ECOTRON: New technologies for one-pot glycolysis of printed electronics

Antonio Dobón López[1], Vicente Cloquell[2], Sergio Clemente Castro[1*] and Miriam Lorenzo Navarro[1]

[1] ITENE, Technology Centre. Packaging, transport and logistics. Technological Park, C/ Albert Einstein, 1, Paterna, Valencia (Spain)
[2] School of Aerospace Engineering and Industrial Design. Polytechnic University of Valencia. Camino de Vera, s/n. Valencia (Spain)

[1*]*Corresponding author. Email: sergio.clemente@itene.com*

Abstract: With the aim of promoting sustainable technologies within conventional electronics and mitigating the environmental impact of electronic devices, technologies for chemical recycling of polymeric substrates of these devices have been developed. The initiative stems from the need for a paradigm shift in electronic design and manufacturing processes to reduce the environmental footprint of electronic products.

In order to evaluate recyclability of polymeric substrates two methodologies were tested: alkaline hydrolysis and catalyzed glycolysis. The hydrolysis process has been performed with the aim of recovering Ag-ink while achieving PET substrate recycling towards terephthalic acid (TPA) in the same process step. Analogously, catalyzed glycolysis processes have been investigated with the aim of recovering Ag-ink and simultaneously PET substrate recycling towards bis (2-hidroxyethyl) terephthalate (BHET). Ag-ink can be easily recovered during a first filtration step, while all the other reactants and products remain on the liquid phase if 100% PET substrate conversion is achieved.

In this study we have first analyzed the chemical recycling processes of PET substrates by means of energy and environmentally friendly technologies, with special focus on the recovery of Ag-inks. Specifically, we began by evaluating alkaline hydrolysis using water and NaOH as the reaction medium (with good ink separation), although we had to be applied by more intensive technologies to obtain good conversions of the PET substrate and high-value products for the industry (BHET). Moreover, glycolysis reaction parameters were optimized with commercial catalysts and different inks and polymeric substrates to evaluate and define the best strategies for the glycolysis process. Characterization of all fractions produced was carried out to check the efficiency of the separation process of the metallic inks, the depolymerization of the PET substrate and the purity of the BHET obtained.

Keywords: PET, solvolysis, BHET, depolymerization, printed electronic

1 Introduction

Polyethylene terephthalate (PET) is a thermoplastic polyester renowned for its excellent barrier properties against moisture, high optical clarity, and superior mechanical performance. It is synthesized through a step-growth polycondensation reaction between terephthalic acid (or its dimethyl ester) and ethylene glycol (1). Due to its favourable physicochemical properties, low production cost, and ease of processing, PET has become one of the most widely used polymers in modern societies. However, its high durability, large-scale manufacturing, and inadequate end-of-life waste management have contributed significantly to a growing global environmental challenge (2).

The vast majority of post-consumer PET waste accumulates in landfills or is released into the natural environment. Due to its high chemical stability and resistance to biodegradation, PET undergoes extremely slow decomposition, often persisting for decades. During this prolonged degradation process, it can break down into low-molecular-weight oligomers and microplastics, which pose potential risks to human health through bioaccumulation in the food chain, particularly via the consumption of marine organisms (3). To address this issue and mitigate the greenhouse gas emissions associated with PET production -predominantly derived from fossil resources- the development and implementation of advanced recycling technologies capable of reintegrating PET waste into the production cycle is strongly recommended.

Chemical recycling through solvolysis represents an alternative and complementary pathway to conventional mechanical recycling, offering solutions to several inherent limitations of traditional PET recycling process. This approach enables the depolymerization of PET into its monomeric or oligomeric constituents, thereby facilitating the recovery and reuse of materials from complex waste streams, such as printed electronics that incorporate PET substrates and are often difficult to recycle due to their intricate designs, conductive inks, and electronic components. As a result, solvolysis holds promise for improving the recycling rates of PET-based complex waste that would otherwise be challenging to process using traditional mechanical methods (4,5).

The depolymerization of PET is essentially the reverse of the polycondensation reaction used to synthesize PET from terephthalic acid (TPA) and ethylene glycol (EG), or from bis(hydroxyethyl) terephthalate (BHET). Depending on the extent of the depolymerization, this process yields either the original of the depolymerization, this process yields either the original monomers or oligomers as products, with complete depolymerization resulting in the recovery of monomers, and partial depolymerization yielding oligomers (6). After purification, these monomers can be reintroduced into the production cycle without compromising the final material´s quality, similar to virgin PET. This is possible because the process involves a chemical attack, where specific polymer bonds are substituted in the presence of a solvent (7).

The nucleophilic reagent (solvent) targets the ester bond (R-CO-OR´), leading to the cleavage of the polymer chain and the formation of monomers and/or oligomers that constitute the polymer (8). Solvolysis depolymerization process can be categorized based on the solvent used, including hydrolysis, alcoholysis, aminolysis, and glycolysis (9).

Hydrolysis can be performed under various conditions, with alkaline and acidic hydrolysis being the most extensively studied methods. In these processes, a water molecule attacks the ester bond in the PET chain, resulting in the cleavage of the chain and the production of TPA and EG (10). The main drawbacks of these methods are the need for highly corrosive chemicals and the requirement for extreme operating conditions.

Alcoholysis uses short-chain alcohols like methanol to produce dimethyl terephthalate (DMT) and EG, which can be reused to obtain recycled PET (r-PET). The main drawbacks are the use of flammable and toxic substances under harsh conditions, as well as the need for two reaction steps to produce PET from DMT and EG (11).

Aminolysis uses various amines and nitrogen compounds (such as ethanolamine or hydrazine) to chemically recycle PET. While this process can produce valuable nitrogen compounds, it requires the use of carcinogenic substances and severe operating conditions (12).

Glycolysis processes are the most extensively studied, as they use glycols (e.g., EG) to depolymerize PET waste. The resulting products are monomers, dimers, trimers, and other oligomers of BHET, depending on whether the depolymerization process is complete or partial (13).

The main advantage of glycolysis is that it operates under moderate conditions (atmospheric pressure and temperatures around 200 °C), significantly lower than those required by other thermochemical recycling methods like gasification or pyrolysis, which operate at temperatures between 450-600 °C. Additionally, the key benefit is the ability to achieve complete depolymerization to obtain the BHET monomer, which can be directly used to produce r-PET, eliminating the need for an extra reaction step (14).

It is important to note that, due to the nature of the solvolysis process, which yields the original monomers or oligomers, and the subsequent purification steps, the resulting building blocks have a low level of impurities. These monomers/oligomers are chemically identical to those derived from petroleum sources.

For these reasons, this study focuses on the development of evaluation of glycolysis processes for the chemical depolymerization of PET substrates in printed electronics.

2 Experimental

2.1 Description of materials

Firstly, an initial identification and classification of the printed electronic containing PET substrate covered by the project was carried out.

Work was performed using various types of printed electronics, primarily ITENE samples composed of a PET substrate, a dielectric layer, and silver-based conducive inks (Figure 1). Some other configurations provided by collaborators are made of: (1) PET substrate: two metal layers and one dielectric on one side (Figure 2), and (2) PET substrate: Ag-based ink sample / White cover layer / transparent PET (Figure 3).

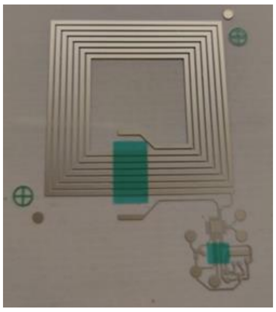

Figure 1. ITENE Printed electronic with PET substrate (base material), dielectric (green) and silver ink (grey)

On the one hand, the printed electronics manufactured by ITENE were characterized using FTIR, ICP-MS, and DSC analyses. In addition, the most representative samples, along with those specifically manufactured by ITENE for these studies, were characterized by DSC and FTIR.

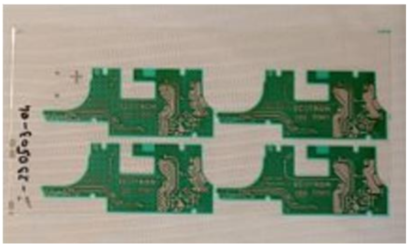

Figure 2. Printed electronic with PET substrate: two metal layers and one dielectric on one side

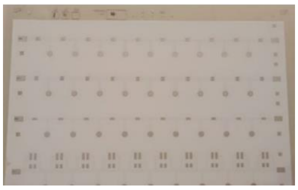

Figure 3. Printed electronic with PET substrate: Ag-based ink sample / White cover layer / transparent PET

Preliminary conditioning and pretreatment studies were carried out on the printed electronic samples received to determine the optimal processing conditions for obtaining a sufficiently reduced, compact material with an adequate PET content to enable effective depolymerization. The process was primarily relied on a shredding unit, whose operating conditions were previously analysed.

2.2 Characterization methods

FTIR analysis was employed to qualitatively assess the molecular structure of the printed electronic substrates materials. The technique requires only a few milligrams of sample and operates in the infrared range of 4000 cm^{-1} to 400 cm^{-1} under nitrogen atmosphere. The structural composition of the analysed material is represented by distinct peaks corresponding to its functional groups and bonding patterns. Spectral interpretation is carried out by comparison with reference spectra from established databases.

Figure 4. Fourier transform infrared spectroscopy equipment.

Differential scanning calorimetry (DSC) is used to determine the temperature at which the polymer chains in amorphous or semi-crystalline materials begin to exhibit segmental mobility – referred to as the glass transition temperature – resulting in an increase in free volume within the polymer matrix. The analysis is conducted under nitrogen atmosphere, following a predefined thermal program. This technique also enables the identification of the melting points of BHET monomers, dimers, and oligomers formed during the PET depolymerization process.

The quantification of metallic elements such a silver (Ag), sodium (Na), potassium (K), and zinc (Zn) was carried out using Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Approximately 0.1 g of the sample was accurately weighed and subjected to acid digestion using a mixture of ultrapure nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) under controlled temperature and pressure in a microwave digestion system. The resulting solution was diluted with deionized water to a final volume of 50 Ml. Calibration curves were prepared using certified multi-element standard solutions. The samples were introduced into the ICP-MS system, and detection was performed under optimized operating conditions. Quantification was achieved by comparing the measured intensities with the calibration curves, ensuring detection limits in the low ppb range.

To assess the purity of TPA and BHET obtained following the depolymerization process, ultra-high-performance liquid chromatography (U-HPLC) was performed using a Waters UPLC-Qtof quadrupole time-of-flight system (EQ182/ITN) equipped with a C19 column (Figure 5). Initially, a commercial BHET standard (98% purity) was used for calibration. Approximately 10 mg of the standard were weighed and dissolved in methanol to prepare a stock solution, from which calibrations standards at concentrations of 50, 100, 200, 400, 600, 800, and 1000 µg/L were prepared.

Figure 5. Water UPLC-Qtof quadrupole time-of-flight system (EQ182/ITN) equipped with a C19 column.

For the analysis of TPA samples, 10–15 mg of each sample were weighed and dissolved in 10 mL of a 1:1 (v/v) methanol–water mixture to enhance solubility. The solution was subjected to ultrasound for 15–20 minutes, with gentle heating if necessary, to ensure complete dissolution, and subsequently diluted (1:10 to 1:100) with the same solvent mixture. Chromatographic separation was performed using a reverse-phase C18 column with a mobile phase composed of water and methanol containing 0.1% formic acid, at a flow rate of 0.3 mL/min.

For the analysis of BHET samples obtained post-depolymerization, 10-15 mg of each sample were weighed and dissolved in 10 mL methanol. The solution was subjected to ultrasound for 15 minutes to ensure complete dissolution, and subsequently diluted (1:10 to 1:100) with methanol. Chromatographic separation was carried out using a mobile phase composed of water and methanol containing 0.1% formic acid, at a flow rate of 0.3 mL/min.

After the run time, the exact mass of the analyte was extracted by processing the signal obtained from the detector. The signals were compared against the previously established calibration curve, enabling quantification of TPA and BHET concentration in the solution. From this data, the percentage purity of the samples was determined.

2.3 Experimental methodology

Initial experiments involving printed electronics materials were conducted using ITENE-prepared samples and hydrolysis-based depolymerization. Alkaline hydrolysis was selected as the first route to recover terephthalic acid (TPA) and ethylene glycol (EG), effectively reversing the conventional PET synthesis process. During the reaction, silver oxide precipitates as a non-photosensitive black solid. In cases where precipitation does not occur, residual silver is neutralized with sulfuric acid, forming silver sulphate –an off-white compound with low solubility and minimal likelihood of remaining in the recovered TPA.

Following the alkaline hydrolysis reaction, the reaction mixture undergoes an initial filtration step to separate unreacted Ag/PET, silver oxides and hydroxides, along with other insoluble components. The resulting filtrate is then neutralized with sulfuric acid to facilitate the recovery of high purity terephthalic acid (TPA). The neutralized solution is subsequently cooled at 4 °C for 24 hours to promote TPA crystallization. A second filtration step is then performed to isolate the solid TPA from the remaining solution. Figure 6 illustrates the material flow and unit process operations involved in the hydrolysis stage.

Hydrolysis experiments were conducted using of ITENE PET printed electronic samples containing Ag-ink, dispersed in 40 mL of reaction solution. The study focused on evaluating the influence of key process variables, including the ethanol-to-water ratio in the reaction medium, sodium hydroxide concentration, reaction temperature, and reaction time.

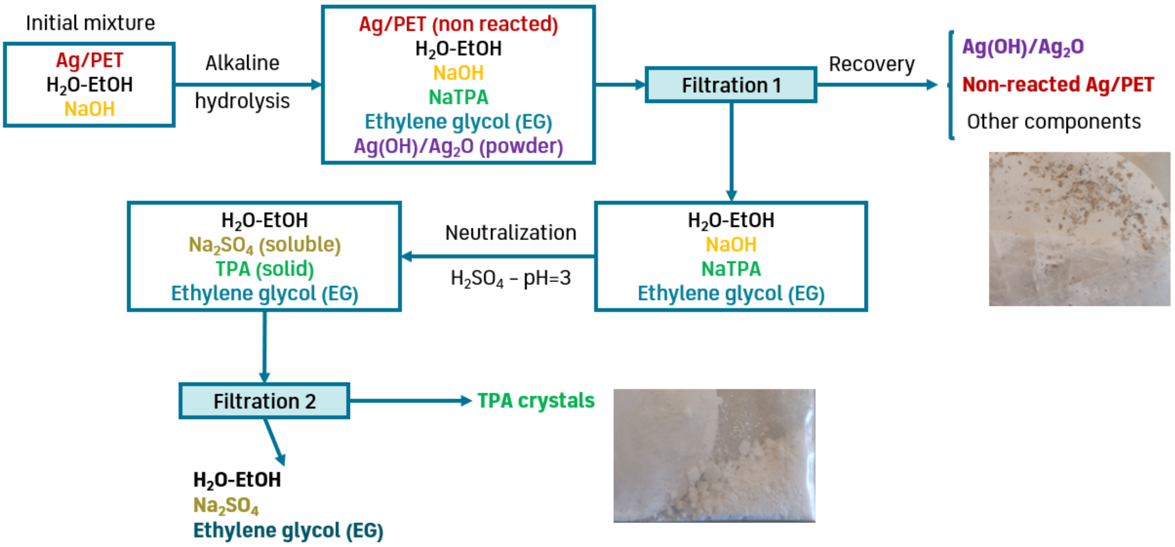

Figure 6. One-pot Hydrolysis process of PET-based supports.

Concerning glycolysis experiments using printed electronics materials, these processes were performed in parallel with hydrolysis, employing ITENE-prepared samples and Na_2CO_3 as catalyst initially, although it was studied either zin acetate, potassium hydroxide, sodium hydroxide and potassium carbonate. This depolymerization pathway leads to the formation of bis(2-hydroxyethyl) terephthalate (BHET) as the primary product. During the process, silver remains unaltered in its metallic form, facilitating its subsequent recovery.

Following the glycolysis reaction, the reaction mixture undergoes an initial filtration step to separate unreacted Ag/PET and other insoluble residues such as dielectric, including metallic silver. The resulting liquid phase, containing bis(2-hydroxyethyl) terephthalate (BHET), is then cooled to 4 °C for 24 hours to promote BHET crystallization. A second filtration step is subsequently performed to isolate the solid BHET from the remaining solution. Figure 7 shows the process flow diagram of glycolysis using printed electronics with Na_2CO_3 as a catalyst.

The process parameters evaluated included reaction temperature, reaction time, type of homogeneous catalyst, catalyst-to-PET ratio, and ethylene glycol-to-PET (EG/PET) solvent ratio.

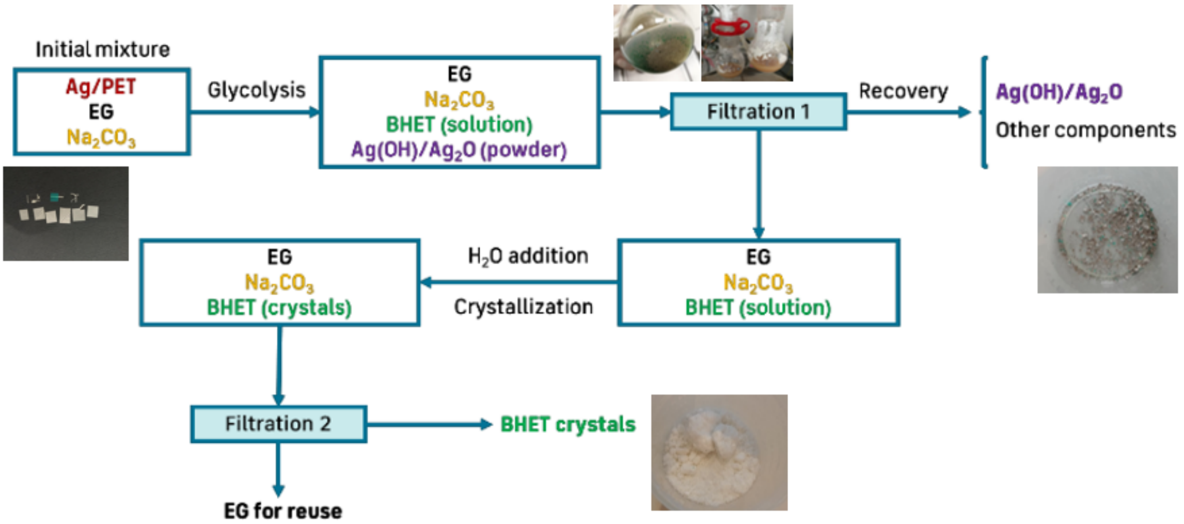

Figure 7. One-Pot Glycolysis of PET-based supports using Na2CO3 as catalyst.

In these studies, three key performance indicators were evaluated: (X) the depolymerization conversion rate, defined as the ratio of reacted PET to the initial PET input; (Y) the solid yield, assuming complete conversion of the product to TPA or BHET; and (Z) the TPA or BHET yield, adjusted based on the purity of the recovered TPA or BHET crystals. These parameters were quantified using Equation (1), Equation (2), Equation (3), Equation (4), and Equation (5), respectively.

$$Conversion\ of\ PET\ (X) = \frac{W_{initial\ PET} - W_{unreacted\ PET}}{W_{initial\ PET}} \qquad \text{Equation (1)}$$

$$Yield\ to\ solids\ (Y) = \frac{W_{TPA}/MW_{TPA}}{W_{PET}/MW_{PET}} = \frac{W_{TPA}/166}{W_{PET}/192} \qquad \text{Equation (2)}$$

$$Yield\ to\ TPA\ (Z) = Yield\ to\ solids\ (Y) \cdot TPA\ purity \qquad \text{Equation (3)}$$

$$Yield\ to\ solids\ (Y) = \frac{W_{BHET}/MW_{BHET}}{W_{PET}/MW_{PET}} = \frac{W_{BHET}/254}{W_{PET}/192} \qquad \text{Equation (4)}$$

$$Yield\ to\ BHET\ (Z) = Yield\ to\ solids\ (Y) \cdot BHET\ purity \qquad \text{Equation (5)}$$

Following the definition of initial operating parameters, and due to better results in preliminary experience with ITENE printed electronic samples, the glycolysis-based depolymerization process was systematically investigated and optimized. An initial series of experiments was conducted to determine the optimal conditions for maximizing PET conversion, reaction selectivity, and product yield, with the objective of obtaining high-purity BHET crystals.

3 Results and discussion

The primary objective of this study was the recovery of silver from printed electronic materials, along with the simultaneous recovery of valuable compounds through chemical recycling of the PET substrate (TPA and BHET monomers than can be repolymerize to r-PET). To achieve this, efforts were focused on adapting the process and optimizing operational conditions to enable the effective depolymerization of PET embedded in multilayer structures and/or complex waste streams that are not suitable for mechanical recycling.

3.1 Characteristics of printed electronic materials

The different printed electronic samples from the project collaborators and ITENE-prepared samples were analysed by FTIR and DSC.

In this spectrum example (Figure 8), it can be seen how there is an overlap between the reference spectra of PET with the spectrum of the sample, this indicates that the sample is composed by PET in high extension. To confirm if the sample has more materials in the intermediate layers, DSC is used as a characterization technique to provide this information.

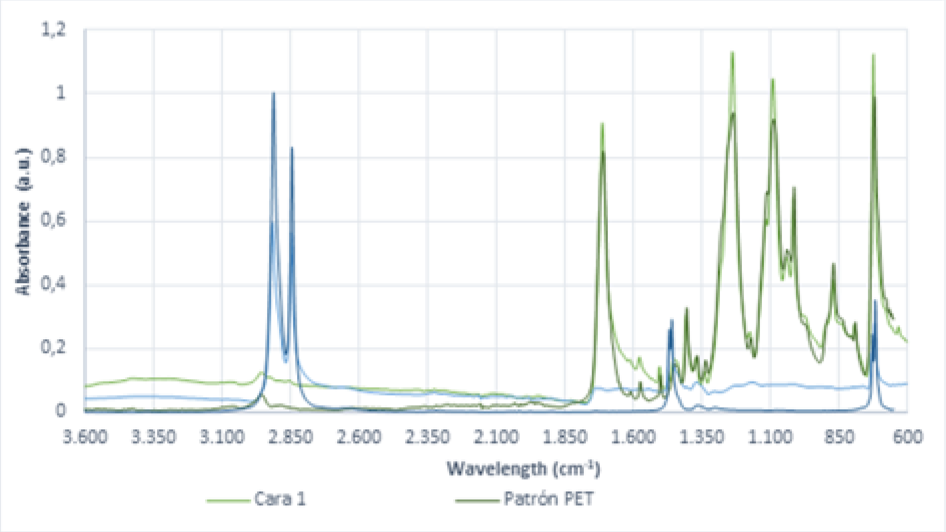

Figure 8. Example of FTIR spectrogram obtained on sample with PET in layer tray of printed electronics.

The DSC analysis (Figure 9) performed on the samples indicates that the material is predominantly composed of PET, thereby refuting the FTIR surface analysis results. Working with clean materials and products of know composition allows for depolymerization with minimal risk of interference. Nevertheless, these preliminary analyses are essential to ensure absence of surface coatings on the PET substrates of printed electronics and to verify that no reaction-interfering substances are present within the material´s internal structure.

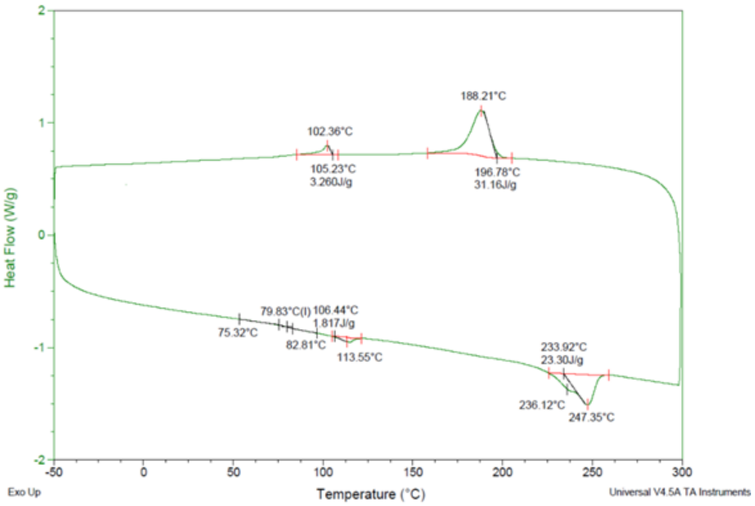

Figure 9. Example of calorimetric curves obtained in DSC of sample of PET substrate in printed electronics.

Regarding ICP-MS analysis, these were performed in samples received at ITENE. The objective was calculated how many silver ink had the materials to recalculate the efficiency of the recovery. In all cases the percentage of silver were variable, but it was situated around 5% in weight, so we chose this amount of silver as representative to recalculate the initial of PET in the samples and perform the PET conversion and monomer selectivity calculations.

3.2 Conditioning and pretreatment methods

The pretreatment of PET-based laminates containing printed electronic components for one-pot chemical recycling requires a precise and controlled mechanical and manual conditioning process. This step is essential to ensure optimal depolymerization efficiency and high product purity. Initially, the laminated substrates are manually cut using precision scissors to obtain a particle size distribution that facilitates effective processing. This also increases the surface area available for the subsequent chemical reaction, ensuring that the PET matrix is adequately exposed to the depolymerizing agents.

During this phase, non-PET materials, such as metallic conductors, encapsulated microelectronic components, and other heterogeneous substances, are carefully removed. This selective extraction is crucial to minimize the presence of inorganic impurities and catalytic contaminants, which could otherwise interfere with the glycolysis process. Properly executed, this preprocessing step maintains the integrity of the chemical reaction, reduces the formation of side products, and enhances the recovery rates of monomers in the recycling pathway, ultimately improving the efficiency of the overall recycling process.

3.3 Selection of high PET content waste materials

Regarding the selection of the most suitable candidates for the depolymerization of printed electronics on PET substrates, an initial screening was carried out using reference samples provided by ITENE. This preliminary stage aimed to gain a better understanding of the materials' behavior under chemical recycling conditions, particularly hydrolysis and glycolysis. The selected samples included a variety of PET-based laminates with different types of conductive inks and printed circuitry. Through these initial trials, key parameters such as PET depolymerization, ink detachment behaviour, and thermal response were evaluated in order to identify which samples were most compatible with downstream depolymerization steps.

Following this assessment, the most promising candidates were selected for process optimization studies, where specific glycolysis conditions—such as reagent ratios, temperature, and reaction time—were adjusted to maximize PET monomer recovery and facilitate separation of the Ag-based conductive inks. These optimized processes were then replicated to validate reproducibility and assess scalability. The final recovered materials, including terephthalic acid (TPA) and Bis(2-hydroxyethyl) terephthalate (BHET), as well as silver inks, were thoroughly characterized to determine purity levels and conversion efficiencies. This comprehensive evaluation enabled the identification of the most effective pathways for both PET repolymerization and Ag-ink recovery within a circular economy framework.

3.4 Analysis and evaluation of depolymerization of PET substrate of printed electronics

3.4.1. Hydrolysis of ITENE printed electronics

Regarding hydrolysis process, a series of alkaline experiences was conducted at laboratory scale to evaluate the recovery of silver ink and the depolymerization efficiency of the PET substrate. The variables investigated included reaction temperatures, sodium hydroxide concentrations, reaction time, and varying ethanol/water ratios under atmospheric pressure. Initial trials were carried out at 70 °C with 1:1 ethanol/water mixture and low NaOH concentrations. While silver ink recovery was successful, the incomplete solubility of NaOH in ethanol and the formation of insoluble Na_2SO_4 during neutralization negatively impacted the purity of the recovered terephthalic acid (TPA).

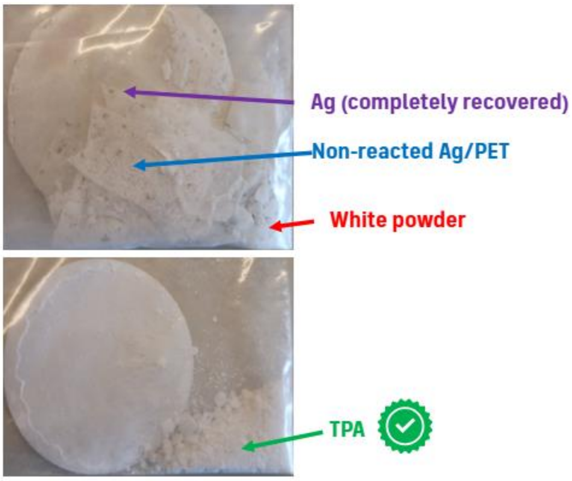

Figure 10. First filter containing Ag recovered, non-reacted Ag/PET and white powder (Ag_2SO_4) (above). Second filter containing TPA monomer product (below).

To address these limitations, subsequent tests were conducted in aqueous-only media at an elevated temperature, close to 95 °C, while maintaining other parameters constant. Under these conditions, first hydrolysis tries (Figure 11, left) with this strategy failed to depolymerize the PET or recover the silver ink. In contrast, optimizing hydrolysis conditions (Figure 11, right) enabled effective silver ink separation through washing, and partial depolymerization of the PET substrate was observed, with TPA yields in low range (5-10% by weight).

Figure 11. Printed electric non-reacted, no recovery Ag-ink and monomers achieved (left). Ag-ink recovered but only 5% of TPA recovered (right).

Further experiments aimed to improve performance by increasing the NaOH concentration while reverting to ethanol/water mixtures at 70 °C. However, neither PET depolymerization nor silver recovery was achieved under these conditions. Final trials using moderate NaOH concentrations and similar ethanol/water ratios, also failed to deliver significant depolymerization or ink recovery (Figure 12).

Figure 12. Non-recovered Ag-ink in no experience and no PET converted (in both images).

Due to these results, by hydrolysis it is only possible to separate silver ink and dielectric in certain scenarios, and the extent of PET depolymerization remained low in all cases. These limitations highlighted the need to shift focus toward an alternative solvolysis pathway—glycolysis—which was subsequently proposed and investigated as a more effective strategy for PET depolymerization and silver recovery.

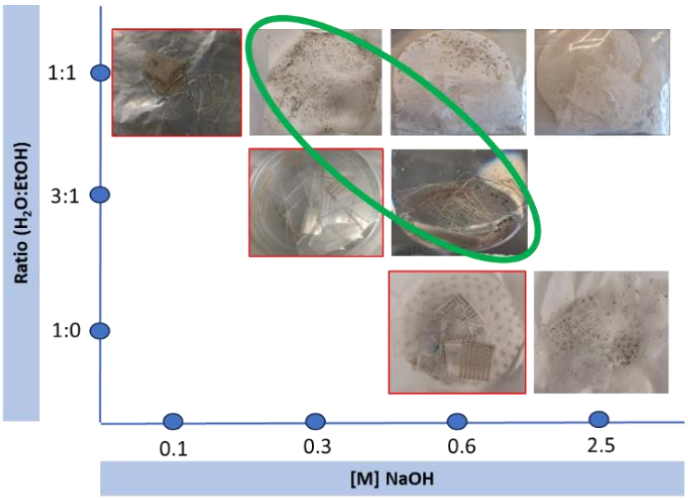

Figure 13. Hydrolysis test under different conditions of water/ethanol ratios and NaOH concentrations

The most successful hydrolysis results are highlighted in Figure 13 within the green oval, alongside a summary of the other trials conducted.

3.4.2. Glycolysis of ITENE printed electronics

Regarding glycolysis, the initial process conditions and parameters were established based on prior experience in PET depolymerization via glycolysis. The studied variables included a reaction temperature of 196 °C under atmospheric pressure, a reaction time ranging from 1 to 3 hours, the use of zinc acetate as a homogeneous catalyst at concentrations between 1–5% by weight PET based, and ethylene glycol to PET mass ratios between 3:1 and 8:1.

Initial glycolysis trials (Figure 14) using printed electronic ITENE samples—comprising a transparent PET substrate with Ag-ink and dielectric layer on one side—were performed to evaluate the process feasibility. The first test, employing zinc acetate and potassium hydroxide as catalysts, resulted in high PET depolymerization rates and BHET yields of approximately 30%. Subsequent experiments using potassium carbonate and zinc acetate as catalysts achieved near-complete PET depolymerization and BHET purities exceeding 90%, with corresponding BHET recoveries of 90% and 80%, respectively.

Figure 14. Solid product (two images on the left) and first filter with unreacted PET and Ag-ink (two images on the right) in first glycolysis ITENE samples.

Subsequent glycolysis experiments were conducted using sodium carbonate as a catalyst to evaluate the influence of reaction temperature. Under optimized conditions, complete depolymerization of the PET substrate was achieved, resulting in a 55% yield of BHET crystals and full recovery of the Ag-ink (Figure 15). However, in a later experiment under similar conditions, no PET degradation was observed, and consequently, silver recovery was not possible.

Figure 15. First filter with Ag-ink and unreacted ITENE's sample (left) and BHET crystals (right).

In the subsequent glycolysis tests, the reaction temperature was kept constant while the reaction time was reduced. Under these conditions, incomplete depolymerization of PET was observed, and the silver remained embedded within the unreacted polymer matrix. Conversely, in a following trial with the same temperature but adjusted conditions, complete PET depolymerization and full recovery of Ag-ink were achieved. In both cases, the BHET crystals obtained exhibited a purity near 100%.

The last glycolysis experiments that were conducted in parallel with hydrolysis trials, involved the use of alternative catalysts such as sodium hydroxide and potassium carbonate (Figure 16). In one case, glycolysis resulted in nearly complete depolymerization of the PET substrate and full recovery of the Ag-ink, although the BHET recovery was limited to approximately 20%. In a subsequent experiment, PET depolymerization was incomplete and Ag-ink recovery was only partial; however, the BHET yield increased to around 50%.

Figure 16. Unreacted PET and Ag-ink using another catalyst in the glycolysis of ITENE samples.

Overall, the glycolysis trials using ITENE's printed electronic samples yielded promising outcomes. In most cases, Ag-ink was effectively separated during the initial filtration step, which is the main objective of the process in the project, achieving recovery rates above 95%. Furthermore, the PET substrate was almost entirely depolymerized into BHET monomer during the second filtration stage, reaching yields exceeding 90%, when using sodium carbonate as catalyst—demonstrating an efficient recovery of both metallic and polymeric components.

3.4.3. Glycolysis of printed electronics with two metal layers and one dielectric on one side

A second series of glycolysis experiments was conducted using printed electronic samples composed of two metal layers and one dielectric on one side. Initial BHET yields were satisfactory when zinc acetate was employed as a catalyst; however, recrystallization of BHET was not achieved under conditions involving both a high EG/PET mass ratio and elevated catalyst concentration. Silver ink separation was effective across all trials, with only slight inefficiencies observed at an EG/PET ratio of 20:1, though still within acceptable performance limits. Following process optimization, the most favourable conditions for these samples were established at 180–200 °C, a reaction time of 1–3 hours, and an EG/PET ratio between 3:1 and 6:1, using zinc acetate as catalyst. Under these conditions, complete Ag-ink separation was achieved along with a BHET crystal yield of approximately 80%.

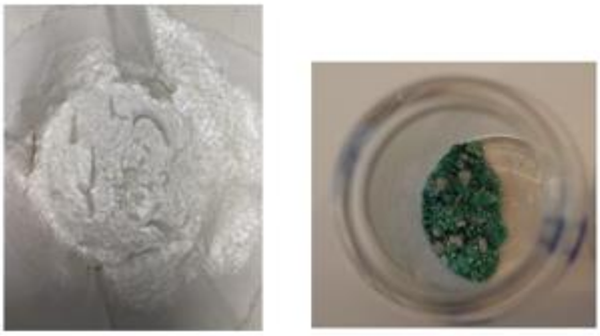

Figure 17. Best experience with printed electronics of two metal layers and one dielectric on one side samples (Left: BHET crystals. Right: separated Ag-ink + dielectric)

3.4.4. Glycolysis of printed electronics with Ag-based ink sample / White cover layer / transparent PET

A third set of glycolysis experiments was carried out using printed electronics with Ag-based ink samples, white cover layer and transparent PET. Depolymerization of PET substrate using Na_2CO_3 as a catalyst proved ineffective in the presence of Ag-based inks, resulting in incomplete reactions and significantly low BHET crystal yields. In contrast, optimal depolymerization was achieved using zinc acetate as the catalyst, with moderate ethylene glycol/PET ratios and reaction times. Under these conditions, approximately 90% BHET recovery was obtained, indicating efficient PET breakdown. Complete depolymerization and higher BHET purity (>95%) were consistently observed with Ag-based samples under these conditions, in comparison to the results obtained from printed electronics, which showed lower performance and impurities issues.

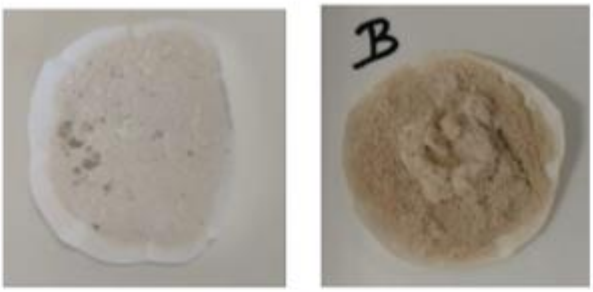

Figure 18. Best experience with printed electronics with Ag-based ink samples, white cover layer and transparent PET (Left: separated Ag-ink + unreacted PET and white cover layer. Right: BHET crystals)

3.4 Characterization of BHET crystals

The BHET crystals obtained from the glycolysis reactions were subsequently characterized by Fourier Transform Infrared Spectroscopy (FTIR) and Differential Scanning Calorimetry (DSC). Figure 19 presents the FTIR spectrum of the BHET samples derived from the glycolysis of printed electronics from ITENE, compared against a reference spectrum of pure PET. A notable increase in the intensity of the absorption band associated with the C–O stretching vibration of alcohols at 1068 cm^{-1} was observed, along with the emergence of multiple absorption bands within the hydroxyl (-OH) region between 3500 and 3200 cm^{-1}. These spectral features confirm the presence of terminal hydroxyl groups in the BHET product, indicating successful depolymerization. The relative intensity and distribution of these hydroxyl-related bands varied depending on the source of the depolymerized PET material, with characteristic peaks identified at 3525 cm^{-1}, 3430 cm^{-1}, and a broader band centered around 3260 cm^{-1}.

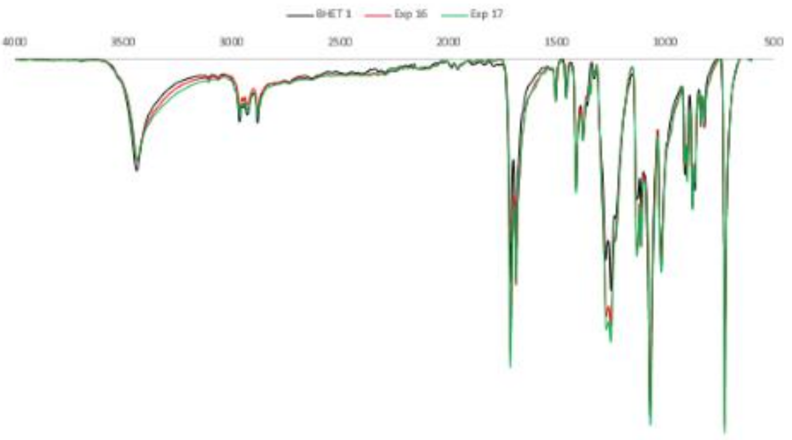

Figure 19. Example of FTIR spectrum of BHET samples (red and green) and reference BHET standard (black)

Variations in the hydroxyl region bands have been interpreted differently in the literature, as they can be influenced by factors such as sample concentration and the specific FTIR instrumentation used. Therefore, the significance of the observed spectral differences remains inconclusive. Furthermore, all products analysed by HPLC showed a purity of 100%, and the differential scanning calorimetry (DSC) profiles did not exhibit any significant deviations, suggesting that these spectral variations may not be analytically relevant ().

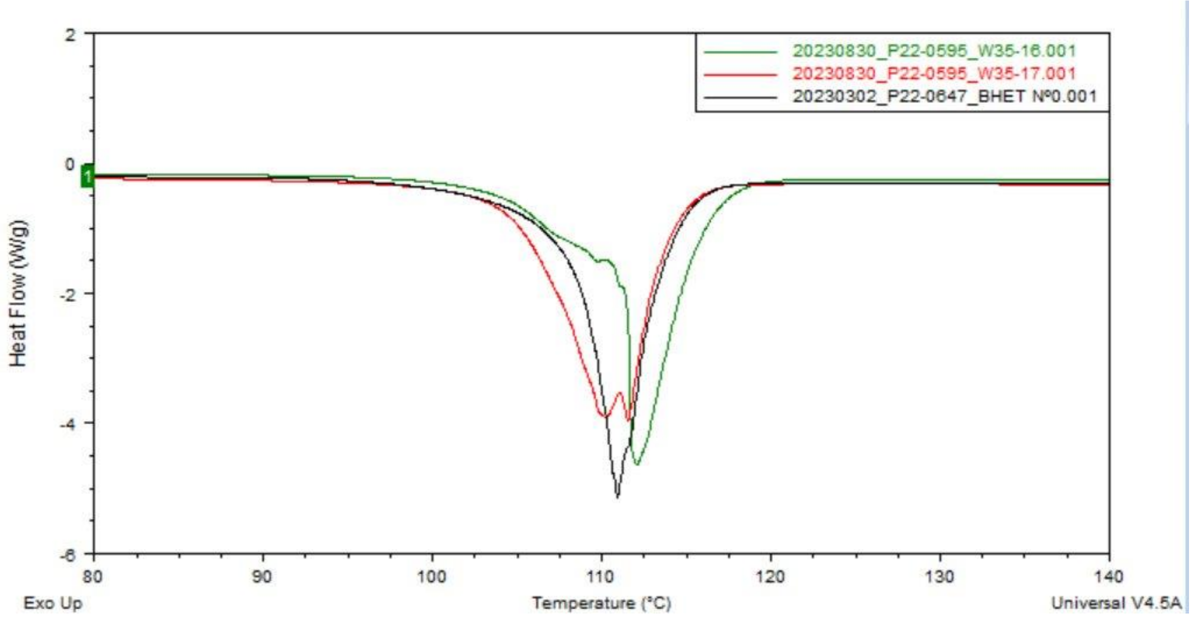

Figure 20. DSC of BHET samples (green and red) and reference BHET standard (black)

4 Conclusions

In this study, chemical recycling of complex printed electronics with PET substrate was carried out to obtain high purity TPA and BHET crystals, at the same time as the silver ink is recovered.

Printed electronics contain various interfering materials such as metal inks, dielectrics, and other components, which can account for up to 25% of the total mass. Additionally, some samples are multilayered and printed in multiple steps, making the recycling process more complex. Analysis has shown that printed electronics typically have a high PET content, while interference from other polymers is negligible. Although the silver content in the inks varies, its recovery presents a promising opportunity for reuse in other applications as recycled silver.

The hydrolysis process for producing TPA and recovering silver ink is only feasible under specific conditions and typically results in low TPA yields. It operates in an alkaline medium with an optimal ethanol/water concentration. Due to its limited efficiency, this method was discarded in favor of glycolysis, which offers higher monomer yields.

The glycolysis process, using ethylene glycol and a catalyst, achieves a high yield of BHET monomers and has proven effective in recovering silver ink in most cases. Preliminary trials with printed electronic samples from ITENE yielded promising results, which led to the application of the process to commercially available printed electronics. In these additional samples, significant amounts of silver were recovered in the first filtration step, and BHET yields remained high. These findings support the conclusion that glycolysis is an effective method for chemically recycling complex materials and structures, enabling the production of recycled polymers such as recycled PET.

FTIR and DSC analyses of the obtained BHET crystals confirm almost 100% purity in all cases, attributed to the use of highly pure PET as the starting material, with no presence of other polymers or spectral interferences.

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References

1. Kambiz Sadeghi, Yukwon Jeon, Jongchul Seo, Roadmap to the sustainable synthesis of polymers: From the perspective of CO2 upcycling, Progress in Materials Science, Volume 135, 2023, 101103, SSN 0079-6425, https://doi.org/10.1016/j.pmatsci.2023.101103.
2. Monika Dokl, Anja Copot, Damjan Krajnc, Yee Van Fan, Annamaria Vujanović, Kathleen B. Aviso, Raymond R. Tan, Zdravko Kravanja, Lidija Čuček, Global projections of plastic use, end-of-life fate and potential changes in consumption, reduction, recycling and replacement with bioplastics to 2050, Sustainable Production and Consumption, Volume 51, 2024, Pages 498-518, ISSN 2352-5509, https://doi.org/10.1016/j.spc.2024.09.025.
3. Jianfu Liu, Liang Zheng, Microplastic migration and transformation pathways and exposure health risks, Environmental Pollution, Volume 368, 2025, 125700, ISSN 0269-7491, https://doi.org/10.1016/j.envpol.2025.125700
4. Luqman Umdagas, Rafael Orozco, Kieran Heeley, William Thom, Bushra Al-Duri, Advances in chemical recycling of polyethylene terephthalate (PET) via hydrolysis: A comprehensive review, Polymer Degradation and Stability, Volume 234, 2025, 111246, ISSN 0141-3910, https://doi.org/10.1016/j.polymdegradstab.2025.111246
5. Stuart Conroy, Xiaolei Zhang, Theoretical insights into chemical recycling of polyethylene terephthalate (PET), Polymer Degradation and Stability, Volume 223, 2024, 110729, ISSN 0141-3910, https://doi.org/10.1016/j.polymdegradstab.2024.110729
6. Marco Rollo, Francesca Raffi, Elisa Rossi, Matteo Tiecco, Elisa Martinelli, Gianluca Ciancaleoni, Depolymerization of polyethylene terephthalate (PET) under mild conditions by Lewis/Brønsted acidic deep eutectic solvents, Chemical Engineering Journal, Volume 456, 2023, 141092, ISSN 1385-8947, https://doi.org/10.1016/j.cej.2022.141092
7. Mahsa Babaei, Milad Jalilian, Kaveh Shahbaz, Chemical recycling of Polyethylene terephthalate: A mini-review, Journal of Environmental Chemical Engineering, Volume 12, Issue 3, 2024, 112507, ISSN 2213-3437, https://doi.org/10.1016/j.jece.2024.112507
8. Rulin Yang, Guangqiang Xu, Wenjie Tao, Qinggang Wang, Yong Tang, Recycled polymer: Green roads for polyester plastics, Green Carbon, Volume 2, Issue 1, 2024, Pages 1-11, ISSN 2950-1555, https://doi.org/10.1016/j.greenca.2024.01.004.

9. Adam McNeeley and Y. A. Liu, Assessment of PET Depolymerization Processes for Circular Economy. 2. Process Design Options and Process Modeling Evaluation for Methanolysis, Glycolysis, and Hydrolysis, Industrial & Engineering Chemistry Research 2024 63 (8), 3400-3424. DOI: 10.1021/acs.iecr.3c04001
10. Myung Jong Kang, Hye Jin Yu, Jonggeon Jegal, Hyun Sung Kim, Hyun Gil Cha, Depolymerization of PET into terephthalic acid in neutral media catalyzed by the ZSM-5 acidic catalyst, Chemical Engineering Journal, Volume 398, 2020, 125655, ISSN 1385-8947, https://doi.org/10.1016/j.cej.2020.125655
11. Minghao Zhang, Yijin Lu, Zhuo Wang, Xiong Gao, Xuanhang Luo, Xin Shen, Weixiang Wu, Qingqing Mei, Targeted conversion of waste PET into dimethyl terephthalate and ethylene carbonate under metal-free conditions, Eco-Environment & Health, Volume 4, Issue 2, 2025, 100139, ISSN 2772-9850, https://doi.org/10.1016/j.eehl.2025.100139
12. Stuart Conroy, Xiaolei Zhang, Theoretical insights into chemical recycling of polyethylene terephthalate (PET), Polymer Degradation and Stability, Volume 223, 2024, 110729, ISSN 0141-3910, https://doi.org/10.1016/j.polymdegradstab.2024.110729
13. Jiayu Xin, Qi Zhang, Junjie Huang, Rong Huang, Quratulain Zahra Jaffery, Dongxia Yan, Qing Zhou, Junli Xu, Xingmei Lu, Progress in the catalytic glycolysis of polyethylene terephthalate, Journal of Environmental Management, Volume 296, 2021, 113267, ISSN 0301-4797, https://doi.org/10.1016/j.jenvman.2021.113267
14. Yongqi Feng, Shi-Wen Lv, Rui Zhang, Xuhua Ren, Juefeng Shen, Yanqing Cong, From waste to wealth: Glycolysis of PET for high-value resource utilization, Waste Management, Volume 200, 2025, 114768, ISSN 0956-053X, https://doi.org/10.1016/j.wasman.2025.114768