

PET Depolymerization to BHET and Purification via Crystallization

Report Monitoring Contamination Levels

Report required by Article 13 of Regulation (EU) 2022/1616

10 October 2024

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1. Introduction

The novel technology -‘Polyethylene terephthalate (PET) depolymerization to bis(2-hydroxyethyl)terephthalate (BHET) followed by a series of purification steps of which the main step is crystallization’ was notified as required under Articles 10(2) and 10(3) of Commission Regulation (EU) 2022/1616 on 7 April 2023.

According to Article 13 of Commission Regulation (EU) 2022/1616 a recycler operating a decontamination installation in accordance with Article 11 of the regulation shall monitor the average contaminant level on the basis of a robust sampling strategy which samples the plastic input batches and the corresponding plastic output batches. On 10 October 2023 and 10 April 2023, a first and second report discussing the monitoring data and the information as required by Article 13(5) have been published.

None of the recyclers operating a decontamination installation applying this Novel Technology produced recycled material that was placed on the market in Europe in the period of 10 April 2024 to 10 October 2024. Therefore, no samples have been submitted for the third monitoring program.

2. Brief description of the novel technology

This novel technology builds on the principle of breaking the polymer matrix to allow more easy access to the contaminants that may be present in the collected PET waste stream and that are usually physically bonded to the polymer and trapped by the matrix. This recycling technology, therefore, counters the difficulties and efficiency limitations often encountered to purify relatively insoluble polymers such as PET when using traditional methods such as extraction and/or partial solubilisation and reprecipitation.

Contrary to mechanical recycling that focuses on the decontamination of the PET polymer, this technology breaks selectively certain chemical bonds of the PET polymer to give a starting molecule - BHET- from which the PET polymer can be remade again. The depolymerization is done through glycolysis by heating the PET waste in a reactor in the presence of an excess of ethylene glycol (EG), a monomer which is also used in the manufacturing of PET (Figure 1).

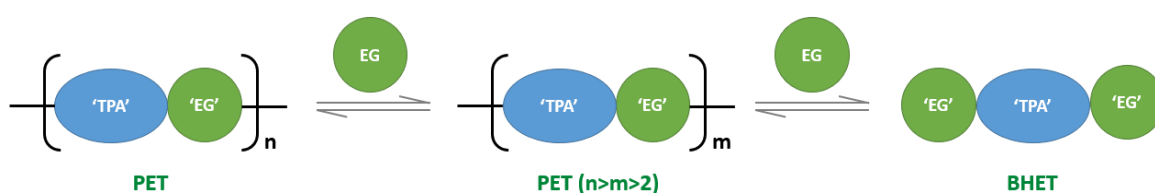


Figure 1. Schematic representation of PET glycolysis; TPA: purified terephthalic acid; EG: ethyleneglycol; BHET: bis(2-hydroxyethyl)terephthalate.

By breaking down the polymer matrix of waste PET, contaminants that may be present in the collected PET waste stream are released into the reaction medium; they are no longer physically bound or difficult to access. This allows impurities to be removed in much higher quantities and more efficiently than with mechanical recycling technologies (Figure 2) by standard physical processes like solid/liquid separation, distillation, active substrate adsorption, crystallisation and washing and drying. Therefore, this novel recycling technology can recycle highly contaminated input materials that cannot be recycled by mechanical recycling processes (Welle, 2021).

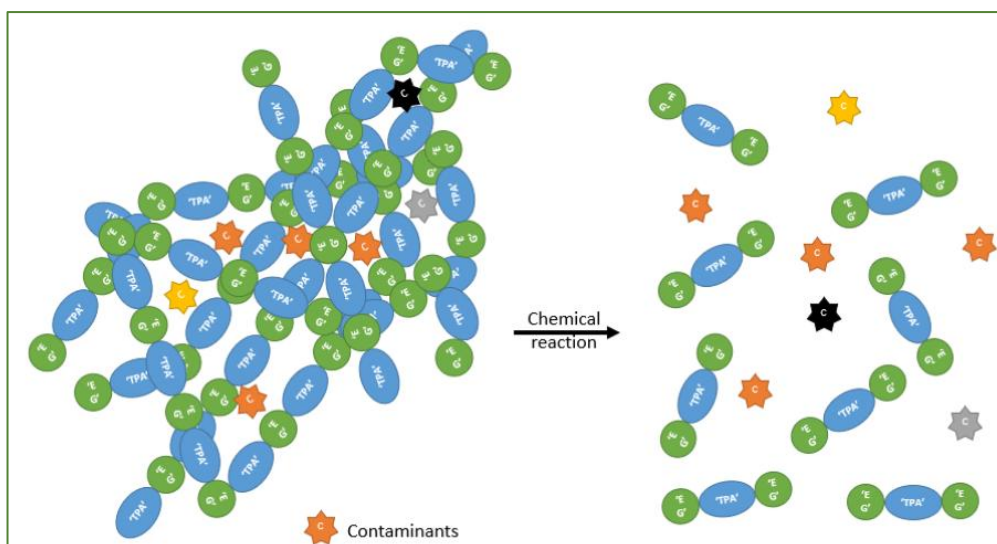


Figure 2. Schematic representation of the release of contaminants during glycolysis.

The novel technology can use 3 different waste streams:

- PET waste originating from packaging: Post-consumer and post-industrial PET packaging waste from food and non-food contact applications.
- Post-consumer and post-industrial polyester textile.
- Post-consumer¹ or post-industrial PET film.

These waste streams can deviate from the requirements for collection and pre-processing of Article 6 of Regulation (EU) 2022/1616 due to the following:

- Compliance with Regulation (EU) 10/2011 cannot always be confirmed.
- The waste not always originates from food contact applications.
- Not all plastic waste might have been subject to separate collection.
- The waste does not always originate from municipal waste or from food retail or other food businesses.

Irrespective of the type, mode of collection and origin of the input material, the output contains minimum 85% of BHET. Together with the identified BHET ‘alike’ co-products like MHET² (CAS# 71949-29-6), BHEI³ (CAS# 3644-99-3), BHEET⁴ (CAS# 65133-69-9), BHET⁵ dimer (CAS# 2144-69-6), EG (CAS# 107-21-1) and diethylene glycol (DEG, CAS# 111-46-6) (Figure 3) that, as indicated by their structure and further confirmed through analysis, will be repolymerized together with the BHET output in the post-processing step, the purity of the obtained output could be considered as 99.9%.

¹ Post-consumer plastic waste as defined in the proposal for a Regulation on packaging and packaging waste, published on 30 November 2022 (European Commission, 2022): ‘post-consumer plastic waste’ means plastic waste that is generated from plastic products that have been placed on the market.

² TPA having reacted with only one molecule of EG

³ isophthalic acid (IPA) having reacted with two molecules of EG

⁴ TPA having reacted with one molecule of EG and one molecule of DEG

⁵ BHET that has reacted with itself

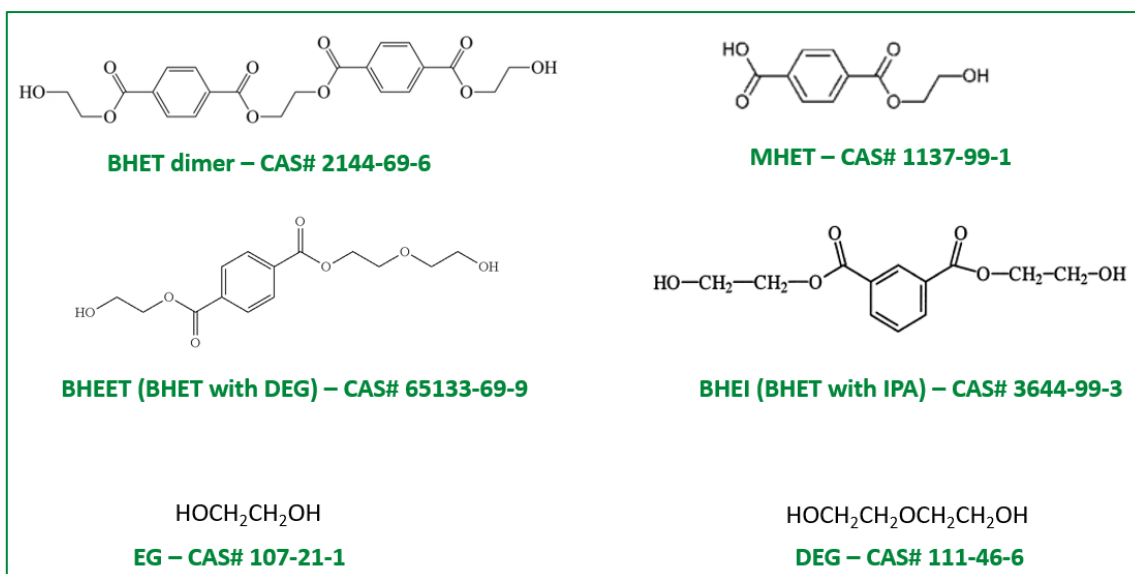


Figure 3. Main co-products formed during the glycolysis depolymerization of PET with ethylene glycol to BHET.

The specifications of the obtained BHET including its impurities are controlled at batch level using High Performance Liquid Chromatography (HPLC)⁶, Gas Chromatography (GC)⁷, Colorimetry⁸, X-Ray Fluorescence Spectroscopy (XRF)⁹ or Inductively Coupled Plasma Mass Spectrometry (ICP-MS)¹⁰ or Atomic Emission Spectroscopy (ICP-AES)¹¹, Karl Fischer Titration¹², optical microscopy and test polymerizations. Other analysis techniques may occasionally be used as well.

The output BHET is used for the manufacture of PET and its common copolyesters. Up to 100% BHET can be used for the production of PET.

The final recycled PET and its common co-polyesters are intended to be used for the manufacturing of materials and articles for contact with similar foods and under similar conditions for which virgin PET is intended:

- All types of foodstuffs;
- Hotfill and/or long term storage at room temperature and below;
- Use for microwaveable applications.

⁶ HPLC: analytical technique for the separation, identification and quantification of non-volatile substances.

⁷ GC: analytical technique for the separation, identification and quantification of volatile and semi-volatile substances.

⁸ Colorimetry: analytical technique for the determination of the concentration of colored compounds in a solution.

⁹ XRF: analytical technique for the qualitative and quantitative determination of the elemental composition of a material.

¹⁰ ICP-MS: analytical technique for the detection and quantification of metals and several non-metals in samples at very low concentrations.

¹¹ ICP-AES: analytical technique for the qualitative and quantitative determination of the elemental composition of a sample.

¹² Karl Fischer titration: classic titration method to determine trace amounts of water in a sample.

3. Summary of the reasoning on the capability of the novel technology and the recycling processes to manufacture recycled plastic materials and articles that comply with Article 3 of Regulation (EC) No 1935/2004 and that are microbiologically safe

The pre-processing, decontamination and post-processing processes of recycling processes applying this novel technology are described in paragraph 2. The novel technology combines the transformation of the polymer into its building block BHET to facilitate the removal of all the impurities by traditional and well recognized purification techniques.

The physical processes that are used (solid/liquid separation; distillation; adsorption on active substrates; crystallization, washing and drying) are common processes used in the chemical industry to purify most of its reagents and products. They rely on robust scientific principles that are compulsory for the obtention of highly purified molecules in the medical devices and pharmaceutical industries. As a result, the BHET obtained by recycling processes that apply this novel technology is of very high purity: BHET + polymerizable co-products >99.9% as confirmed by the analysis conducted on the purified BHET output batches part of this monitoring program.

The decontamination efficiency of a recycling process using this novel technology, assessed by Welle (2021) using a challenge study, was shown to be at least > 99.94% for all surrogate contaminants. Based on this decontamination efficiency, it could be calculated that, for the non-volatile surrogate contaminants, the novel technology is able to reduce contaminant levels of minimum 1300 mg/kg in the input material down to a concentration in the output (C_{mod}) that would not lead to the exceedance of the dietary exposure in infants of 0.0025 µg/kg bw/day¹³ (EFSA, 2011; Appendix III,C). For volatile surrogate contaminants, this calculated maximum contaminant level in the input material is lower mainly because of the limitations of the detection limit of the analytical method and the lower initial contaminant levels in the challenge test. Since the boiling points of these volatile surrogates are below the temperature of the glycolysis process and considering the different decontamination steps that are applied in this technology, it can be assumed that the decontamination efficiency for volatiles is at least as high as for non-volatiles.

Since the submission of the Novel Technology dossier, EFSA published a new scientific guidance. Following this guidance (EFSA, 2024, Appendix D, Table D.1), the concentration of non-volatile substances present in the input material that can be reduced by this novel technology in the worst case scenario A is minimum 1200 mg/kg (based on C_{mod} for methyl salicylate). For volatile surrogate contaminants, this calculated maximum contaminant level in the input material would be 67 mg/kg (based on C_{mod} for toluene) but the real maximum contaminant level is likely to be much higher for the same reasons as indicated above.

Contaminant levels in post-consumer food contact PET waste is conservatively set by EFSA (2011) at 3 mg/kg PET. For non-food contact applications, Franz and Welle (2020) demonstrated that contaminants can be present in concentrations ranging from sub-mg/kg up to around 30 mg/kg on average with some exceptionally high levels of ethanol of up to 1100 mg/kg in some individual samples. These levels are lower than the contaminant levels that this novel technology can handle.

¹³ human exposure threshold value for chemicals with structural alerts raising concern for potential genotoxicity (Kroes *et al.*, 2004),

Even though the waste used by the recyclers originates from the EU market and PET production industry states that PET produced in Europe complies with Regulation (EU) 10/2011, compliance of the waste materials input with Regulation (EU) 10/2011 cannot be entirely demonstrated since a small fraction of the plastic waste originates from non-food contact applications that do not have compulsory and specific compositional regulatory requirements. However, the obtained BHET output is 99.9% pure and is an di-ester of EG and TPA which is naturally formed during the normal production of PET. Both EG and TPA are authorized substances in Regulation (EU) No 10/2011 with identification number FCM227 and FCM785, respectively.

The BHET output produced by this novel technology can manufacture recycled plastic materials and articles that are microbiologically safe. Temperatures used during the glycolysis process and during post-processing –respectively, minimum 195°C and for minimum 1 hour and above 245°C for several hours– are more stringent than the typical conditions for sterilization used in the medical, pharmaceutical and food and beverage industry as reported by Jildeh *et al.* (2021).

Disclaimer

PETCORE Europe is providing this report and related information solely as the entity representing certain business operators (the “**Business Operators**”) in accordance with Articles 10.1, §2 and 13.4-5 of the Commission Regulation (EU) 2022/1616 on recycled plastic materials and articles intended to come into contact with foods (the “**Regulation**”), in the name and on behalf of said Business Operators.

PETCORE Europe has aimed to report, consolidate and accurately provide the necessary data and materials as provided in article 13.5 of the Regulation on behalf of the Business Operators (the “**Information**”). However, PETCORE Europe makes no representations or warranties regarding the completeness, accuracy or validity of such Information and does not warrant the good performance of the Business Operators’ obligations under the Regulation.

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The work and this document have been prepared following the recommendations of the Pierstone Memorandum to PETCORE Europe '**Assessment of the publication of data pursuant to Regulation 2022/1616 under competition law** '.

ABBREVIATIONS

| | |
|---------|---|
| BHEI | bis(2-hydroxyethyl)isophthalate |
| BHEET | 2-hydroxyethyl[2-(2-hydroxyethoxy-)ethyl]terephthalate |
| BHET | bis(2-hydroxyethyl)terephthalate |
| Cmod | Modelled concentration |
| DEG | diethylene glycol |
| Ea | activation energy |
| EG | ethylene glycol |
| GC | gas chromatography |
| HPLC | high performance liquid chromatography |
| ICP-MS | Inductively Coupled Plasma Mass Spectrometry |
| ICP-AES | Inductively Coupled Plasma Atomic Emission Spectroscopy |
| IPA | isophthalic acid |
| MHET | mono(2-hydroxyethyl)terephthalate |
| NIAS | non-intentionally added substances |
| PE | polyethylene |
| PET | polyethylene terephthalate |
| PP | polypropylene |
| PVC | polyvinyl chloride |
| SML | specific migration limit |
| TPA | terephthalic acid |
| XRF | X-ray fluorescence spectroscopy |

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