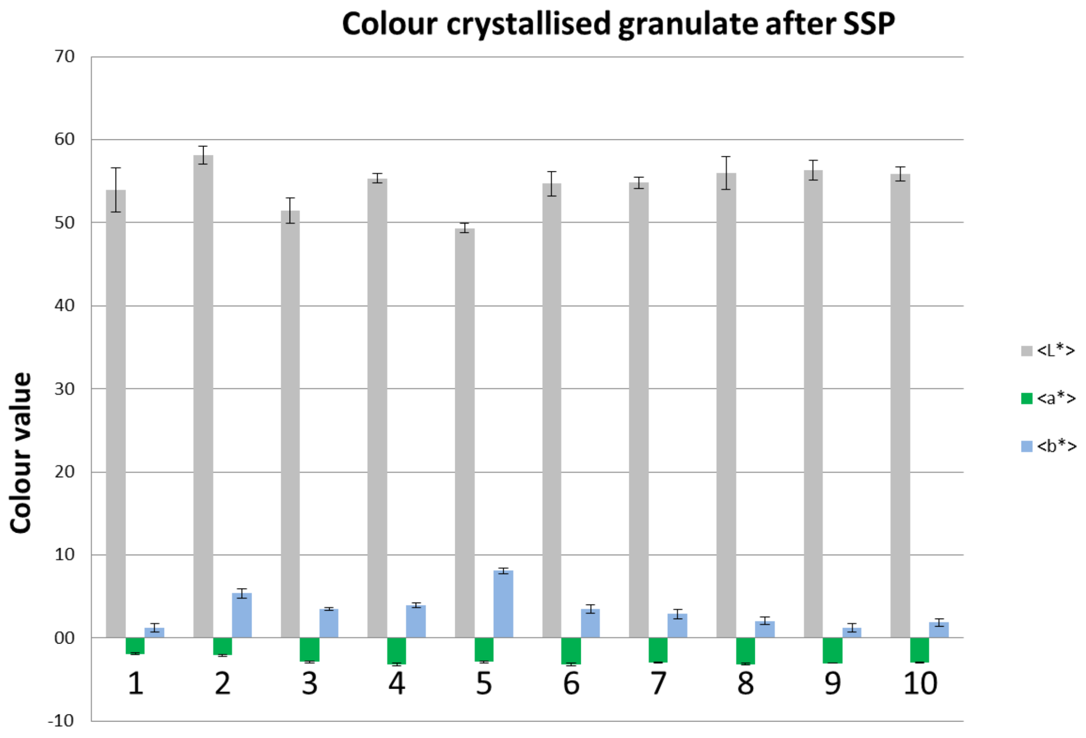

Figure 27: The CIELAB colour values for the crystallised granulates after SSP treatment.[20]

These colour values show that there are clear differences between PET products made from different origins with the standard recycling process (1, 2, 3, 5). The two PET granulates from the two different deposit refund systems already differ. The PET product of the SRN origin has lower L* value and b* as compared to the values of the PET product of the other deposit refund system. The lower L* value for the product from the SRN system could perhaps be explained by the use of reheat additive (carbon black) by a part of the concomitant producers. The higher b* value for the product from the other deposit refund system is likely to be caused by the presence of a limited amount of barrier bottles in this system that cause yellowing.

The PET products made from bottles originating from separate collection (3) and mechanical recovery (5) with the standard recycling process have lower L* values and relatively high b* values. When subjecting these feedstocks to the advanced recycling process (4, 6) the L* value is raised and the b* value is reduced slightly. Adding the PET bottles from the deposit refund systems and sorting the PET products to the 325 specification improves these colour values only marginally further. The presence of barrier bottles in the feedstocks is likely to be the prime cause of the relative high b* values.

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[20] *CIELAB technology is explained in https://nl.wikipedia.org/wiki/CIELAB*

The L*-values in task 3 are lower than those in task 2 (comparing tables 15 and 26). The cause is not precisely known, but could relate to the mechanical recycling process itself, the incomplete removal of attached product residues and the types of PET present in the bottles.

3.3.8 *Partisol*

The particle contamination inside the produced rPET granulates after SSP were studied with Partisol. The total amount of counted particles per 10,000 images and the approximated amount per gram of rPET are listed in Table 30. The particle contamination in the rPET products made from the deposit refund bottles with the standard process (1, 2) are already relative high compared to the industrial reference point (15,000/10,000 images for rPET and 370/10,000 images for virgin PET), but the samples made from separately collected and mechanically recovered bottles recycled with the standard process (3, 5) are much higher. When both feedstocks are treated with the advanced process the particle contamination drops considerably (4, 6). By adding large PET bottles from the deposit refund system to both feedstocks the particle contamination drops again (7, 9). Implementing the stricter sorting protocol hardly affects the particle contamination (8, 10).

Table 30: The main results of the Partisol measurements.

Code	Nature of the solution	Total particle count per 10000 images	Estimated amount of particles per gram, [millions per gram]
1	White	130,570	33 ± 5
2	Hazy	58,454	15 ± 5
3	Grey-white	1,162,175	291 ± 5
4	Clear	123,976	31 ± 5
5	Hazy	695,396	174 ± 5
6	Clear	51,872	13 ± 5
7	Clear	48,391	12 ± 5
8	Clear	45,300	11 ± 5
9	Clear	41,907	11 ± 5
10	Clear	52,330	13 ± 5

The particle distribution is shown in Figure 28. This clearly shows that the smaller particles (1-3 µm) dominate in all rPET product, but especially in those that are heavily contaminated (3,5). Such a particle size distribution was expected, since the granulates were well-extruded and subjected to melt-filtration, which would exclude particles with a size larger than 50 µm.

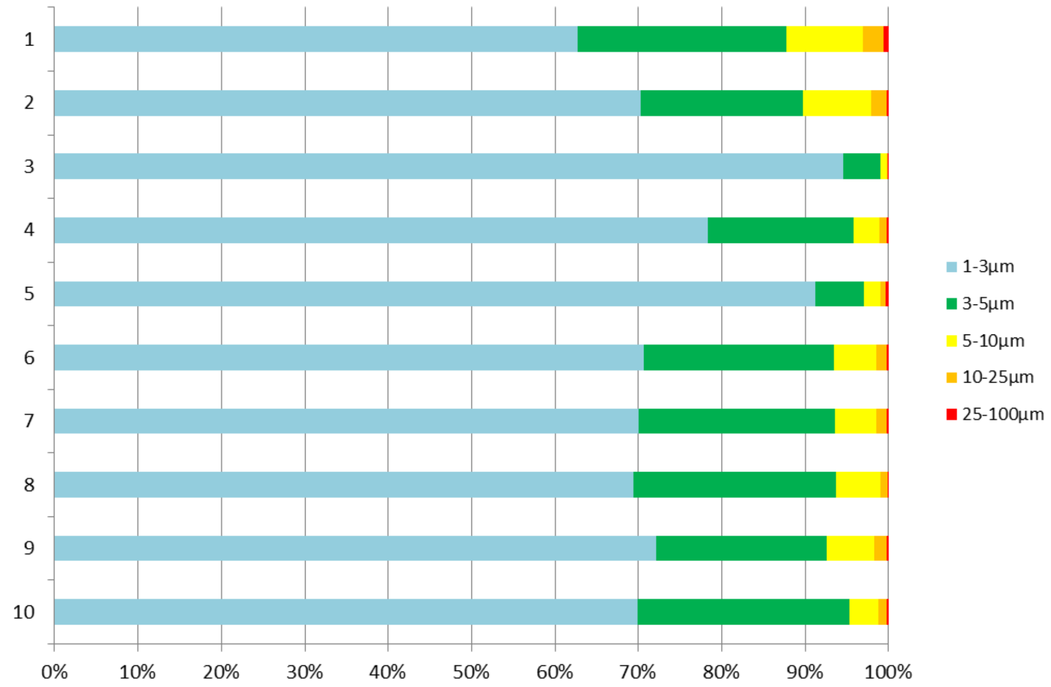

Figure 28: Normalised particle size distribution of the PET samples studied in task 3.

3.3.9 *Haze*

The results of the haze measurements are listed in Table 31. The particle contamination (as determined by Partisol measurements) and the haze measurements were found to correlate reasonably well (also see Figure 29). The samples with the most particles also had the highest haze values. Although none of the test samples reached haze values as low as 13% (industrial purchase condition for 3 mm thick plates), it is clear that the advanced recycling process greatly reduces the particle contamination and the haze values. Nevertheless, samples made with the advanced process were found to possess lower haze values than those made with standard process. The lowest haze values were recorded for samples 6, 8, 9 and 10.

Table 31: Haze results of the PET samples made in task 3, measured with Haze guard.

Code	Haze, [%]	Transmission, [%]
1	45.1 ± 0.5	51.2 ± 1.1
2	39.0 ± 0.7	51.7 ± 0.3
3	87.7 ± 0.6	21.6 ± 0.1
4	41.0 ± 1.1	52.8 ± 0.2
5	84.4 ± 0.3	18.0 ± 0.2
6	29.6 ± 0.4	55.5 ± 0.1
7	33.0 ± 0.6	51.6 ± 0.3
8	29.6 ± 0.4	55.2 ± 0.1
9	30.6 ± 0.6	52.7 ± 0.1
10	30.0 ± 1.0	55.9 ± 0.2

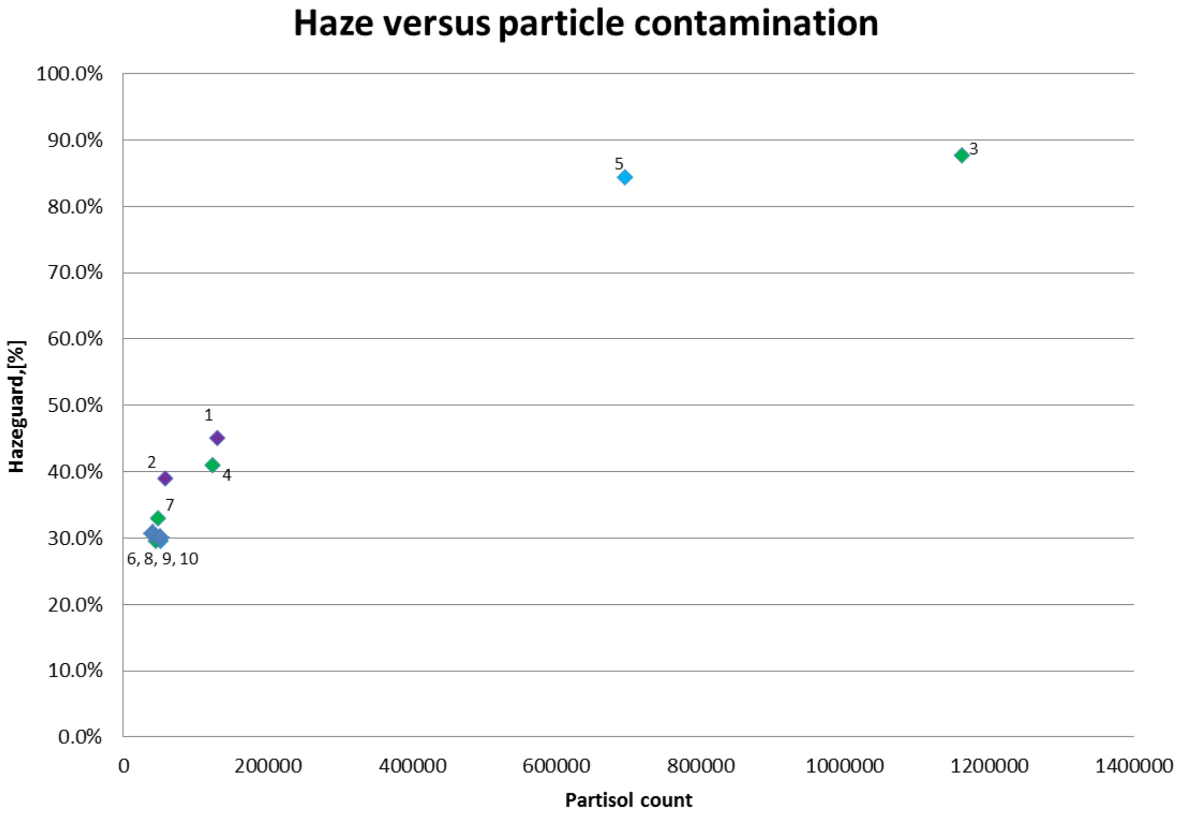

Figure 29: The relationship between the particle counts and the determined haze values. The numbers indicate the combinations of feedstock and process studied.

3.3.10 *IV and GPC*

The intrinsic viscosities of the PET granulate were measured prior to SSP treatment and after SSP treatment, see Figure 30. For both PET bottle feedstocks originating from deposit refund systems (1, 2) the intrinsic viscosity clearly exceeded the 0.8 or 0.78 dl/g purchase conditions of the beverage industry. Also the PET bottles originating from the separate collection system that were subjected to the advanced recycling process (4) resulted in a rPET with a sufficient high intrinsic viscosity value after SSP. For all other combinations of feedstock and recycling process (3, 5-10) the intrinsic viscosity after SSP did not reach the desired level. The main culprit appears to be the lower IV prior to SSP, since the change in IV during SSP treatment is still more or less constant, with a few interesting exceptions. Unfortunately, no clear relationship could be found between the estimated concentration of contaminants in the rPET products (Appendix L) and the IV prior to SSP.

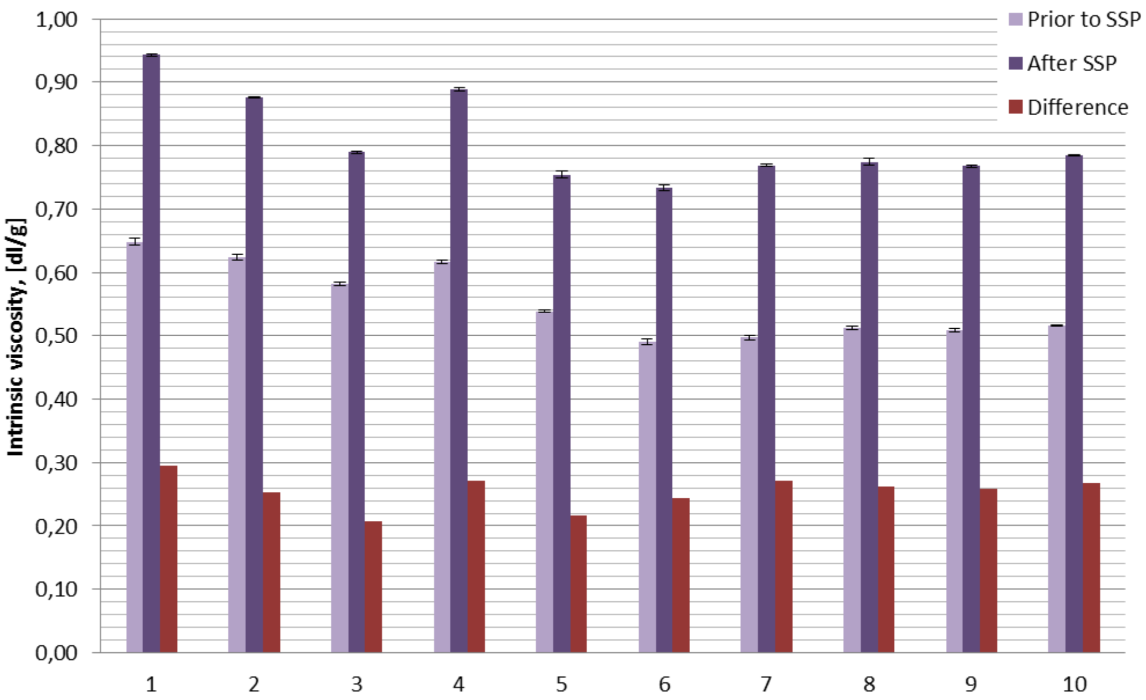

Figure 30: The intrinsic viscosities of the ten combinations of feedstock and process before and after the SSP treatment.

When comparing samples 1 and 2 (feedstock from the two different deposit refund systems treated with the standard process) the SSP treatment renders an increase in IV of almost 0.3 for sample 1 and only 0.25 for sample 2. This could be caused by a one hour longer residence time for sample 1 as compared to the other samples in the SSP reactor, as the laboratory assistant reported.

The difference between the IV prior and after SSP treatment was the smallest for the two most contaminated samples (3 and 5). This reconfirms the hypothesis that contaminants can hamper the recovery of IV during SSP treatment.

The molecular weights were determined with gel partitioning chromatography for all samples prior and after SSP treatment, the results are shown in Table 32. These results are very similar to the IV results. Apparently the molecular weight distributions broaden for all rPET samples, but no sample possesses a shoulder indicating excessively a low or high molecular weight. This indicates that the amount of EVOH from barrier bottles is apparently very low in these samples.

Table 32: The molecular weights determined by gel partitioning chromatography.

Code	Prior to SSP				After SSP			
	M_n	M_w	M_z	M_w/M_n	M_n	M_w	M_z	M_w/M_n
1	25739	44397	75165	1.725	35703	65066	96213	1.837
2	26163	42963	70344	1.643	33469	63059	99978	1.887
3	25920	41035	65976	1.584	32542	57131	92548	1.764
4	25759	43957	76111	1.706	35522	64024	104019	1.821
5	23681	38523	65138	1.627	34888	59753	99635	1.762
6	20931	34556	54580	1.655	32178	55068	86656	1.748
7	22054	34839	61810	1.582	30685	55610	85721	1.837
8	21677	35303	60144	1.628	30580	54894	86826	1.808
9	20935	34897	61443	1.668	30310	55156	89385	1.827
10	20873	35222	53998	1.688	25736	50795	81209	1.998

3.3.11 *DSC*

Most DSC thermograms of the ten PET samples appeared fairly similar. A typical thermogram of sample 5 is shown in Figure 31.

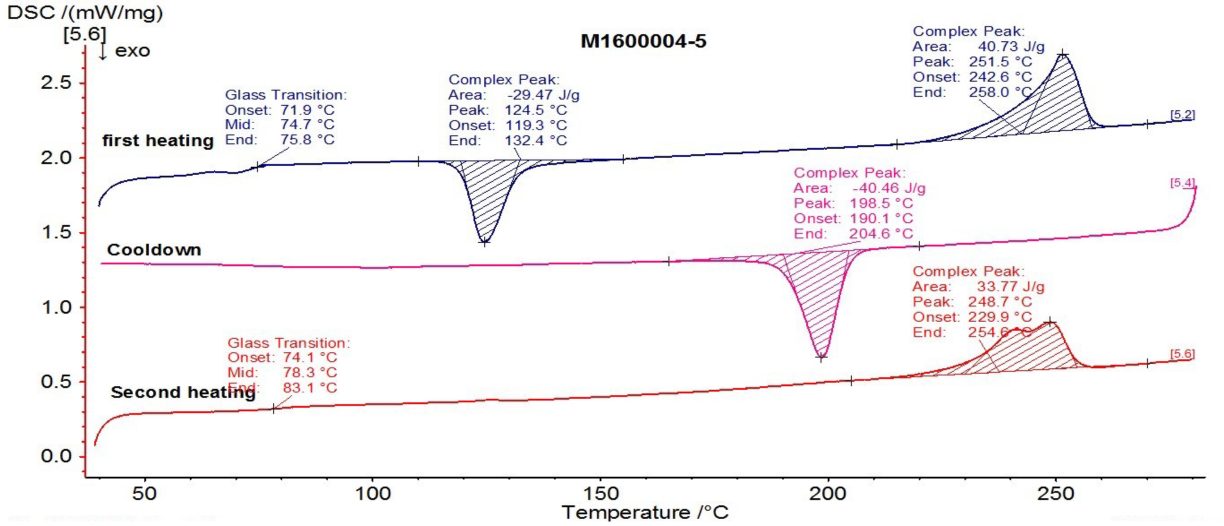

Figure 31: Thermogram of DSC measurements on 4.7 mg of sample 5.

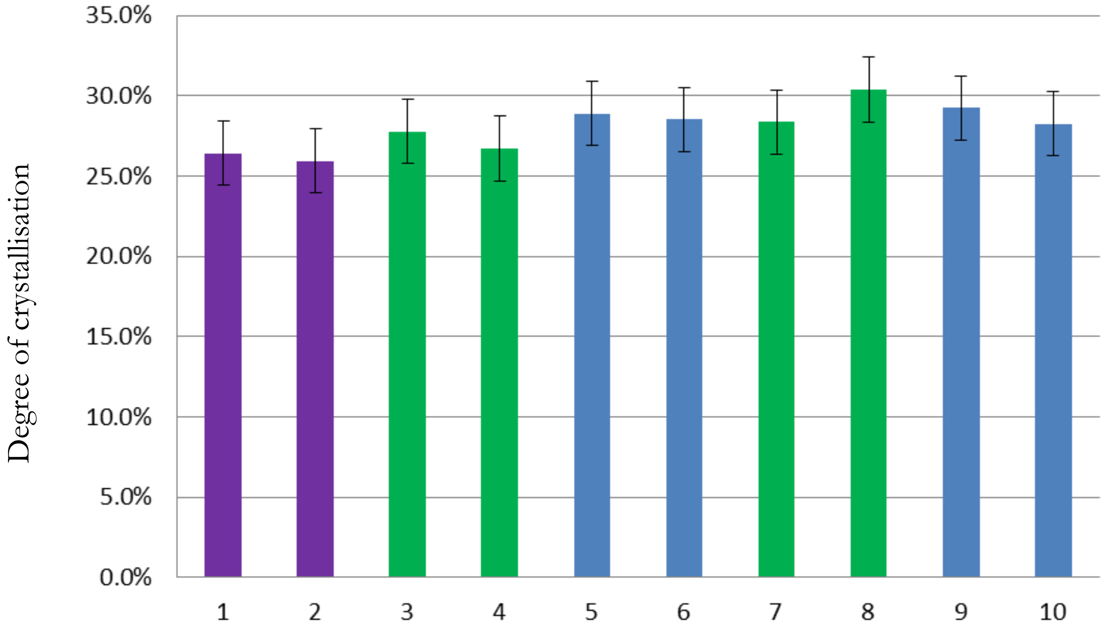

Figure 32: The degree of crystallisation of the 10 types of PET deduced from the melting peak in the second heating run. The colours indicate the collection systems: purple for deposit-refund systems, green for separate collection systems and blue for mechanical recovery systems.

The degrees of crystallisation were deduced from the area under the melting peak in the second heating run. These values were compared to the theoretical melt enthalpy for PET of 140 J/g. The derived degrees of crystallisation are shown in Figure 32. These degrees of crystallisation vary between 26 and almost 30%. The relationship between the degree of crystallisation and the origin and treatment of the samples in not very clear. But because these are single measurements, the differences are likely to be not relevant.

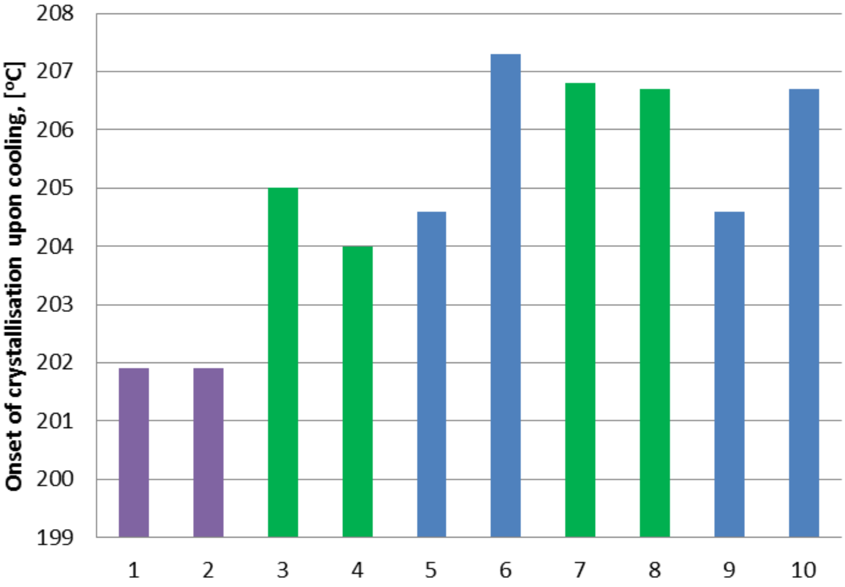

Figure 33: Onset of crystallisation derived from the cooling run [°C]. The colours indicate the collection systems: purple for deposit-refund systems, green for separate collection systems and blue for mechanical recovery systems.

The onset of crystallisation derived from the cooling run (the temperature at which the rPET starts to crystallise) is shown in Figure 33. This temperature is relevant for the injection-moulding process of the preforms. The lower this onset temperature is, the larger the chance that crystallisation during injection-moulding of the preforms is avoided. The samples originating from deposit refund systems (1 and 2) possess to lowest onset temperatures, which indicates that these samples contain the least contaminants that act as nucleating agents. All rPET samples originating from separate collection (3, 4, 7, 8) and mechanical recovery (5, 6, 9, 10) have higher onset temperatures and are hence likely to contain more contaminants that act as nucleating agents.

The onset of crystallisation during the first heating run is shown in Figure 34. This temperature is relevant for the bottle-blowing process. The higher this onset temperature is, the larger the chance that crystallisation during bottle-blowing is avoided.
There is a marked difference between the standard and the advanced process (3 versus 4 and 5 versus 6) in this onset temperature. This onset temperature is much lower for the advanced process. This could indicate that the advanced process adds particles to the PET matrix, that are invisible with the Partisol measurement, perhaps because they are smaller than 1 µm or because they dissolve in the solvent used for Partisol measurements.

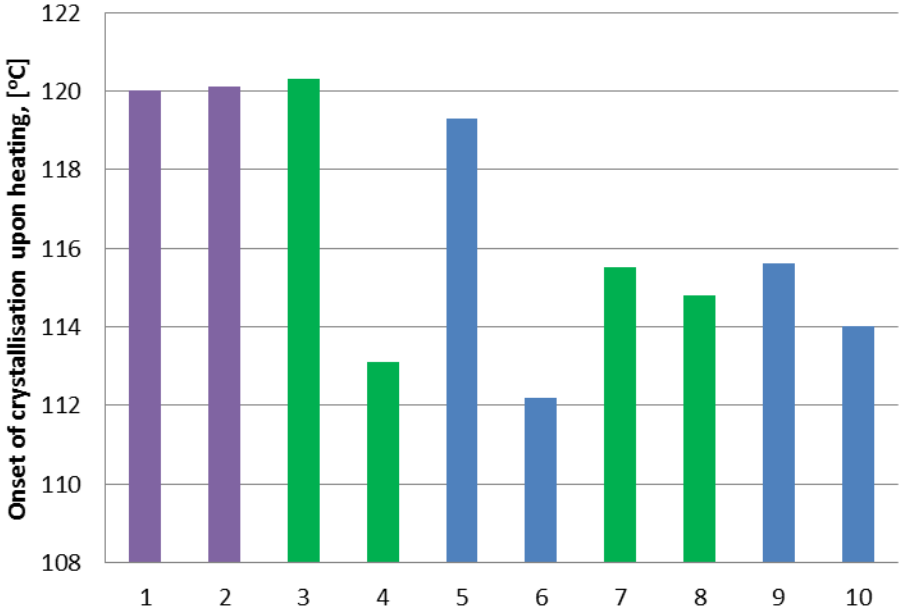

Figure 34: Onset of crystallisation upon heating the rPET, derived from the first heating run [ºC]. The colours indicate the collection systems: purple for deposit-refund systems, green for separate collection systems and blue for mechanical recovery systems.

3.3.12 *GC*

The volatile components in PET as measured using GC are listed in Table 33. Acetaldehyde amounts are in the same range as the measurements in task 2. From Table 33 it can be seen that sample 3 and 5 show elevated levels of benzene emission. This could indicate that these materials contain PVC. Benzene traces are still measurable after SSP. After further cleaning and sorting (samples 6-10) these benzene emissions drop to levels similar as PET from refund systems. This indicates that PVC can be removed using additional sorting processes. Also see paragraph 4.2.

Table 33: Results of the headspace GC analysis of the rPET granulates.

Code	GC prior to SSP, [µg/g or ppm]			GC after SSP, [µg/g or ppm]		
	Acetaldehyde	Benzene	Limonene	Acetaldehyde	Benzene	Limonene
1	18.2	0.004	0	3.0	0.0000	0
2	13.7	0.005	0	2.3	0.0000	0
3	14.7	**0.167**	0	2.9	**0.0011**	0
4	14.7	0.005	0	3.2	0.0000	0
5	16.6	**1.602**	0	3.1	**0.0009**	0
6	13.0	0.009	0	3.4	0.0000	0
7	12.7	0.004	0	2.9	0.0000	0
8	11.1	0.008	0	2.4	0.0000	0
9	11.5	0.003	0	2.5	0.0000	0
10	11.3	0.002	0	2.2	0.0000	0

3.3.13 *Hexane extraction on washed milled goods.*

The hexane soxhlet extraction of washed milled PET goods originating from mechanical recovery and treated with the standard recycling process (code 5) gave 0.3% extractable material as result. Since this process sample is considered to be the least clean, and this result was so low, no further samples were extracted with hexane. The experiment proves that sufficient hot-melt residues and fatty food residues are removed with the standard recycling process.

3.3.14 *Conclusions work package 3*

None of the rPET granulates produced in task 3 comply with the purchase conditions of major beverage producers. Also the PET products made from bottles originating from deposit refund systems with the standard recycling process do not comply with specifications such as L*>67, b*<3 and haze <15%. This contrasts with the industrial practise. Apparently subtle differences in processing conditions between the industrial processes and the laboratory process have large ramifications on the properties of the products made. Especially the moisture content of the washed milled flakes and the level of vacuum in the extruder (paragraph 2.2.3) are difficult to mimic in a laboratory. The comparison between the industrial practise and our laboratory results indicates that products made in our laboratory have a systematic lower quality than those in industry. This implies that the results of these laboratory studies cannot be used in a quantitative manner but only in a qualitative, comparative manner.

Standard recycling process

The quality of the rPET products made with the standard process from both deposit refund systems is clearly different. rPET made from SRN bottles (code 1) is darker in comparison to rPET made from bottles from the other deposit refund system (code 2) which is more yellow and has a lower intrinsic viscosity. These differences can be related to the material composition of the PET bottles in both deposit refund systems. One of the major beverage producers of which the bottles are collected via the SRN system uses reheat additive (carbon black) in their PET bottles, hence darkening the colour of the rPET produced via the SRN system and causing a higher particle contamination (Partisol count). Whereas the PET bottles in the other deposit refund system do not use reheat additive and hence possess a less dark colour and a lower Partisol count. On the other hand, in the other deposit refund system 0.6% of barrier bottles are present, which explains that this rPET is more yellow and has a slightly lower intrinsic viscosity after SSP treatment.

The rPET products made from separately collected and mechanically recovered PET bottles that have been recycled with the standard process (codes 3 and 5) are relatively low quality products in comparison to those made from deposit refund bottles with the same process (codes 1 and 2). These rPET products are darker, more yellow, more hazy, have higher levels of particle contamination and release benzene as degradation product. Additionally, the recovery of mass was much lower for separately collected and mechanically recovered PET bottles that were recycled with the standard process in comparison to the PET bottles originating from deposit refund systems.

Advanced recycling process

rPET products made from separately collected and mechanically recovered PET bottles that have been recycled with the advanced process have far better properties than the rPET products made from the same bottles with the standard recycling process (code 4 in comparison to 3 and code 6 in comparison to 5). This is apparent from properties such as colour, haze, Partisol counts and GC analysis. With respect to these properties, the rPET products made from separate collection and mechanical recovery with the advanced process are comparable to the rPET products made from deposit refund bottles with the standard process (codes 4 and 6 in comparison to 1 and 2). However, with respect to the intrinsic viscosity, molecular weights and the onsets of crystallisation in PET products, the situation is less clear. For the PET products made from the separately collected bottles with the advanced process (code 4) these properties are still reasonably well comparable to the properties of the products made from the deposit refund bottles with the standard process (codes 1 and 2). But for the PET products made from the mechanically recovered PET bottles (code 6) these properties are inferior to the properties of the products made from deposit refund bottles with the standard process (codes 1 and 2).

This result is not completely understood, since this appears to be contradictory. On the one hand the Partisol measurements and the Haze measurements indicate that samples 4 and 6 contain less particular contamination than samples 3 and 5. On the other hand the onset temperatures of crystallisation shows that samples 4 and 6 tend to crystallise faster than 3 and 5, implying the presence of more nucleating agents, in other words particular contamination.

Possible explanations could be an increase in sub–micrometre particular contamination, which would be invisible for Partisol. Alternatively, contaminants could be present that would be invisible for Partisol since they would also dissolve and would not cause light scattering.
A major difference between the standard and the advanced recycling processes is the difference in recovery of mass. The losses of material are much larger in the advanced process in comparison to the standard process, as expected due to the larger number of process steps.

System amalgamation and more precise sorting

In case PET bottles from the deposit refund systems are added to the PET bottles from the separate collection system and the mechanical recovery system and these mixtures are processed with the advanced process, rPET products are obtained that have roughly the same properties as the PET products made from the separately collected and mechanically recovered PET bottles with the advanced process (codes 7 and 9 in comparison to 4 and 6). The amounts of contaminants that originate from the separate collection system and the mechanical recovery system both as sorting mistakes and as integral bottle components are much larger in comparison the amounts of contaminants that originate from the deposit refund bottles and hence dominate the results. The recoveries of mass do improve slightly, due to the addition of the large PET bottles from the deposit refund systems.

There is hardly any impact of more precise sorting (from DKR 328-1 to DKR 325) on the properties of the PET products made (comparison between codes 7 and 8 and 9 and 10). Although the level of contaminants is reduced slightly in the feedstocks due to the more advanced sorting procedure, the amount of contaminants remaining is still fairly high. A more advanced sorting specification aimed at reducing PET bottles with a negative impact on the rPET quality (non-food flasks, barrier bottles) is expected to render a more clear positive impact on the quality.

4 Discussion

4.1 Deviations from original research plan

The project execution deviated in two instances from the original research plan (Appendix M). First of all, task 1.4 was dropped due to lack of sufficiently detailed and relevant industrial data on the variance in the composition of PET bottle products. Secondly the tasks devoted to modelling (task 2.5 and task 3.3) were cancelled, since this proved to yield insufficiently reliable quality predictions.

4.1.1 *Industrial data*

With respect to the data for the PET recycling industries only data of the Belgium Fost-Plus was made available by one of the members of the industrial board. This was compositional data of the PET bottle fraction from 2014-2015 analysed on eight different days. These samples did not contain wood, stones, paper & board or glass. The majority of the objects present were indeed PET bottles, in Table 34 some data on undesired fractions and contaminants are listed.

Table 34: Average composition of sorted PET bottle products from the Belgium Fost-Plus system in 2014-2015, based on eight different samples.

Objects	Average	Standard deviation
Non-food PET flasks	5.3%	2.4%
Trays	0.7%	0.4%
PE and PP	0.1%	0.3%
PVC	0.8%	0.5%
PS	0.01%	0.01%
Beverage cartons	0.09%	0.07%

Although these numbers give some insights in the level of impurities and their variance in time, these numbers are related to the Belgium system of separate collection of only bottles and flasks. In the Netherlands there are two different collection systems for PET bottles: a deposit refund systems for large PET bottles for water and soda beverages and a separate collection and mechanical recovery system for all other post-consumer plastic packages. Due to difference in collection systems and the likely differences in consumption behaviour these industrial values were deemed to be non-relevant for the Dutch situation. Due to the lack of industrial data no variances could be calculated for Dutch PET bottle products and task 1.4 was dropped.

4.1.2 *Modelling*

Several attempts were made to predict the properties of rPET based on the contaminants present. First the data of task 2 was analysed in terms of concentration dependant property parameters. So, for all ten deliberately added contaminants in task 2, the change in property was related to their concentration, see for example Equation 9.

Equation 9: Example of calculating the concentration dependant property parameters.

$$A_{PVC}^{b*} = \frac{b^*_{PVC-task2} - b^*_{reference-task2}}{C_{PVC\ added\ in\ task\ 2}} = \frac{1.28 - 1.17}{0.001} = 110$$

For each contaminant studied in task 2 the concentrations present in the samples of task 3 were estimated based on the data in Table 31 and a list of estimated removal efficiencies for both recycling processes (Table 35). This gave the estimated concentrations of the contaminants present the samples of task 3 after recycling. Subsequently an additive array-calculation was preformed of all contaminants, in which the estimated concentrations after recycling were multiplied with the concentration dependant parameters.

Table 35: Estimated removal efficiencies for both mechanical recycling processes used to convert the compositional data prior the recycling to the compositional data after recycling.

Process	Barriers	PE	PP	PVC	PS	Residual plastics	Residual waste
Standard	0%	95%	95%	0%	0%	0%	95%
Advanced	90%	99%	99%	90%	90%	90%	99%

The relation between measured property in task 3 and predicted, calculated property from the model was poor, see for example Figure 34.

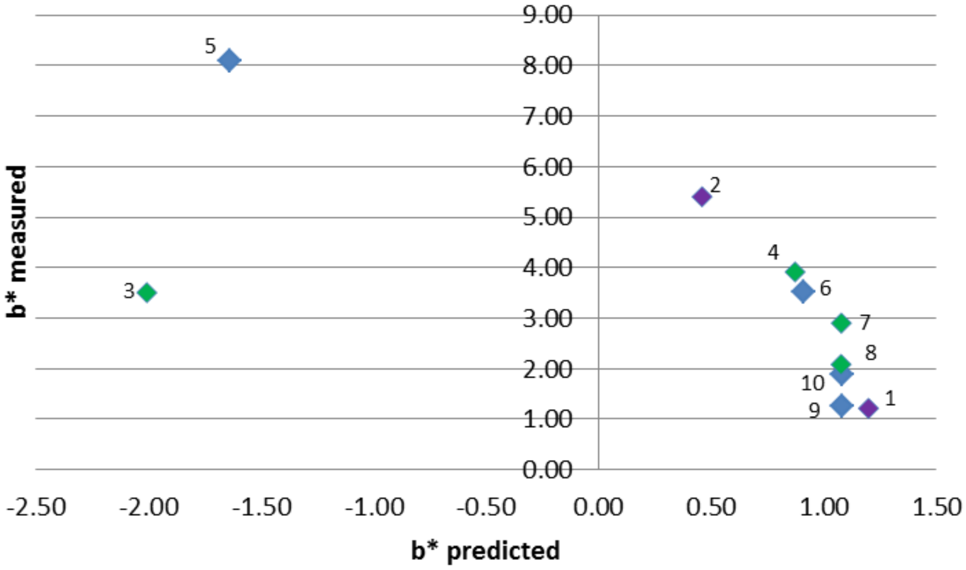

Figure 35: example of modelling b* with the derived property parameters and the concentrations of contaminants present in the samples of task 3 after recycling. The colours indicate the collection systems: purple for deposit-refund systems, green for separate collection systems and blue for mechanical recovery systems.

The poor agreement between the modelled properties and the measured properties are attributed to the following aspects:

1. The error in the derived concentration-dependant property parameters is relatively large due to the fact that change in parameter is relatively small for the contaminants studied.
2. The actual concentration in the samples after recycling will probably have differed from the concentration predicted.
3. The assumed linear relationships between the concentration of contaminants and the properties is probably too simplistic and incorrect.
4. The division between the type of barriers used is unknown. In task 1 the total amount of barrier bottles was studied, whereas in task 2 three different barriers were studied as contaminants that often yielded opposing effects on properties.
5. It is likely that contaminants have synergetic impacts on the properties, whereas this model suggests that all influences of contaminants on the quality of rPET are independent.

Based on these poor modelling results, it was decided that this activity had no added-value and was cancelled. It was decided to analyse the impact of contaminants in a different manner. To predict the concentrations in the recycled rPET and to try and find correlations between those concentrations and the measured properties.

4.2 Correlation analysis

The concentrations of contaminants present in mechanically recycled rPET were estimated from their concentrations in the feedstocks (Table 31) and crudely estimated removal efficiencies for both recycling processes. This is similar as done for the modelling in 4.1 but the list of contaminants is slightly different, see Table 36.

Table 36: Estimated removal efficiencies for both mechanical recycling processes used to convert the compositional data prior the recycling to the compositional data after recycling.

	Standard	Advanced
PET	0%	0%
PP	95%	99%
PE	95%	99%
PS	0%	90%
PVC	0%	90%
Paper	95%	99%
Metal	99%	99.9%
Glass	0%	90%
PA/POM/ABS, black, etc.	0%	90%
Silicon	0%	90%
Undefined/rest, incl. textiles	95%	99%

The estimated concentrations of contaminants present in the samples of task 3 are listed in Appendix L. Several attempts were made to correlate the properties of the rPET samples measured in task 3 to these concentrations. Three reasonable correlations were found between:

1. Haze and the total concentration of contaminants present,
2. Lightness value L* and the total concentration of contaminants present,
3. Benzene emission and PVC present,

The correlation between the measured haze for the samples from task 3 and the total concentration contaminants estimated to be present in these samples is shown in Figure 36. Linear regression between both parameters shows a r^2 of 0.91, which is reasonable good given the estimations made.

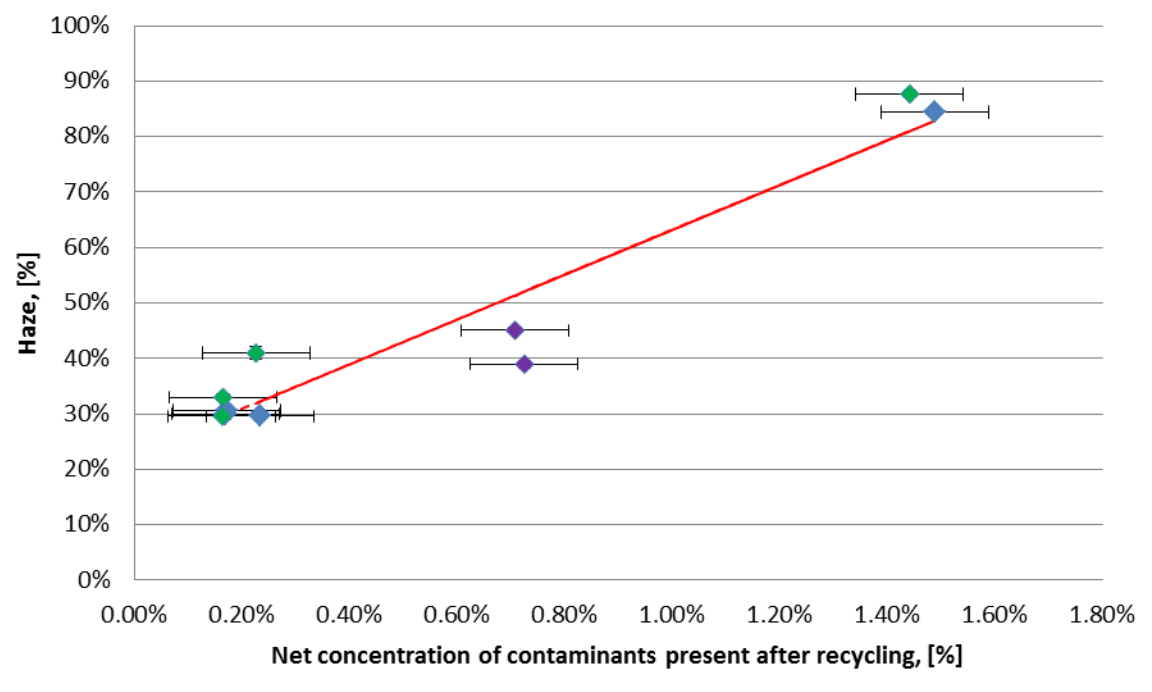

Figure 36: The correlation found between the measured haze for the samples of task 3 and the total concentration of contaminants estimated to be present in these samples. The colours indicate the collection systems: purple for deposit-refund systems, green for separate collection systems and blue for mechanical recovery systems.

The correlation between the measured lightness value (L*) for the samples from task 3 and the total concentration contaminants estimated to be present in these samples is shown in Figure 37. A reasonable correlation was found, with one outlier. This data point is of sample 2 from task 3 (the other deposit refund system). Since this sample is the only one without reheat additive, which is known to have a large impact on the lightness value, this could indeed be treated as an outlier. Once removed, a linear relation with a correlation of r^2 is 0.91 was found, which is again reasonable good given the estimations made.

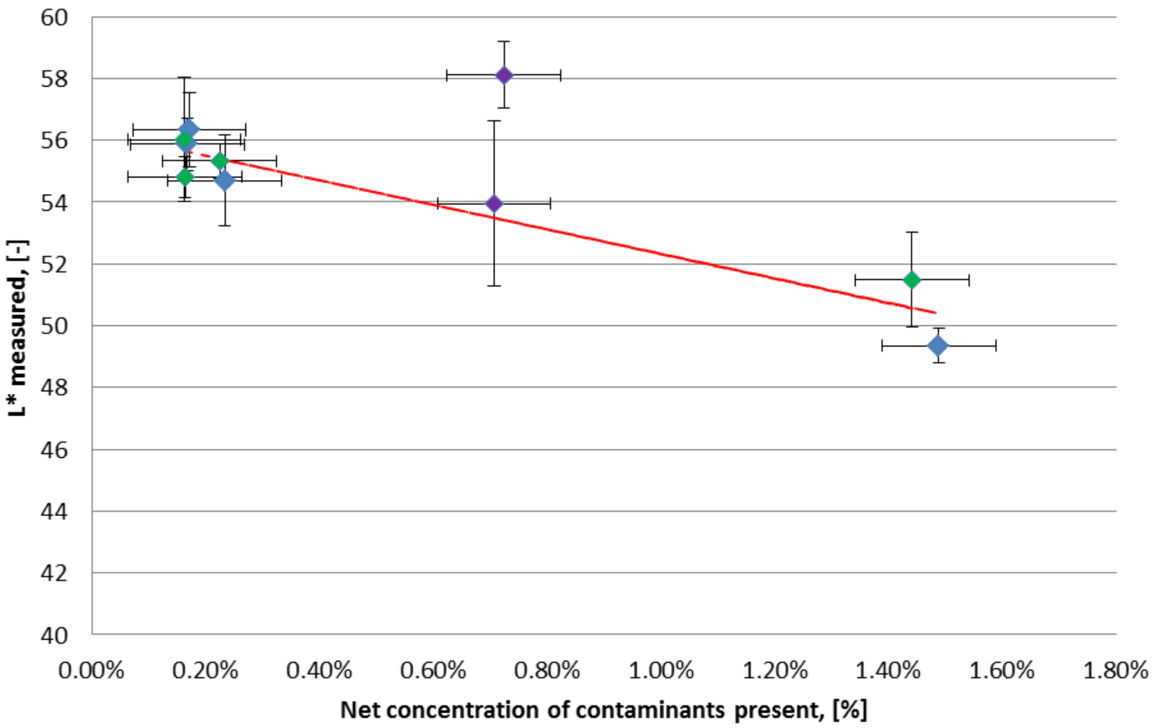

Figure 37: The correlation found between the measured L* lightness value for the sample of task 3 and the total concentration of contaminants estimated to be present in these samples. The colours indicate the collection systems: purple for deposit-refund systems, green for separate collection systems and blue for mechanical recovery systems.

Also a very good correlation could be found between the amount of benzene vapour present in the headspace over the rPET granulates after SSP of task 3 and the PVC concentration in these samples. The regression coefficient was even 0.997.

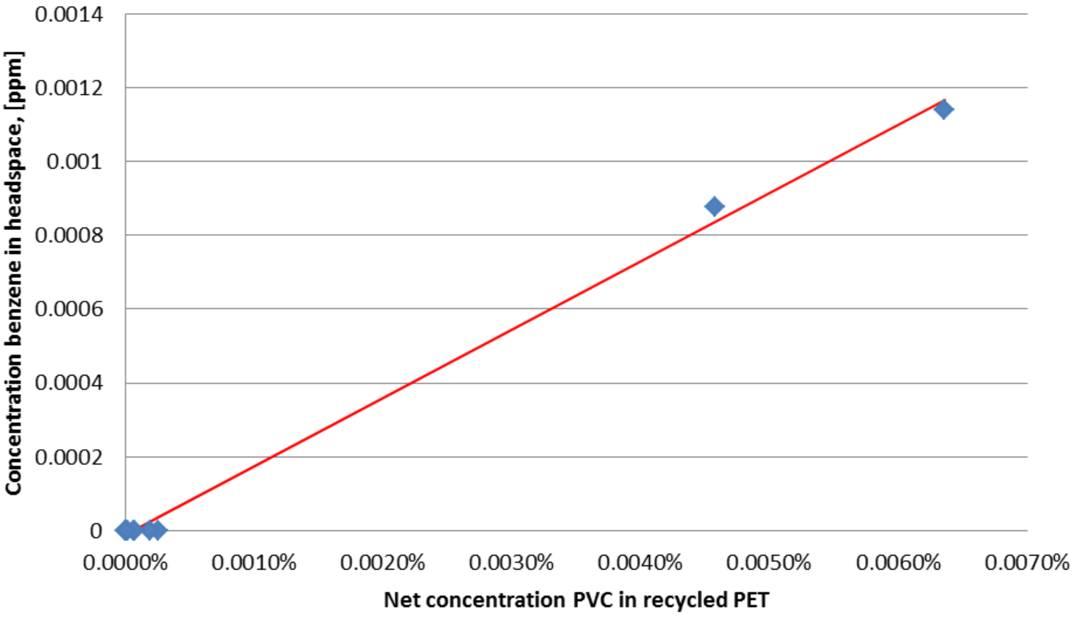

Figure 38: The correlation found between the measured benzene emission values in the headspace after SSP treatment in the samples of task 3 and the estimated concentration of PVC in these samples.

Finally, the data of task 2 and task 3 were combined to find correlations between technical parameters. Such a correlation was found for amount of particles measured with Partisol and the haze measured. Since, the property haze is by definition equal to the amount of light scattered, this relation was not surprising.
A reasonable mathematical correlation was the function as described below.

$$Haze = A \times PC^{B}$$

Parameter	**Meaning**	**Unit**
Haze	Haze-value	[%]
PC	Partisol count	[#/10000 images]
A	Pre-factor = 0.004 ± 0.002	
B	Power-factor = 0.41 ± 0.07	[-]

The correlation only gave a correlation coefficient of 0.64. This low correlation can be attributed to a few clear outliers, such as the data points for PS and PE from task 2. Apparently these samples generate more haze than what would be expected only on the amount of particles present in the rPET. Furthermore, since some contaminants will dissolve in the Partisol-solvent, a complete correlation was also not expected. In case only the data-points from task 3 are used, a much better correlation was found with a coefficient of 0.966. Then the pre-factor amounted 0.008 ± 0.001 and the power-factor amounted: 0.34 ± 0.02.

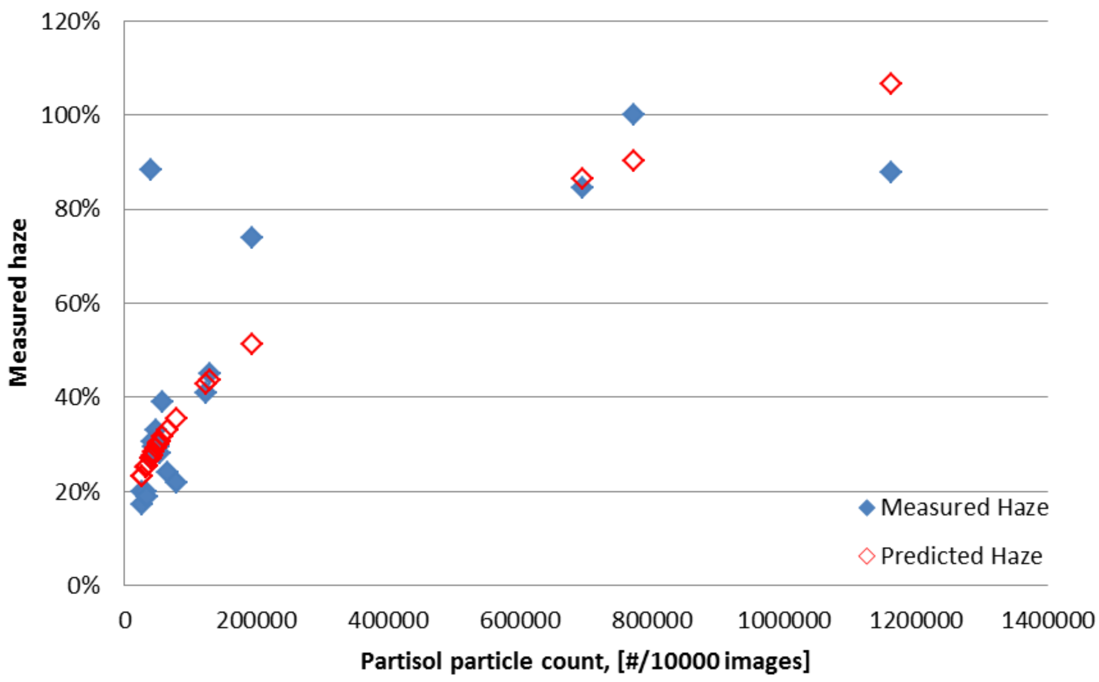

Figure 39: correlation between the measured haze values for the samples of task 2 and 3 and the measured partisol counts for the same samples.

4.3 Recommendations for stakeholders

The recommendations towards the stakeholders have been written from the perspective of maximising the amount of recycled PET at a high quality level.

The major hurdle for closed loop recycling (bottle-to-bottle) of Dutch PET bottle feedstocks from source separation and mechanical recovery systems is the large content of PET non-food flasks. In PET bottle products from these systems this amounts to 17-24%, whereas 5% is the legal limit set by EFSA for food grade recycling. Although reviewer Welle argues that this 5% limit is not evidence-based, but rather a precautionary measure of EFSA, it is nevertheless, still the current reality that mechanical recyclers have to comply with in order to produce food-grade rPET. Since it is impractical to sort these non-food-flasks manually at sorting facilities, there is a great need for mechanical sorting processes which would allow the automatic identification and mechanical removal of non-food flasks at high speeds. Hence, several companies and research groups investigate the use of tracer technology. We are aware of three initiatives: Polymark[21], Prism[22] and rare-earth metals (Bezati et al. 2011). The choice for a marking technology and the marking code should preferably be made on a supranational (European) level since PET bottle products are traded internationally. Since every tracing technology has its own unique sets of pros and cons for each group of stakeholders making this choice a difficult one. Critical technical demands are the speed of recognition (milliseconds or seconds), the accuracy of subsequent removal and the level at which the tracer can be removed during the mechanical recycling process (to avoid faulty sorting in future recycling cycles). In case such a marking technology is implemented, this technology could potentially also be used for the automatic recognition of PET-bottles with undesired components (design related contaminants) and for non-bottle PET packages. Hence, the most important recommendation to stakeholders is to select and implement such a tracer technology as soon as possible.

The implementation of such a tracer technology in the PET recycling chain will have major ramifications throughout the chain. New sorting specifications will emerge, which will reflect the new technological possibilities and demand less than 5% of non-food flasks and perhaps even less than 1% of non-bottle PET, etc. Furthermore, some organisation needs to decide on the actual use of the tracers for individual packages. This can encourage brand-owners further to comply with the EPBP design guidelines.

Once the issue of automatic sorting non-food flasks is solved, the control of contaminants in rPET is the most important remaining issue. Contaminants can have profound negative impacts on the rPET quality when insufficiently managed. They originate from either bottle-components (design related) or from sorting faults. The resilience of recycling systems varies for different contaminants. Some contaminants can easily and efficiently be removed (such as PE and PP) with standard separation processes such as float-sink-separation. Other contaminants cannot be

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[21] *http://www.polymark.org/*

[22] *'Gone with a trace' Plastics in Packaging 174, April 2016 p20-21*

removed with standard separation processes and require additional separation techniques such as sieving, flake-sorting and colour sorting technologies (such as PVC, PS, POM, glass, Silicone, barriers, etc.). Once non-food-PET-flasks are sufficiently removed from the feedstocks, the concentration of some of these contaminants (glass, POM) will lower as well. However, other contaminants such as barriers, metal, silicone rubber, ink, PS and PVC will remain present in the feedstock, as these originate from either other PET bottle categories or from sorting faults.
One of the most critical contaminants appears to be PVC. In the Dutch situation, PVC only originates from sorting faults, since no PET bottles with PVC components were found on the Dutch market. The PVC sorting faults found in PET bottle products were mostly PVC non-packaging objects and some PET meat trays with PVC stretch films. It is unlikely that the presence of PVC non-packaging objects can easily be reduced further, with sorting instructions to civilians. The use of PVC based stretch wrap in combination with PET trays for meat and meat products could be discouraged. Nevertheless, most effort to reduce the concentration of PVC in rPET should be focussed on the sorting facilities and recycling facilities.

EVOH is a barrier material (packaging component) of which the precise use in the Netherlands is unclear. The list of manufacturers that bring PET-barrier bottles on the Dutch market is limited and direct persuasion between authorities and these producers appears the best approach to limit the use of especially EVOH but preferably all barriers. Although several food products cannot be packaged in PET bottles without a barrier (wine, beer) and still have sufficient shelf life. A potential solution for the recycling industry is to use marking technologies also for PET bottles and flasks with barriers and other components that are not compatible for the recycling industry. The other critical contaminants (silicone rubber, POM and glass) originate from both sorting faults and bottle design. Such contaminants can only be managed with an integrated approach (design, sorting & recycling process).
Optionally, PET flasks with undesired packaging components could be redesigned to have bottle body's made out of PE instead. Contamination with heavy foreign plastics (POM, ABS, Silicon rubber) is less of a problem for PE recycling than for PET recycling since the majority of these contaminants can be removed with a sink-float separation.

The quality of rPET remains a complicated shared responsibility of beverage manufacturers (bottle designs), collection services (collection portfolio), sorting facilities (specifications) and recycling facilities (recycling techniques and process configuration) as shown in Figure 1. Once marking technology has been implemented to allow for an automatic sorting of the non-food PET flasks and additional design improvements have been implemented (as discussed above) the relative contribution of contaminants will partially shift towards sorting faults and the focus will hence also shift towards more stringent sorting specifications (e.g. DKR 325 instead of DKR 328-1).
In the end, the quality of rPET is a multi-stakeholder responsibility with strong interactions between the efforts done by the various stakeholders. No matter what type of collection & recycling system will be developed in the future, the individual responsibilities in relation to the

total system achievement (rPET quality) will need to be managed to be able to achieve high qualities of rPET in high volumes.

In summary the recommendations to the stakeholders, from the perspective of maximising the amount of recycled PET at a high quality level, are:

1. To select and implement a tracer technology to allow for the fast, mechanical sorting of non-food PET flasks from PET bottle products,
2. To simultaneously use this tracer technology to mark other PET packages and simplify the automatic sorting of PET barrier bottles, opaque PET packages, PET trays etc.,
3. To actively encourage the complete chain[23] to comply with the design guidelines of EPBP,
4. To continue encouraging sorting facilities to produce PET bottle feedstocks with a minimum amount of sorting faults and residual waste components and to develop new stricter specifications that are in agreement with the newly implemented tracer technology,
5. To study and redesign PET non-food-flasks with hand pumps and spray guns in such a manner that less or preferably no glass, metal, POM, silicon rubber, ABS or any other plastic with a density of more than 1 g/ml are being introduced in the recycled material.

4.4 Recommendations for scientists

For scientists which would like to study the mechanical recycling of PET bottles and the impact of contaminants on this recycling system (or other plastic recycling systems in general) we have several advices:

1. To include virgin PET material in laboratory mechanical recycling experiments as an internal standard to assess the impact of laboratory set-up on the plastic quality,
2. To study the impact of reheat additive on the properties of rPET,
3. To perform multiple measurements to assess the impact of contaminants on the properties of rPET, both repetition measurements and measurements at different concentrations,
4. To perform contaminant-impact-studies first on virgin PET and then secondly on rPET, so a more clear indication of the impact of the contaminant is obtained and subsequently also an idea is given of this impact in a much more complex matrix such as rPET,
5. To assess the impact of drying PET milled goods on the quality of rPET in more detail.
6. We would like to encourage other scientists to adopt the applied method of compositional analysis, which is based on the object-wise sorting of plastic feedstocks in packaging categories and the material analysis per packaging category. With this method the origin of contaminants can be established.

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[23] *With the complete chain is referred to all companies that influence the design of PET packages, like design agencies, production companies, purchase offices, sales offices and retailers.*

5 Conclusions

The technical quality of recycled PET made from sorted Dutch PET bottles has been studied in a scientific comparative manner. First of all, the composition of the sorted PET bottle products has been analysed in detail on object-level in terms of categories of packages and on component-level in terms of average material composition per category of packages. This approach not only rendered information on the material composition of sorted PET bottle products but also gave information on the origin of contaminants; either being design related (originating from PET bottle / flask components) or being sorted faults. Major differences in the composition of PET bottle products were found especially between PET bottle products from different collection systems. But even between the PET bottle products from two different Dutch deposit refund systems some difference was found; one containing 0.2 % coloured bottles and no barrier bottles and the other containing 4.9% coloured bottles and 0.6% barrier bottles. In general, however, PET bottle products from Dutch deposit refund systems contained few contaminants, since the design of nearly all the bottles complied with the EPBP design guidelines and the products contained small amounts of sorting faults. Dutch PET bottle products that originate from separate collection and mechanical recovery contained much more contaminants and also non-food PET flasks, barrier bottles, opaque PET bottles and non-bottle PET. A large fraction of the PET bottles and flasks present do not comply with the EPBP design guidelines and introduce contaminants. Furthermore, these PET bottle products also contain faulty sorted objects (packages from different plastics and other materials).

Secondly, the impact of contaminants on the quality of rPET was studied systematically. rPET was made from clean beverage bottles by milling, extrusion and SSP treatment. Ten different likely contaminants were added prior to extrusion in concentrations that were considered relevant. The impact of these contaminants on the quality of the rPET was monitored with various analysis methods: colour, haze, particle contamination, DSC, DMTA, IV, GPC and GC. Almost all contaminants were found to affect the quality of rPET in some manner negatively, yielding discoloration, particle contamination, haze formation, shortening of the average polymeric chain lengths and subtle alteration of the thermal properties. The precise impacts were strongly dependant on the type of contaminant tested. Three contaminants were found to impact the PET in a special manner. EVOH caused cross-linking with PET, which could possibly result in gel-formation. PVC was found to yield benzene vapour, most likely due to an acid-catalysed degradation of PET. Hotmelt glue residues were found to strongly discolour the rPET yellow.

Thirdly, the PET bottle products were mechanically recycled into rPET granulates with two processes, a straight-forward standard process and an advanced process in a systematic and comparative manner. The quality of the rPET produced was analysed with the same analytical techniques as in task 2. Fairly good qualities of rPET could be obtained from deposit refund PET bottles with the standard process, in terms of colour, haze, particle contamination, molecular chain lengths and thermal properties. One type of deposit refund system yielded darker rPET, which could be attributed to the use of reheat additive by one of the beverage producers. The other type of deposit refund system yielded more yellow rPET which could be attributed to the use of about 0.6% of barrier bottles. PET bottle products originating from separate collection and mechanical recovery that were recycled with the standard process yielded an inferior quality of rPET. This could be attributed to the higher concentration of contaminants present and the inability of the standard process to remove these sufficiently. The same PET bottle products could, however, be recycled with the advanced process to rPET qualities that were far better. These qualities of rPET are in some quality aspects comparable to or better than the rPET made from deposit refund systems with the standard process; haze, colour, particle contamination. But in other quality aspects (molecular weights and crystallisation behaviour) slightly less good. This is not completely understood yet and could possibly be influenced by the laboratory equipment and procedures used for these comparative tests.

Nevertheless, these tests did show that the amount of contaminants in rPET can be controlled with the advanced recycling process and a relatively high quality rPET can be attained. Although the net recovery of mass and the net PET yield are much lower for the advanced process in comparison to the standard process. These research results verify the hypothesis (page 15) that the needed effort at the end of the recycling chain (sorting and recycling) is strongly dependent on the effort that has been made at the start of the recycling chain (collection method and design of the present bottles).

Additionally, several PET bottle products were recycled that would mimic the composition after a potential merger of collection systems (adding the PET bottles from the deposit refund systems to the separate collection and mechanical recovery systems). When these PET bottle products were recycled with the advanced process rPET recyclates with similar quality aspects were obtained, only showing small improvements in colour and haze. Finally also the impact of more intense sorting (from DKR specification 328-1 to 325) on the quality of the produced rPET was tested and this impact proved to be small. The likely explanation is that although this increased sorting effort reduces the level of contaminants originating from faulty sorted objects, still more than sufficient remain that originate from the PET bottles themselves.

In the end, the quality of rPET is influenced by multiple stakeholders with strong interactions between the efforts done by the various stakeholders. No matter what type of collection & recycling system will be developed in the future, the individual responsibilities in relation to the total system achievement (rPET quality) will need to be managed to be able to achieve high qualities of rPET.

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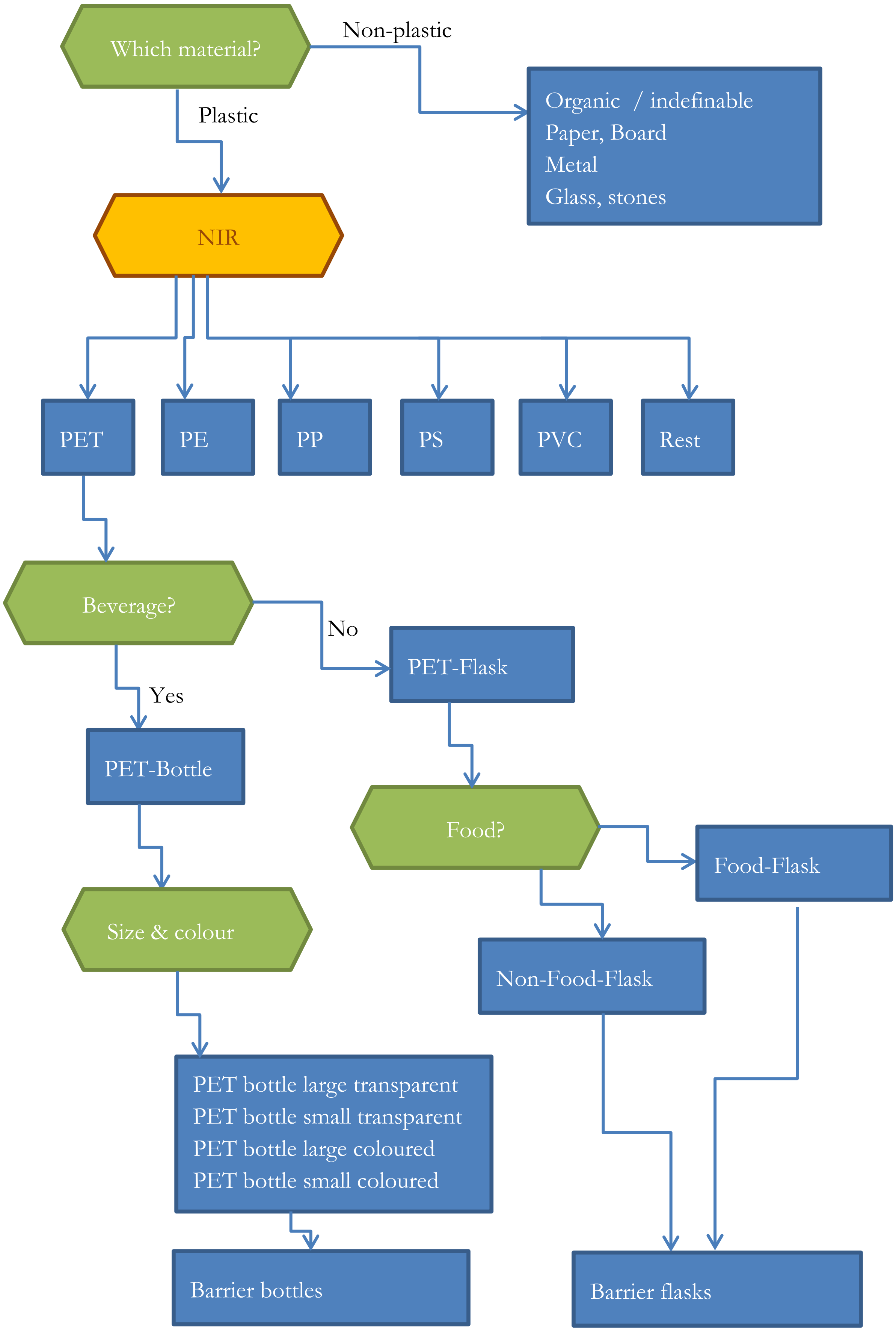
Appendix A: Glossary

Mechanical recovery	Mechanical recovery of plastics from municipal solid refuse waste. Post sorting systems, integral collection systems. Dutch: Nascheiding German: Mechanische Kunststoffabtrennung bei MBT-anlagen
Separate collection	The collection of plastic packaging waste, that is separated by consumers. E.g. Plastic Heroes collection. Dutch: Gescheiden inzameling / bronscheiding German: Getrennte Erfassung
Deposit refund system	Dutch: Statiegeldsysteem German: Pfandsystem

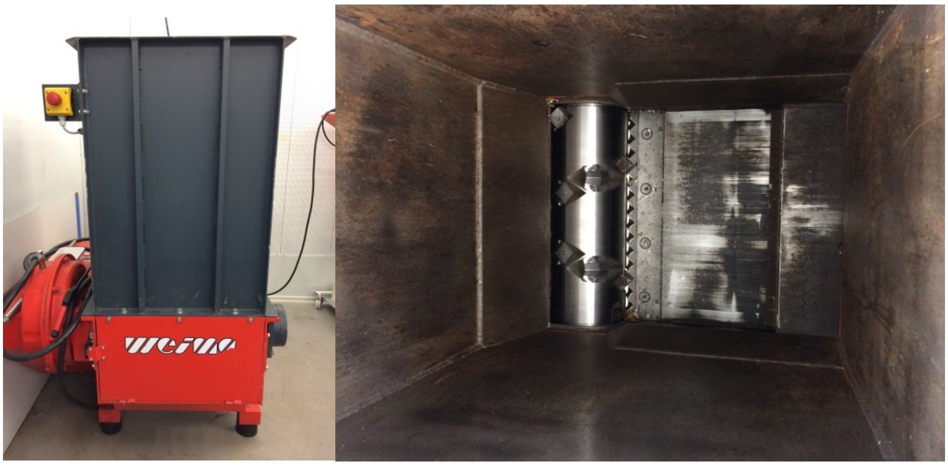
Abbreviations

DEG	Diethylene glycol
DKR	Deutsche Gesellschaft für Kreislaufwirtschaft und Rohstoffe mbH
DMTA	Dynamic mechanical thermal analysis
DSC	Differential scanning calorimetry
EPBP	European PET Bottle Platform
EVOH	Ethylene vinyl alcohol
GC	Gas chromatography
GPC	Gel partitioning Chromatography
HDPE	High Density Polyethylene
IR	Infrared
IV	Intrinsic viscosity
LDPE	Low Density Polyethylene
NIR	Near infrared
PA	Polyamide
PE	Polyethylene
PE	Polyethylene terephtalate
PLA	Polylactic acid
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinylchloride
SRN	Stichting retourverpakkingen Nederland
SSP	Solid State Poly Condensation

Appendix B: Sorting decision tree PET bottle fractions

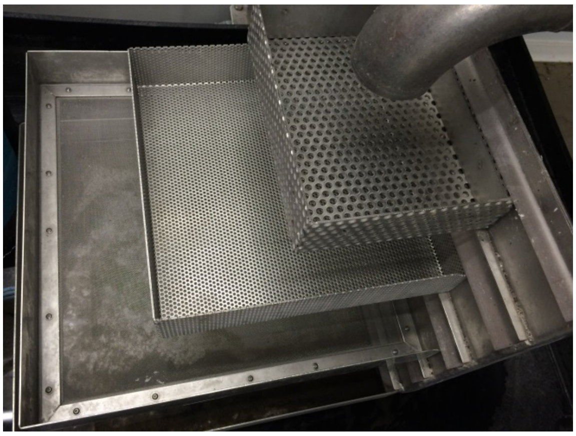


Appendix C: Photos of lab-equipment

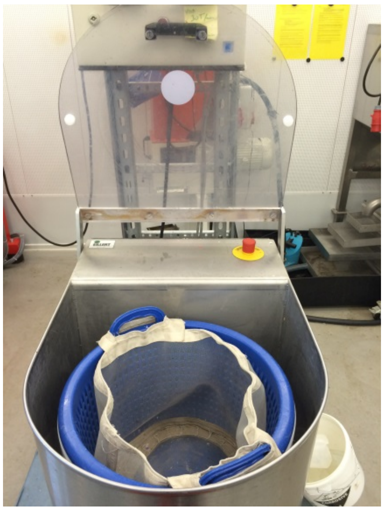


Weima schredder (outside and inside)

Washing mill (outside and inside) , including heater. Heater is removed before washing process.



Sieving plates (for washing water)



Centrifuge

Appendix D: Detailed composition of packaging types

PET bottle clear transparent ≤ 0,5 L (n=114)

	PET	PP	PE	PS	Paper	Metal	PA
Weighted mass average	84.9%	3.5%	10.2%	0.5%	0.6%	0.1%	0.1%
Standard deviation	3.9%	4.4%	5.5%	1.6%	1.5%	0.9%	1.0%
Min	73.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%
Max	90.9%	17.0%	20.5%	8.3%	6.3%	6.8%	12.0%

PET bottle clear transparent > 0,5 L (n=42)

	PET	PP	PE	PS	Paper	Metal
Weighted mass average	90.1%	2.3%	6.5%	0.2%	0.8%	0.0%
Standard deviation	2.9%	2.5%	2.9%	1.1%	2.1%	0.1%
Min	82.9%	0.0%	0.0%	0.0%	0.0%	0.0%
Max	93.6%	10.8%	13.4%	7.2%	8.0%	0.6%

PET bottle coloured ≤ 0,5 L (n=16)

	PET	PP	PE	PS	Paper	Metal
Weighted mass average	87.7%	1.4%	9.8%	0.4%	0.3%	0.4%
Standard deviation	3.6%	2.4%	4.1%	1.7%	0.9%	1.6%
Min	79.7%	0.0%	1.2%	0.0%	0.0%	0.0%
Max	91.5%	9.2%	17.7%	7.0%	3.0%	6.7%

PET bottle coloured > 0,5 L (n=11)

	PET	PP	PE	Paper
Weighted mass average	91.6%	1.5%	5.8%	1.1%
Standard deviation	3.7%	1.5%	3.2%	2.3%
Min	80.8%	0.0%	0.3%	0.0%
Max	95.5%	4.3%	12.4%	6.8%

PET bottle opaque ≤ 0,5 L (n=5)

	PET	PP	PE
Weighted mass average	75.5%	5.0%	19.4%
Standard deviation	5.3%	3.6%	5.1%
Min	72.0%	1.7%	14.5%
Max	83.8%	10.5%	25.7%

PET food flask (n=39)

	PET	PP	PE	PS	Paper	Silicon
Weighted mass average	79.2%	15.8%	2.7%	0.1%	2.1%	0.1%
Standard deviation	8.0%	11.3%	4.0%	0.6%	2.5%	0.3%
Min	58.5%	0.0%	0.0%	0.0%	0.0%	0.0%
Max	91.2%	38.4%	12.7%	4.5%	11.7%	1.4%

PET non-food flask (n=36)

	PET	PP	PE	PS	Paper	Metal	Glass	Silicon	Other plastics (PA, POM, etc.)
Weighted mass average	76.9%	17.6%	3.8%	0.1%	1.3%	0.3%	0.0%	0.0%	0.1%
Standard deviation	8.9%	9.2%	5.2%	1.0%	1.6%	0.7%	0.1%	0.0%	0.3%
Min	58.6%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%
Max	90.0%	35.1%	33.1%	6.4%	5.2%	2.9%	0.4%	0.1%	1.2%

Appendix E: Materials/impurities found in PET bottle categories

	N	# of bottles with barrier (discoloured in oven)	# of bottles with ink on body	# of bottles with part of metal	# of bottles with part of glass	# of bottles with part of other plastics (PA, silicone, ABS, POM, etc.)	# of bottles with part of PS
PET bottle clear transparent ≤ 0,5 L	114	11	50	4	0	1	10
PET bottle clear transparent > 0,5 L	42	1	14	1	0	0	1
PET bottle coloured ≤ 0,5 L	16	Not tested	4	1	0	0	1
PET bottle coloured > 0,5 L	11	Not tested	4	0	0	0	0
PET bottle opaque ≤ 0,5 L	5	Not tested	4	0	0	0	0
PET food flask	39	2	13	0	0	8	1
PET non-food flask	36	0	9	6	5	3	1

Appendix F: Processing conditions washing steps in mechanical recycling process

	Process conditions washing steps						
	Prewash		Hot wash				
	Temperature at start [°C]	Temperature after washing [°C]	Temperature at start [°C]	Temperature after washing [°C]	Caustic soda [M]	Conductivity, before adding PET [mS/cm]	Conductivity, after washing PET [mS/cm]
1. SRN deposit, standard	52.5 +/- 1.6	51.5 +/- 1.6	80.5 +/- 0.4	76.7 +/- 0.1	0.1	35.4 +/- 0.8	35.2 +/- 1.1
2. Other deposit, standard	49.3 +/- 1.9	48.5 +/- 2.6	80.6 +/- 0.1	76.6 +/- 0.2	0.1	38.8 +/- 0.4	37.6 +/- 0.4
3. Source separated, standard	51.7 +/- 1.2	50.3 +/- 1.3	80.5 +/- 0.3	76.5 +/- 0.6	0.1	39.0 +/- 0.3	37.8 +/- 0.1
4. Source separated, advanced	50.9 +/- 1.3	50.1 +/- 1.6	80.7 +/- 0.0	77.0 +/- 0.1	0.1	38.2 +/- 1.5	37.0 +/- 1.1
5. Recovered, standard	51.8 +/- 1.1	50.8 +/- 0.7	80.5 +/- 0.1	76.8 +/- 0.3	0.1	37.4 +/- 0.6	36.3 +/- 0.4
6. Recovered, advanced	52.2 +/- 1.4	51.3 +/- 1.3	80.4 +/- 0.4	76.5 +/- 0.4	0.1	38.9 +/- 0.1	37.7 +/- 0.1
7. Source separated + deposit, advanced	51.2 +/- 0.2	50.4 +/- 0.0	80.4 +/- 0.5	76.7 +/- 0.7	0.1	39.1 +/- 0.2	37.9 +/- 0.1
8. Source separated + deposit, DKR325, advanced	53.0 +/- 1.0	52.1 +/- 0.8	80.7 +/- 0.0	77.2 +/- 0.4	0.1	39.2 +/- 0.5	38.2 +/- 0.5
9. Recovered + deposit, advanced	51.8 +/- 0.4	50.6 +/- 0.3	80.5 +/- 0.1	76.8 +/- 0.2	0.1	39.5 +/- 0.4	38.5 +/- 0.6
10. Recovered + deposit, DKR325, advanced	54.3 +/- 2.4	53.0 +/- 2.0	80.8 +/- 0.2	77.3 +/- 0.8	0.1	38.8 +/- 1.5	38.3 +/- 0.8
Average	51.8	50.8	80.5	76.8	0.1	38.4	37.4

Appendix G: Material content per packaging category

	PET [%]	**PP [%]**	**PE [%]**	**PS [%]**	**PVC [%]**	**Paper [%]**	**Metal [%]**	**Glass [%]**	**PA/POM/ ABS,etc [%]**	**Silicon [%]**	**Ondefin./rest [%]**	
PET bottle transparant ≤ 0,5 liter	85%	4%	10%	1%	0%	0%	0%	0%	0%	0%		Project measurements
PET bottle transparant ≤ 0,5 liter + Barrière	85%	0%	11%	0%	0%	2%	1%	0%	0%	0%		Project measurements
PET bottle lightblue ≤ 0,5 liter	87%	1%	11%	0%	0%	0%	0%	0%	0%	0%		Project measurements
PET bottle coloured ≤ 0,5 liter	88%	1%	10%	0%	0%	0%	0%	0%		0%		Project measurements
PET bottle coloured ≤ 0,5 liter + Barrière	88%	1%	10%	0%	0%	0%	0%	0%		0%		Project measurements
PET bottle opaque ≤ 0,5 liter	76%	5%	19%	0%	0%	0%	0%	0%		0%		Project measurements
PET bottle transparant > 0,5 liter	90%	2%	6%	0%	0%	1%	0%	0%		0%		Project measurements
PET bottle transparant > 0,5 liter + Barrière	90%	1%	8%	0%	0%	0%	0%	0%		0%		Project measurements
PET bottle lightblue > 0,5 liter	90%	2%	6%	0%	0%	1%	0%	0%		0%		Project measurements
PET bottle coloured > 0,5 liter	92%	2%	6%	0%	0%	1%	0%	0%		0%		Project measurements
PET bottle coloured > 0,5 liter + Barrière	92%	2%	6%	0%	0%	1%	0%	0%		0%		Project measurements
PET bottle opaque > 0,5 liter	92%	2%	6%	0%	0%	1%	0%	0%		0%		Project measurements
Bottle PE	0%	0%	96%	0%		4%	0%					Data from W2P project
Bottle PP	0%	92%	4%			4%						Estimate
Bottle PS			4%	94%		2%						Estimate
Bottle other			4%		92%	4%						Estimate
Flask PET food	79%	16%	3%	0%	0%	2%	0%	0%		0%		Project measurements
Flask PET food + Barrière	76%	20%	0%	0%	0%	3%	0%	0%		1%		Project measurements
Flask PET non-food	77%	18%	4%	0%	0%	1%	0%	0%	0%	0%		Project measurements
Flask PE	0%	0%	96%	0%	0%	4%	0%	0%	0%	0%		Estimate
Flask PP	0%	0%	4%	94%	0%	2%	0%	0%	0%	0%		Estimate
Flask other			4%	96%								Estimate
PET thermoformed	98%					2%						Estimate
PE thermoformed			98%			2%						Estimate
PP thermoformed		98%				2%						Estimate
PVC thermoformed					98%	2%						Estimate
PS thermoformed				98%		2%						Estimate
PET rigid	100%											Estimate
PE rigid			100%									Estimate
PP rigid		100%										Estimate
PVC rigid					100%							Estimate
PS rigid				100%								Estimate
Carrier bags (PE) > A4			100%									Estimate
Carrier bags (PE) < A4			100%									Estimate
Film PET > A4	100%											Estimate

Film PET < A4	100%											Estimate
Film PE > A4			100%									Estimate
Film PE < A4			100%									Estimate
Film PP > A4		100%										Estimate
Film PP < A4		100%										Estimate
Film PVC > A4					100%							Estimate
Film PVC < A4					100%							Estimate
Film PS > A4				100%								Estimate
Film PS < A4				100%								Estimate
Other plastics, not NIT sortable, rigid	18%	31%	36%						15%			Previous WUR-FBR Project
Other plastics, not NIT sortable, film > A4			100%									Estimate
Other plastics, not NIT sortable, film < A4	50%		50%									Previous WUR-FBR Project
Other plastics (PC, PLA, etc.)									100%			Estimate
Laminate PET	95%						4%		1%			Estimate
Laminate PE			95%				4%		1%			Estimate
Laminate PP		95%					4%		1%			Estimate
Laminate PVC					95%		4%		1%			Estimate
Laminate PS				95%			4%		1%			Estimate
Blister strips PET	92%						8%					Estimate
Blister strips PE			92%				8%					Estimate
Blister strips PP		92%					8%					Estimate
Blister strips PVC					92%		8%					Estimate
Blister strips PS				92%			8%					Estimate
Styrofoam trays				100%								Estimate
Styrofoam blocks				100%								Estimate
Silicone tubes												Wil not be in these samples
PET no packaging	100%											Only material
PE (rigid) no packaging			100%									Only material
PE (film > A4) no packaging			100%									Only material
PE (film < A4) no packaging			100%									Only material
PP no packaging		100%										Only material
PVC no packaging					100%							Only material
PS no packaging				100%								Only material
Plastic Hero collection bags (PE)			100%									Only material
Organic / ondefinable											100%	Only material
Paper, Board, Beverage cartons, etc						100%						Only material
Metal < 100 g							100%					Only material
Metal > 100 g							100%					Only material
Glass								100%				Only material
Textiles											100%	Only material

Appendix H: The composition of the samples as input in the lab mechanical recycling process

	1	2	3	4	5	6	7	8	9	10
	[gross g]	[gross g]	[gross g]	[gross g]	[gross g]	[gross g]	[gross g]	[gross g]	[gross g]	[gross g]
PET bottle transparant ≤ 0,5 liter			4510.00	4529.00	3911.00	3925.00	1553.00	1562.00	1500.00	1519.00
PET bottle transparant ≤ 0,5 liter + Barrière			105.10	105.50	133.00	133.00	36.21	36.41	51.02	51.64
PET bottle lightblue ≤ 0,5 liter			607.00	609.00	510.00	512.00	209.00	210.00	196.00	199.80
PET bottle coloured ≤ 0,5 liter		0.00	4.17	4.19	4.00	4.29	1.43	0.02	1.63	0.02
PET bottle coloured ≤ 0,5 liter + Barrière			0.09	0.09	0.13	0.13	0.03	0.00	0.05	0.00
PET bottle opaque ≤ 0,5 liter			1.00	1.00	0.34	0.34	0.34	0.00	0.13	0.00
PET bottle transparant > 0,5 liter	9349.00	9667.00	2181.00	2190.00	2072.00	2078.00	6626.00	6663.00	6267.00	6346.00
PET bottle transparant > 0,5 liter + Barrière		57.00	1.06	1.06	3.00	3.19	7.05	7.09	7.44	7.54
PET bottle lightblue > 0,5 liter	648.00	239.00	65.42	65.70	44.00	44.03	378.43	381.07	348.28	352.53
PET bottle coloured > 0,5 liter	0.24	6.00	0.34	0.34	1.00	0.98	0.86	0.01	1.06	0.01
PET bottle coloured > 0,5 liter + Barrière			0.10				0.00	0.00	0.00	0.00
PET bottle opaque > 0,5 liter			0.11	0.11	0.24	0.24	0.04	0.00	0.09	0.00
Bottle PE					3.60	1.44	0.00	0.00	0.55	0.13
Bottle PP			0.16	0.07	1.27	0.51	0.02	0.01	0.20	0.05
Bottle PS			0.26	0.10			0.04	0.01	0.00	0.00
Bottle other							0.00	0.00	0.00	0.00
Flask PET food			1653.00	1660.00	2542.00	2552.00	569.00	572.00	976.00	988.00
Flask PET food + Barrière			142.35	143.96	85.00	83.23	49.38	49.66	31.81	32.21
Flask PET non-food			489.00	491.00	488.00	491.00	498.00	495.00	552.00	495.00
Flask PE			2.50	1.00	5.00	2.18	0.34	0.09	0.83	0.21
Flask PP			1.09	0.44	7.00	2.64	0.15	0.04	1.01	0.26
Flask other							0.00	0.00	0.00	0.00
PET thermoformed			3.78	3.79	17.00	15.67	1.30	0.34	5.99	0.61
PE thermoformed			0.30	0.12	1.60	0.64	0.04	0.01	0.25	0.06
PP thermoformed			0.11	0.04	0.34	0.14	0.02	0.00	0.05	0.01
PVC thermoformed							0.00	0.00	0.00	0.00
PS thermoformed			0.13	0.05			0.02	0.00	0.00	0.00
PET rigid			151.50	152.14	117.00	116.91	52.19	13.80	44.68	4.52
PE rigid	1.06	25.00	40.53	16.28	4.00	1.61	7.00	1.85	1.94	0.49
PP rigid			1.80	0.72	8.00	3.30	0.25	0.07	1.26	0.32
PVC rigid			0.01		0.10	0.04	0.00	0.00	0.02	0.00
PS rigid			1.62	0.65	0.25	0.10	0.23	0.06	0.04	0.01
Carrier bags (PE) > A4							0.00	0.00	0.00	0.00
Carrier bags (PE) < A4			0.11	0.04	0.03	0.01	0.02	0.00	0.00	0.00
Film PET > A4					0.33	0.33	0.00	0.00	0.13	0.01
Film PET < A4			0.08	0.08	0.96	0.97	0.03	0.01	0.37	0.04

Film PE > A4			0.03	0.01	0.44	0.18	0.00	0.00	0.07	0.02
Film PE < A4			4.24	1.70	1.00	0.50	0.58	0.15	0.19	0.05
Film PP > A4		3.00	0.08	0.03	0.20	0.08	0.14	0.04	0.16	0.40
Film PP < A4	0.14		0.22	0.09	1.39	0.56	0.06	0.02	0.24	0.06
Film PVC > A4							0.00	0.00	0.00	0.00
Film PVC < A4					0.07	0.03	0.00	0.00	0.01	0.00
Film PS > A4							0.00	0.00	0.00	0.00
Film PS < A4			0.02	0.01	0.01		0.00	0.00	0.00	0.00
Other plastics, not NIT sortable, rigid		2.00	7.05	7.08			2.64	2.66	0.52	0.40
Other plastics, not NIT sortable, film > A4							0.00	0.00	0.00	0.00
Other plastics, not NIT sortable, film < A4			0.03	0.03	0.13	0.13	0.01	0.01	0.05	0.01
Other plastics (PC, PLA, etc.)			0.61	0.62	5.65	5.67	0.21	0.21	2.17	0.55
Other plastics, undefined sample (too small to sort)			4.02	4.04			1.39	1.39	0.00	0.00
Laminate PET					0.62	0.62	0.00	0.00	0.24	0.60
Laminate PE			0.13	0.05	0.18	0.07	0.02	0.00	0.03	0.01
Laminate PP			0.41	0.16	0.46	0.18	0.06	0.01	0.07	0.02
Laminate PVC							0.00	0.00	0.00	0.00
Laminate PS							0.00	0.00	0.00	0.00
Blister strips PET							0.00	0.00	0.00	0.00
Blister strips PE							0.00	0.00	0.00	0.00
Blister strips PP			0.02	0.01			0.00	0.00	0.00	0.00
Blister strips PVC			0.07	0.03			0.01	0.00	0.00	0.00
Blister strips PS							0.00	0.00	0.00	0.00
Styrofoam trays			0.01				0.00	0.00	0.00	0.00
Styrofoam blocks			0.24	0.10	0.04	0.02	0.03	0.01	0.01	0.00
Silicone tubes							0.00	0.00	0.00	0.00
PET no packaging			0.74	0.74	2.49	2.50	0.26	0.26	0.96	0.24
PE (rigid) no packaging			1.01	0.41	0.20	0.08	0.14	0.04	0.03	0.10
PE (film > A4) no packaging							0.00	0.00	0.00	0.00
PE (film < A4) no packaging			0.02	0.01			0.00	0.00	0.00	0.00
PP no packaging			2.60	1.04	2.58	1.04	0.36	0.09	0.40	0.10
PVC no packaging			0.47	0.19	0.22	0.09	0.06	0.02	0.03	0.01
PS no packaging			3.95	1.59	0.56	0.22	0.54	0.14	0.09	0.02
Plastic Hero collection bags (PE)							0.00	0.00	0.00	0.00
Organic / ondefinable			5.72	2.30	7.59	3.05	0.79	0.21	1.16	0.29
Paper, Board, Beverage cartons, etc	0.15		2.10	0.84	15.78	6.43	0.29	0.09	2.45	0.62
Metal < 100 g			0.50	0.05	0.07	0.07	0.05	0.00	0.03	0.00
Metal > 100 g							0.00	0.00	0.00	0.00
Glass			0.51	0.21			0.08	0.02	0.00	0.00
Textiles			0.09	0.03			0.01	0.00	0.00	0.00
Total	9998.59	9999.00	9998.61	9997.84	9999.87	9995.44	9998.15	9997.92	9998.74	10001.97

Appendix I: The molecular masses determined by GPC prior and after SSP [g/mole].

Averages of measurements with independent duplicates and standard deviation of ca. 5%.
M_n= number average molar mass, M_w= mass average molar mass, M_z= z-average molar mass

	GPC prior to SSP				**GPC after SSP**			
	M_n	**M_w**	**M_z**	**M_w/M_n**	**M_n**	**M_w**	**M_z**	**M_w/M_n**
0	26840	45381	70242	1.69	36066	64589	108762	1.79
1	23348	37524	60047	1.61	35423	62511	115420	1.76
2	28582	43519	68469	1.52	38831	68040	131574	1.75
3	30007	45834	87904	1.53	38841	66853	119019	1.72
4	27835	44549	70031	1.60	38223	68850	122514	1.80
5	26136	41832	63005	1.60	38186	69866	115059	1.83
6	27819	46663	80637	1.68	35555	73139	189045	2.06
7	26342	42142	76516	1.60	38879	68564	111667	1.77
8	29910	45905	76360	1.54	38272	69444	118183	1.81
9	27544	44013	66981	1.60	37350	66974	118206	1.79
10	28582	43519	68469	1.52	36735	65490	116216	1.78

Appendix J: Measurements Emmtech (requested by Stefan Morssinkhof) on samples from work package 2 before SSP treatment

Measurement 1

	L	a	b	transparency	IV	AA [mg/kg]	Benzene [mg/kg]	Limonene [mg/kg]
0, rPET	35.76	-0.15	4.05	yes	0.64	7.8	0.06	0.02
1, ink	37.84	-0.98	2.62	??	0.56	4.1	0.09	0.03
2, Amosorb	37.39	-1.49	6.71	yes	0.63	5.9	0.10	0.02
3, PS	42.17	-2.78	3.48	no	0.62	5.1	0.11	0.02
4, PP	29.69	1.53	0.01	no	0.62			
5, PLA	43.03	-2.66	1.41	moderate	0.63	4.1	0.07	0.01
6, EVOH	36.01	-0.87	2.04	yes	0.65	5.9	0.16	<0,01
7, PE	42.72	-2.19	2.08	no	0.63	3.5	0.09	0.02
8, Nylon	38.73	-1.07	4.30	yes	0.65	3.8	0.07	0.01
9, Hotmelt	37.58	-1.07	9.13	yes	0.63	4.3	0.07	0.03
10, PVC	36.94	0.32	7.30	yes	0.61	5.2	6.39	0.02

Measurement 2

	L	a	b	transparency	IV	AA [mg/kg]	Benzene [mg/kg]	Limonene [mg/kg]
0, rPET	36.20	-0.14	4.25	yes	0.64	8.3	0.07	0.02
1, ink	36.18	-0.98	2.61	??	0.58	4.3	0.10	0.01
2, Amosorb	37.03	-1.45	6.34	yes	0.63	6.1	0.11	0.01
3, PS	42.29	-2.79	3.72	no	0.62	5.6	0.11	0.02
4, PP	28.92	1.41	-0.21	no	0.61	3.9	0.30	0.11
5, PLA	42.87	-2.68	1.83	moderate	0.63	3.9	0.10	0.01
6, EVOH	36.29	-0.85	1.84	yes	0.66	5.8	0.16	<0,01
7, PE	43.18	-2.25	2.16	moderate	0.58	3.1	0.10	0.03
8, Nylon	38.33	-1.12	4.49	yes	0.65	3.9	0.07	<0,01
9, Hotmelt	37.00	-0.98	8.95	yes	0.63	4.2	0.07	0.02
10, PVC	37.41	0.29	7.53	yes	0.61	5.6	6.81	0.02

Appendix K: Origin of impurities in the samples, based on the samples after modelled pre-sorting

		PP [%]	PE [%]	PS [%]	PVC [%]	Paper [%]	Metal [%]	Glass [%]	PA/POM/ABS, etc. [%]	Silicon [%]	Undefined/rest [%] incl. textiles
1	Design	100%	100%	100%	0%	100%	0%	0%	0%	0%	0%
	Sorting	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
2	Design	98%	96%	100%	0%	100%	0%	0%	0%	0%	0%
	Sorting	2%	4%	0%	0%	0%	0%	0%	100%	0%	0%
3	Design	99%	94%	81%	0%	98%	87%	30%	72%	100%	0%
	Sorting	1%	6%	19%	100%	2%	13%	70%	28%	0%	100%
4	Design	99%	97%	91%	0%	99%	98%	51%	72%	100%	0%
	Sorting	1%	3%	9%	100%	1%	2%	49%	28%	0%	100%
5	Design	98%	98%	79%	0%	86%	97%	100%	40%	100%	0%
	Sorting	2%	2%	21%	100%	14%	3%	0%	60%	0%	100%
6	Design	99%	99%	91%	0%	94%	97%	100%	40%	100%	0%
	Sorting	1%	1%	9%	100%	6%	3%	0%	60%	0%	100%
7	Design	100%	99%	95%	0%	100%	98%	73%	73%	100%	0%
	Sorting	0%	1%	5%	100%	0%	2%	27%	27%	0%	100%
8	Design	100%	100%	99%	0%	100%	100%	92%	73%	100%	0%
	Sorting	0%	0%	1%	100%	0%	0%	8%	27%	0%	100%
9	Design	99%	99%	95%	0%	97%	98%	100%	42%	100%	0%
	Sorting	1%	1%	5%	100%	3%	2%	0%	58%	0%	100%
10	Design	100%	100%	99%	0%	99%	99%	100%	73%	100%	0%
	Sorting	0%	0%	1%	100%	1%	1%	0%	27%	0%	100%

* Indicative results, based on measurement, estimations and calculations.

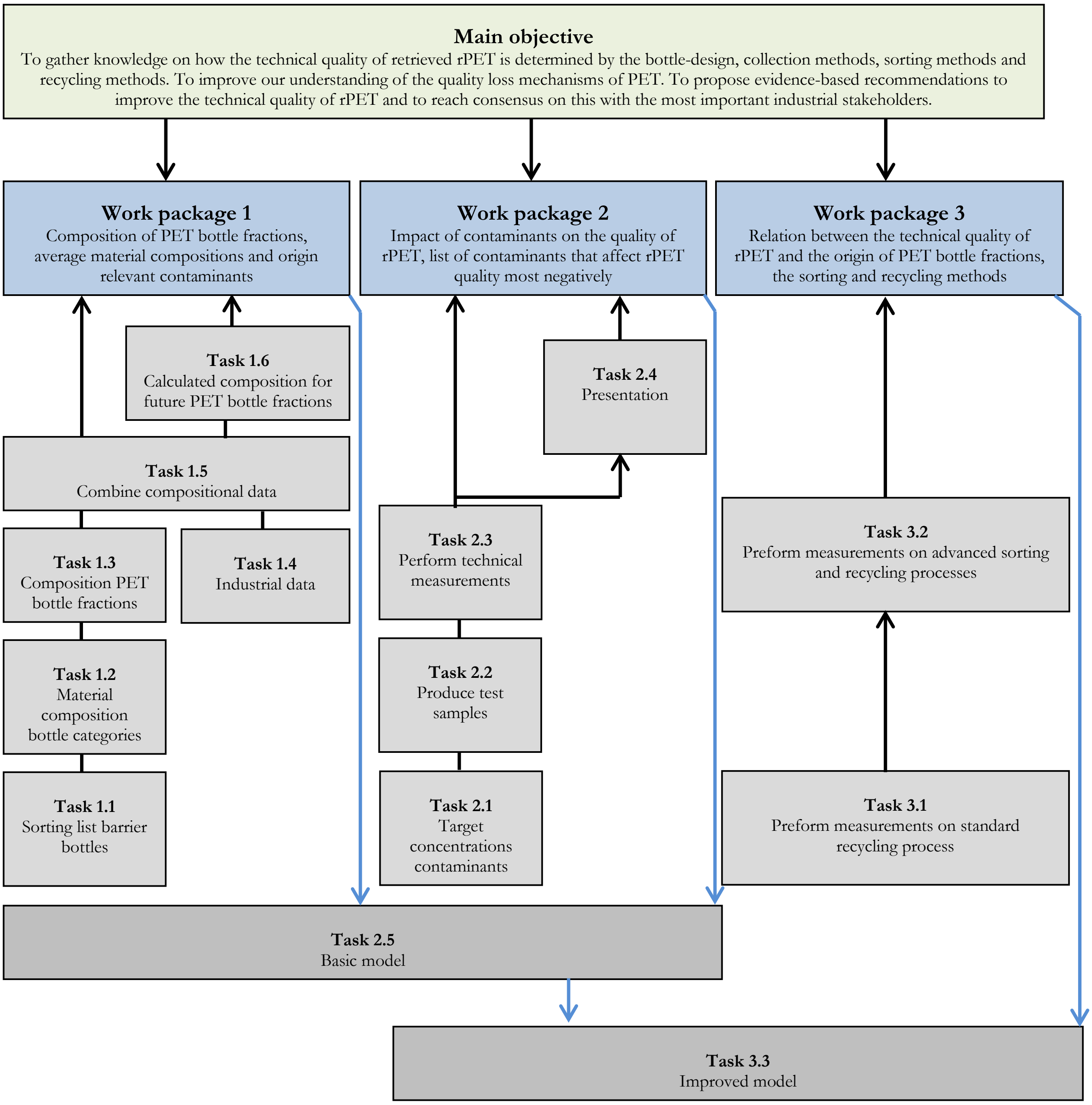
Origin of the impurities in the samples. Contaminants can have two origins: design related and sorting faults. This table provides a calculated estimate of the origin division for every sample, and every contaminant in this sample.

Appendix L: Estimated concentration of PET and contaminants in the rPET products of task 3

	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**
PET	99.293%	99.275%	98.556%	99.774%	98.509%	99.765%	99.836%	99.837%	99.828%	99.832%
PE	0.131%	0.132%	0.362%	0.073%	0.426%	0.085%	0.048%	0.048%	0.055%	0.054%
PP	0.357%	0.371%	0.465%	0.091%	0.416%	0.083%	0.077%	0.077%	0.075%	0.076%
PS	0.173%	0.172%	0.438%	0.039%	0.412%	0.036%	0.025%	0.024%	0.024%	0.024%
PVC	0.000%	0.000%	0.006%	0.000%	0.005%	0.000%	0.000%	0.000%	0.000%	0.000%
Paper	0.047%	0.046%	0.054%	0.011%	0.071%	0.013%	0.010%	0.010%	0.011%	0.011%
Metal	0.000%	0.000%	0.000%	0.000%	0.000%	0.000%	0.000%	0.000%	0.000%	0.000%
Glass	0.000%	0.000%	0.008%	0.001%	0.003%	0.000%	0.000%	0.000%	0.000%	0.000%
POM / ABS / PA	0.000%	0.003%	0.070%	0.007%	0.111%	0.011%	0.003%	0.003%	0.004%	0.003%
Silicone	0.000%	0.000%	0.034%	0.003%	0.041%	0.004%	0.001%	0.001%	0.002%	0.002%
Indefinable	0.000%	0.000%	0.003%	0.000%	0.004%	0.000%	0.000%	0.000%	0.000%	0.000%

This table provides a calculated estimate of the percentage of certain materials in the samples. These concentrations will vary in practice and should not be considered as fixed values. Nevertheless they are based on our measurements and give insight in the presence of contaminants within these feedstocks.

Appendix M: Schematic overview (flow scheme) of original project plan



To gather knowledge on how the technical quality of retrieved rPET is determined by the bottle-design, collection methods, sorting methods and recycling methods. To improve our understanding of the quality loss mechanisms of PET. To propose evidence-based recommendations to improve the technical quality of rPET and to reach consensus on this with the most important industrial stakeholders.

Appendix N: Comments of the industrial board and the responses of the authors

Comments of Stefan Morssinkhof of Morssinkhof Plastics

Comment 1.
P101. ... [*"PET bottle products originating from separate collection and mechanical recovery that were recycled with the standard process yielded an inferior quality of rPET. This could be attributed to the higher concentration of contaminants present and the inability of the standard process to remove these sufficiently. The same PET bottle products could, however, be recycled with the advanced process to rPET qualities that were far better. These qualities of rPET are in some quality aspects comparable to or better than the rPET made from deposit refund systems with the standard process; haze, colour, particle contamination. But in other quality aspects (molecular weights and crystallisation behaviour) slightly less good. This is not completely understood yet and could possibly be influenced by the laboratory equipment and procedures used for these comparative tests."*]

Voor de rapportage mogen we ervan uitgaan dat overal de basis waslijn min of meer dezelfde performance geeft. Het verschil zit hem voornamelijk details van de (nu handmatige) bottle & flake sortering. Het kan NOOIT zo zijn dat een bronscheiding systeem na intensive sortering beter scoort dan een statiegeld systeem. Dus de "not completly understood" moet wel anders geformuleerd worden ; beter nog deze gehele alinea heroverwegen want maak het onderzoek niet sterk zoals het er nu staat.

1 Translation of comment 1
For the report we may assume that the standard processing method will perform everywhere similarly. The main differences are the details of the (currently manual) bottle and flake sorting. It is impossible that a recycling system based on separately collected bottles will perform better than a recycling system based on a deposit refund system.
Hence "not completely understood" has to be phrased differently; even better would it be to reconsider the whole paragraph, because it does not strengthen the research as it is phrased now.

1 Response to comment 1
There is no public scientific literature on how the quality of rPET depends on the origin of the bottles and the type of collection system. Hence we cannot verify or refute on forehand that certain qualities of rPET are better or worse than others depending on their origin. The lack of this knowledge in the scientific literature was the reason to start this research. Therefore, we studied samples of sorted PET bottle products, determined rPET qualities and drew conclusions afterwards.
The phrase 'not completely understood' refers to a comparison between rPET made from deposit refund system bottles with the standard process and rPET made from separately collected bottles with the advanced process. Measurements revealed that some of the rPET properties were slightly better and some were slightly worse. These are recorded facts, which we as scientists do not completely understand.

Comment 2.
P101. ... [*"Nevertheless, these tests did show that the amount of contaminants in rPET can be controlled with the advanced recycling process and a relatively high quality rPET can be attained. Although the net recovery of mass and the net PET yield are much lower for the advanced process in comparison to the standard process.*"]

Uiteindelijk kunnen we alles onder controle brengen, inderdaad zoals gesteld met als resultaat dat de yield behoorlijk lager is. Er wordt nergens gesproken over CapEx / Opex praten in de conclusie? Maw, de verhouding tussen investering en opbrengst werkt niet, zelf niet met hele lage prijzen van de feedstock. Zie de UK als voorbeeld met vele missers faillissementen , massa geld erin gepompt > nog steeds geen kwaliteit als ook geen business model dat werkt. Ook hier geldt dat de sorterig in het onderzoek handmatig is gedaan. In praktijk zijn er afgezien van barcode scanning vooralsnog geen geautomatiseerde systemen die sortering kwalitatief voldoende goed kunnen uitvoeren voor B2B applicatie met een economische haalbare yield.
In de discussie/conclusies ontbreekt deze belangrijke nuancering van de sorteermethode op lab schaal en die in de praktijk.

2 Translation of comment 2
Eventually we can control everything, indeed as stated, resulting in a considerably lower yield. CapEx/Opex is not mentioned in the conclusion? The ratio between investment and return is problematic, even with very low prices for the feedstock. See also the UK as an example for many bankruptcies, a lot of money pumped into it > yet still no quality or business model that works. Here again the sorting in the research has been done manually. In practice there are, apart from barcode scanning, still no automated systems that perform sufficiently well in sorting for B2B applications with an economically viable yield. In the discussion/conclusion, this important nuancing of the sorting methods on lab scale and in practice is missing.

2 Response to comment 2
This is a technical study, not an economical study. Hence a business cost analysis is outside the scope of this study.
The composition of the feedstocks were based on two sets of data; 1) the average composition of sorted PET bottle products as determined in task 1 and 2) the removal efficiencies of the pre-sorting step as advised by the industrial board. As discussed in the industrial board, the most difficult part of the pre-sorting is the manual removal of PET-non-food-flasks. Currently, there is no automatic sorting process available and manual sorting is laborious and difficult. Nevertheless we followed the advice of the industrial board to choose the removal efficiency of the non-food-flasks in such a manner that the concentration in the output stream is just below 5% as is legally required for food-grade recycling.
To emphasize the difficulty of the manual sorting of non-food-flasks we include an additional clause in the first paragraph of paragraph 2.3.2.

Comment 3.
Mbt de 0,6% barrier bottles. Ik mis de opmerking dat dit een snelgroeiend percentage is, er komen steeds meer technische moeilijk te scheiden verpakkingen op de markt. De 0,6% is dus

best case scenario.

3 Translation of comment 3
Referring to the 0.6% barrier bottles; I miss the remark that this is a rapidly growing percentage, as there is introduced more and more packaging that is difficult to separate technically. The 0.6% is a best case scenario.

3 Response to comment 3
Based on this study and the available public literature we cannot confirm or refute that the percentage of PET-barrier bottles is strongly rising. We can only report how much PET barrier bottles we have found in the various PET bottle products. Note that 0.6% barrier bottles are found in the other deposit refund system. In the source separated system 2.5% barrier bottles were found, in the recovery system 2.3%. (See section 3.3.2)

Comment 4.
Dat het verschil tussen de 325 en 328 sorteerkwaliteit van DKR zo weinig verschil oplevert is enkel een gevolg van het niet rekening houden met de in praktijk onmogelijke sortering tussen food en non food en het feit dat de lab schaal handmatige sortering beter is dan de industriële sortering. Handmatige sortering van alleen de voor B2B geschikte flessen geeft bij alle typen inzameling systeem vergelijkbare resultaten. Het gaat er nu juist om dat sortering op deze wijze (nog) niet mogelijk is in de praktijk. Met name schaaltjes en multi layer flessen spelen hierbij een rol. De conclusie over het verschil tussen deze twee sorteernormen is derhalve onzorgvuldig cq behoeft nuancering. Bewijs is dat zelfs de meest geavanceerde PET was fabrieken met voor en nageschakelde (her) sortering de huidige sorteerkwaliteit van bron en nascheiding voor maximaal 10% inzetten voor B2B, en zelfs dan nog kwaliteitsproblemen optreden.

4 Translation of comment 4
The fact that the difference between sorting to DKR specification 325 and 328 results in so little distinction is only a consequence of not taking the, in practice, impossible sorting between food and non-food into account, and the fact that sorting is better on manual lab scale compared to industrial sorting. Manual sorting of the only for B2B suitable bottles leads to similar results for all types of collection systems. At stake is now the fact that sorting in this way is not (yet) possible in practice. In particular, trays and multi-layer bottles play a role. The conclusion on the difference between those two sorting standards is therefore improper and requires nuancing. The proof is that even most advanced PET mechanical recycling facilities with a subsequent (re) sorting deploy the current quality of separately collected and mechanically recovered PET bottle sorting products for maximal 10% for B2B, and even then quality problems occur.

4 Response to comment 4
We do acknowledge that at this moment non-food flasks cannot be sorted out mechanically and have to be sorted manually. This is hence also written in the report in paragraph 2.3.2 and 4.3. It has been the choice of the industrial board to mimic this manual sorting step with removal efficiencies that would result in a concentration non-food-flasks of just under 5% in the output stream. The research in WP3 of this report has been conducted in this manner and hence the results obtained are a consequence of this chosen research method. Both feedstocks (sorted to

comply with DKR 328-1 and 325) were subjected to this chosen research method and hence contained both less than 5% of non-food flasks. Furthermore, relative clean large PET bottles from the deposit refund systems were added to both feedstocks and hence both feedstocks were already relatively comparable, and therefore the actual difference between both feedstocks is relatively small in terms of types of packages present, see also Appendix G (compare feedstocks 7 and 8 and 9 and 10). It is therefore not a very large surprise that the quality of the rPET produced from both feedstocks is also relatively similar. We will make sure this is clearly addressed in the text.
The industrial information that recycling facilities only accept maximally 10% of separately collected and / or mechanically recovered PET bottle sorting products in their input was read with interest. However, we as scientists cannot confirm or refute this information based on literature or results and as far as we know there could be multiple reasons for these policies.

Comment 5.

Beginnen door te zeggen dat rPET uit statiegeld heel aardig is en uit andere systemen minderwaardig is onjuiste beeldvorming. Statiegeld en Fostplus zijn de enige inzamelsystemen met het hoog percentage hergebruik in flessen. Alle andere in de markt beschikbare kwaliteiten PCW flessen komen in praktijk niet of veel minder in fles hergebruik terecht. Voor statiegeld kwaliteit worden enorme prijzen betaald, voor Fostplus 2/3 daarvan en voor bronscheiding PET materiaal slechts een fractie daarvan. Statiegeld is dus superieur op dit moment en Plastic Hero materiaal is zeer laagwaarwaardig. In de huidige formuleringen wordt teveel waarde toegekent aan NIR en IR sorteertechniek. Hoe representatief is de labscheiding voor industriele schaal sortering komt ook bij dit punt weer om de hoek kijken; hier mot dus echt een stuk discussie over worden opgenomen. Op lab.-basis haal je er vrijwel alles uit, op industriële schaal slechts 90%? Het is juist die resterende 10% die industriële schaal sortering er in laat zitten, die nog altijd destructieve effecten heeft. Als IR en NIR sortering zo effectief is, waarom zijn er dan geen voorbeelden van bedrijven die een afdoende bottle-to-bottle kwaliteit produceren, ondanks dat ze wel de genoemde apparatuur geïnstalleerd hebben. Met handmatige sortering is het bovendien logisch dat de impact van statiegeld flessen toevoegen aan de bronscheiding stroom niet veel verschil zal maken. De praktijk is echter geen handmatige sortering en dat maakt het een veel complexer thema.

5 Translation of comment 5

Starting with saying that rPET from deposit refund systems is quite well and rPET from other systems is inferior leads to misperceptions. Deposit refund and Fostplus are the only collection systems with a high percentage in reuse of bottles. All other on the market available qualities of PCW bottles in practice end up far less often or even never in the reuse of bottles. Enormous prices are paid for deposit refunded bottles, for Fostplus 2/3 thereof and for separately collected PET material only a fraction thereof. Deposit refund is thus superior on the moment and Plastic Hero material is of very low value. NIR and IR sorting techniques are valued too high in the current formulation. The representativeness of lab sorting compared to industrial sorting is again a discussion point, and should thus be included in the discussion part. Based on lab sorting, you get out almost everything, on industrial scale only 90%? Especially those remaining 10% in industrial scale sorting still have destructive effects. If IR and NIR sorting is so effective, why are there no examples of companies that produce an adequate bottle-to-bottle quality, although they do have installed the listed equipment. With manual sorting, it is also logical that the impact of adding deposit-refund bottles to separately collected PET bottles will not make much difference. However, this is a much more complex issue, as in practice there is no manual separation.

5 Response to comment 5

This is a scientific report with conclusions that are phrased in scientific terms. Therefore we will conclude in terms of reported properties and the comparisons between those properties. In task 3 (paragraph 3.3.14) we found that the properties of rPET made from deposit-refund bottles with the standard process were either better or comparable to rPET made from separately collected PET bottles with the advanced process. This is a scientific conclusion based on the comparison of properties. We deliberately do not interpret these results further in terms of superiority and inferiority of feedstocks.

Regarding the ‘contaminants with destructive properties’, these were present in the feedstocks that we have used in task 3 for the recycling experiments. As discussed with the industrial board these feedstocks first received a pre-sorting step in which these contaminants are partially removed with removal efficiencies that have been discussed and established in the industrial board. Hence we did not clean the feedstocks by manual sorting, we mimicked in the laboratory an industrial pre-sorting step.

Comment 6

Terecht wordt gesteld dat aan dat de huidige feedstock uit bron- en nascheiding 10 – 20% non-food artikelen bevat. Het is noodzakelijk voor een juiste beeldvorming daarbij stellen dat de samenstelling daardoor per definitie afvalt voor opwerking tot Food Grade rPET, gezien de Europese regelgeving. Wat gebeurt er met dit percentage na vermenging met statiegeld? Het aandeel non-food verpakkingen in PET neemt toe; hoe kan ook toekomstig zeker gesteld worden dat het aandeel non-food verpakking in de ingezamelde stroom onder de 5%-norm blijft?

Er is geen enkele sorteertechniek nog in staat food en non food te scheiden.

6 Translation of comment 6

It is correctly stated that the current feedstock received from resource- and subsequent separation contains 10-20% non-food items. For a clear rending, it is necessary to state that hence the composition is not suitable to process to food grade rPET, considering the European regulations. What happens to this percentage after system amalgamation of the deposit refund system and source separation/recovery systems? The share of non-food packaging in PET increases; how can it be ensured that also in future the proportion of non-food packaging in the collected flow will stay under the 5% standard?

There is no sorting technique available that separates food and non-food.

6 Response to comment 6

In the report it is clearly mentioned on in paragraph 1.1.2 and 4.3 that PET-non-food-flasks can currently not be sorted out by machines. Hence, the potential for separate collected and recovered PET to be used in bottle to bottle purposes is dependant of the development of these sorting technologies. In the potential case of system amalgamation this will be an issue that needs to be resolved.

Comment 7.

Zoals in een eerdere email met wat eigen interpretaties van de amorfe monsters na extrusie gesteld zijn de metingen op chemische componenten alleen voor vergelijking te gebruiken in mijn opinie. De absolute waarde is vaak niet juist bepaald en geeft zker bij bijvoorbeeld AA verwarring. Bovendien heb ik op basis van de de kleur al de nodige interpretaties voorgesteld, daarmee is naar onze smaak te weinig gedaan. Kleur is naast chemie essentieel voor mogelijk hergebruik in food flessen. Dat is waar we mee moeten vergelijken. Niet met Sheet of met Fiber hergebruik. Het gaat hier om een onderzoek voor de applicatie terug in food flessen, zoals nu voor 100% ook met alle statiegeld flessen gebeurd. Nogmaals : kleur is essentieel (kleurloos transparant als doel) voor hergebruik en flessen en dus voor dit onderzoek. Conclusies op bais van kleur zijn ten eerste niet alle juist en niet duidelijk genoeg gesteld.

7 Translation of comment 7

As stated in a previous email, including personal interpretations of the amorphous samples after extrusion, in my opinion the measurements on chemical components are only to be used as a comparison. The absolute value often is not correctly determined and causes, especially in example AA, confusion. Moreover, based on the colour I have proposed interpretations, which have not been addressed according to our liking. Colour is, next to chemistry, essential for potential reuse in food bottles. That's what we have to compare. And not with Sheet or Fiber reuse. It concerns a research for the application in food bottles, as done for 100% of the deposit refund bottles. Again: colour is essential (colourless transparent as aim) for the reuse of food bottles and thus for this research. Conclusions based on colour are firstly not all correct and secondly not plain enough.

7 Response to comment 7

The results of the control measurements of the samples of task 2 have been added in appendix I, with a description in section 3.2.9 . We notice that different stakeholders use different acceptance criteria for rPET quality and hence derive to different conclusions of which rPET is acceptable and which is not. As far as we know there is no broadly accepted generic acceptance criteria for rPET. Therefore, we measured in task 2 the impact of adding impurities to rPET in terms of changes in properties and reported that in paragraph 3.2 and hence refrained from conclusions in terms of 'adding contaminant A to rPET results in rPET qualities that are acceptable for the market'.

Comment 8.

Een lagere IV door door inkt is niet zo relevant en niet erg waarschijnlijk, kleur en food grade destructie is veel belangrijker.

8 Translation of comment 8
A lower IV because of ink is less relevant and not very likely, colour and food grade destruction is more important.

8 Response to comment 8
It was noticed that the presence of ink resulted in slightly lower IV, M_n, M_w and M_z of the rPET prior to SSP treatment, see paragraph 3.2.6. SSP treatment, however, successfully restores these indicators, so the final impact on the rPET is indeed negligible. The impact of the presence of ink on the colour was determined to be negligible (paragraph 3.2.4.). The impact of the presence of ink on the food safety aspects of the rPET was not studied, as explained in paragraph 1.2.2.

Comment 9.
Morssinkhof graag vervangen door *Morssinkhof Plastics*

9 Translation of comment 9
Please replace *Morssinkhof* by *Morssinkhof Plastics*

9 Response to comment 9
The name has been corrected accordingly.

Comment 10.
In SRN zit 0% gekleurde flessen. Tabel 11 stelt iets anders ?
Tabel 12 PAMD voor deposit klopt niet. Dirt is verwaarloosbaar en moisture is maximaal 3% (rest inhoud fles).

10 Translation of comment 10
SRN contains 0% coloured bottles. Figure 11 suggests something different?
Figure 12 PAMD for deposit is incorrect. This is negligible and moisture is maximum 3% (rest content bottle).

10 Response to comment 10
We also thought that the SRN deposit refund bottles would only contain transparent and light-blue PET bottles, however, in reality we did find a few coloured bottles and hence we reported that.
Relating the PAMD, this is the actually measured amount.

Comment 11.
Tabel 13 PVC geeft hoge druk=> is onwaarschijnlijk; PVC geeft degradatie, dus lage IV dus dun materiaal, dus lage druk. PVC en de ongewenste afbraakproducten passeren het filterpakket gewoon en dat is nu juist het probleem.
De hoge druk voor het filterpakket is dus niet uit te leggen en in strijd met ander onderzoek.
Advies is dus hier nog eens aandachtig naar te kijken.

11 Translation of comment 11
Figure 13 PVC creates high pressure ->seems unlikely. PVC gives degradation, thus low IV and thin material, thus low pressure. PVC and the unwanted degradation pass through the filter pack and that is precisely the problem. The high pressure for the filter pack is hence inexplicable and in contradiction with other research.
It is advised to investigate this issue carefully.

11 Response to comment 11

The pressures in figure 13 are recorded observations. PVC indeed causes PET degradation, as is apparent from the formation of benzene vapour and the lowering of the IV. This degradation should, in principle, result in a decrease of the pressure in the extruder. However, at a processing temperature of 265°C, PVC itself is not stable and will dehydrohalogenate, form polyenes, crosslink and form volatile degradation products. This results in a black, foamed tar-like substance that partially clogs the melt filter (50 µm mesh), which subsequently causes the pressure to increase in the extruder. Even with a very low dosage of PVC, this latter phenomenon appears.

Comment 12.

Op pagina 58 wordt gesteld dat wellicht hoge % rPET in de verwerkte flessen (47.5%) een verklaring zijn voor slechte resultaten.

Goede Rpet kan wel oneindig voor een veel hoger % steeds weer opnieuw worden hergebruikt.

70-80% is een algemeen geaccepteerd percentage voor GOEDE rpet.

Inderdaad is fast-reheat een ongewenste ontwikkeling in de markt, maar met veel rpet in de fles (maakt fles al iets donkerder) is fast reheat eigenlijk net meer nodig.

Fast reheat is misschien wel een groter probleem voor de kleur op de lange termijn dan een hoog rpet percentage in de flessen.

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12 Translation of comment 12

Page 58 states that possibly the high % rPET in the processed bottles (47.5%) explains poor results. High quality rPET can be reused infinitely for a much higher %.

70-80% is a generally acceptable percentage for qualitative good rPET.

Fast reheat is a non-desired development in the market, yet with much rPET in the bottle (colours the bottle slightly darker) fast-reheat is actually not required any more.

Fast-reheat might, in long term, be a bigger problem for the colour than a high percentage rPET in bottles.

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12 Response to comment 12

In case we would have known on forehand that fast reheat additive is so important for the rPET quality, we would have studied this additive separately in task 2 and we would perhaps also have chosen a different feedstock for task 2. But since we have not done so, we can only confirm that we have heard this opinion of this member of the industrial board and hope that the impact of this additive on rPET can be studied in the near future scientifically as well.

Secondly we acknowledge that there are many different qualities of rPET on the European market, in colour terms ranging from transparent to almost brown granulates. Hence, with good qualities rPET high recycled contents can be achieved and with low qualities only low levels of recycled content. Nevertheless, the rPET we produced was relatively dark and hence had a relatively lower quality. Our statement only referred to our quality of rPET and was not intended as a generic statement regarding all rPET qualities.

Appendix O: Comments of the scientific review board and the responses of the authors

Sigbritt Karlsson Högskolan in Skövde

Comment 1

In general, you need to go through all Figures and Tables legends so that they are understandable without instructions in text,
Some examples:
Fig 8 what is given on the x- and y-axis?
Table 15: what are L*, a* and b*, give this in the legend of the Table.
Fig. 9: unclear what the percentages in the boxes refer to (11%, 2% ...)
Several others need additions in the legends.

1 Response to comment 1

Captions have been adjusted accordingly.

Comment 2

Page 61 second paragraph where you talk about PVC and formation of hydrochloric acids. Please add some information about temperature when the onset of formation of hydrochloric acids and the amount in order to cause splitting of ester bonds in PET.

2 Response to comment 2

Formation of HCl from PVC starts above 70°C and once it starts, it is an autocatalytic process. The process can be retarded using heat stabilizers and these are the most crucial additives for PVC. Also plasticisers retard degradation and HCl formation. As a consequence the actual degradation and HCl formation depends on how well PVC is stabilized and the specific type of PVC compound. Still, even if PVC is well stabilized it should not be heated above 220°C[24]. PET is processed at temperatures well over 250°C and more commonly around 275°C. At these temperature degradation of PVC and autocatalytic HCl formation is unavoidable.
The effect of (traces of) HCl on PET depend on the presence of water. In case the PET recyclate is not dried (mimicking common practice), and degassing is used to control excessive degradation (Some degradation is desirable and SSPC is used to get the PET at the needed IV) than traces of HCl will catalyze hydrolysis of PET.

Comment 3

Page 62 first paragraph, last sentences: have you observed gel in the analysis?

3 Response to comment 3

In the analysis we have not observed a gel and found a recovery rate of about 98% in all samples both prior to and after SSPC. The EVOH concentration was very low. Still at this low concentration we found a large increase in the Mz value. At higher EVOH concentration or due to insufficient mixing gel formation can occur easily.

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[24] *PVC Degradation and Stabilization (Third Edition), edited by George Wypych, ChemTec Publishing, Boston, 2015.*

Comment 4

Are there any recommendations for further studies to improve knowledge or means to increase the recycling and subsequent reuse of mechanically recycled PET?

4 Response to comment 4

Indeed we have two sets of recommendations, one for researchers and one for stakeholders.

For researchers which pursue this type of research we would strongly advice to:
-include virgin PET in laboratory mechanical recycling research as an internal standard,
-to research the impact of reheat additive on the properties of rPET,
-to perform more measurements to assess the impact of contaminants on the properties of rPET, both repetition measurements and measurements at different concentrations.
-to assess the impact of drying the PET milled goods on the quality of rPET

For stakeholders we stress:
-the need for the development, implementation and international standardisation of sorting techniques to mechanically sort and remove undesired objects, such as: non-food flasks, barrier PET bottles and non-bottle PET packages (trays, tubs, blisters, etc.).
-to maintain the effort on ‘design for recycling’ guidelines for PET bottles and to extent that towards PET flasks and also achieve acceptance for the design-guidelines from these producers.

All recommendations are listed in paragraph 4.3 and 4.4

Frank Welle, Fraunhofer Institute for Process Engineering and Packaging (IVV)

Comment 1

1.1.2.

It can be mentioned here, that the PET raw materials for non-food containers is also "food-grade" PET. From a food law compliance point of view a higher amount of non-food PET container might be possible (e.g. in the US up 100% is approved for several recycling processes (chemical containers are excluded, however). I think EFSA introduced the 5% limit as precaution because higher contamination levels might occur in non-food containers.

1 Response to comment 1

We thank you for making that point and we have elaborated the text accordingly.

Comment 2

1.1.3.

1995? I thinks it's from 2005.

2 Response to comment 2

Adjusted accordingly.

Comment 3

1.3.4.

The correct name of our Institute is Fraunhofer Institute for Process Engineering and Packaging (IVV)

3 Response to comment 3

Adjusted accordingly throughout entire report.

Comment 4

2.1.1.

The SiOx layers are typically removed during caustic soda wash.

4 Response to comment 4

We were not aware of that. Thanks for this information. Since we have no literature reference for it, we will add a footnote in the text with this information.

Comment 5

2.2.2. Table 2.

The correct name is Amosorb. It should be corrected in the whole document.

5 Response to comment 5

Adjusted accordingly throughout entire report.

Comment 6

2.2.4. Question on the PET samples used for GC analysis.

Is the PET milled before weighting? Or do you have used pellets/flakes?

6 Response to comment 6

The samples used for GC analysis were the finely milled pellets. This has been clarified text.

Comment 7

2.2.4. Figure 4

I missed the ethylene glycol peak here in the chromatogram.

7 Response to comment 7

No ethylene glycol peak was expected, due to the absorbing inlet column. In case ethylene glycol was present in similar concentrations as other volatile molecules, then it would have been absorbed on the inlet column and elute only slowly at elevated temperatures and would probably not show as a single peak but rather as a raised baseline.

For GC analysis of polar components like ethylene glycol and glycerol another type of column is required then the apolar RXI-5ms column we used. Additionally, ethylene glycol (Mwt =62 g/mole) is known to show mostly as mass fragments of Mz =31 g/mole in our GC-MS system and this system has threshold for fragments below 35 g/mole, to exclude interference with the air gases (nitrogen and oxygen Mz= 28 and 32 g/mole) , hence even with a dedicated polar column it is unlikely that ethylene glycol will show up in the GC-MS spectrum.

Comment 8

2.3.4.

There are several food grade processes established in France. Can you describe the simulated process a little bit more detailed. Maybe you can refer also the EFSA opinion on this process.

8 Response to comment 8

The process descriptions were established in a discussion with the industrial reviewer. He analysed several ‘advanced’ processes for PET bottle recycling and in a discussion we decided how we could mimic such industrial processes in the best manner in the laboratory. Therefore the ‘advanced process’ we chose was a compromise between several industrial processes and the laboratory capabilities. Therefore, I think it would not be correct to refer to one specific ‘advanced’ process.

Edward Kosior, Nextek

Comment 1
p9. …[*In order to be able to identify barrier bottles, first a new barrier bottle detection method was developed.*]

the method of sorting should be outlined along with its reliability

1 Response to comment 1
The newly developed method for identifying PET barrier bottles is described in paragraph 2.1.1. A thickness-normalised discolouration factor is measured after the PET-bottle-wall-cutting has been subjected to a standard oven propagation test. The derivation of the error propagation law is given to calculate the standard deviation in this normalised discolouration factor.

Comment 2
p9. …[*This comprehensive analysis of the PET bottle feedstocks revealed that in bottle feedstocks from deposit refund systems almost exclusively PET-bottles are present that were designed according to the EPBP design guidelines.*]

this work is unique

2 Response to comment 2
Thank you very much.

Comment 3
p10. …[*A major difference between both recycling processes are the yields; both the net recovered masses as the net PET yields are lower by 5 to 10% for the advanced process in comparison to the standard process. This loss in yield can mostly be attributed to the additional flake sorting step.*]

the advanced process might be standard to PET recycling operations

3 Response to comment 3
The standard PET recycling process is the standard process in the Netherlands. Your valid point is, that it is not so common and standard in other European countries. Therefore, we understand that what we call ‘advanced process’ is in fact the ‘standard process’ in other European countries. Thanks for bringing that perspective to our report.

Comment 4
p26. …[*In case the difference in thickness normalised b* colour values was larger than the threshold value of 1.6, the bottle was identified as a barrier bottle.*]

this is a generalisation as the colour change could be due to additives such as AA absorber

4 Response to comment 4
Precisely, the barrier test method is intended to identify those PET bottles that discolour more than average during an oven provocation test and of which it is likely that this discoloration is caused by a barrier functionality of some sort (additional layer, additive, coating, etc.). The AA absorber is one of the barrier function that would be identified.

Comment 5

p28. …[*Roughly 140 kg of the cleaned PET bottles were milled with a WEIMA WLK04 shredder and a 2 cm sieve plate, yielding milled goods of 2-5 cm size. These milled goods proved too large for direct extrusion and hence this milled goods was additionally milled with a Wanner C17.26 shredder and a 0.8 cm sieve plate to sub-centimetre size milled goods, which could be extruded.*]

this seems odd as 5 mm is small flake for most extruders

5 Response to comment 5

Unfortunately, the 2-5 cm flakes proved too large for the feeding hopper of our Berstorff ZE 25 extruder and we had to force them in with a broom stick. This was, however, both cumbersome and undesired. So, we decided to mill the PET flakes one size class finer with the Wanner mill and then the milled goods did flow in.

Comment 6

p29, table 2

Contaminant	*Origin*	*Concentration*
PVC	*Sorting fault*	*1‰*

Too high

6 Response to comment 6

We apologise for the confusion relating to the use of the premille symbol (‰). We now understand the use of the permille symbol is uncommon in Anglo-Saxon countries and will change it to percent, with a conversion factor of 0.1. So we have changed it to 0.1%.

Comment 7

P29, table 2

PP Labels as integral part of PET bottles 19‰

hard to see why this is so high?

Response to comment 7

We now understand the use of the permille symbol (‰) is uncommon in Anglo-Saxon countries and will change it to percent, with a conversion factor of 0.1. So we have changed it to 1.9%.

Comment 8

p29….[*The PVC concentration was set to be 1‰, just like the concentration limit in specification DKR 328-1. Pure granular PVC powder was chosen, hence, without additives.]*

unrealistic

8 Response to comment 8

We now understand the use of the permille symbol (‰) is uncommon in Anglo-Saxon countries and will change it to percent, with a conversion factor of 0.1. So we have changed it to 0.1%. Relating to the use of PVC powder, instead of stabilised PVC packages the following considerations had guided to choose for PVC powder:

In the Netherlands the most common PVC contamination in PET bottle products are stretch wrapping film and non-packaging objects. Dosing and mixing in the wrapping film would have been very difficult practically. Moreover, the film would have contained plasticiser so the amount of actual PVC in the material would have been different/more difficult to assess. Addition of pure well-stabilised PVC would not give a completely honest picture since typically stabilisers are used during thermal processing and the amount “left” in a final product is not known.

Moreover PVC stabilisers are effective to about 220 degrees Celsius and certainly not at the 275 degrees we used during PET processing.

As a conclusion addition of PVC powder allows accurate PVC dosing and it is expected that the effect is similar to addition of PVC products.

Comment 9

p29. …[*The concentration was set at 3.3‰ since 10 kg bottles roughly contain 333 bottles with 0.1 gram of hot-melt per bottle.]*

should be 0.3%

9 Response to comment 9

We now understand the use of the permille symbol (‰) is uncommon in Anglo-Saxon countries and will change it to percent, with a conversion factor of 0.1. So we have changed it to 0.33%.

Comment 10

p30….[*The concentration was set at 7‰ since the DKR 328-1 specification allows for maximally 10% PET-trays which are maximally composed off 93% PET and 7% PE*.]

0.7%

10 Response to comment 10

We now understand the use of the permille symbol (‰) is uncommon in Anglo-Saxon countries and will change it to percent, with a conversion factor of 0.1. So we have changed it to 0.7%.

Comment 11

p30….[*The PP concentration was set at 19‰, roughly reflecting the bottle to label weight ratio of 50:1.]*

2%

11 Response to comment 11

We now understand the use of the permille symbol (‰) is uncommon in Anglo-Saxon countries and will change it to percent, with a conversion factor of 0.1. So we have changed it to 1.9%.

Comment 12

p30….[*The concentration was set to 3‰, reflecting the contamination of 10 kg of rPET with one tray of 30 grams of PLA*.]

0.3%

12 Response to comment 12

We now understand the use of the permille symbol (‰) is uncommon in Anglo-Saxon countries and will change it to percent, with a conversion factor of 0.1. So we have changed it to 0.3%.

Comment 13

p31….[*Compounding extrusion was performed using a Berstorff ZE 25 (25 mm, 40D) co-rotating twin screw extruder with degassing unit melt filter and strain pelletizing.]*

what was melt temperature?

13 Response to comment 13

The melt temperatures were measured at several locations in the extruder and hence a temperature profile was determined. Highest temperatures were measure at temperature zone 9 or 10 (of the 12 zones) and typically range from 275°C to 285°C depending on the sample. (set-point at that zone was 275°C.

Comment 14

p31….[*It was the mutual consensus that for this study the washed milled goods would not be dried before processing.]*

why is this done???

14 Response to comment 14

PET processing was discussed in detail with one member of the industrial board (Mr. Morssinkhof). He advised us not to dry the PET milled goods prior to extrusion but to degas, since this would mimic the Dutch standard process the best. In Dutch industrial practise PET milled goods are often not-dried but instead the residual moisture is removed during degassing in the extruder. This lowers the IV after extrusion, but the subsequent SSPC restores it again. This is done deliberately, to have a longer residence time in the SSPC reactor and hence to remove more volatiles. The target IV after extrusion was about 0.65 and SSPC is used to boost the IV to above 0.8 and typically during SSPC volatile contaminants are removed (even allowing production of PET that is food grade/bottle grade). With this procedure (non-dried PET) it is possible to study the effect of contaminants on the SSPC process.

We now understand that from your comments that this process (not drying, but instead degassing and SSPC) is not common practise outside the Netherlands. In hindsight, a reference process run with and without drying could better have been performed to evaluate the impact of this process method.

Comment 15

p31….[*Melt temperature (measured) : 265-267 °C.*]

PET melts at 265 C

15 Response to comment 15

See the response to comment 13.

Comment 16

p32….[*During this SSP treatment the rPET granulates were flushed with 0.04 ml nitrogen gas per minute*] what was final IV?

16 Response to comment 16
The final IV values after SSP-treatment are shown in figure 16 for task 2 and in figure 29 for task 3. The time period for SSP treatment was set to achieve a target IV of 0.8 for the first sample and this time period was kept as much as possible constant for the rest of the samples.

Comment 17
p32….[*One type of test specimen (bone shaped rods/tensile bars) was intended for DMTA and 50 x 50 x 3 mm plates were intended for Haze measurements.*]

very thick for this test

17 Response to comment 17
We re-measured the Haze-plates and they are 2 millimetre thick. We will change the text accordingly.

Comment 18
p32….[*Prior to injection moulding materials were dried at 120°C for 4 hours using a desiccant dryer, to prevent hydrolysis.*]

too low temp

18 Response to comment 18
We did not observe any problems with degradation or hydrolysis after drying at these conditions. The procedure was tested using references (virgin PET). Apparently the moisture content was sufficiently low.

Comment 19
p34….[*The peak at r.t. 11.62 min. with a mass of 136 g/mole with a more elaborate fragmentation pattern.*]

limonene

19 Response to comment 19
Correct, we have changed the text accordingly.

Comment 20
p41….[*After washing the flakes were rinsed very well with cold water to rinse off all the alkaline solution from the flakes.*]

was pH tested?

20 Response to comment 20
We did not measure the pH value, but instead the ionic conductivity of the effluents. Our tap water has an ionic conductivity of approximately 300-400 µS/cm and our 0.1 M NaOH solutions of about 38 mS/cm. Normally, the first effluent (the waste water) has still high ionic conductivities of 80-90% of the hydroxide solution and after one full rinse with 150 litre of cold water the effluent already returns to tap-water levels of ionic conductivity. For these experiments

we even rinsed twice with 150 litre cold water, so all the hydroxide should be rinsed off.

Comment 21

p53….[*These parameters were weight-averaged with respect to the material composition of all the samples and subsequently weight-averaged to overall parameters for PET bottles and flasks from the three main types of origin, see Table 12.]*

how much was moisture and how much dirt as the moisture is not an issue.

21 Response to comment 21

It had been decided to measure combined moisture and dirt contents and not separate moisture contents and dirt contents, so the separate numbers are not available. But your preference is noted for future research.

Comment 22

p58….[*This only significantly changes for PE and PP. The addition of these contaminants results in even more small particles.*]

the behaviour of dispersed phase particles in a PET melt will be as viscous melts and they will pass through a mesh of any size

22 Response to comment 22

Your comment sounds plausible, nevertheless, our plastic technologists have measured and reported pressure variations for extruding rPET that was deliberately contaminated with either PE and PP. These pressure variations were not recorded for any other contaminant (besides PVC, but here the pressure develops due to degradation). Their explanation of these small pressure variations recorded for PE and PP was the passing of molten dispersed polyolefines through the melt filter, we still cannot think of other plausible explanations. We agree with you that our suggestion that the melt filter (50 um) has caused the dispersion to break up in smaller particles is likely to be incorrect and we have rephrased the sentence accordingly.

Comment 23

p59, table 15.
what colour were the shrink labels

23 Response to comment 23

The PP labels were mostly black (Coca Cola Zero printed labels). These were normal PP labels not shrink labels.
The PS shrink labels were mostly blue and red (Dubbelfriss printed shrink labels).

Comment 24

p59, table 15.

why not add natural pp not black labels

24 Response to comment 24

A mix of different PP labels or transparent PP film could also have been used instead. We chose the PP labels that were present, since there are many different grades of PP polymer for films and we wanted to mimic the impact of real labels as close as possible.

Comment 25

p59….[*Compared to for instance the purchase specifications of Coca-Cola (L* >67 for crystallised pellets), all rPET-granulates are too dark.]*

this is a big shame as the data would have been very helpful otherwise

25 Response to comment 25

This also was a disappointment for the researchers involved.

Comment 26

p60….[*The reference rPET has a haze-value of 24% and is too hazy in comparison to purchase specifications that demand that the haze should be less than 13%.]*

this is a bad choice

26 Response to comment 26

In hindsight this is the correct conclusion. At the time this decision was made, it appeared to be an obvious choice, to use one of the most common PET bottles on the Dutch market.

Comment 27

p60….[*With the exception of the specimen with PVC, this can be explained by the drop in particle contamination as determined with the Partisol measurement.]*

the added levels are too high and the results so severe that differences are masked

27 Response to comment 27

We excuse ourselves for using the premille symbol and causing the confusion. Now, we have changed all into percent, I am confident that you will agree that the levels are not too high.

Comment 28

p64….[*The melt peaks in the second heating runs were found to deviate too strong (systematically smaller) and were not used.] (sentence is now re-written)*

the first scan is related to heat history and the second scan related to material properties. the virgin resin should be the standard as there is a high heat additive in the rPET

28 Response to comment 28

We followed your advice and changed the tables and figures regarding DSC to the results of the second heating run.

Comment 29

p65, table 17

surprising PP and PE do not have greater impact as they are used to nucleate PET in CPET

29 Response to comment 29

We presented the data as it was measured. The answer is perhaps related to your comment 30, that the relative high concentration of reheat additive has masked the impact of the deliberately added contaminants.

Comment 30

p65…[*It drops with less than 10% and this drop is probably not significant.]*

data is masked by the high heat additive and the high level of additives

30 Response to comment 30

Your suggestion regarding the high concentration of heat additive sounds plausible, hence we changed the text accordingly.

Comment 31

p65…[*This could potentially result in crystallisation too early in the blowing process and is therefore undesired.]*

there would be better clarity if the second cycle data was used.

31 Response to comment 31

The second heating run doesn't show a crystallisation peak at about 120°C anymore, hence we could only use the onsets from the first heating run.

Comment 32

p71….[*It does make the rPET slightly more yellow and it does increase the speed of crystallisation.]*

this was the worst

32 Response to comment 32

You are correct, we changed the text accordingly.

Comment 33

p71….[*PVC makes the rPET more yellow, increases the speed of crystallisation.]*

and also more red

33 Response to comment 33

You are correct, we will change the text accordingly.

Comment 34

p75, table 23

Amasorb often added to the PET or in mid layer

34 Response to comment 34

Yes we agree, that is why we mention at 'wind sifting' that Amosorb can be removed 'if loose', meaning if it is not mixing in the PET, but present in a separate mid-layer.

Comment 35

p75, table 23

MXD can be in a mid-layer or added to PET matrix

35 Response to comment 35

Yes, we agree, MXD is often used in a separate mid-layer, hence we will change the text in the table accordingly.

Comment 36
p75, table 23…[Nylon conclusion]

depends on hot wash

36 Response to comment 36
We were not aware that the hot wash alone could remove (a part of) the nylon layer, but we will indicate that as an option.

Comment 37
p75….[*PVC originates only from faulty sorted objects (mainly PVC from non-packaging objects), as PVC is no part of the design of PET bottles and flasks.]*

sleeves on bottles can be from PVC

37 Response to comment 37
Indeed can PET bottle sleeves be of PVC, but we have not found these in the Netherlands. So, bottle sleeves might be a potential source of PVC in foreign PET bottle products, but for Dutch PET bottle products, the only source of PVC was found to be faulty sorted objects.

Comment 38
p88….[*Also the PET products made from bottles originating from deposit refund systems with the standard recycling process do not comply with specifications such as* $L^*>67$, $b^*<3$ *and haze* $<15\%$*.]*

this is a big gap in lab to industrial practice

38 Response to comment 38
That is correct and is also a major concern for the researchers. Your suggestion to incorporate a virgin PET resin as a reference in future research is in our perspective the right approach (see comment 53).

Comment 39
P100….[*Major differences in the composition of PET bottle products were found and even between the PET bottle products from two different Dutch deposit refund systems; one containing 0.2 % coloured bottles and no barrier bottles and the other containing 4.9% coloured bottles and 0.6% barrier bottles.*]

this difference would not be significant

39 Response to comment 39
We have changed the text accordingly.

Comment 40
P100….[*PVC was found to yield benzene vapour, most likely due to an acid-catalysed degradation of PET.*]

the impact of hot melt glues is significant and should be mentioned

40 Response to comment 40
Hotmelt is now also mentioned in the conclusion.

Comment 41
P101…[*The likely explanation is that although this increased sorting effort reduces the level of contaminants originating from faulty sorted objects, still more than sufficient remain that originate from the PET bottles themselves.]*

results could be masked by the reference rPET used.
Virgin PET should be included as a reference material in this study

41 Response to comment 41
We agree that it would be wise to include virgin PET as a reference material in this study. This would have clarified the influence of the laboratory mechanical recycling approach on the quality of rPET. Hence this is now also one of our recommendations.
This task 3, however, is not effected by the poor choice of the reference rPET, that is task 2.

Comment 42
Overall the project is a very positive and detailed investigation that has been very well planned scientifically.
Generally the execution of each part of the project has been done in great detail and the data in the report will be of great value to the scientific community due to the unique nature of the investigation and the importance of the results as initially planned.

42 Response to comment 42
Thank you, that is much appreciated.

Comment 43
There have been specific instances where decisions have been made in the actualisation of the investigation that have reduced the detailed relevance elf the results to industrial recycling operations.
Examples are given below this point:
- selection of unstabilised PVC against the type of PVC used in packaging,

43 Response to comment 43
See our response to comment 8.

Comment 44
There have been specific instances where decisions have been made in the actualisation of the investigation that have reduced the detailed relevance elf the results to industrial recycling operations.
Examples are given below this point:
- the selection of dark preforms as the reference material,

44 Response to comment 44
We used the common Coca-Cola bottles (not preforms) as input material, since we needed a substantial amount (150 kg empty and clean bottles) of precise the same quality. We agree that in hindsight, we could better have used another type of PET-bottle source with less or no reheat additive.

Comment 45
There have been specific instances where decisions have been made in the actualisation of the investigation that have reduced the detailed relevance elf the results to industrial recycling operations.
Examples are given below this point:
- the absence of virgin resin as a reference material,

45 Response to comment 45
We agree that it would be better for understanding the difference between the results of the industrial mechanical recycling and the mechanical recycling in the laboratory to include virgin PET as an internal standard. Also see our response to comments 38 and 41.

Comment 46
There have been specific instances where decisions have been made in the actualisation of the investigation that have reduced the detailed relevance elf the results to industrial recycling operations.
Examples are given below this point:
- the processing of PET without pre-drying as a reference,

46 Response to comment 46
See our response to comment 14.

Comment 47
There have been specific instances where decisions have been made in the actualisation of the investigation that have reduced the detailed relevance elf the results to industrial recycling operations.
Examples are given below this point:
- the level of additives put into the rPET was single point not multi point to measure the sensitivity of contamination to the process, the level of additives was artificially high and not reflecting the normal ranges seen in recycling operations which should have been readily accessible via the industry advisor,

47 Response to comment 47
We excuse ourselves for using the premille symbol (‰) and causing the confusion. Now, we have changed all into percent, I am confident that you will agree that the levels are not too high.

Comment 48
There have been specific instances where decisions have been made in the actualisation of the investigation that have reduced the detailed relevance elf the results to industrial recycling operations.
Examples are given below this point:
- the use of thick plaques for colour and haze measurement compared to actual products made from rPET ,

48 Response to comment 48
See our response to comment 17.

Comment 49
There have been specific instances where decisions have been made in the actualisation of the investigation that have reduced the detailed relevance elf the results to industrial recycling operations.
Examples are given below this point:
- inadequate drying of the materials before moulding,

49 Response to comment 49
See our response to comment 14.

Comment 50
There have been specific instances where decisions have been made in the actualisation of the investigation that have reduced the detailed relevance elf the results to industrial recycling operations.
Examples are given below this point:
- misinterpretation of the way PET melts behave during filtration has led to unsubstantiated conclusions being made on the section on Partisol particle analysis (this section can be reviewed and the conclusions re-written),

50 Response to comment 50
See our response to comment 22

Comment 51
There have been specific instances where decisions have been made in the actualisation of the investigation that have reduced the detailed relevance elf the results to industrial recycling operations.
Examples are given below this point:
- the DSC measurements were all based on first scan that reflect the recent process history whereas the second cycle scan would reveal the material characteristics (this section needs to be reviewed).

51 Response to comment 51
We have changed the interpretation of the DSC measurements according to your suggestion to the second heating run.

Comment 52
There have been specific instances where decisions have been made in the actualisation of the investigation that have reduced the detailed relevance elf the results to industrial recycling operations.
Examples are given below this point:
- The classification that hot melt makes rPET slightly yellow when the data shows that it is the worst influence

52 Response to comment 52
We have changed that according to your comment.

Comment 53
There have been specific instances where decisions have been made in the actualisation of the

investigation that have reduced the detailed relevance elf the results to industrial recycling operations.

Examples are given below this point:

- The lack of understanding why the lab recycling results were far worse than industrial practice. This could have been tested by initial calibrations of the recycling process with virgin resin or clear bottles

53 Response to comment 53

In hindsight that is correct. In case we had the opportunity to redo the research we would indeed have used virgin PET and/or clear bottles as an internal reference.

Comment 54

The limitations of the experiments has meant that the authors advise that the results are not directly applicable to industrial practice but can be seen as providing relative data is very disappointing since this project had the potential the be a landmark investigation in the science and practice of recoiling.

54 Response to comment 54

We agree as researchers. Nevertheless, progress has been made and future researchers can learn from this study to be more effective in the future.

Comment 55

Despite this comment the report has good data that is valuable to many people in the industry however I suggest that where possible the sections indicated are reviewed and edited to make the whole report a reliable and trusted document for the Governments and Industry to use as a reference document to make very important decisions.

Without this review the report may either mislead researchers and governments or be ignored by industry.

55 Response to comment 55

We take the comments from the industrial board and the scientific review committee very serious and will respond to all their comments and improve all parts where possible.