Summary

Recycled PET in new bottles

The effect on migration, discoloration, and bottle strength

Ulphard Thoden van Velzen, Fresia Alvarado Chacon, and Marieke Brouwer from Wageningen Food & Biobased Research investigated the influence of the origin and concentration of recycled PET (rPET) in PET bottles on migration, discoloration, and strength. The insights from this research can be used by the packaging industry in their sustainability strategy.

Polyethylene terephthalate (PET) is a widely used packaging material for beverage bottles. The main research question was whether, and to what extent, the use of rPET - with different origins and concentrations (compared to virgin) - has an impact on:

- the degree of migration of substances from the bottle into its content;
- the degree of haze and discoloration of the bottle;
- the stress cracking resistance of the bottle.

The results are important for the conditions under and extent to which recycled PET can be used in the production of PET bottles. In addition, the study also provides insight into conditions that will enable us to achieve a more circular economy as well as where the possible limits lie.

A general description of the research

Three types of recycled PET available on the market with different qualities were collected for the research. Using these, hundreds of bottles were created in a systematic way at a small production location. In addition to virgin PET bottles, bottles with different concentrations of rPET were also produced, specifically: 25%, 50%, 75%, and 100% rPET. The researchers studied the migration from these bottles to the water to detect "non-intentionally added substances" (NIAS). Substances that were expected, such as ethylene glycol, acetaldehyde, limonene, and oligomers were found, but also very small concentrations of unexpected substances such as acetone, furan, benzene, and styrene. In addition, haze (a technical term for the optical transparency of the bottle) and the colour of the bottles were researched to identify the degree of greying and discolouration. Thirdly, stress cracking tests were conducted to study the stress cracking resistance of the bottles.

The general conclusion is that the quality of rPET in particular influences the material characteristics. Poor quality has a disproportionately large (negative) influence. There seem to be limitations on the proportions recycled content that can be used in PET bottles with respect to the amount of virgin PET. At relatively high percentages of rPET per bottle, the material characteristics decline.

Statutory framework and primary reason for this research

There are extensive regulations within the European Union on the use of recycled material for the packaging of food items, including beverages. These regulations are aimed to maintain a high level of food safety. The production of soft drink bottles containing rPET must comply with European Food Contact Material legislation (FCM). Moreover, the European Commission will soon (possibly in 2019) issue approval statements for authorised processes of European recycling companies, during which both the recycling process and the bottle feedstock used are examined (description of the collection system wherefrom the bottles originate). Very small quantities of NIAS can migrate from bottles containing rPET, making beverage companies reluctant to use high levels of recycled content in their bottles. This was the prime motivation to start this study and explore the impact of rPET quality and recycled content levels (in a bottle) on factors such as the migration of unwanted substances from the bottles to their content (in this case, bottled water). Given the goal of a circular economy on the one hand and to guarantee food safety on the other, the results of the research can be used to find the optimum level of recycled content for a PET bottle.

This rPET research is part of the scientific research programme of the Netherlands Institute for Sustainable Packaging and the Top Institute Food and Nutrition (2014-2019). The research was supervised by an industrial advisory board (IAB) with representatives from three soft drink manufacturers and the FWS branch organisation. The IAB provided samples of rPET and supported the researchers in developing the research method and the interpretation of the results.

The small-scale production of PET bottles

Based on the rPET samples submitted by the members of the IAB, three types of rPET are distinguished: one from a recycler that processes deposit bottles (mono-collection) and two from a recycler that recycles sorted and recycled bottles from mixed packaging waste (co-collection). Higher and lower qualities of rPET were specifically requested for this research. Indorama RamaPET N180, provided by Indorama, was used as reference virgin material.

The samples, varying in origin and composition, and the virgin material were analysed by WFBR and Fraunhofer according to industry-standard analysis methods. The test bottles themselves were created as accurately as possible (in accordance with industry practice), with five types of bottles produced per type of rPET material, varying in the rPET percentage of 0:100%, 25:75%, 50:50%, 75:25% to 100:0%.

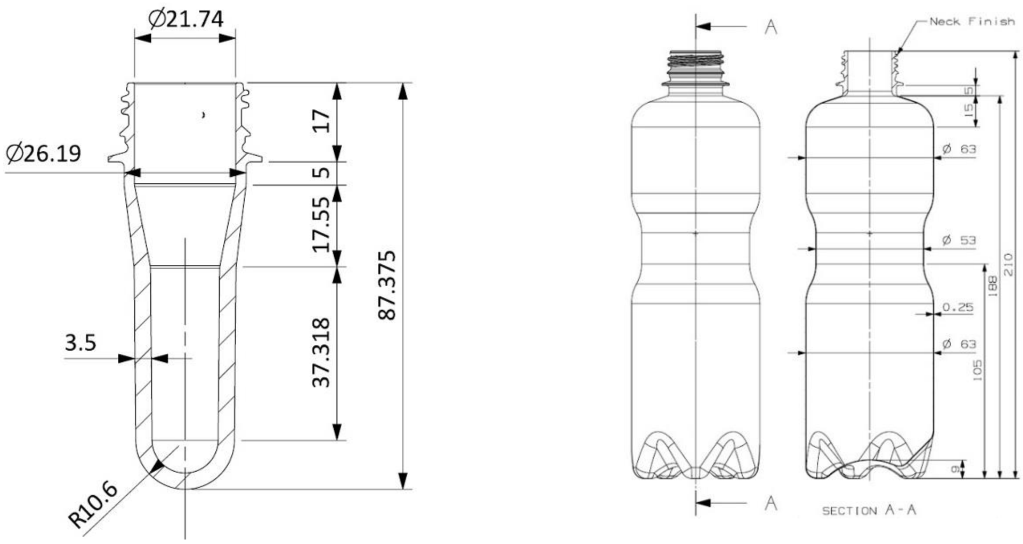

Figure 1. The technical design of the bottle

The migration of unwanted substances was measured under lab conditions at one of the soft drink manufacturers, using standard measurement methods. The rPET material itself was also examined for volatile substances and oligomers. The expected migration model was calculated using this data. These model-based calculations appear to correspond well with the results found in the experiment.

Results of migration analyses: a few topics

The elemental analysis (i.e. the analysis of chemical elements from the periodic table) shows that more cobalt is found in virgin material compared to rPET granulate; this is attributed to the PET production method. Chlorine, hardly present in PET, is found in rPET granules and especially in rPET from a co-collection system. The probable cause of this is the contamination of the collected PET material by PVC or PVdC.

With regard to volatile molecular contaminants, four substances were examined: acetaldehyde, 2-methyl-1,3-dioxolane, benzene, and ethylene glycol. The different types of recycled and virgin PET pellets show slight differences in the presence of these volatile contaminants. During the injection moulding of the preforms and the blowing of the bottles, more volatile contaminants are formed. There are no discernible differences in acetaldehyde, 2-methyl-1,3-dioxolane, and ethylene glycol in the different bottles (% and type of rPET). Benzene shows a different pattern; there are clear differences between the bottles of different types of rPET and different compositions (% rPET). rPET from mono-collection in a relatively low proportion (25%) scores comparably to virgin. Especially rPET from a co-collection system, but also higher percentages of rPET in the bottle give less favourable results.

Migration to the contents (water) has been tested after the bottles have been kept in a test room for 10 days at 40 °C. Apart from 2-methyl -1,3-dioxolane, benzene, and styrene, there were no striking results observed between qualities and compositions. Remarkably, 2-methyl-1,3-dioxolane is present in higher concentrations in water in virgin bottles than in water in bottles containing recycled content. This is attributed to differences in production methods and in particular, to the solid state post-condensation process during recycling. Benzene and styrene are perceived in very low concentrations in mono-collection rPET and increase as the recycled content increases. The concentration is higher in rPET from a co-collection system. Contamination from polystyrene is probably the cause of the presence of styrene in the rPET from a co-collection system.

Results from discoloration, greying analysis and stress cracking

PET bottles with recycled content contain more contaminating particles. This amount increases with a higher percentage of rPET; especially when mixing virgin PET with rPET originating from a co-collection system. This contamination can arise through various causes, such as sand and other contaminants that are scratched into the rPET material during mechanical treatment (baling), by recycling other materials with the PET bottles from incorrectly sorted objects or packaging components, such as paper, PS, PE, PP, etc., or by additives in the rPET material. This contamination has effects on haze and discoloration

Haze

The haziness of the bottles strongly correlates with particle contamination; greater contamination leads to less transparent bottles. This is clearly visible in the various percentages of rPET used in the bottles and is independent of the origin of the rPET.

Discoloration (greying or yellowing)

Some discoloration aspects correlate strongly with the amount of recycled content and the origin of the rPET. More rPET means a greyer bottle (L* value) and more yellowing (b* value). Greying is not affected by the origin of the rPET. For yellowing (b* value), however, both the origin and the recycled content appear to affect it. At the same time, yellowing is not only caused by particle contamination, but can also be corrected by producers by adding blue pigment.

Stress cracking

The analysis shows that bottles with a higher intrinsic viscosity (IV) are less likely to leak or crack during the stress cracking test. However, no clear relationship was found between the IV values and the amount or origin of rPET. All granules meet IV standards set by the market. For blown bottles whose IV was higher than the other bottles, the risk of stress cracking decreased.

Small-scale production conditions versus practice

The PET bottles had to be produced at a small-scale production facility, which in itself can result in over-estimations of the studied effects in comparison to the current industrial practice, because the residence times in the production equipment is longer than what is common in the industrial reality. Therefore, additional tests were conducted using commercially produced bottles, to determine the extent to which the lab analysis is in agreement with the analysis of commercially produced bottles. This additional analysis shows that the findings overlap. As a result, the migration analysis of bottles produced on a small scale provides a good approximation of the material produced on a commercial scale (granulate and bottles).

Business relevance of the research

The presence of benzene in rPET is probably caused by contamination of the PET stream by PVC or PVdC, for example through the use of PVC labels or PVC non-packaging in the PET sorted product. PVC can contribute to the formation of benzene both directly and indirectly through the catalyst effect on PET. The research discovered a proportional relationship between the migrated amount of benzene from a PET bottle and the chlorine content of the rPET granulate. The quantities of benzene that migrate are very small. From a toxicological perspective, such exposure has no priority for public health.

The question whether or not the exposure to benzene complies with the FCM legislation is more difficult to answer. This requires a risk assessment. Such a risk assessment can be conducted with multiple consumption patterns and body weights and hence not yielding one single result. But one can compare the exposure to the WHO guidance values for drinking water. Then it follows that the highest measured concentration of benzene (migrated from a bottle entirely made with rPET from a co-collection system) is less than half the amount of the permitted concentration of benzene in drinking water.

In any case it is wise to act with care when it comes to using rPET and to follow the ALARA (As Low As Reasonably Achievable) principle as much as possible; which means aiming for the lowest possible exposure. With controlled (mono-)collection streams, a relatively high percentage of rPET seems justified, depending on the acceptance limits of a manufacturer. In the case of co-collection (and therefore stronger accumulation of contaminants in the rPET), it is advisable to use lower percentages.

The ALARA principle also applies because there is still insufficient insight into the accumulation of contaminants in practice, especially when the origin of rPET streams is not entirely certain. Contamination responsible for the formation of benzene and other substances can accumulate during successive recycling cycles. Depolymerisation techniques, where the contaminants are filtered out during the depolymerisation process, could potentially result in clean PET. This has not been investigated in this study.

High-quality rPET can be achieved by only working with a strictly controlled mono-collection system (deposit refund or other closed collection systems), in which circular design and high quality recycling standards are assured. A parallel option is to improve the quality of the rPET from co-collection systems, by producing cleaner sorted products with reduced pollution levels for PVC, PS, and other polymer and particle contamination as much as possible.
Moreover, unwanted substances also occur in virgin PET bottles and high-quality rPET bottles, so that continuous attention is needed for monitoring the quality of the PET and the packaged contents.

At the same time, the issue deserves a nuanced perspective. The low exposure to benzene as a result of the use of high-quality rPET in the production of PET bottles is not a priority from a public health perspective. However, on the basis of this exploratory research and the political and social desire to move to a circular design, it is worth considering a more thorough risk assessment when it comes to a more intensive use of rPET in food packaging.

Academic publications (titles in accordance with publications)

Alvarado Chacon, F. (WFBR), Brouwer, M.T., Thoden van Velzen, E.U. (TiFN & WFBR), 2019. Effect of recycled content and rPET quality on the properties of PET bottles, part I: optical and mechanical properties.

Thoden van Velzen, E.U., Brouwer, M.T. (TiFN & WFBR); Stärker, C. en Welle, F. (Fraunhofer Institut für Verfahrenstechnik und Verpackung IVV) 2018. Effect of recycled content and rPET quality on the properties of PET bottles, part II: Migration.

Brouwer M.T., Thoden van Velzen, E.U. (TiFN & WFBR), Alvarado Chacon, F. (WFBR), 2019. Effect of recycled content and rPET quality on the properties of PET bottles, part III: modelling of repetitive recycling.