Development of a Food Grade PET recycling Process.

Literature Review and Feasibility Study

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1 Introduction

In November 2003, it was reported that 22% of households in the UK have the opportunity to participate in kerbside collection that include plastic bottles. However only 25,000 T per annum or 5.5% of plastic bottles used in the home are recycled.\(^1\)\(^1\) There clearly needs to be a continued increase in this percentage in order to meet UK Government and European goals. In order to close the loop on recycling of plastic bottles there will need to be a significant increase in the recycling of PET into high value applications.

The estimated demand for Recycled PET (RPET) in Western Europe of 660 KT pa, of which 50% is estimated to be in Food Contact applications.

The increased useage of RPET in food contact applications, is being driven by a number of factors including:

- the imminent requirements of European Directives,
- the desire to meet for environmental sustainability goals,
- the potential for reducing the cost of bottle grade PET resin
- the need to develop high volume and high value applications for RPET\(^2\)

European reprocessors report that UK sourced PET bottles is more highly contaminated than material collected through mainland European collection schemes. Contamination arising from PVC labels and cap liners being a particular problem.

In its favour, UK sourced PET generally has lower levels of coloured bottles.\(^3\)

Clearly UK sourced rPET has different characteristics from that collected through mainland Europe, hence the cost efficiency of rPET recycling in the UK requires comprehensive understanding of the output quality requirements and the input quality requirements.
2 Quality Requirements for Food Grade PET

2.1 Regulatory Background

UK Regulations require materials in contact with food to meet broad requirements which ‘...aim to keep food safe and wholesome by requiring that contact materials and articles, such as packaging do not transfer their constituents into food so as to endanger health or affect food quality’.4

Plastics that are to be used in food contact applications are always evaluated for any migration that might occur when in contact with food material. By definition, any migrating substances are considered to be indirect food additives and need to be evaluated against relevant regulations for the approval of Food Packaging or standard Food Additive Approval procedures.

Food safety is the paramount consideration in use of recycled plastics in food contact applications. Regulatory requirements have been a major factor controlling the development of recycling processes that can demonstrate that they can produce recycled plastics suitable for food contact applications. Without such regulations it is not possible to gain approval or “non objection” for any process that recycles PET back into food contact applications.

In the USA, the FDA have three main safety concerns associated with the use of recycled plastics in food-contact applications: i) transfer of adventitious contamination from the recyclate to the foodstuff, ii) incorporation of material not regulated for food contact, and iii) the presence of adjuvants that may not comply with regulations for food contact use in the recycled plastic.

The FDA has specified a “Challenge Test” procedure to allow recycling processes to be validated as being capable of removing severe contamination from bottles that have been deliberately contaminated in a controlled way. This provides regulatory bodies with assurance that the much lower level of contamination present in collected bottles will be readily removed to levels that present negligible risks to consumers.

The existence of this procedure has meant that a large number of technologies and processes have evolved in the USA and other countries (such as Australia) that refer to USFDA for standards in Plastics in food contact applications. From February 1990 to August 2003 58 “letters of non-objection” have been issued to 17 chemical processes and 41 physical recycling processes for recycling PET back into food grade applications5.

The regulatory position with regard to chemically recycled plastic is clear. Monomers and starting substances derived from feedstock recycling have to comply with EC Directive 90/128/EEC.
The USFDA have in the past issued letters on non-observation to processes that use chemical recycling methods since the processes have been shown to be very effective in removing contaminants. The USFDA Centre for Food Safety and Applied Nutrition (CFSAN) website October 2003 states “surrogate contaminant testing is no longer considered necessary to demonstrate that post-consumer recycled (PCR) polyethylene terephthalate (PET) or polyethylene naphthalate (PEN) produced by a tertiary recycling process is suitable for food-contact use. Because FDA has determined that tertiary recycling processes produce PCR-PET or PEN of suitable purity for food-contact use, the Agency no longer sees a need to evaluate tertiary recycling processes for PET or PEN or to issue individual opinion letters for them”.

Physical recycling, ie secondary recycling, processes have outnumbered the chemical processes especially in the last seven years when 30 physical process and 8 chemical processes received letters of non objection from the USFDA. The commercial recycling operations for PET have focussed on physical recycling due to the smaller capital cost, lower operating cost and the smaller material inputs required to operate such a process.

### 2.2 Recent EU Regulatory Developments.

International Life Sciences Institute (ILSI) Packaging Material Task Force, (Recycling of Plastics for Food Contact Use) based in Belgium, has developed guidance for the provision of chemical safety of recycled plastics for food contact use as well as procedures for quickly assessing the likelihood of residual contamination. ILSI adopt the position that if recovered material, originally of food contact grade, is processed so as to reduce concentrations of likely contaminants to levels below the regulatory threshold it should be considered acceptable, as it is demonstrably equivalent to virgin plastic in terms of food safety. This guidance however, whilst based upon an EU-funded research projects and guidelines developed by the USFDA and other organisations, currently has no legal status.

Nevertheless, as a technical document that describes the current state of the art, it is the basis for due diligence purposes and is the principal procedure used to “challenge” recycling processes within the EU. Comparing the USFDA and ILSI approach, the final end point in both instances ie. The Threshold of Regulation and the non-detectable migration limit are in fact the same value for PET recycling (10 ppb or µg/kg).

This document discusses the both approaches in further detail below.

The working definition of Food grade PET varies for different parts of the world. For Europe it is PET plastic of a suitable standard for food applications manufactured in compliance with Commission Directive EU 2002/72/EEC (and future amendments).

Commission Directive EU 2002/72/EEC builds on the Commission Directive EU 82/711/EEC which lays down rules for testing migration of the constituents of plastic materials and articles and Council Directive 86/572/EEC establishes the list of simulants to be used in the migration tests. Article 2 defines the overall migration limit as “Plastic Materials and articles shall not transfer their constituents to foodstuffs in quantities exceeding...
10 milligrams per square decimetre of surface area of material or article “or 60 milligrams per kilogram of food for containers with a capacity of 0.5 to 10 litres.

The EU Commission is actively developing further guidance and controls in the area of Food Safety- Chemical Safety of Food- Chemical Safety. The harmonisation at EU level of the legislation on food contact materials fulfils two essential goals: the protection of the health of the consumer and the removal of the technical barriers to trade.

In October 2004 the new Framework Regulation (EC) 1935/2004 was introduced. This establishes rules on materials and articles intended to come into contact with food and repeals Directives 80/590/EEC and 89/109/EEC. The principle underlying this regulation is that any material or article intended to come into contact with food must be sufficiently inert to preclude substances from being transferred to food in quantities large enough to endanger human health or bring about an unacceptable change in the composition of the food or a deterioration in its organoleptic properties.

The Framework Regulation sets up general requirements for all food contact materials whereas Specific Directives cover single groups of materials and articles listed in the Framework Directives. The Regulation further states that as a matter of principle, “The use of recycled materials and articles should be favoured in the Community for environmental reasons, provided that strict requirements are established to ensure food safety and consumer protection. Of the 17 listed groups..., priority should be given to the harmonisation of rules on recycled plastic materials and articles as their use is increasing and national laws and provisions are lacking or are divergent.”

Frank Koelewijn, from the industry organisation, Petcore welcomes this Framework, stating “On the one hand, this step could strengthen the food industry and consumers’ confidence in PET and recycle PET. On the other hand, a clear regulation with minimum standards would protect PET recyclers who had invested in sophisticated technology against unfair competition. Clearly the ongoing development of EU Frameworks and Directives is a major driving force for member states and those companies who are investing in the production of high quality food grade RPET.

2.3 Regulatory and Recommended Requirements regarding Migration.

In order to comply with US Food and Drug Administration (USFDA) requirements, Code of Federal Regulations: 21 CFR parts 5, 25, 170, 171 and 174, the migrating substances must either only be present in dietary concentration at very low levels ie., less than 0.5 ppb, or that the level is less than 1% of the acceptable daily intake (ADI) for those substances.

In the case of recycled plastics that are to be evaluated for food contact use, the recycling process must be capable of removing a wide range of categories of materials so that should consumers misuse PET containers i.e. place used engine oil in them, then the recycling process will still be able to adequately reduce the concentrations to acceptable low levels to avoid food adulteration.

The specification of a lower limit for the threshold level for dietary concentration allows the permissible level of contaminants in recycled plastics to be calculated.
The dietary concentration of a contaminant is related to the concentration of the migrating contaminant (M) into a food simulating solvent, i, (where i represents the four simulated food types: aqueous, acidic, alcoholic, and fatty foods), the food distribution factor (fT), and the consumption factor (CF) in the following manner.

\[
\text{dietary concentration} = \text{CF} \times \sum_{i=1}^{4} M \times fT
\]

For the case of PET, with a food distribution factor of 1.0, and a consumption factor (CF) of 0.05 (as per Points to Consider for the Use of Recycled Plastics in Food Packaging: Chemistry Considerations document of USFDA), the maximum concentration of the migrating contaminant (M) can be calculated, given the maximum dietary concentration of 0.5 ppb.

\[
0.5 \text{ ppb} = 0.05 \times M \times 1.0
\]

i.e. \( M = \frac{0.5 \text{ ppb}}{0.05} = 10 \text{ ppb} \)

This result means that if less than 10 ppb is extracted from a PET bottle into a food simulant (such as 10% aqueous ethanol) then the dietary concentration (M) will be less than 0.5 ppb assuming the above consumption patterns. This would mean that the contaminant would not pose concerns to public health and safety.

In the case of physical reprocessing, it is important that the recycling process is able to demonstrate its efficiency in decontaminating the PET. This will establish its ability to cope with consumer misuse of the bottles and inadvertent inclusion of bottles from non-food sources.

2.3.1 Surrogate Contaminant Testing

Consumer misuse can be simulated by exposing plastic packaging (either in container form or as flake) to selected surrogate contaminants. Following exposure of the polymer to the surrogate contaminants, the polymer would be subjected to the recycling process. Subsequent analysis of the polymer for those contaminants would demonstrate the efficacy of the recycling process.

The way in which a process is validated by the USFDA for its efficiency is to subject the PET recycling process to a “Challenge Test” in which PET is spiked with a cocktail of toxic substances that represent the various categories of chemicals that could be absorbed by PET.

Even though the number of articles likely to be contaminated will be only a small proportion of the total number of articles being recycled, the “Challenge Test” stipulates that 100% of the articles should be contaminated with surrogate solutions to provide a substantial safety factor. The objective of contaminating the recycled articles is to simulate the very worst-case scenario that kerbside collection of PET could induce.
reality the number of bottles, which might be affected by consumer misuse would typically be very low and certainly less than the number of non-food PET bottles.

The materials used for the simulation of consumer misuse should bracket a variety of chemical and physical properties. Model contaminants should be “common” materials accessible to the consumer and include.

A volatile, non-polar organic substance
A volatile, polar organic substance
A non-volatile, non-polar substance
A non-volatile, polar organic substance.

Examples of such materials are toluene, chloroform, lindane, and diazinon respectively. Toluene and chloroform are components of cleaning solvents, while lindane and diazinon are common insecticides. A toxic salt, such as disodium monomethylarsenate (crabgrass killer), would complete the range of potential contaminants.

Due to the toxic nature of the above chemicals, alternative less toxic “surrogate” chemicals that behave in a similar manner can be used in the “Challenge Test”. The concentration of surrogate chemicals for the contamination of PET is based on the examination of typical formulations for domestic products and a 10-fold safety factor over “user strength” is applied. Examples of recommended surrogates are given in Table 1; only one surrogate per category needs to be included in the testing.

<table>
<thead>
<tr>
<th>Volatile Polar</th>
<th>Non-Volatile Polar</th>
<th>Volatile Non-Polar</th>
<th>Non-Volatile Non-Polar</th>
<th>Heavy Metal</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chloroform</td>
<td>Benzophenone</td>
<td>Toluene</td>
<td>Tetracosane</td>
<td>Copper(II) 2-ethylhexanoate</td>
</tr>
<tr>
<td>Chlorobenzene</td>
<td>Methyl salicylate</td>
<td></td>
<td>Lindane</td>
<td></td>
</tr>
<tr>
<td>1,1,1-Trichloroethane</td>
<td>Diethyl ketone</td>
<td></td>
<td>Methyl stearate</td>
<td></td>
</tr>
<tr>
<td>1-Phenyldecane</td>
<td>Phenylcyclohexane</td>
<td>2,4,6-Trichloroanisole</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 1  Examples of surrogates used in Challenge Tests

Chloroform and toluene are components of cleaning solvents; benzophenone is a suitable substitute for non-volatile polar pesticides such as Diazinon; and tetracosane is a good representative for the long-chain hydrocarbons that comprise motor oil. A heavy metal salt such as copper(II) 2-ethylhexanoate, would represent a substitute for the toxic salts commonly used in herbicides.

In the case of PET, it is no longer necessary to include a heavy metal salt in surrogate testing. In the PET recycling submissions reviewed by the USFDA over the past decade, migration of the heavy metal surrogate has never been detected in food simulants. The
data indicate that the metal salts do not sorb as readily into PET as do small organic molecules and that the salts are more easily washed out of PET, probably because the salts simply adsorb to the PET surface.

Containers or flaked virgin plastic would then be exposed to the selected contaminants, again either “neat” or with “at use” concentrations. A mixture, or “cocktail,” of the contaminants is usually used. In this case, the components of the cocktail should not react with each other. Examples of minimum concentrations of surrogates that should be present in the cocktail are shown in Table 2 below.

<table>
<thead>
<tr>
<th>Contaminant</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chloroform (volatile polar)</td>
<td>10% v/v&lt;sub&gt;a&lt;/sub&gt;</td>
</tr>
<tr>
<td>Toluene (volatile non-polar)</td>
<td>10% v/v&lt;sub&gt;a&lt;/sub&gt;</td>
</tr>
<tr>
<td>Benzophenone (non-volatile polar)</td>
<td>1% v/v&lt;sub&gt;a&lt;/sub&gt;</td>
</tr>
<tr>
<td>Tetracosane or Lindane (non-volatile non-polar)</td>
<td>1% w/w&lt;sub&gt;b&lt;/sub&gt;</td>
</tr>
<tr>
<td>Hexane or Heptane (as overall solvent for cocktail)</td>
<td>68% v/v&lt;sub&gt;a&lt;/sub&gt;</td>
</tr>
</tbody>
</table>

v/v<sub>a</sub> – volume of contaminant per unit volume of entire cocktail
w/w<sub>b</sub> – mass of surrogate per unit mass of entire cocktail

Table 2. Examples of Minimum Concentrations of Contaminants in a Surrogate Cocktail

The bottles or flakes are stored sealed for two weeks at 40 ºC with periodic agitation. After the contaminants are drained and the bottles or flakes are rinsed, the contaminated polymer is then subjected to the proposed recycling process, and regenerated components or packaging material formed from the reprocessed polymer should be analyzed for residual contaminants. This approach represents a worst-case scenario, i.e., all material entering the recycling stream is assumed to be contaminated.

2.3.2 Plastic Containers from Non-Food-Contact Applications as Feedstock

Containers for non-food substances (“non-food containers”) are likely to sorb large amounts of chemicals from their contents over very long shelf lives. When such containers are included in the post-consumer feedstock on a regular basis (i.e., from kerbside collection programs), the level of contamination in recycled containers is likely to be higher than if the feedstock were restricted to food containers.

Although the probability that 100% of the post-consumer plastic feedstock might consist of non-food containers is low, worst-case contamination assumptions must be made for non-food articles entering the recycling stream.

A mathematical model, based on Fick’s law of diffusion, predicts the amount of a contaminant (represented by the surrogate contaminants described above) that will sorb into a PET bottle after one year at 25 ºC, the shelf life and use temperature of a typical non-food substance packaged in PET. The PET sorption values for several surrogates are summarized in Table 3 below.
### Table 3. Sorption of Surrogate Contaminants into PET after 365 Days at 25 °C

These values establish the minimum concentrations of each surrogate that must be present in the challenged PET flake at the beginning of surrogate testing in order to conclude that the resultant exposure estimates are applicable to a recycling process that includes non-food PET in the feedstock.

### 2.3.3 ILSI Guidelines for Recycling Plastics for Food Contact Use.

These guidelines\(^\text{11}\) share many of the same scientific principles established by the USFDA guideline and have some differences that have developed from new scientific knowledge and also differences in approach to the same problem.

These guidelines were initially developed for food grade plastics and use the same concept of a “Challenge Test” conducted with surrogate substances. The specific substances and concentrations are listed in Table 4, and while similar to those used by USFDA, they differ in the detail and concentration. The key difference is the use only 1% of a polar, volatile penetrant instead of 10%.

### Table 4. Surrogate Chemicals for use in Challenge Tests.

<table>
<thead>
<tr>
<th>Penetrant</th>
<th>Substance</th>
<th>Concentration %</th>
</tr>
</thead>
<tbody>
<tr>
<td>polar, volatile</td>
<td>trichloroethane:</td>
<td>1</td>
</tr>
<tr>
<td>polar, non-volatile</td>
<td>benzophenone:</td>
<td>1</td>
</tr>
<tr>
<td>non-polar volatile</td>
<td>toluene</td>
<td>10</td>
</tr>
<tr>
<td>non-polar, non-volatile</td>
<td>chlorobenzene</td>
<td>1</td>
</tr>
<tr>
<td>non-polar, non-volatile</td>
<td>phenylcyclohexane</td>
<td>1</td>
</tr>
<tr>
<td>organometallic compounds</td>
<td>methyl palmitate (or methyl stearate)</td>
<td>1</td>
</tr>
</tbody>
</table>

\(^\text{11}\) ILSI Guidelines for Recycling Plastics for Food Contact Use.
The recent report on FAIR-CT98-4318 by Franz et al (2003) recommends the same surrogates as above but without the use of trichloroethane. This report also describes the migration modelling of substances based molecular weight and shows that the inverse relationship between the amount migrated after 10 days at 40 °C and molecular weight reaches a limit beyond 350 g/mol. Beyond this level substances are virtually immobilised in the PET matrix.

A key difference from the USFDA protocol is the way the surrogates are mixed with the PET. The USFDA uses a solvent based cocktail whereas the ILSI approach uses a direct mixing and sorption by mixing at 50 °C for 7 days which was found to be functionally equivalent but simpler to prepare. The level of surrogates in the PET flake should be between 500 to 350 ppm for all except for toluene which should approach 500 ppm. The contaminated flake is then directed straight into the Supercleaning process. These levels are quoted as being equivalent to 1750 to 2500 ppm for systems that first undergo washing and drying steps, before Supercleaning12.

The normal requirements of the challenge test are that 100% of the test articles are contaminated with surrogates and the results from the cleaning processes must demonstrate “not detectable migration” at the limit of detection of the analytical methodology. The limit of detection at which reliable analytical measurement can be made is stipulated to be 10 µg/kg, including any analytical tolerance.

2.3.4 Coca Cola Standards for Decontamination

In order for a process to gain approval for use for Coca Cola products it must meet the requirements specified by the specialists of The Coca Cola Company13. The focus is on being sure that not only is the process capable of removing contaminants as specified by USFDA, but also being capable of removing organic molecules such as fragrances and flavourants to low levels in the flake state so that bottles made with the resin do not influence the flavour of subsequent beverages. The following requirements must be met:

1. Decontamination process:
The process must have obtained a Letter of Non Objection from the FDA and appropriate regulatory clearance for the process in the intended use country

1.1 Surrogate Testing:
Surrogate testing needs to be performed in compliance with the FDA's “Points to Consider Document”. Surrogates will consist of the following compounds:
Chloroform 10%
Toluene 10%
Benzophenone 1%
Lindane 1%
Copper II ethylhexanoate 1%

1.2 Decontamination levels
The removal of the surrogates is measured when the PET is in the final flake state before processing into a melt. Volatile compounds (chloroform and toluene) need to be removed
to below a level of 3 ppm and the non-volatile compounds (benzophenone and lindane) need to be removed to below a level of 10 ppm.

1.3 Migration studies
For all compounds not removed to below a level of 215 micrograms/kg, migration testing needs to be performed using validated analytical techniques (detection levels @ 5 ppb). Migration testing should be done in 10% ethanol for a period of 30 days at 40 °C for ambient fill approval. Migration levels must not exceed a level of 10 ppb. Samples should be taken at intervals of 0, 3, 10, 20 and 30 days of testing. Recommendation is made to also perform migration testing for hot-fill conditions.

1.4 Process material analysis
PET flake from the process will be need to be tested by either thermal desorption or heated dynamic purge and trap GC/MS analysis. These analyses are performed by Coca Cola in their Atlanta laboratory.

1.5 Process description
There is a requirement for full disclosure of the details of the PET decontamination process under non-disclosure agreements with limited distribution of the information.

1.6 Audit:
The cleaning facilities and process must be inspected and approved by Coca Cola’s specialists to show that the process is repeatable and consistent. The Coca Cola QA requirements must be met.

2. Taste Test Approval
Bottles produced from the material by in the appropriate blend ratio with virgin resin need to be filled with product and taste tested over a 10 week period.

3. Bottle Physical Performance
During production of taste test samples, bottles will also be produced for physical performance testing. This includes the typical PET authorization tests such as top load, fill point drop, drop impact, carbonation retention, and stress crack resistance.

2.4 Input Quality Standards

Significant barriers to achieving the cost effective recycling of PET are the inconsistency of the sourced material and the high level of contaminants. Steps toward improving this situation are the introduction of standard terminology and specifications for recycled plastics. The introduction of the Reprise Specification for baled PET bottles will begin to achieve greater consistency amongst recyclers. The Publicly Available Specification 103 (PAS 103) classification and grading system for the quality of collected plastics packaging intended for recycling, was introduced in 2004. Through these systems, the value of the baled plastic bottles can be better identified, controlled and therefore increased. Closed Loop London has developed a detailed raw material specification, in Table 5, which forms the basis for a quantitative input quality standard.
**Purpose**

The purpose of the quality specifications is to establish uniformity and consistency in the PET and HDPE bottles recovered from suppliers from Mixed Bottle bales so that the PET and HDPE can be manufactured back into new products.

**Bottle Composition**

The composition of mixed bottles from kerbside collections will be:

(a) PET * 40%
(b) Natural HDPE * 40%
(c) Other bottles (Coloured HDPE, PP) 20%

The tolerance on all bottle specifications is +/- 5% except for PET which is -5%

Prices for mixed bottles will be agreed once bale audit details have been submitted by the vendor. Prices may be adjusted if the actual compositions vary from the agreed compositions.

**Contamination Definitions**

The quality specifications are designed to identify the levels of contamination that inhibit the recycling process. The types of contamination are listed below in two categories, with the allowable percentages per contaminant, per item quoted on a dry basis;

**"Prohibited Materials"** means materials which are difficult to remove through the recycling process including (but not limited to):

(a) Plastics film < 0.1%
(b) Plastics other than bottles < 0.5%
(c) Aluminium cans & closures < 0.02%
(d) Sand, dirt, gravel, stones < 0.02%
(e) Glass bottles & glass fragments < 0.01%
(f) Metal objects of any kind nil.%
(g) Hypodermic syringes nil.%
(h) Hydraulic oil, or other oil based products < 0.01%

**"Out throws"** means materials which can be tolerated or removed during the recycling process, including (but not limited to):

(a) Liquid residues in bottles < 1.5%
(b) Detergent bottles containing residue < 1.5%
(c) Food residues < 1.5%
(d) Household chemical bottles < 2.0%

The above specifications apply to all containers. Closed Loop London is prepared to work with their suppliers to rectify any excessive contamination levels, however if we cannot reduce the levels of contamination, the Mixed Bottles would be deemed unacceptable.

* Based on information from London Remade and Cleanaway, UK.

Table 5. Mixed plastics bottles quality specifications for recycling
2.5 Output Quality Standards

Specifications have been developed for the desired quality of the food grade rPET supplied to processors. Closed Loop London has developed specifications from those currently provided by raw material suppliers around the world. The specification below is achievable commercially with available technology and is needed to ensure that the rPET material always meets the critical customer’s requirements for extrusion, thermoforming and injection stretch blow moulding.

**Source:** The raw material will be from Post Consumer beverage bottle collections.

**Applications:** To be processed into sheet for thermoforming or for injection stretch blow moulding into bottles.

<table>
<thead>
<tr>
<th>PROPERTY</th>
<th>TEST METHOD and UNITS</th>
<th>TARGET VALUE</th>
<th>Tolerance</th>
</tr>
</thead>
<tbody>
<tr>
<td>Intrinsic Viscosity</td>
<td>solution or melt equivalent dl/g</td>
<td>0.76</td>
<td>+/- 0.2</td>
</tr>
<tr>
<td>Colour L value</td>
<td>Colour spectrophotometer</td>
<td>75</td>
<td>+/- 10</td>
</tr>
<tr>
<td>Colour a value</td>
<td>Colour spectrophotometer</td>
<td>-2</td>
<td>+/- 1</td>
</tr>
<tr>
<td>Colour b value</td>
<td>Colour spectrophotometer</td>
<td>2.5</td>
<td>+/- 1.5</td>
</tr>
<tr>
<td>Colour YI value</td>
<td>Colour spectrophotometer</td>
<td>5</td>
<td>+/- 2</td>
</tr>
<tr>
<td>Average Pellet size</td>
<td>Measurement of length, diameter in mm.</td>
<td>3</td>
<td>+/- 1</td>
</tr>
<tr>
<td>Fines less than 0.5mm</td>
<td>sieve analysis</td>
<td>0.1</td>
<td>+/- 0.05%</td>
</tr>
<tr>
<td>Bulk Density</td>
<td>mass/volume kg/m3</td>
<td>840</td>
<td>+/- 40</td>
</tr>
<tr>
<td>Moisture Content</td>
<td>weight loss</td>
<td>0.3%</td>
<td>+/- 0.1%</td>
</tr>
<tr>
<td>Foreign Materials-PVC, polyolefins, metals, other materials</td>
<td>ppm</td>
<td>&lt;1 ppm in total</td>
<td></td>
</tr>
<tr>
<td>Black Specs, using filtration &lt;80 microns</td>
<td>Microscopy Black specs above 100 microns</td>
<td>None</td>
<td></td>
</tr>
</tbody>
</table>

Table 6. Specification for Crystallised Food Grade PCR PET Pellet
3 Status and Economics of Food Grade recycling

3.1 Background on the characteristics of PET

PET has several unique characteristics which are critical to successful processing and recycling of PET:

**Drying:**
In order to maintain the molecular weight and intrinsic viscosity, PET must be thoroughly dried to achieve a moisture level of less than 50 ppm before melt processing. This is usually achieved by drying in a dehumidifier drier with a dew point of -40 ºC for at least five hours at 170 ºC.

**Morphology:**
The morphology or molecular structure of PET is such that when heated above its glass transition temperature of 80 ºC, it changes from a rigid glass state into an elastic, pliable form.

All thermoplastic melts have an amorphous morphology. This means that the macromolecules have a randomly oriented structure. During processing, the polymer melt is cooled to solidify in the shape of the bottle. During cooling, some polymers, including PET, have sufficient molecular chain mobility that they are able to form crystalline regions, where the macromolecules are aligned or oriented, forming highly ordered regions. Polymers can never be fully crystalline, but can be semi-crystalline, with crystalline and amorphous regions present.

Crystallinity can also be induced by molecular orientation. This can occur by random molecular rotation at elevated temperatures or by mechanically stretching the polymer while it is heated above its glass transition temperature. The mechanically induced crystallinity greatly strengthens and stiffens the polymer. This property is exploited in the manufacture of fibres, film and stretch blow moulded bottles.

In the latter case, by stretching test tube shaped preforms, tough, clear, low gas permeability bottles are produced.

PET bottles have two distinct morphologies:
- **The thicker neck and base section of the bottles.**
  These regions have an amorphous structure since the moulded preform is rapidly cooled from the melt state and there is insufficient time for molecular realignment in to crystallites and hence limited opportunity for crystallinity to develop.
- **The thinner wall sections of the bottles.**
  These regions are formed by the biaxial orientation of the preform above the Glass Transition temperature. The preform is stretched vertically by an internal pin and
horizontally by compressed air. The mechanical stretching and then cooling of the macromolecular chains creates biaxially oriented crystalline phases. The physical and permeability properties of these two morphologies are significantly different. The more crystalline biaxially oriented phase is much tougher and less permeable than the amorphous phase (which is more susceptible to absorption of low molecular products).

**Additives and Diffusion:**
The desired properties for packaging applications are achieved from the intrinsic properties of PET. Therefore additives, such as are needed for PVC, are not needed and so cannot migrate out of the plastic. The low diffusion coefficient of PET makes it much more suitable than other plastics materials for use as a recovered recycled material. Its low diffusion coefficient means that it is 100 times more difficult for chemicals to diffuse into or out of PET than for some other materials.

These material characteristics are particularly important when considering PET recycling.

### 3.2 Approaches to Food Grade Recycling

Three approaches have been developed for the production of food grade PET from recyclate:
- Chemical or Feedstock recycling
- Multilayer processing
- Superclean or Physical recycling

The current status of each of these approaches will be reviewed. This is essential in order to determine the benefits of further process development.

### 3.3 Chemical or Feedstock recycling

PET (a condensation polymer) may be depolymerised to produce chemical precursors, which may then be re-polymerised to PET.
Feedstock recycling may be subdivided into two categories; processes that involve breaking down mixed plastics to form a ‘naphtha’ feedstock for petrochemical processing, which could include production of PET, and processes that depolymerise PET feedstock to produce a more limited range of chemical precursors, which are then purified and re-polymerised to produce PET. PET specific processes are considered here. PET may be depolymerised by a range of chemical agents and processing conditions, the most important of which are summarised below. A range of factors is involved in process selection including the quality of feedstock available and demand for the intermediates formed.
3.3.1 Methanolysis

PET may be depolymerised to produce dimethyl terephthalate (DMT) and ethylene glycol (EG) by reaction with methanol, under pressure at around 200ºC. The DMT produced may be purified by distillation and crystallisation to give a high quality intermediate, which may be used to produce new PET. Once refined the EG may be used for a variety of applications, including antifreeze and PET production.

3.3.2 Glycolysis

PET may be depolymerised to produce bis-hydroxyethylene terephthalate (BHET) and short chain polymers of just a few repeat units (oligomers). The BHET produced may be purified by melt filtration under pressure to remove physical impurities and treated with carbon to remove chemical impurities. Recent recipients of FDA letters of non-object for glycolysis based processes include: Hoechst Celanese (1995), Wellman Inc. (1996), Innovations in PET Pty Ltd. (1996), The Eastman Chemical Co. (1997 / 2000), Teijin, NanYa, and AIES (2001), Roychem, OHL and Mitsubishi (2003).

3.3.3 Hydrolysis

PET may be hydrolysed by treatment with water, acids or caustic soda to give terephthalic acid (TA) and ethylene glycol (EG), which may be repolymerised following purification. PET hydrolysis is less well commercially established than glycolysis or methanolysis. RecocPET / Technochim Engineering in France has developed a patented caustic hydrolysis process to recycle PET.

3.3.4 Chemical Recycling Summary

The first generation of recycling processes issued with FDA non-objects were based on methanolysis. More recent successful applications utilise glycolysis or glycolysis / methanolysis. Methanolysis, hydrolysis and saponification are able to remove at least some colour from coloured PET feedstock; glycolysis may not remove all the impurities and a certain amount of degradation and yellowing may occur. Glycolysis plants are frequently integrated with conventional PET production plants where the recovered BHET can be blended with new BHET to give the required end product quality. Glycolysis is most suited to recovered material of known history and high quality. Conversion costs of glycolysis may be less than those of methanolysis since the intermediate formed (BHET) is further down the PET production chain than produced by methanolysis (DMT). Feedstock recycling has greater flexibility over the composition of the feed material and greater tolerance to impurities than multilayer processing and Superclean recycling. Feedstock recycling is, however, relatively capital intensive and may only be commercially viable where comparatively large throughputs are possible. The competitiveness of feedstock recycling is improving as new technologies are developed.
3.4 Multilayer Processing

Multilayer bottles have a functional barrier between the recyclate and the foodstuff. A 25 micron virgin layer either side of an internal recycled content layer has been demonstrated as being sufficient to provide a functional barrier to prevent migration of any potential contaminants to the foodstuff substance.

High quality mechanically recycled food grade polymer, derived from sources such as limited refill bottles and bottles recovered through deposit or kerbside collection schemes may be sandwiched between layers of virgin polymer. Acceptance of multilayer bottles for food contact applications was dependent upon the demonstration of the functional barrier, preventing the migration of chemical contamination present in the recyclate to the foodstuff. Multilayer bottles generally incorporate 40-60% R PET.

Extensive testing involving the deliberate introduction of a range of model contaminants into the pcr PET middle layer of multilayer PET bottle, followed by testing under specified conditions has demonstrated the action of PET as a functional barrier. Multilayer PET bottles are now well established, having received ‘non-object’ for food contact applications in several markets including the UK. The USFDA have issued twelve letters of ‘non-object’ to processes based on physical processing of recyclate for use in non-contact multilayer food packaging, recipients include:

- Continental PET Technologies Inc. (1992, 1994) - soft drink bottles
- Union Carbide (1995) - bottles
- PET Technologies Ltd. (1998) – bottles
- Ivex Packaging Corp (2000)

Altoplast and Hofstetter (both Swiss companies) have patented variations on preforms with recycled content.

3.5 Superclean or Physical Recycling

Several organisations have developed processes using mechanical and non-mechanical procedures to recycle high quality post consumer material, producing polymer suitable for use in mono-layer applications i.e. use in direct contact with food.

Superclean processing involves the removal of volatile contaminants with maintenance and raising of intrinsic viscosity (IV) to bottle grade (0.75-0.82 dl g⁻¹). The processes are proprietary, but where details are known they generally involve combinations of standard mechanical recycling processes with non-mechanical procedures such as high temperature washing, high temperature and pressure treatments, use of pressure and catalysts and filtration, to remove polymer entrained contaminants and raise IV. Commercially significant European examples include:

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3 A functional barrier is defined as any integral layer, which under foreseeable conditions of use reduces the migration of components from any layer beyond the barrier to technologically unavoidable and analytically insignificant levels (ILSI, 1998). FDA have specified 0.025 mm (1 mil) of virgin PET as a functional barrier (http://www.fsan.fda.gov/~dms/opac-recy.html).
3.5.1 **SupercycleTM PET (Schmalbach-Lubeca PET Containers)**

Developed by Johnson Controls Industries, now part of Amcor (previously Schmalbach-Lubeca) PET Containers, SupercycleTM PET was the first mono-layer ‘Superclean’ mechanically reprocessed PET to obtain FDA non-object for direct food contact in 1994. SupercycleTM PET has been produced at the 9,000 tonne/year Novi plant at Michigan, US since 1994, for use in the US, Australia and Europe. A second SupercycleTM plant opened in Beaune, France with a capacity of 6,000 tonnes/year in 1998. This plant now also operates the more advanced Buhler process with an output of 20,000 tonnes per year.

3.5.2 **EcoclearTM (Wellman)**

Wellman received non-object status from the USFDA in 1996 for EcoclearTM recycled PET, or use at up to 100% for food contact applications. EcoclearTM is manufactured by Wellman using a proprietary process based upon automated sorting and chemical/thermal cleaning of food grade bottles from collection systems. EcoclearTM recycled content (25% RPET:75% virgin PET) PET resin and PET sheet for beverage containers is produced at Wellman’s 30,000 tonne/year Emmen site in Holland. EcoclearTM has received regulatory approvals for use in many European markets and is widely used in Scandinavia and countries such as Holland and Belgium.

3.5.3 **OHL Apparatebau and Verfahrenstechnik GmbH**

OHL supply a range of products including: extrusion and solid-state post condensation equipment, in a range of sizes from 5,000 – 30,000 tonnes/year. The FDA approved OHL Stehning PET recycling process has been used in Germany since 1999. OHL report plant costs for food contact processing as follows: 5,000 tonnes/year DM 8-9 million, 10,000 tonnes/year DM 10-11 million; 15,000 tonnes/year DM 12-13 million.

The OHL Stehning bottle to bottle process consists of the following main stages:

- Washing and grinding: hot water/steam pre-wash, manual inspection (optional), automatic sort system (aluminium cans and caps, PVC and colour sorting), produce a PET bottle stream for wet grinding and several intensive washing operations (involving high temperature friction washing, flotation and centrifugation) before flash drying. The flakes then enter a further grinding stage for final size reduction.

- The clean dry flake feed is fed to modified extrusion system (vented double screw extruder) where diffused volatile components are partially eliminated and solid contaminants are filtered out by melt filtration before regranulation.

- Solid State polycondensation (SSP) in a batch reactor (vacuum rotary dryer) eliminates volatile by-products such as glycol, acetaldehyde and oligomers, raises the IV of the recyclate to a level suitable for bottle production i.e. from (~0.70 dl-1 to ~0.82 dl-1) and converts the amorphous PET to crystalline PET.

- The OHL system allows control over IV up to 1.1 dl-1, hence recyclate could be used for a range of high performance products such as engineering applications, strapping, fabric for filters, tire cord etc.

3.5.4 **Erema Plastic Recycling Systems**

The Erema TE-VSV (VacuRema) process converts clean dry bottle flake to crystallised pellets meeting food safety requirements and suitable for use as raw material for bottle or sheet production. It can also produce a melt for direct extrusion to a finished product without the need for pelletisation.
The VacuRema was granted an initial ‘non-object’ by the USFDA in November 2000 for production of pcr PET for use in food contact applications from post consumer soft drink and mineral water bottles. The Erema system has subsequently received food contact approval from Pepsi USA and in Germany, Switzerland and Austria, with food contact approval applied for in Canada, Hungary and Brazil.

The VacuRema process comes in a number of variations. In its most advanced process, clean dry bottle flake from a wash plant is fed to the Erema crystallisation dryer, which may be under vacuum if required, where material is pre-heated, dried and crystallised in one operation. Heated flake is passed to the next vacuum “VacuRema” reactor and vacuum vented extruder where it is melt filtered and pelletised. Residual heat in the strands is used to crystallise the pellets. The high temperatures and vacuum employed effectively removes any moisture and volatile contaminants from the PET flake and can increase viscosity up to virgin material levels e.g. typically to 0.77 to 0.80 dlg-1.

Newer versions of the VacuRema use optional solid stating on the final pellets for further control over the IV.

The VacuRema system is currently being used at a number of sites in Europe (30 units sold by 2004) for food grade applications as well as in-line processing leading to production of a range of non-food products such as sheet, staple fibres and strapping.

Fig. 3 Erema Process diagrams
3.5.5 Bühler Process

The Bühler process produces bottle grade pellets from PC PET bottles. The process is reported to be competitive with virgin PET, to cut energy consumption associated with production of virgin PET by 10%, to below 150 KWh/tonne.

Fig. 4  Bühler Process diagram

The key process stages are: decontamination, filtration, granulation, drying, crystallisation and solid state polycondensation. The technology involves the Bühler Ring Extruder and continuous Solid State Polycondensation (SSP) processes. The Bühler process was awarded an FDA letter of ‘non-object’ in 2001. The first Bühler food contact PET reprocessing line was installed by Amcor (then Schmalbach Lubecca) in Beaune, France in 2001. The plant has an annual output capacity of 20,000 tonnes per year.
3.5.6 Starlinger Recostar IV Plus

The Starlinger recycling system is similar to the Erema process in that it starts with washed PET flake which is extruded and melt filtered by an integrated unit that preheats the flake before feeding it into the extruder. The pelletised resin then is fed to a modular crystalliser and vertical vacuum solid stating unit that allows the pellet to be decontaminated, reduced in Acetaldehyde levels and increased in IV up to virgin levels (depending on the residence time in the unit). The solid stating unit is built in modules of 200kgs per hour allowing flexibility in capital cost and product output specifications. Multiple units could each operate at different residence time to suit different markets eg water bottle and CSD simultaneously.

![Starlinger Recostar Plus](image)

3.5.7 VISY Industries

This process was approved in 2001 by USFDA and utilises commingled plastics bottles or sorted PET bottles as input. The bottles are pre-washed and then ground to flake and washed in a hot alkaline wash with specific detergents. The PET flakes are then dried in a conventional drier to below 100 ppm of moisture and then extruded in either a single or twin screw extruder fitted with triple vacuum venting. The extruder has a special profile and high levels of vacuum applied to the extruder vent ports to decontaminate the melt which is processed at 270 to 300 °C. The final material has an IV of 0.76 to 0.78. The special advantage of this process is the simplicity of the process and the use of standardised equipment for the processing steps.
3.5.8 United Resource Recovery Corporation (URRC) process.

The process starts with sorted PET bottles. In the first stage, the bottles are separated out and ground into flakes. Any remaining contents, labels, closures, and foreign matter are also taken out. This is achieved using conventional, dry and wet-operation separating techniques as well as via a washing technology. In the second stage of the process, both adherent and migrated impurities are removed. At the same time, the surface of the flakes is detached in a reaction using caustic compounds. In a rotary tubular kiln, residual foreign substances and odours are effectively removed using heat treatment where the flakes and caustic compounds are kept at high temperatures (up to 200 °C) for up to 5 hours. In the third stage, the cleaned PET is rinsed and freed of coloured flakes with colour sorting. Any PVC remnants also change colour in the rotary tubular kiln and are detected and expelled in the colour sorting process. In this operation, any remaining foreign particles and tiny impurities can also be removed.

The process can cope with highly contaminated PET as well as making a final product in flake form that can be used directly or extruded before use. The final IV is in the range 0.76 to 0.77. Plants are currently in use in Europe in Switzerland, (Recypet in Fraunfeld), and Germany, (Cleanaway, ex SKP GmbH, in Rostock), and in Mexico (2005).

Figure 6 Tubular reactor for treating flake and caustic mixture.
3.5.9 Phoenix PTI Process

The process begins with washed flake with a controlled specification. The flake is ground to a controlled particle size distribution typically 1 to 4 mm with an average of 2 mm. This flake is then placed into a vacuum solid stating device for decontamination and IV building to levels of 0.86 to 0.88. The principle behind the process is that the smaller flake size allows an exponentially faster decontamination of the PET flakes. The solid stating device used is typically a batch wise rotary vacuum vessel which takes between 8 to 10 hours per cycle with a critical condition of 4 hours at or over 200 °C being always maintained as a condition of the FDA approved (1999) process. The flakes are then extruded in a single screw extruder after passing through a drier. The resultant pellets have an IV of 0.76 to 0.78. Plants are in operation at Phoenix Recycling in Bowling Green, Ohio USA and Visy Industries, Prestons, NSW, Australia

Figure 7. OHL vacuum vessel used in Phoenix process in Australia at Visy Prestons plant.

3.6 Comparison of Status of Food Grade Recycling approaches

Chemical recycling is currently being conducted Teijin in Japan, after previous trials by DuPont and Eastman were closed down in late 1990s. The key issue has been that the process costs have meant that the final price has exceeded virgin price. The capital investment for these plants is typically on excess of US$10m and the plants need to be located adjacent to polymerisation operations. So despite having ready approval from USFDA, higher cost such a process has inhibited its commercial exploitation.

Multilayer co-injection techniques have largely been supplanted by monolayer blends due to the technical simplicity and the lower capital cost of the machines and moulds for perform injection.

Superclean or Physical Recycling processes were investigated for their safety in the major EU Project FAIR-CT98-4318. It was concluded that “...from the project results and from many other findings and considerations it can be concluded that modern Superclean
technologies can safely reprocess rPET into new materials and articles for direct food applications which are indistinguishable from virgin food grade PET.\(^\text{18}\)

The feedstock for the early “Superclean” processes was typically bottles collected under deposit and refund schemes to control the purity of the incoming PET bottles. From 2000 onwards the majority of the approved processes have used kerbside collection sources for PET bottles and have demonstrated that they can achieve the high levels of purity required to meet the USFDA and EU requirements.

The FAIR study referred to above showed that only 0.03 to 0.04 % of bottles were chemically “misused” and the maximum levels of consumer exposure to misuse chemicals such as solvents would be less than 50 ppb per day. This data justifies the inclusion of bottles that have been used for non food applications and from less controlled sources such as kerbside collection.

All the Physical Recycling approaches that have been evaluated by the USFDA and been granted letters of “Non Objection”. However, of all of the physical processes only three have approval from the Coca Cola company.

1. Phoenix Recycling
2. URRC Process
3. Buhler Process

The approval from Coca Cola is based on the ability of the process to remove residual flavourants such as limonene and cineole that may be within the flakes of recycled PET from previous applications. Removal of these flavourants usually ensures the removal of other potential contaminants.

Typically all of the processes will need to utilise a high quality washing process for the PET flakes to achieve Bottle-to-Bottle quality standards. This will usually involve a prewash of bottles followed by bottle sorting and grinding to 10 to 14 mm flake and then a hot (>80 °C) wash using caustic soda and low foaming detergents. At the next stage there is a decontamination process either before or after extrusion which involves exposure to at least 4 hours at 200 °C with the application of vacuum or within an inert atmosphere such as nitrogen.

Processes that achieve decontamination in extrusion processes have had difficulty in gaining Coca Cola approval (since it specifies the contaminant level in flakes prior to melt processing) with the exception of the original Supercycle process and the subsequent improvement into the Buhler process.

The development of a number of Food Grade approved processes has meant that any new plant will need to consider the balance between the need for Coca Cola approval, process efficiency, automation and capital cost. So far all of the processes developed cannot deliver the best of performance in each of those sectors consequently making a final choice a matter of compromise and preference.
4 Process development of rPET using sortation and vented extrusion.

Commercial in Confidence

4.1 Process Economics using sortation and vented extrusion technology

As discussed in section 3 there are many recycling processes for PET and up to 60 have USFDA approval for use with food contact applications however fewer than that number are in use due to the economics being less favourable compared to virgin resin.

In the last quarter in UK of 2004 the price of virgin resin varied between £800 and £1000 per tonne and the price of baled clear PET bottles varied from £70 to £120 per tonne. Given that a recycling company may have to sell at least a proportion of its output at 70% of the price of virgin resin this means that there is a margin of approximately £440 to £580/tonne available for a recycling operation to make a profitable business out of recycling PET back into food grade resins on a reliable basis. The best margins would be achieved by supplying the resin back into beverage containers such carbonated soft drinks. Coca Cola is the dominant global company in this field and it would almost certainly be a target customer for any sales of recycled PET. To do this the recycled resin would need to have Coca Cola approval.

These requirements for a potentially successful PET operation can be expressed as the following equation:

\[
\text{PET Bottle cost} + \text{Process cost} + \text{Coca Cola approval} < £560 \text{ to } £700/\text{t}
\]

This means the Process cost should be < £440 to £580/t.

Process costs of current recycling processes have been £400 to £600/t which would allow only a small residual profit to pay back the capital which could be in the order of £6 to 10 million for a plant operating at 2 tonnes per hour.

Recycling processes may not be viable when the market forces up the price of collected PET bottles or if the cost of energy and water increase making a process more expensive.

The target for this Project is to develop a new process with a cost of less than £350/t and a capacity to meet the most stringent quality standards including Coca Cola approval. Such a process should remain economically viable under a range of market conditions.

This new process will employ a new flake separation principle that will potentially reduce the contamination level by a factor of 2 to 5. It will eliminate a Solid Stating process by the use of High Temperature Drying under Nitrogen.
It will use specially configured triple vacuum vent Twin Screw extruders to further decontaminate and increase the IV of the rPET to 0.76 to 0.78. This process is compared with conventional processes in Table 8.

<table>
<thead>
<tr>
<th>Steps and Cost/ Tonne</th>
<th>Conventional Processes</th>
<th>New Process</th>
</tr>
</thead>
<tbody>
<tr>
<td>De-Baling</td>
<td>Y</td>
<td>Y</td>
</tr>
<tr>
<td>Whole Bottle Wash</td>
<td>Y</td>
<td>Y</td>
</tr>
<tr>
<td>Sorting and Metal Removal</td>
<td>Y</td>
<td>Y</td>
</tr>
<tr>
<td>Grinding</td>
<td>Y</td>
<td>Y</td>
</tr>
<tr>
<td>Hot Detergent Wash</td>
<td>£150 for the above steps</td>
<td>£150 for the above steps</td>
</tr>
<tr>
<td>Sorting by destoning</td>
<td>N</td>
<td>Y</td>
</tr>
<tr>
<td>Drying</td>
<td>Y</td>
<td>Y</td>
</tr>
<tr>
<td>Nitrogen Drying</td>
<td>N</td>
<td>Y</td>
</tr>
<tr>
<td>Extrusion and pelletisation</td>
<td>£250 for the above steps</td>
<td>N</td>
</tr>
<tr>
<td>Decontamination/ Solid Stating</td>
<td>£100 for this step</td>
<td>£50 for this step</td>
</tr>
<tr>
<td>Extrusion/Decontamination/ IV Build/pelletisation</td>
<td>N</td>
<td>£150 for this step</td>
</tr>
</tbody>
</table>

**Indicative Total Cost/Tonne**

|                     | **£500** | **£350** |

Y = Yes, this step is part of the process; N = No, this step is not part of the process

Table 8. Steps of conventional superclean processes versus the new process

**4.2 Sortation Technology to reduce contamination**

Recent work from Swinburne University\(^1^9\) reported detailed analysis of the washed contaminated flake. They found that there were significantly more contaminants in the smaller particles than in the larger particles, even though all of the measurements were in the 10 to 1000 ppb range. Fig 8 shows the levels extracted from small particles <300 µm over a period of extraction time. Figures 9 and 10 shows the 2 to 5 fold variation of contaminants with particle size going from < 300 µm (small) to 300-425 µm (medium) and 425-700µm (large).
Fig 8 Equilibrium levels of residual contaminants

Fig 9 Extraction of residual contaminants (low level) with differing particle size, small, medium and large.
Fig 10. Extraction of residual contaminants (higher level) with differing particle size, small, medium and large.

In seeking an explanation for this phenomenon, the authors measured the crystallinity of the particles by X-Ray diffraction (XRD) and also of differing sections of bottles. The data shown in Tables 9A and 9B shows that the particles, which were made by grinding flakes, have the character much more like the amorphous sections of the bottles.

<table>
<thead>
<tr>
<th></th>
<th>Mid section of bottle crystallinity (75.4% by mass in total sample)</th>
<th>Top and bottom of bottle crystallinity (24.6% by mass in total sample)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ground to 0-300 µm</td>
<td>48.41 %</td>
<td>19.61 %</td>
</tr>
<tr>
<td>Ground to 300-425 µm</td>
<td>51.85 %</td>
<td>19.22 %</td>
</tr>
<tr>
<td>Ground to 425-700 µm</td>
<td>48.93 %</td>
<td>12.94 %</td>
</tr>
</tbody>
</table>

Table 9A Percentage Crystallinity of different sections of bottles and ground PET particles.
Table 9B Percentage crystallinity of two batches of unsegregated ground flake

<table>
<thead>
<tr>
<th>Ground Size</th>
<th>Batch 1</th>
<th>Batch 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-300 µm</td>
<td>20.6 %</td>
<td>22.8 %</td>
</tr>
<tr>
<td>300-425 µm</td>
<td>20.9 %</td>
<td>22.2 %</td>
</tr>
<tr>
<td>425-700 µm</td>
<td>23.0 %</td>
<td>23.6 %</td>
</tr>
</tbody>
</table>

If it assumed that the crystallinity of Base and neck (amorphous PET) is 19% 
Bottle wall (crystalline PET) is 49% 
Then the particles in the 0-300 µm range with 20.6 to 22.8% crystallinity are largely amorphous and the particles in 425-700 µm range with crystallinity of 23.0 to 23.6% contains slightly more crystalline content. This difference in crystallinity is attributed as being responsible for a 2 to 5 fold reduction in contamination due to the lower absorptivity of the more crystalline component of the PET bottle.

The method of preparing the ground PET was to place the flakes into a grinder and to allow the blades to impact the flakes before the powder is removed. The powder seems to be generated by the minor phase of the more brittle amorphous fragments present in the flakes. This suggests that separation of the crystalline and amorphous sections could reduce the level of contaminants in the more crystalline fraction by a factor of 2 to 5 times depending on molecular weight and diffusivity in PET. The thinner, more crystalline sections will absorb less contamination. Therefore by separating this fraction, it can become the feed material for a cleaner higher value stream. This separation can be achieved by the use of sorters typically called destoners or air classifiers.

4.3 Separation Technology

Destoners and Air classifiers are widely used commercially in the food processing industries. They are also used in the plastic recycling and processing areas to separate out fines (dust) and oversize particles. The standard function of the destoner is to effectively separate heavier-than-product debris, such as glass, stones and metal from a large amount of light product. The principle is that an inclined plate is vibrated and under set pressure the lighter flakes move down the plate. For example, Forsberg Destoners, shown in Fig. 11, are constructed of heavy gauge steel and driven by a unique eccentric drive which is counterbalanced to ensure minimum maintenance operation.
Air classifiers work by using the principle of terminal velocity for a specific product to classify and separate particles as in Fig. 12 and 13. When particles have different terminal velocities due to different size, density or shape, some will fall, while lighter particles will be lifted by an air stream. By using multiple aspirators, and variable air flow rates, appropriate separation can be achieved.

Each pass lifts approximately 75% of the fine particles that can be lifted at a set velocity. This leaves a balance of 25% of the fines in the through stock. On a dual pass unit the second pass would remove 75% of the remaining fines (25%), leaving 6.25% fines in the thru stock, which figures 93.75% efficiency. By using multiple passes the efficiency of collecting the fine material is improved dramatically without “over aspirating” and inadvertently pulling heavy material.

The air supply is drawn by suction into the aspiration zone through the hollow slide, with the stock cascading down from the slide directly above. As the stock drops off the end of the slide, it is contacted by high velocity air. This tends to lift all the particles, but due to the expanding shape of the suction area near the air outlet slots, the air velocity slows down and the “hi-terminal velocity” particles tumble to the next slide, exposing any "trapped" lightweight particles to re-aspiration. Only the "low-terminal velocity" particles are lifted through the low-velocity zone and are conveyed away with the aspiration air. Recent trials have indicated success in separation of thick from thin flaked PET as shown in Fig 14.
Fig 12. Principle of the four pas aspirator air classifier, showing particles entering from the top and air entering from the right. Light particles are lifted to the cyclone, whereas heavy particles become the “thru stock”.

Fig 13. Multiple Aspiration Equipment (6 pass) from the Kice Industries Wichita laboratory.
Fig 14. Examples of the showing the separation of post consumer PET bottles into light (left) and heavy (right) fractions.

5 Conclusions

- The extensive use of PET as a packaging material and the greater emphasis by communities on recycling packaging materials has led to the development of regulations, commercial processes and procedures to ensure the safe recycling of PET back into food grade application sourced from post consumer collection systems.
Recent EU directives and regulations encourage the use of recycled PET for food contact materials through the development of new guidelines and legislation.

The quality requirements for food contact materials are well researched and specified in challenge test guidelines developed by USFDA, ILSI and Coca Cola.

Commercially, physical recycling is the most feasible approach due to the development of energy and resource efficient processes.
6 References and End Notes

9 Framework Regulation (EC) 1935/2004 “Materials and articles intended to come into contact with food”, pages 1 and 3
13 Coca Cola Private correspondence
15 PAS103, 2004 BSI, BPF, WRAP Report - Collected waste plastic packaging, page 1

WRAP