Abstract:
The present invention relates to a process for recycling post-consumer polypropylene (PP) packaging to Food Grade recycled PP (rPP). The process comprises the steps of: (i) providing an amount of polypropylene comprising at least about 99% food grade polypropylene; (ii) extruding the polypropylene, employing a residence time of from about 1 minute to about 3 minutes for the polypropylene in a vacuum zone of the extruder; and (iii) exposing the polypropylene to a temperature of from about 130°C to about 170°C for about 3 to about 8 hours.
Recycling of Polypropylene

The present invention relates to a process for recycling post-consumer polypropylene (PP) packaging to Food Grade recycled PP (rPP).

Recycling of waste products has become increasingly common practice in the last couple of decades, and the recycling of plastic materials is one of the most important and widely carried out with many industries and households around the world actively involved. A multitude of everyday consumer items are made from plastic materials, such as bottles, bags and product packaging.

However, it is necessary to monitor and ensure the purity of the recycled plastics in order to substantially remove the presence of any contaminants before the plastics can be re-used as packaging for food products.

In 1992 and 1995 the US Food and Drug Administration (FDA) published guidelines for industry dealing with post-consumer plastics for direct food contact applications. These guidelines provide recommendations about testing of the cleaning efficiency of the investigated recycling process and the maximum content of post-consumer recyclate in packaging materials as well as threshold limits for migration. These guidelines have now been updated to include new knowledge, mainly for PET, and recommendations are given for feedstock material from non-food applications, which are intended to be recycled into food packaging.

The International Life Sciences Institute (ILSI) published a report in 1998 where it was concluded that materials manufactured from 100% recycled content can successfully be recycled through three cycle operations (100% recycled). These materials are polyethylene terephthalate (PET), polystyrene (PS), polyethylene (PE)
and polypropylene (PP) materials which had undergone processing, such as washing, melting and re-extrusion etc. No significant effect of these multiple reprocessing operations was found on levels of overall migration, molecular weight distribution of the polymer, monomer migration levels, additive migration levels, the presence of oligomers in the plastics, and the formation and levels of additive degradation products.

It can thus be concluded that recycled plastics per se, with respect to intrinsic physical properties and their migration behaviour, can be regarded for all practical purposes as equivalent to virgin materials. This conclusion is based on a maximum average number of three cycle operations.

The only significant difference between recycled and virgin materials is the previous use of the material being recycled. Concern then centres on any contaminants from this previous use that might be introduced into the recycled materials that may potentially lead to contamination of food products which are brought into contact with the recycled material.

The ILSI report states that 'because any contamination episode is likely to be rare and sporadic, it can be argued that every production batch should be tested'. This would be difficult and indeed impractical due to the possible unknown nature of any adventitious contamination that might be introduced. To ensure a sufficient safety margin, the analytical limits need to be set at levels below those that might reasonably be achieved in a routine testing laboratory. This conventional approach of ensuring safety with emphasis on the end product therefore seems to offer less assurance than demonstrating that the process itself has certain protective capabilities.
The subsequent clean-up stages undertaken during recycling are aimed at eliminating substances that may contaminate the surface and small, mobile molecules that may penetrate the plastic. In contrast, ingredients of plastics (additives and processing aids) and their degradation products may be less likely to be removed by the cleaning process. All of these additives must themselves be approved for food contact and be on the list of approved substances that can be used in food contact as published by FSA and USFDA. The effects of dilution will further reduce the potential impact of these substances.

Thus, any recycling operation must demonstrate the capability to select and use only food-grade feedstock. This task is made easier than it may initially seem by selecting the in-feed packaging formats such as bottles, tubs and trays which by their very nature will be made from food grade plastics. The incidence of non-food grade plastics is then very low. The required minimum efficiency of any sorting operation to remove plastics that have been used for non-food applications would have to be decided on an individual case-by-case basis, which would take into account the likely presence and levels of non-permitted additives in particular plastics and the level of any absorbed substances from non-food ingredients. However, even for a polymer such as PET, which is essentially made only in grades for food contact, it was judged that a recommended level of effectiveness of 99% should be met even though this level is not specified in any regulations relating to the recycling of plastics to food grade quality. Expert authorities judge this level as a guide to the capacity of the recycling process overcoming challenges in removing any potential contaminants. The aim is therefore to reach a high level of purity in the final product by employing
separation steps after the input stage to further improve the quality of the separation so that levels approaching 100% purity are reached in the final resin.

By the term 'food grade plastics' used above, it is meant herein materials which have already been used in contact with or as packaging for foods and can demonstrate that they comply with the relevant food contact regulations of EFSA and USFDA.

In 2005, a complete HDPE bottle-to-bottle recycling process was developed, challenge tested and validated. This showed that HDPE (high density polyethylene) milk bottles used in the UK could safely be mechanically recycled and the resultant polymer remoulded into milk bottles at an addition rate of up to 50%.

In 2007, this process was scaled up to see if it would be stable enough to produce desirable results on a consistent basis. This decontamination technology demonstrated its efficiency via challenge tests that show it is capable of decontaminating HDPE to a 'super-clean' state that meets food-packaging standards.

Due to the fact that most of the detected suspicious compounds were related to flavour and fragrance compounds, this process recommended organoleptic tests with milk using bottles manufactured from super-clean recycled post-consumer HDPE. Also, 'overall migration' tests were performed, and it was found that the overall migration was significantly lower than the 60mg/kg limit set by the EU Plastics Directive 2002/72/EC.

Based on the results obtained from overall and specific migration tests on bottles produced with 50% recycled HDPE manufactured using the described cleaning and decontamination processes it was possible to prove that the bottles satisfy Article 3 of EC Directive 1935/2004, which states that:
Materials and articles, including active and intelligent materials and articles, shall be manufactured in compliance with good manufacturing practice so that, under normal or foreseeable conditions of use, they do not transfer their constituents to food in quantities which could:

(a) Endanger human health; or
(b) Bring about an unacceptable change in the composition of the food; or
(c) Bring about deterioration in the organoleptic characteristics thereof.

All tests performed in this project demonstrated that the recycled HDPE bottles fully satisfy all of the above-described general requirements.

However, replacing the HDPE in this process with PP to try and recycle PP the same way was unsuccessful, because in the case of HDPE only milk and juice bottles were used with a low risk of contamination and in the case of PP there are many more product categories which led to a greater exposure to flavour and fragrance compounds.

It would therefore be desirable to devise a successful process for the recycling of food grade PP. The new process of the invention offers an efficient route to pure recycled food grade PP.

Therefore, in accordance with the present invention, there is provided a process for the recycling of polypropylene, the process comprising the steps of:

i) providing an amount of polypropylene comprising at least about 99% food grade polypropylene;
ii) extruding the polypropylene, employing a residence time of from about 1 minute to about 3 minutes for the polypropylene in a vacuum zone of the extruder;

iii) exposing the polypropylene to a temperature of from about 130°C to about 170°C for about 3 to about 8 hours.

The attempts to recycle PP did help to identify that the types of contaminants encountered were of a higher molecular weight than those used in the typical challenge tests and specifically, two materials were often encountered from shampoos and mouthwash, namely isopropyl myristate and hexyl salicylate. These compounds were difficult to remove in the process used for HDPE and it was clear that a more intensive process was needed to achieve the required level of decontamination. This is provided by the above-defined process of the invention.

The key steps in a process for recycling post consumer packaging to food grade quality is comprised of several distinct stages.

Firstly, the post consumer packaging separation and sorting must be carried out in order to ensure that the material is essentially all food grade PP in the initial application. It should be free of other polymers and contamination. According to the guidelines published by the European Food Safety Authority (EFSA) in Regulation 10/2011, the input material should be at least 99% food grade PP or higher. This sorting can be achieved by manual sorting, sorting by physical principles such as specific gravity over the narrow range that will isolate PP from other materials, or alternatively by using automatic sorting by techniques such as Near Infrared analysis in either large packaging format or a granulated flake format.
Typically, the PP is in a granulated flake form after being sorted and reduced in size by grinding equipment. While other physical forms of the PP may also equally be used in the process of the invention, reference will be made herein to PP flakes for convenience.

Optionally, after the sorting step, the flakes of post consumer PP packaging may then be subjected to a hot washing step. While this step is optional and is not essential for the process of the invention, it can help to improve the results even further, as it would especially remove labels, glues and some of the inks on the packaging as well as the volatile components even further. The conditions for this step would typically be washing the PP at about 80°C in an alkaline solution with a concentration of caustic soda of about 1-2% with an anionic detergent with a concentration of about 0.1-0.3% with a residence time of between 5 to 10 minutes. The flakes would then typically be dried to a moisture level of less than 3%, more typically less than 0.5%, by a combination of mechanical drying in a centrifuge and drying in warm air.

After the sorting step and any hot washing step, the PP is typically subjected to extrusion. The PP material, which may be e.g. in a flake form, may be extruded into another form, such as a pellet form.

The extruder will typically possess provision for vacuum venting and extended residence time in a specific zone thereof. Examples of such extruders include the Gneuss MRS extruder or the Extricom 12 screw planetary extruder. As an example, the MRS extruder has an in-feed consisting of a single feed section which moves the PP material after melting to a special mixing section where the material is effectively moved back towards the feed section by a special arrangement of five screws running
in the opposite direction to the feed screw. The net effect is that the material is
delayed by up to 3 minutes in that zone depending on the conditions. The MRS
extruder also provides a high degree of control over the residence time, vacuum level
and temperature.

The Extricom extruder provides a very high surface area, extended residence
time during extrusion, and controlled vacuum pump performance and melt
temperature conditions.

The vacuum in this zone is less than about 1000 Pa (10 millibar) and typically
less than about 200 Pa (2 millibar) to ensure that any volatile components are
removed during the residence time in this zone. The temperature of the PP during this
condition is above its melting point of about 170°C, and is typically between about
240°C to about 280°C.

The throughput of the extruder can be from about 20 kg/hr to about 1500 kg/hr
provided the mixing section is suitably sized to have an average residence time of
about 1-3 minutes, typically about 2 minutes, provided the vacuum provided can
maintain the specified levels as mentioned above.

After devolatilisation of the melt, the polymer is typically pumped out of the
extruder into a pelletising system where the PP is cooled into granules that are
cylindrical or spherical with major dimensions of about 1 mm to about 4 mm, and
typically about 3 mm.

Vacuum devolatilisation of the solid state granules is carried out at a
temperature below the melting point of PP for an extended time. This vacuum
devolatilisation step is conducted at an elevated temperature which is typically greater
than about 130°C, more typically about 140°C or higher, but below the melting point of PP of about 170°C.

The process can be conducted in either a batch process in equipment as manufactured by OHL engineering or a continuous process such as made by Buhler or Starlinger. In all of these arrangements the material is typically held at its specific temperature for a defined residence time. The residence time will typically be in the range of about 3 to about 8 hours, more typically about 4 to about 6 hours.

The vessel will be either be under a pressure of less than about 600 Pa (6 millibar, i.e. essentially a vacuum), or under atmospheric pressure under a stream of air or inert gas (such as nitrogen) that typically passes through the vessel at the rate of about 1 cubic foot per minute per kilogram of PP material. At the end of the specified residence time the material is typically cooled under atmospheric pressure to ambient conditions.

The vacuum devolatilisation step (iii) may be carried out either before or after the extrusion step (ii), as desired. Typically, however, the steps are carried out in their defined order (i)-(iii).

At the end of this sequence of steps the material will comply with the food grade conditions required by EFSA and the USFDA. This is ultimately tested by analysis of the decontaminated material by the testing of migration from the moulded rPP product into a specified food simulant that represents a specific food type such as aqueous, acidic or oily types of food.

Of course, the recycled PP material made according to the process of the invention must be proven to be safe and not to transfer any contaminant materials to the foods. One way to achieve this is to process the feedstock of polymer through a
cleaning procedure to remove any such contaminants. To establish that the contaminants are removed by the cleaning procedure, materials are artificially impregnated with quantities of contaminant compounds before being subjected to the cleaning procedure. The cleaned material is then analysed for the presence of the contaminant compounds. If levels of the contaminant compounds are sufficiently low, no further analysis is needed. If, however, residual levels of the contaminant compounds are detected, then migration testing is carried out on the finished articles to determine the degree of migration of the contaminant compounds from the PP to the foods which may occur. For a satisfactory level of migration, less than 10 ppb is typically desired.

As contaminant compounds, toluene, chlorobenzene, phenylchlorohexane, benzophenone, n-hexyl salicylate and isopropyl myristate were used, in concentrations of approximately 500-700 mg/kg.

Protocols have been drawn up for this procedure for PET in the 'Guidance and criteria for safe recycling of post consumer polyethylene terephthalate into new food packaging applications', R. Franz, F. Bayer and F. Welle, EU Contract No. FAIR-CT98-4318: RECYCLABILITY. These protocols have been followed herein and applied to PP, except that methyl stearate has been replaced as one of the contaminant compounds by n-hexyl salicylate and isopropyl myristate.

The sample of contaminated PP flakes and pellets was made up in triplicate. In order to analyse the levels of contaminant compounds, samples of the treated and cleaned PP were extracted in 25 ml of tetrahydrofuran by heating in sealed vials at 60°C for 3 days. After shaking, the supernatant liquid was filtered and transferred to a 2 ml volumetric flask to which 20 µl of the internal standard (containing toluene-dg,
chlorobenzene-d5, and benzophenone-dio) had already been added. After mixing, the solution was transferred to a vial and injected for GC/MS analysis.

Decontamination Tests and Challenge Test

Below is a summary of the Decontamination Tests and Challenge Test steps carried out in accordance with the process of the invention to decontaminate the PP:

1. Preparation of 165 kg of challenge test PP flake from a controlled source using white-pigmented material.

2. Preparation of trial quantities of process flake PP from natural regrind.

3. Post processing the flake to pellet at Gneuss in the vacuum vented single screw extruder (MRS) at 240-260°C and vacuum levels of less than 1000 Pa (10 millibar) on the vented section.

4. Analyse the decontaminated flake after 6 hours of residence time in the OHL reactor at a maximum temperature (140°C) and vacuum of 500 Pa (5 millibar).

5. Alternatively, process 50 kg the challenge test PP at OHL Engineering in the vacuum reactor.

6. Processing the challenge flake to pellet in a single step in the in the Extricom vacuum vented multi-screw Ring extruder or in the Gneuss MRS extruder.

7. Complete chemical analysis of the PP pellet for decontamination efficiency.

8. Preparation and conduct of PP challenge test and measurement of decontamination performance and suitability for food contact applications.
In the above process detail, steps 5 and 6 are presented as alternative steps to steps 3 and 4, and are not performed in addition to steps 3 and 4.

The analysis results of the amounts of the contaminant compounds obtained from the triplicate samples cleaned according to the process of the invention is shown in Table 1:

<table>
<thead>
<tr>
<th>Replicates</th>
<th>Toluene</th>
<th>Chlorobenzene</th>
<th>Benzophenone</th>
<th>Phenylcyclohexane</th>
<th>Hexylsalicylate</th>
<th>Isopropylmyristate</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>8.7</td>
<td>1.4</td>
<td>21</td>
<td>49</td>
</tr>
<tr>
<td>2</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>9.3</td>
<td>1.7</td>
<td>22</td>
<td>41</td>
</tr>
<tr>
<td>3</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>9.1</td>
<td>1.9</td>
<td>21</td>
<td>39</td>
</tr>
<tr>
<td>Mean</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>9.0</td>
<td>1.7</td>
<td>22</td>
<td>41</td>
</tr>
<tr>
<td>Standard deviation</td>
<td>0.1</td>
<td>0.1</td>
<td>0.3</td>
<td>0.2</td>
<td>0.5</td>
<td>1.7</td>
</tr>
</tbody>
</table>

These results show that high levels of decontamination were achieved using the process, with levels of toluene and chlorobenzene being virtually undetectable, and phenylchlorohexane at an acceptably low level.

Inputting the residual levels of the benzophenone, n-hexyl salicylate and isopropyl myristate into a recognised migration model (Migratest Lite) gave the following results in Table 2 using olive oil as a food simulant.
Table 2

<table>
<thead>
<tr>
<th>Contaminant</th>
<th>Migration mg/kg at each test condition</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>10 days at 5°C</td>
</tr>
<tr>
<td>Benzophenone</td>
<td>0.021</td>
</tr>
<tr>
<td>Isopropyl myristate</td>
<td>0.058</td>
</tr>
<tr>
<td>n-hexyl salicylate</td>
<td>0.041</td>
</tr>
</tbody>
</table>

For butter and margarine as the foodstuff, a reduction factor of 2 can be applied to these results (EU Regulation 10/2011). As the migration model is designed for compliance purposes it will overestimate migration levels, so these values will necessarily be higher than the actual migration which would be observed.

In summary, the process of the invention provides for a new and efficient process for recycling PP and removing the contaminant compounds which may be found associated with PP products in order to produce food grade recycled PP.

**Food Grade Decontamination Trials**

Large scale food grade decontamination trials were carried out on representative post-consumer UK household PP packaging waste using the process of the present invention.

Nine tonnes of post-consumer PP packaging was sourced from ECO Plastics Limited, who sort mixed plastics from UK MRF's using near infrared (NIR) sorting equipment. This nine tonnes of PP was sorted manually at UKBF - The Plastics
Recycling Centre, into two prior food-use packaging fractions, clear and coloured, and also into a non-food grade packaging fraction.

Using conservative manual sorting it was found that there was a 40:60 split between food and non-food PP and approximately a 50:50 split between clear and coloured food use materials. The prior food use purity at this stage was approximately 95%. This meant that a further sort was needed prior to granulation to achieve the required minimum of 99% purity.

The two discrete streams, clear and coloured PP, were processed in a food grade HDPE recycling process that included a further (negative) sort at reduced throughput rates, grinding into flakes, washing in a hot caustic solution, processing through a sink-float separator, drying, and colour sorting for the clear fraction. The flake was bagged and transported to Gneuss in Germany for the first stage of decontamination.

Audits on composition established that the prior food use purity for clear PP was 98.1% and the coloured food PP was 99.3%. The coloured packaging material was easier to hand-sort since most of the coloured items were recognisable due to decoration with labels and printing and conversely it was harder to recognise all of the items in the clear stream. Some of the errors could well have been made from materials on the positive list such as pet food containers, mouth wash, tooth-paste, hand-wash, etc. However, in order to err on the safe side they were listed as non-food use. The clear fraction included approximately 7% of white flakes derived from caps and sorting errors since many of the natural (non-pigmented) PP containers do have a white appearance. White (43%) and yellow (16%) flakes dominated the coloured fraction, with smaller amounts of other colours.
Decontamination of PP:

The decontamination of the clear and coloured PP was conducted in two stages using the Gneuss MRS extruder at 250°C with 100 micron melt filtration followed by rotary vacuum reactor processing the PP pellets at 140°C for 6 hours at PET Processors in Dumfries, Scotland who had suitably sized reactors. These two processes were conducted without difficulty.

Physical Properties of rPP resins:

The resulting rPP resins were expected to behave more like PP copolymer due to the wide range of grades in the mixtures, such as homopolymer, random copolymer and block polymer PP as well as a low level (<1.5%) of high-density polyethylene (HDPE).

Physical tests conducted on test bars made from 100% rPP showed that the clear and the coloured rPP resins both had properties comparable to virgin PP copolymer resins. This allowed them to be used in blends for the manufacture of PP products without a major effect on the finished packaging properties except for applications that demanded a high level of clarity. The flow behaviour of the two resins differed from each other but both were blended with virgin and underwent successful manufacturing trials with three high-speed processes at 25% and 50% levels with very minor adjustment in settings. The properties of the two rPP resins and a virgin copolymer are shown in Table 3:
Table 3:

<table>
<thead>
<tr>
<th>Property</th>
<th>Method</th>
<th>Clear rPP</th>
<th>Coloured rPP</th>
<th>LyondellBasell Virgin Copolymer Moplen EP548P</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melt Flow Rate (230°C/2.16kg)</td>
<td>ISO/DIS 1133-1:2009</td>
<td>7.8</td>
<td>18.5</td>
<td>16</td>
<td>g/10min</td>
</tr>
<tr>
<td>Tensile Strength</td>
<td>BS EN ISO 527-2/1A/1/5</td>
<td>24.91</td>
<td>23.21</td>
<td>28</td>
<td>MPa</td>
</tr>
<tr>
<td>Young's Modulus</td>
<td>BS EN ISO 527-2/1A/1/5</td>
<td>1.52</td>
<td>1.73</td>
<td>1.55</td>
<td>GPa</td>
</tr>
<tr>
<td>Unnotched Charpy Impact Energy</td>
<td>Part 1: (ISO 179:2010)</td>
<td>197.01</td>
<td>175.8</td>
<td>No Break</td>
<td>KJ/m²</td>
</tr>
<tr>
<td>Melting Point by Differential Scanning Calorimetry (DSC)</td>
<td>Mettler Toledo DSC Second cycle melting peak, 12.5 °C/min</td>
<td>163.4</td>
<td>166.8</td>
<td>Not available.</td>
<td>°C</td>
</tr>
</tbody>
</table>

**Manufacturing Trials using rPP:**

Manufacturing trials were completed at three different locations using different techniques to assess how well post-consumer recovered polypropylene would perform on large scale processing equipment. In this project clear and coloured rPP had been segregated at the manual sorting stage so that they could be treated separately as it was considered that there would be significant demand for the clear PP and more limited markets for the coloured PP.

1100 ml clear pots were injection moulded with 25% and 50% of the clear recycled PP with very minor changes to standard settings in moulding parameters (change in cycle time of 0.2 seconds). The 25% and 50% clear rPP showed a slight tint and increased haze. All dimensions and physical properties including impact were within specification.
A range of snack noodle pot samples with standard levels of 1% white masterbatch were also moulded. The pots were moulded with 25% and 50% of both the clear and coloured rPP with very minor changes in moulding parameters. All dimensions and physical properties including impact were satisfactory. The clear grades were deemed to be suitable for white applications with minor adjustments to masterbatch level.

rPP resins were also processed into food trays using a thermoforming process. Adjustments to operating conditions were successfully made to adjust for the higher melt flow rate of the rPP resins compared to the virgin resins used for this process. The trays all moulded well. The 50% coloured rPP was further coloured with brown masterbatch to match an existing product without any special adjustment.

The three processing trials all ran successfully and encountered no significant problems in adapting the setting to make products within acceptable technical specifications.

Food Contact Compliance:

The compliance of rPP with EU food contact requirements is predicated on the achievement of a sorted post-consumer stream of prior food use PP packaging that is later processed according to the present invention.

The materials found within the processed rPP consisted of substances that are typically compounded into virgin PP resin. The three substances that required further examination by conducting specific migration tests were di-ethyl hexyl phthalate (DEHP), Tinuvin 326 (a UV absorber/antioxidant) and 3,4 di-methyl benzaldehyde (residue of clarifier). The conditions for the migration were 30 minutes at 121°C.
followed by 10 days at 50°C. The tests were conducted on 100% rPP moulded tensile bars. The first two substances easily met the specific migration limits for all food types at 30 minutes at 121°C. The last substance is a degradation by-product of the clarifier molecule, 1,3:2,4-bis (3,4-dimethylbenzyldiene) sorbitol that is on the EU approved list for food contact materials and is widely used in clear PP tubs, trays and cups. While there is EU approval for this clarifier at all levels and the by-product at low concentrations, there may be a need to re-validate the food contact approval at higher levels of the 3,4 di-methyl benzaldehyde seen in the specific migration studies.

The clear and coloured rPP resins have shown that they comply with EU Regulations 1935 and 10/2011 under a range of conditions which are defined in the Table 4 below. In the case of fatty foods, the compliance with the migration limits needs to refer to the migration correction factors when using vegetable oil as the food simulant. The clear rPP showed lower migration levels than the coloured rPP in all cases.

Overall the rPP resins show compliance for:

- Absence of odour and Taint (tested at 50% rPP level);
- Overall migration for 100% clear and coloured rPP at 1 hour at 121°C or 2 hours at 100°C in food contact applications with all aqueous (water based) foods and all acidic foods;
- Overall migration for 100% clear and coloured rPP levels when used under the relatively severe conditions of simulated use of 1 hour at 121°C or 2 hours at 100°C in food contact applications with many fatty and oily foods (shown in the table below for migration correction factors of 3, 4 and 5).
• Further overall migration compliance conditions with fatty and oily foods exist for slightly less aggressive heating conditions as specified in Table 4 below.

### Food contact applications for rPP:

In order to better define the food types that can be used with clear and coloured rPP at the highest heating and food contact applications as described above, Table 4 below presents the fatty foods with the correction factors ranging from 5 to 2 (summarised from Annex 111, Table 2 of Regulation 10/2011).

Table 4 - Food contact applications for rPP with fatty and oily foods

<table>
<thead>
<tr>
<th>Migration correction factor</th>
<th>Description of fatty food types</th>
<th>Migration conditions Clear rPP</th>
<th>Migration conditions Coloured rPP</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>Fried potatoes; Powdered or dried soups and sauces with fatty character; sandwiches, toasted bread, pizza with a fatty surface; dried foods with a fatty surface</td>
<td>1 hr @121°C Olive oil</td>
<td>1 hr @121°C Olive oil</td>
</tr>
<tr>
<td>4</td>
<td>Meat of all types, fresh, chilled, salted, smoked; processed meat products such as ham, salami, bacon, sausages; fried or roasted meat;</td>
<td>1 hr @121°C Olive oil</td>
<td>1 hr @121°C Olive oil</td>
</tr>
<tr>
<td>3</td>
<td>Dry and fresh Pastry, biscuits, cake, bread with fatty surface; Chocolate products; Solid confectionary products with a fatty surface; Fish, fresh, chilled processed, salted or smoked; Preserved meat in fatty medium; Cheeses without rind or with edible rind; soups and sauces with fatty character; Mustard; Cocoa paste;</td>
<td>1 hr @121°C Olive oil</td>
<td>1 hr @121°C Olive oil</td>
</tr>
<tr>
<td>2</td>
<td>Paste confectionary products with a fatty surface; Margarine, butter and oils made into water emulsion;</td>
<td>2 hrs @100°C Olive oil</td>
<td>10 days @40°C Olive oil</td>
</tr>
<tr>
<td>1</td>
<td>Fruit preserved in an oily medium; Nuts</td>
<td>10 days</td>
<td>Not</td>
</tr>
</tbody>
</table>
In summary, the results of the material analysis and migration tests show that the 100% clear and coloured rPP is suited for a very wide range of food contact applications for aqueous and acidic foods under the most severe heating conditions and prolonged storage at room, chilled and frozen temperatures.

The clear and coloured rPP are suited to a wide range of fatty foods where the migration correction factors are 3 to 5. Both clear and coloured rPP can be used for margarine and butter as they both comply with the 10 days at 40°C condition. The clear PP can be used in applications involving even higher temperatures (2 hours at 100°C).

The 100% clear rPP is suited for use with 100% oils and fats as well as all of the other foods in Table 4 at the 10 days at 40°C condition. The coloured rPP cannot be used in applications involving 100% fats and oils at ambient conditions. It may also be able to be used at frozen conditions.

Conclusions:

The tests investigated the suitability of the decontamination process of the present invention to decontaminate real post-consumer PP packaging from UK households to levels suitable for use of the material again in food packaging.

The following conclusions can be drawn from the observations and tests described above:
• It was found that 99% prior food use purity could be achieved if there was positive sorting to greater than 95% purity followed by negative sorting;

• The sorting of the PP packaging into clear and coloured fractions had a profound effect on the Melt Flow Rate (MFR) properties with the clear rPP showing a value in the region of 8 and the coloured rPP in the region of 18. These values are made up of many components that would have MFR's ranging from 3 to 40. However, the finished products did not show variability in processing due to the blending that occurred at the rotary vacuum decontamination step. The colour-sorting step could be one way of tailoring the flow properties of the finished rPP products;

• The use of the widely ranging MFR values did not prevent the two rPP resins from being used in widely varying processes such as in-line extrusion/thermoforming and injection moulding when used in blends up to 50% with only very minor adjustments in processing conditions demonstrating that this recycling process has the potential to produce high quality resins that can be reliably used in high speed production equipment;

• The physical properties of the clear and coloured rPP were typical of virgin PP copolymer resins that are used in the manufacture of packaging. This means that there was no deterioration of properties such as stiffness, strength, impact toughness and temperature resistance when these resins were used in blends with virgin at levels of 50% or more;

• The separation of the rPP into clear and coloured fractions was very successful in producing a "clear" rPP that could be readily used in white PP products at
levels of at least 25%. It could also be used in clear products with a reduction of clarity due to the presence of copolymer and some white PP;

- The coloured rPP fraction could be readily re-coloured into darker colours (brown, blue, green) by the use of normal levels of pigment masterbatch making this fraction suited for many applications;

- The clear and coloured PP resins that were sorted and processed within the trial were able to demonstrate that they imparted no taint or odour to food when used at 100% levels. This signifies that the combined effect of the sorting of the PP and the decontamination process steps were very effective in handling these important aspects of converting real post-consumer waste PP back into food contact applications;

- The analysis of the rPP resins showed that the only substances present besides oligomers of PP were based on the additives originally put into PP packaging resins such as lubricants, antioxidants, clarifiers and into labels such as plasticisers (found in adhesives or inks). The analysis of the specific migration of these substances under aggressive heating conditions (30 minutes at 121°C followed by 10 days at 50°C) with water based, acidic and fatty foods showed that there were no migration issues except for the decomposition fragments of the clarifier molecule (3,4 di-methyl benzaldehyde) used in clear packaging;

- The compliance of the clear and coloured rPP with the requirements to meet the limits for overall migration into food simulants expressed in EU regulation 10/2011 was very impressive. The initial tests were conducted on 100% rPP specimens for 1 hour at 121°C, which covers all heating (including microwave
heating) and storage conditions at ambient chilled and frozen conditions, showed that for all aqueous and acidic foods the 100% coloured and clear rPP samples met the migration limits;

- In the case of the overall migration performance of the clear and coloured rPP in contact with fatty foods under the same aggressive heating conditions of 1 hour at 121°C, both resins showed different behaviour with the clear resin showing lower levels of migration which means that the food types and heating conditions that could be used were different to those for the coloured rPP. This was attributed to the lower molecular weight of the coloured rPP as identified by the higher MFR;

- At the heating conditions of 1 hour at 121°C, the clear and coloured rPP could both be used at 100% levels for a wide range of specific foods such as: fried potatoes; powdered or dried soups and sauces with fatty character; sandwiches, toasted bread, pizza with a fatty surface; dried foods with a fatty surface, meat of all types, fresh, chilled, salted, smoked; processed meat products such as ham, salami, bacon, sausages; fried or roasted meat; dry and fresh pastry, biscuits, cake, bread with fatty surface; chocolate products; solid confectionary products with a fatty surface; fish, fresh, chilled processed, salted or smoked; preserved meat in fatty medium; cheeses without rind or with edible rind; soups and sauces with fatty character; mustard; cocoa paste;

- At the migration heating conditions of 10 days at 40°C (which also allows heating to 70°C for 2 hours or heating up to 100°C for 15 minutes) both clear and coloured rPP could be used at 100% levels for margarine and butter which
would represent a large potential market for the rPP either as a blend or a middle layer;

- Clear rPP could be used at 100% levels under the migration heating condition of 10 days at 40°C for more aggressive fatty and oily materials such as animal fats and oils, mayonnaise, and preserved foods in oil due to the lower migration rate it displays compared to the coloured material which could not be used for these food stuffs due to too high migration rates;

- It is apparent from the specific nature of the recommendations in contact with oily foods that while a very wide range of applications are open to both clear and coloured rPP, the precise migration behaviour will be important in finalising the virgin resin used in the blend and the level of rPP being used as well as the MFR of that batch of rPP.

It is of course to be understood that the present invention is not intended to be restricted to the foregoing examples which are described by way of example only.
Claims

1. A process for recycling polypropylene, the process comprising the steps of:
   i) providing an amount of polypropylene comprising at least about 99% food grade polypropylene;
   ii) extruding the polypropylene, employing a residence time of from about 1 minute to about 3 minutes for the polypropylene in a vacuum zone of the extruder;
   iii) exposing the polypropylene to a temperature of from about 130°C to about 170°C for about 3 to about 8 hours.

2. A process according to claim 1, wherein the step (iii) may be carried out before the extrusion step (ii).

3. A process according to claim 1 or claim 2, wherein the extrusion step further comprises vacuum venting on the extruder.

4. A process according to claim 3, wherein the vacuum in the feed zone is less than about 1000 Pa.

5. A process according to any preceding claim, wherein the polypropylene is exposed to temperatures of above 170°C in step (ii).

6. A process according to claim 5, wherein polypropylene is exposed to temperatures of between about 240°C to about 280°C.
7. A process according to any preceding claim, wherein step (iii) is carried out in a vessel which is either under a pressure of no more than about 600 Pa, or in a vessel which is at atmospheric pressure under a stream of air or inert gas.

8. A process according to claim 7, wherein the stream of air or inert gas passes through the vessel at the rate of 1 cubic foot per minute per kilogram of material.

9. A process according to any preceding claim, wherein, prior to step (ii), the polypropylene is subjected to hot washing in alkaline solution.

10. A process substantially as described herein in the description and drawings.
INTERNATIONAL SEARCH REPORT

A. CLASSIFICATION OF SUBJECT MATTER

INV. B29B7/84 B29B13/00 B29B17/02 B29K105/06 B29C47/76

ADD.

According to International Patent Classification (IPC) and both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

B29B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category* Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No.


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Further documents are listed in the continuation of Box C.

See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"A" document member of the same patent family

Date of the actual completion of the international search

21 June 2012

Date of mailing of the international search report

01/08/2012

Name and mailing address of the ISA

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Authorized officer

Kujat, Christian
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