INNOVATIVE DIGITAL WATERMARKS AND GREEN SOLVENTS FOR THE RECOVERY AND RECYCLING OF MULTI-LAYER MATERIALS

Funding scheme: European Union's Horizon 2020 Research and Innovation programme

Call identifier: H2020-SC5-2020-2

Theme: CE-SC5-24-2020: Improving the sorting, separation and recycling of composite and multi-layer materials

Grant Agreement: 101003532

Project start date: June 1st, 2021

Duration: 36 months

DELIVERABLE N°5.4

Report on the Analysis of Isolated Polymers

Due date of deliverable: 31/08/2023	Actual submission date: 02/11/2023	Lead Beneficiary: TWI

Table of Contents

1 Introduction

SolRec 2 project targets the development and implementation of ground-breaking strategies for improving the sorting, separation and recycling of pharmaceutical blister packs and flexible laminate packaging waste which comprise multiple layers of plastics, adhesives, inks and aluminium. Although widely used for the protection and preservation of pharmaceutical and food produce across the globe, such packaging materials present significant challenges for established recycling infrastructure and, therefore, our future with a circular economy.

Experience from the field of ionic liquids (ILs) and deep eutectic solvents (DESs) has been leveraged to develop a toolbox of novel green solvent systems that delaminate multilayer packaging materials to facilitate separation and recovery of high purity commodity plastics, such as, poly(ethylene) (PE), poly(ethylene terephthalate) (PET) and poly(vinyl chloride) (PVC), as well as the valuable barrier materials such as aluminium (ALU) foils. In parallel, innovative digital watermark technologies are being further developed and progressed through to successful demonstration of rapid and efficient sorting of multilayer packaging; a key step towards proper inventory management of complex mixed waste streams.

Work Package 5 (WP5) of Sol-Rec^2 project has centred on the scale-up of processes and procedures from laboratory bench-top studies to a 1 L laboratory reactor vessel. The activity has enabled partners to produce, study and improve both the quantity and quality of delaminated materials and manage the recovery and recycling of solvents.

In Deliverable D5.4 we present characterisations of the plastics recovered via delamination and sorting processes.

2 Methods

2.1 Multilayer Packaging

Samples of primary industrial waste packaging materials were collected and supplied by our project partners, Plastigram and Mikrolin, as shown in Figure 1. These included pharmaceutical blister packs based on PVC/PE/ALU and three common multilayer packaging films of PE/ALU, ALU/PE/PAPER and PE/ALU/PET.

(a) (b) (c) (d)

Figure 1: From left to right: (a) pharmaceutical blister pack, (b) PE/ALU, (c) ALU/PE/PAPER and (d) PE/ALU/PET packaging materials provided by partners Mikrolin and Plastigram.

2.2 Sol-Rec² Processes

2.2.1 Deep Eutectic Solvents and Ionic Liquids

Partners developed and short-listed three solvent systems for the scale-up of Sol-Rec² processes in WP5 as a result of earlier studies in Work Package 4 (WP4). For reference, the nomenclature for the solvent systems is given in Table 1.

Table 1 Short-list of deep eutectic solvents and ionic liquids established in WP4.

Partner	Type	Solvent System
TWI	DES	DES 1 – TWI
ULEIC	DES	DES 2 – ULEIC
SOL	IL	IL 3 – SOL

2.2.2 Delamination

A brief summary of the delamination experiments (see Deliverable D5.1) performed at TWI using a 1 L reactor vessel with the aforementioned solvent systems and packaging materials is shown in Table 2. The experimental conditions in terms of temperature and duration were chosen to ensure complete delamination of the respective packaging materials.

Table 2 Table of Experiments A-G detailing solvent system, packaging material, size fraction of shredded material denoted by screen size, solids loading in the solvent and number of replicate trials for each set of experiments.

REF	SOLVENT SYSTEM	PACKAGING MATERIAL	SHREDDED SIZE FRACTION (screen aperture) (mm)	SOLIDS LOADING (g L^{-1})	NO. OF TRIALS
A	IL 3 – SOL	PE/ALU	25	30	5
B		PE/ALU/PET	5	30	5
C		PVC/PE/ALU	9	150	1
D	DES 2 – ULEIC	PE/ALU	5	30	5
E		PE/ALU/PET	25	30	5
F	DES 1 – TWI	ALU/PE/PAPER	25	30	5
G		PE/ALU/PET	25	30	5

2.2.3 Sorting

After completion of each series of delamination trials using the 1 L reactor vessel, the mixtures of materials were sent from TWI to AIMPLAS for separation and recovery of the individual components via sorting trials using electrostatic methods. A detailed account of the sorting processes was given in Deliverable D5.3.

Only the 25 mm size fraction of delaminated PE/ALU, ALU/PE/Paper and PE/ALU/PET were readily separated by sorting methods, the smaller 5 mm materials were not. The 9 mm size fraction of blister packaging (PVC/PE/ALU) was easily separated.

The materials recovered after delamination and sorting are those described in this report where characterisations have been performed to assess the quality of the isolated polymers: PE, PET and PVC.

2.2.4 Additional Samples for Characterisation

A separate set of film samples were prepared using the established delamination methods before being provided to University of Leicester for analysis using Fourier Transform Infra-red (FTIR) spectroscopy.

Pieces of PE/ALU, ALU/PE/PAPER and PE/ALU/PET film were cut in dimensions of 25 mm × 25 mm. The blister pack samples (which had been industrially shredded with a 9 mm screen) varied in size and shape, typically, less than 10 mm.

Specimens of the packaging materials were immersed in a batch of each solvent system (100 g) at 70 °C for 60 min under gentle stirring conditions (400 rpm). Table 1 summarises the aforementioned film information as well as the used solvent systems.

Table 3 Details of solvents used to delaminate and prepare materials for FTIR analysis.

#	Solvent System	Mass of Solvent (g)	Packaging Material	Size of Pieces
1	DES 1-TWI	100	PE/ALU	25 mm x 25 mm
2			ALU/PE/PAPER**	
3			PE/ALU/PET**	
4	DES 2-ULEIC	100	PE/ALU	25 mm x 25 mm
5			PE/ALU/PET**	
6	IL 3-SOL	100	PE/ALU	25 mm x 25 mm
7			PE/ALU/PET	
8			PVC/PE/ALU	< 10 mm

Three of the materials, which are indicated in Table 3 with **, showed a partial delamination (~50%), as presented in Figure 2 for PE/ALU/PET. However, FTIR analysis was performed successfully, as the edges of the samples were fully delaminated. It is noteworthy that manual delamination of the different films was possible.

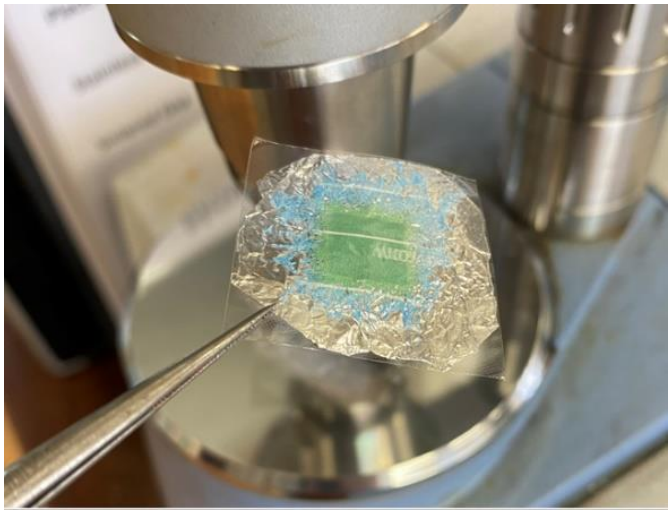


Figure 2 Partially delaminated PE/ALU/PET sample with the printed PET film still attached to the ALU layer.

2.3 Characterisations

2.3.1 Mass Measurements

Mass measurements of the components of delaminated packaging laminates were taken before and after electrostatic sorting to assess the quality of the separation process for PE, PET, paper, PVC and aluminium.

After taking representative samples from the bulk of delaminated material for each packaging type, the different components were inspected and manually separated into individual fractions which could then be measured.

A Cobos A-220-CSI balance, with sensitivity to ± 0.1 mg, was used to weigh the separated films.

2.3.2 Ash Content

An alternative approach to the above for binary mixtures of plastic and aluminium was to determine the residual ash content after placing a sample of delaminated or sorted material in a furnace to burn-off the organic polymers at 600 °C.

Again, a Cobos A-220-CSI balance, with sensitivity to ± 0.1 mg, was used to weigh the residual ash.

2.3.3 Fourier Transform Infra-red Spectroscopy

Fourier Transform Infra-red (FTIR) Spectroscopy is an analytical technique for evaluating the chemical composition of materials. Typically, polymers will have their own characteristic infra-red spectrum which reflects the combination of chemical groups in the material, such as, C-H, C=O, C-O-C and O-H. Therefore, the FTIR spectrum can be used to confirm a material or identify the presence of other materials. In the case of packaging materials, adhesives and inks also used in the assembly of the laminate structures will contribute to FTIR spectra if they remain on the surface of the PE, PET, paper, PVC and aluminium foils.

An Alpha Platinum ATR FTIR Spectrometer (Bruker Corporation) located at University of Leicester (Department of Chemistry) was used to evaluate the separated plastics and aluminium. The number of scans for each sample was three and the scan range was 4000-400 cm^{-1}. The obtained spectra for the three different points were in good agreement, hence one of them is presented in the results. The data were collected and later processed in MS Excel.

Additional FTIR analysis was performed in parallel by AIMPLAS using a Frontier FTIR Spectrometer (Perkin Elmer) equipped with the GladiATR diamond crystal accessory. A total of 4 scans were acquired over the range 4000-400 cm^{-1}. The complementary spectra and results are presented in Appendix A.

2.3.4 Differential Scanning Calorimetry

Differential Scanning Calorimetry (DSC) is a thermal analysis technique to detect and quantify changes in the morphology of materials when heated and cooled, for example, to measure glass-transition temperatures, melting and crystallisation transitions in polymers.

Experiments on the recovered plastics were performed by AIMPLAS using a POLYMA 214 DSC (Netzsch GmbH) under an inert atmosphere of N_2 purge gas (50 mL min^{-1}; 99.998 %). Calibrations of the instrument temperature and heat flow profiles were made using high-purity standards of indium (99.99998%), tin (99.99998%) and zinc (99.99998%). Samples of each plastic film were tested and analysed by employing heat-cool-heat cycles and followed international test standards (UNE-EN ISO 11357-1:2017; UNE-EN ISO 11357-3:2018).

Full details of the test conditions are given in Table 4.

Table 4 Conditions of DSC experiments.

Scheme	Analysis of PE and PET	Analysis of PVC
1st Heating	40 °C – 300 °C	0 °C – 300 °C
1st Cooling	300 °C – 40 °C	300 °C – 0 °C
2nd Heating	40 °C – 300 °C	0 °C – 300 °C
Temperature Ramp Rate	20 °C min^{-1}	20°C min^{-1}
Isothermal	1 min pause between heat/cool/heat cycles	1 min pause between heat/cool/heat cycles
Purge Gas	N_2 (50 mL min^{-1}; 99.998 %)	N_2 (50 mL min^{-1}; 99.998 %)

Evaluation of the DSC thermograms to evaluate glass-transition temperatures, melting and crystallisation behaviours was performed using Proteus® software (Netzsch GmbH).

3 Results

3.1 Sorting Quality

3.1.1 PE/ALU

Table 5 records the composition of the delaminated and sorted fractions of PE/ALU laminate film. The sorted PE fraction contains 96.29% w/w of PE with 3.71% w/w of aluminium and demonstrates an improvement in PE content over the delaminated mixture. Although the sorted aluminium fraction contains 63.76% w/w aluminium and 36.24% w/w of PE, this represents a significant shift, as only 11% w/w of the original mix was due to aluminium.

The results are presented in Figure 3.

Table 5 Composition of delaminated and sorted fractions of PE/ALU packaging laminate.

Reference	Description	Material	Sample Mass (g)	Component	Purity (% w/w)
A	PE/ALU (25mm)	PE/ALU (delaminated)	< 2.9	PE	88.93
				Aluminium	11.07
A Sorted	PE/ALU (25mm)	PE (sorted)	11.01	**PE**	**96.29**
				Aluminium	3.71
		Aluminium (sorted)	7.90	PE	36.24
				Aluminium	**63.76**

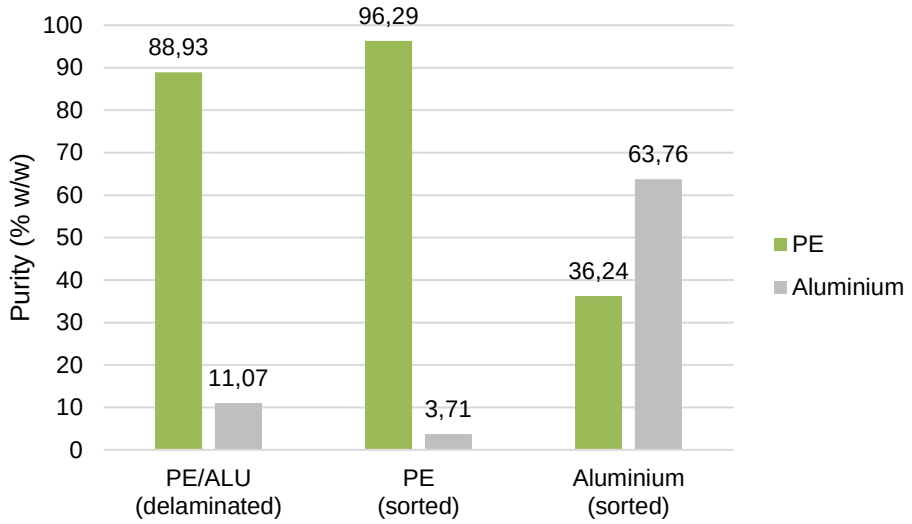


Figure 3 Plot comparing the composition of delaminated and sorted fractions of PE/ALU packaging laminate.

3.1.2 ALU/PE/PAPER

Mass measurements from the delaminated and sorted fractions of ALU/PE/Paper are given in Table 6.

Table 6 Composition of delaminated and sorted fractions of ALU/PE/Paper packaging laminate.

Reference	Description	Material	Sample Mass (g)	Component	Purity (% w/w)
F	PE/ALU/Paper (25mm)	PE/ALU/Paper (delaminated)	2.98	PE	15.04
				Paper + Aluminium	84.96
F Sorted	PE/ALU/Paper (25mm)	PE (sorted)	0.72	**PE**	**79.80**
				Paper + Aluminium	20.20
		Paper + Aluminium (sorted)	7.174	PE	12.33
				Paper + Aluminium	87.67

The PE component of the delaminated mixture is far less than for the PE/ALU and PE/ALU/PET laminates as it forms a much thinner interlayer between paper and aluminium. Nevertheless, the sorted PE fraction was shown to contain 79.80% w/w of PE with a residual 20.20% w/w mix of paper and aluminium which demonstrates a reasonable level purity.

The results are plotted in Figure 4.

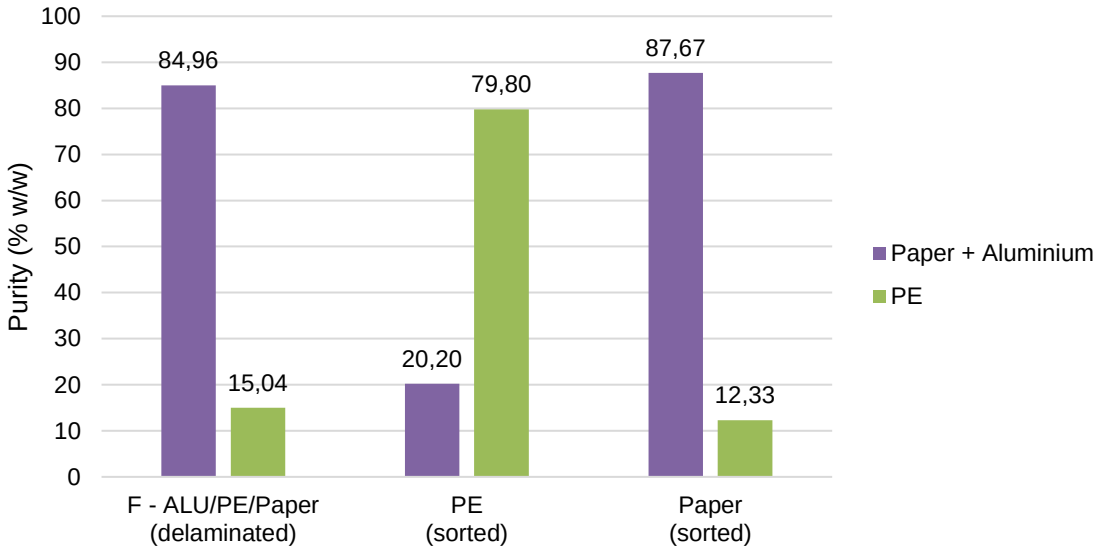


Figure 4 Plot comparing the composition of delaminated and sorted fractions of ALU/PE/Paper packaging laminate.

3.1.3 PE/ALU/PET

Table 7 records the composition of the delaminated and sorted fractions of PE/ALU/PET laminate film.

Table 7 Composition of delaminated and sorted fractions of PE/ALU/PET packaging laminate.

Reference	Description	Material	Sample Mass (g)	Component	Purity (% w/w)
E	PE/ALU/PET (25mm) DES2-ULEIC	PE/ALU/PET (delaminated)	0.52	PET	13.76
				PE	69.39
				Aluminium	16.85
G	PE/ALU/PET (25mm) DES1-TWI	PE/ALU/PET (delaminated)	0.30	PET	8.81
				PE	78.51
				Aluminium	12.68
E/G Sorted	PE/ALU/PET (25mm)	PE (sorted)	0.76	PET	7.94
				PE	**87.55**
				Aluminium	4.51
		Aluminium (sorted)	0.320	PET	4.31
				PE	19.08
				Aluminium	**76.61**
		PET (sorted)	0.189	**PET**	**44.01**
				PE	44.28
				Aluminium	11.71

The sorted PE fraction contains 87.55% w/w of PE with 4.51% w/w of aluminium and 7.94% w/w PET. Again, this demonstrates an improvement in PE content over the delaminated mixture. The sorted aluminium fraction contains a higher 76.61% w/w aluminium component which is a significant improvement. The sorted PET fraction proved to be

the most challenging as approximately equal components of PE and PET were obtained at 44.28% w/w and 44.01% w/w, respectively.

The results are presented in Figure 5.

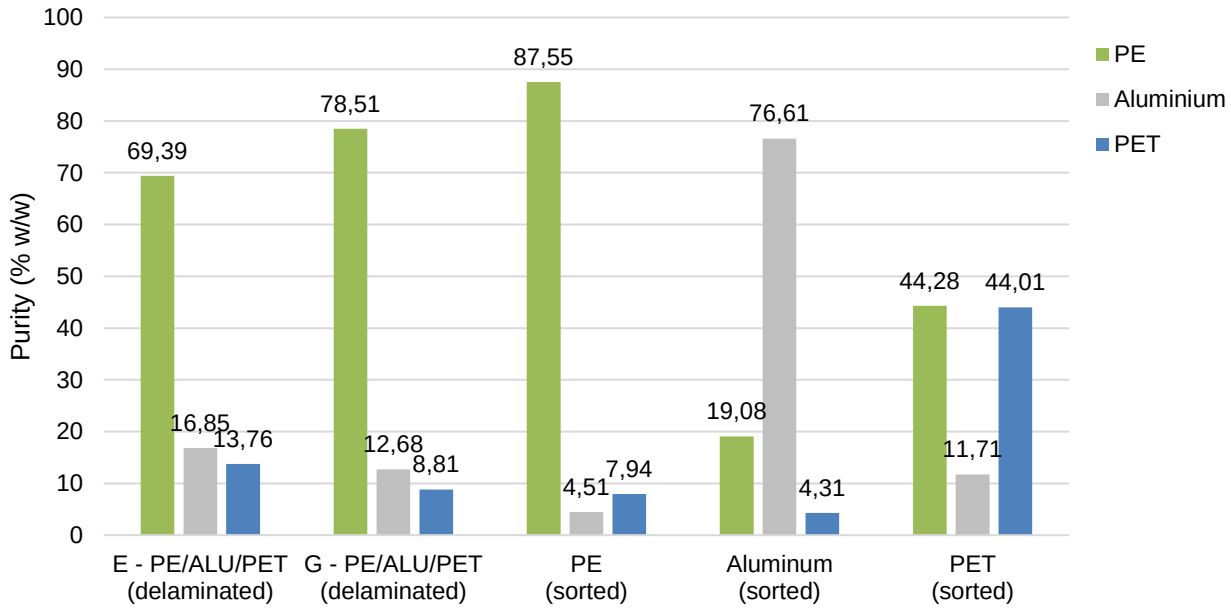


Figure 5 Plot comparing the composition of delaminated and sorted fractions of PE/ALU/PET packaging laminate.

3.1.4 Blister Packaging

The higher mass and rigidity of PVC and aluminium flakes from the blister packaging material helped to ensure a very high purity of each sorted fraction. As can be seen in Table 8, the sorted fractions of aluminium, PVC and PE were determined to be ~99% pure.

The results are plotted in Figure 6.

Table 8 Composition of delaminated and sorted fractions of PVC/PE/ALU blister packaging.

Reference	Description	Material	Sample Mass (g)	Component	Purity (% w/w)
C Sorted	Blister Packaging (9 mm)	Aluminium (sorted)	1.9	PVC + PE	0.31
				Aluminium	**99.69**
		PVC (sorted)	5	**PVC**	**99.67**
				Aluminium	0.33
		PE (sorted)	5	**PE**	**98.86**
				Aluminium	1.14

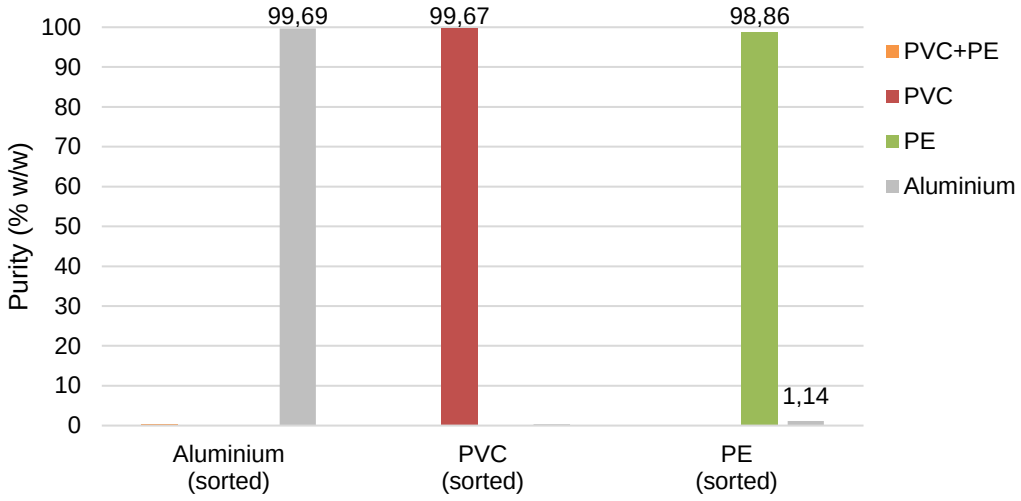


Figure 6 Plot comparing the composition of delaminated and sorted fractions of PVC/PE/ALU blister packaging.

3.2 Thermal Analysis

3.2.1 PE/ALU

The analysis of PE film on first and second heating cycles is shown in Figure 7. The broad melting behaviour observed between 50 – 120 °C can be attributed to various crystalline phases of PE.

The two main PE crystalline peak melting temperatures were measured at 110.1 °C and 118.3 °C in the first heating cycle and 110.6 °C and 120.2 °C in the second cycle. These features are attributed to melting of two different crystalline phases associated with low-density and high-density PE.

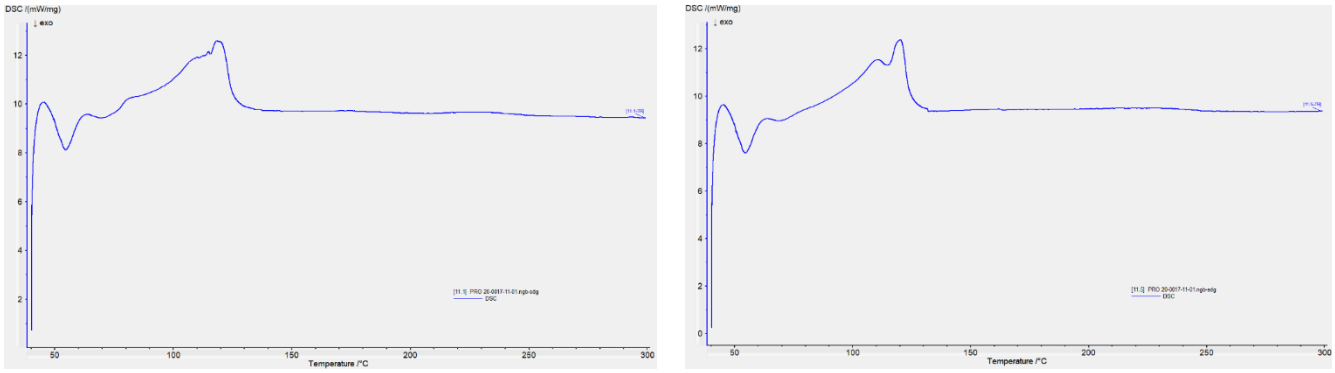
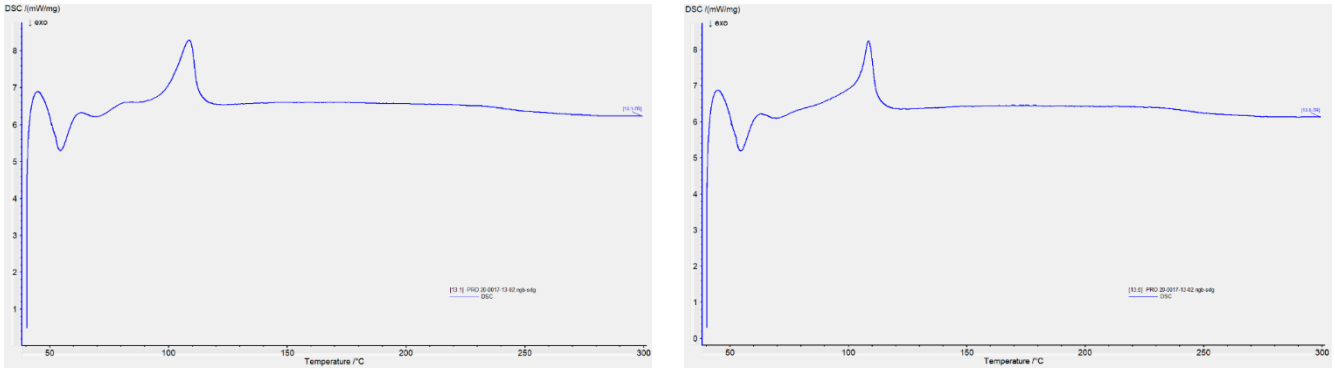


Figure 7 Comparison of (a) first and (b) second DSC heating cycles on PE film recovered from PE/ALU.

In the first heating cycle a small peak was also observed at 85.4 °C, which was not apparent in the second cycle. This may have been due to the evaporation of a volatile component, perhaps residual solvent or moisture, or could be associated with residual adhesive on the film surface.

3.2.2 PE/ALU/ PAPER

The analysis of the thin PE film interlayer between aluminium and paper is presented in Figure 8. From the melting behaviour of the PE, where a single peak appears at 108.5 °C, which corresponds to low-density PE. We can infer that a different grade of polymer has been used compared to PE/ALU and PE/ALU/PET laminates.



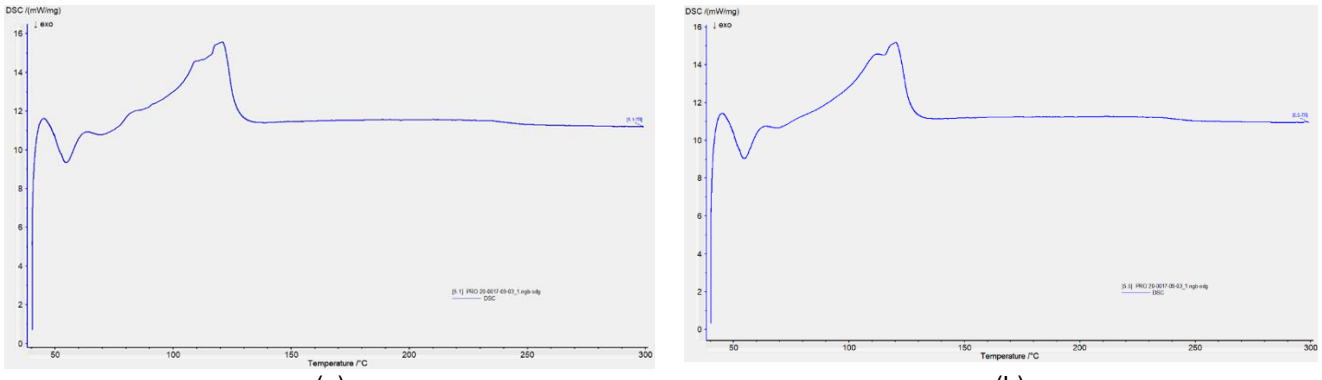
(a) (b)

Figure 8 Comparison of (a) first and (b) second DSC heating cycles on PE film recovered from PE/ALU.

The small peak in the first heating, again, at 85.4 °C is not apparent in the second cycle. The position of the peak in temperature implies a similar origin to that observe in PE from PE/ALU and PE/ALU/PET.

3.2.3 PE/ALU/PET

Figure 9 presents the first and second heating cycles from PE film. The results are very similar to those above in Figure 7 (for PE from PE/ALU laminate) and correspond to the same broad melting behaviour of different crystalline phases between 50 – 120 °C.



(a) (b)

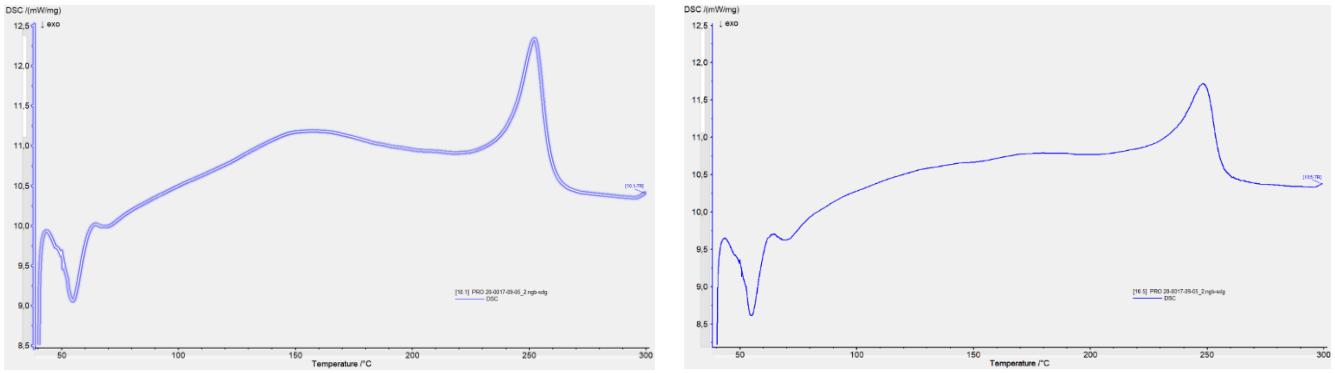
Figure 9 Comparison of (a) first and (b) second DSC heating cycles on PE film recovered from PE/ALU.

The peak melting temperatures associated with low-density and high-density PE were measured at 112.0 °C and 120.7 °C, respectively, during the first heating cycle and 112.3 °C and 120.2 °C on the second cycle.

As previously mentioned, the small peak in the first heating, this time, at 82.0 °C, is not apparent in the second cycle.

The analysis of PET film in Figure 10 clearly highlights the melting behaviour of crystalline PET with single peaks at 252.1 °C and 248.1 °C on first and second heating cycles. The glass-transition of PET associated with the amorphous phase, normally, observed as step-wise change in heat-flow at ~ 85 °C, is not clearly defined in the measurement.

A broad feature observed in the first heating cycle centred at 159.7 °C may be due to a residual volatile component in the polymer, such as solvent.

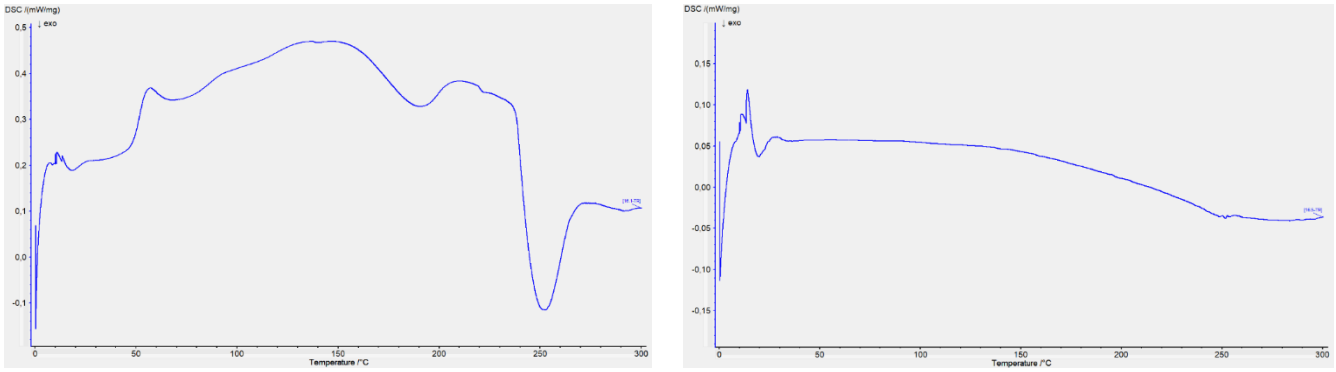


(a) (b)

Figure 10 Comparison of (a) first and (b) second DSC heating cycles on PET film recovered from PE/ALU/PET.

3.2.4 Blister Packaging

The analysis of PVC is presented in Figure 11. The first heating displays a number of features; the step-wise increase in heat-flow at 50.6 °C can be attributed the glass-transition temperature of PVC. The high temperature inverted peaks at ~180 °C and ~ 250 °C are not consistent with characteristic thermal behaviour of PVC and maybe be due to polymer degradation and loss of volatile material as the temperature increases towards 300 °C. The second heating cycle is largely featureless which would be consistent with degradation of the polymer. A substantial 46% reduction in sample mass was measured at the conclusion of the tests and again supports this conclusion (albeit a consequence of the test conditions).



(a) (b)

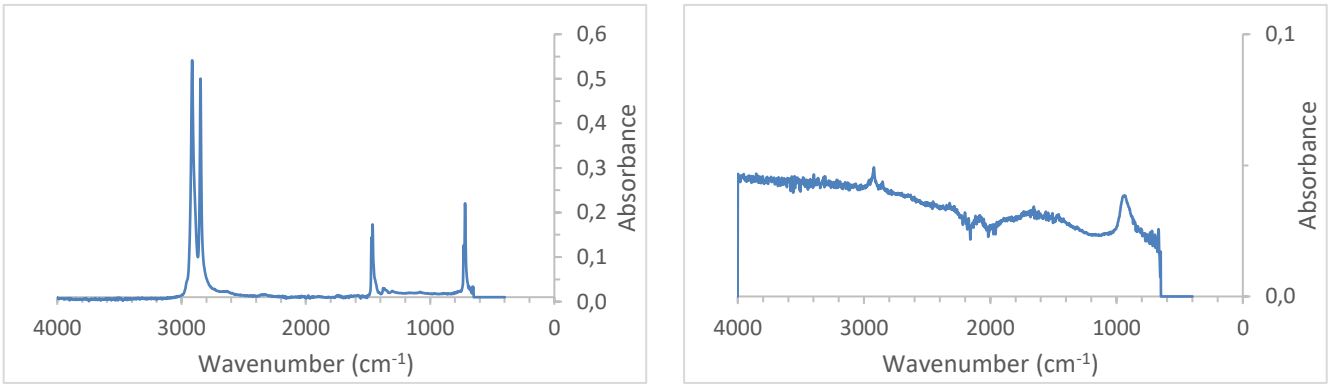
Figure 11 Comparison of (a) first and (b) second DSC heating cycles on PVC recovered from blister packaging.

3.3 Fourier Transform Infra-red Spectroscopy

3.3.1 FTIR Analysis of Initial Packaging Materials

The FTIR spectra of the initial packaging materials are presented in this section. These were obtained for comparison with those of the delaminated films. All spectra were also compared with and referenced against known examples of material spectra available on SpectraBase® (John Wiley & Sons, Inc.).

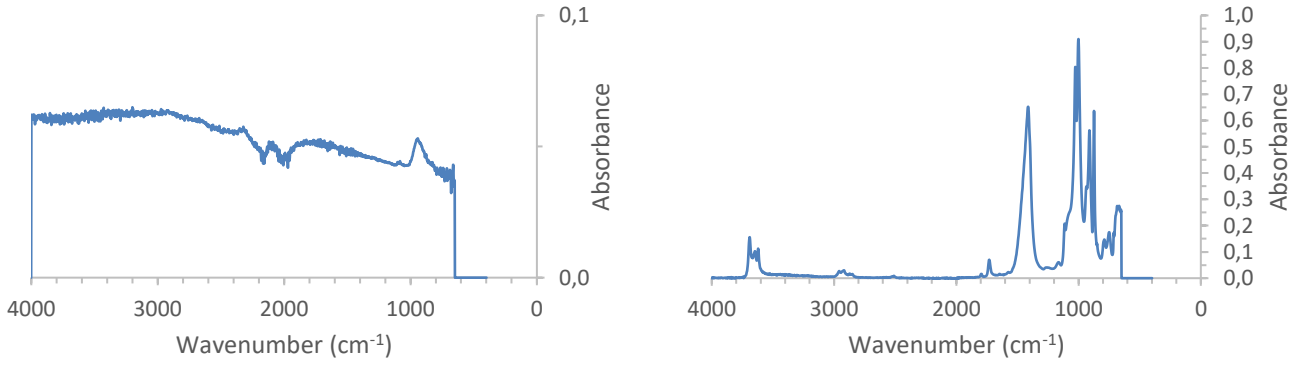
Figure 12 shows the two outer surfaces of PE/ALU. Figure 12(a), as expected, shows the characteristic FTIR spectrum of PE. The spectrum obtained from the outer aluminium surface in Figure 12(b) is noisy with no clearly identifiable spectral bands.



(a) (b)

Figure 12 FTIR analysis of the two outer surfaces of PE/ALU: (a) PE and (b) ALU.

The outer surfaces of ALU/PE/PAPER are shown in Figure 13. The aluminium spectrum in Figure 13(a) is similarly noisy whilst the spectrum in Figure 13(b) is consistent with a cellulosic material (i.e., paper).



(a) (b)

Figure 13 FTIR analysis of the two outer surfaces of ALU/PE/PAPER: (a) ALU and (b) PAPER.

The spectra of PE and PET are presented in Figure 14 (a) and (b), respectively, as the outer surfaces of PE/ALU/PET laminate.

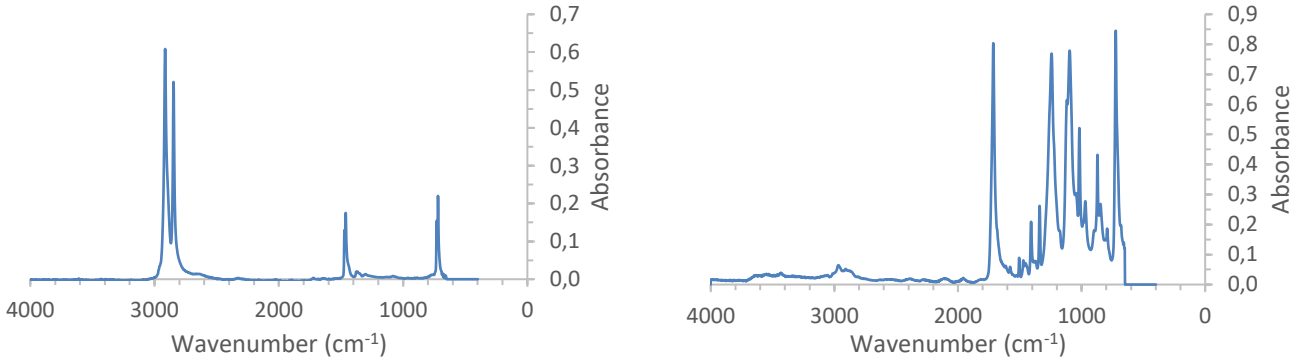


Figure 14 FTIR analysis of the two outer surfaces of PE/ALU/PET: (a) PE and (b) PET.

Finally, in Figure 15(a) and (b), respectively, the spectra obtained for the blister packaging were consistent with PVC and an acrylate-based ink printed on the top surface of the aluminium.

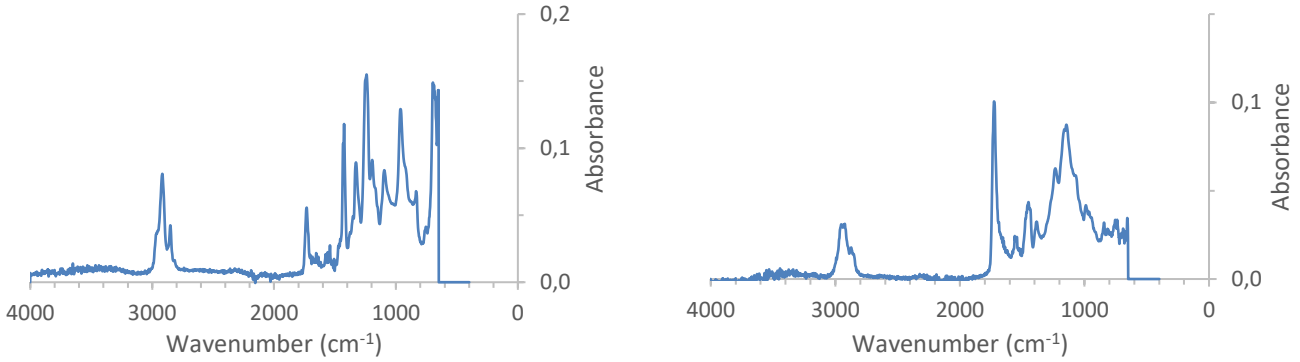


Figure 15 FTIR analysis of the two outer surfaces of pharmaceutical blister pack: (a) PVC and (b) printed ink on ALU.

3.3.2 FTIR Analysis of Delaminated PE/ALU

The three solvent systems were successful in fully delaminating the PE/ALU film. FTIR analysis was performed for both sides of PE, inner and outer surfaces, for each of the three samples. Figure 16 shows the films of polyethylene and aluminium after separation.

Figure 16 Images of PE/ALU film delaminated using: (a) DES 1 – TWI, (b) DES 2 – ULEIC and (c) IL 3 – SOL.

Figure 17 contains the FTIR spectra of PE. The outer (left) and inner (right) surfaces of PE are in good agreement between the different tests. The inner surfaces contain residues of adhesives, used for attaching the aluminium film. The similarities in the obtained spectra confirm that the DES 1 – TWI, DES 2 – ULEIC and IL 3 – SOL solvent systems were successful in the recovery of PE without any significant chemical interaction or absorption.

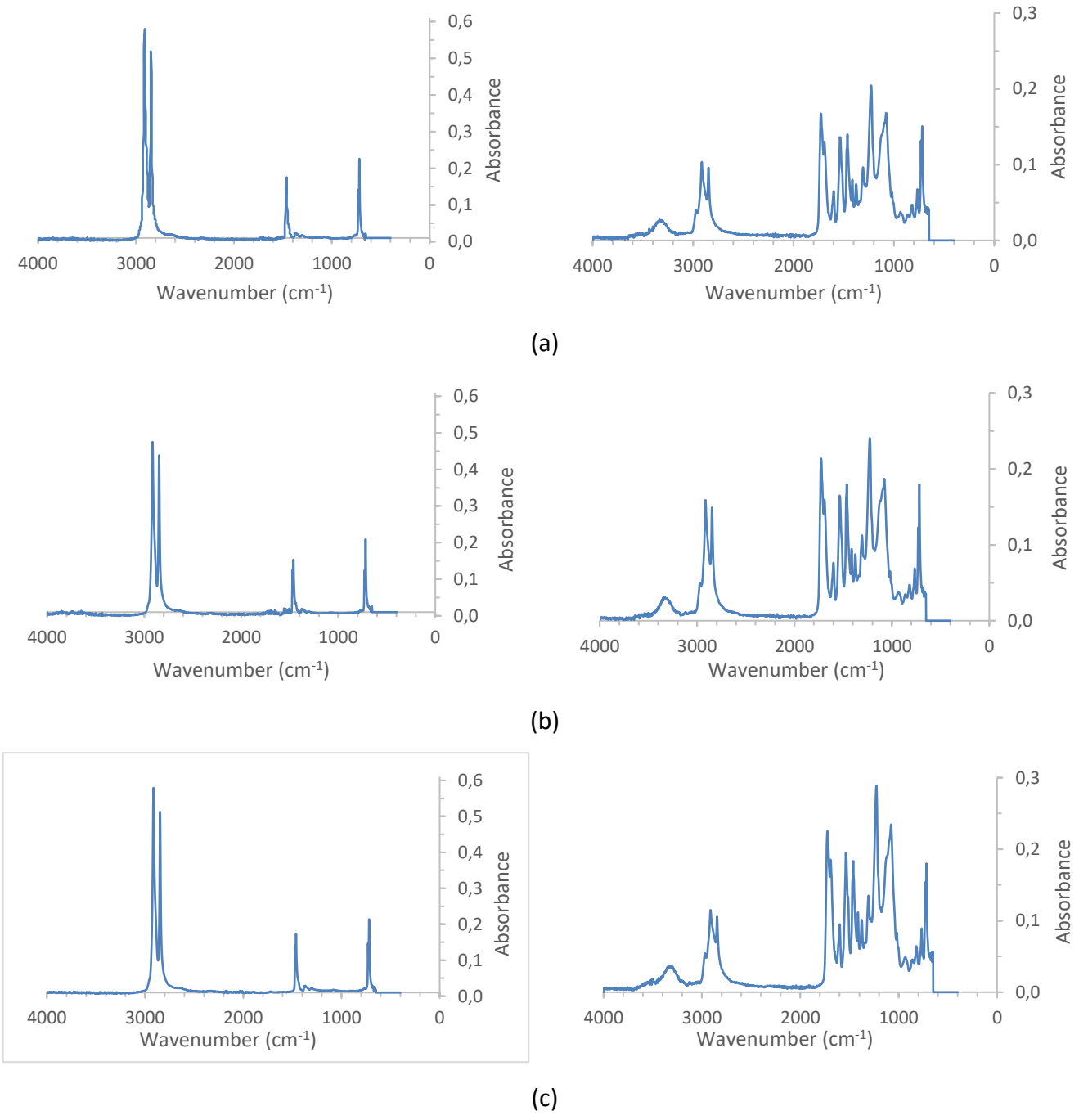


Figure 17 FTIR analysis of the outer (left) and inner (right) surfaces of PE film delaminated using: (a) DES 1 – TWI, (b) DES 2 – ULEIC and (c) IL 3 – SOL.

The aluminium film spectra are also in good agreement, as observed in Figure 18. Generally, aluminium gives noisy spectra. There were no obvious residues of adhesive or PE.

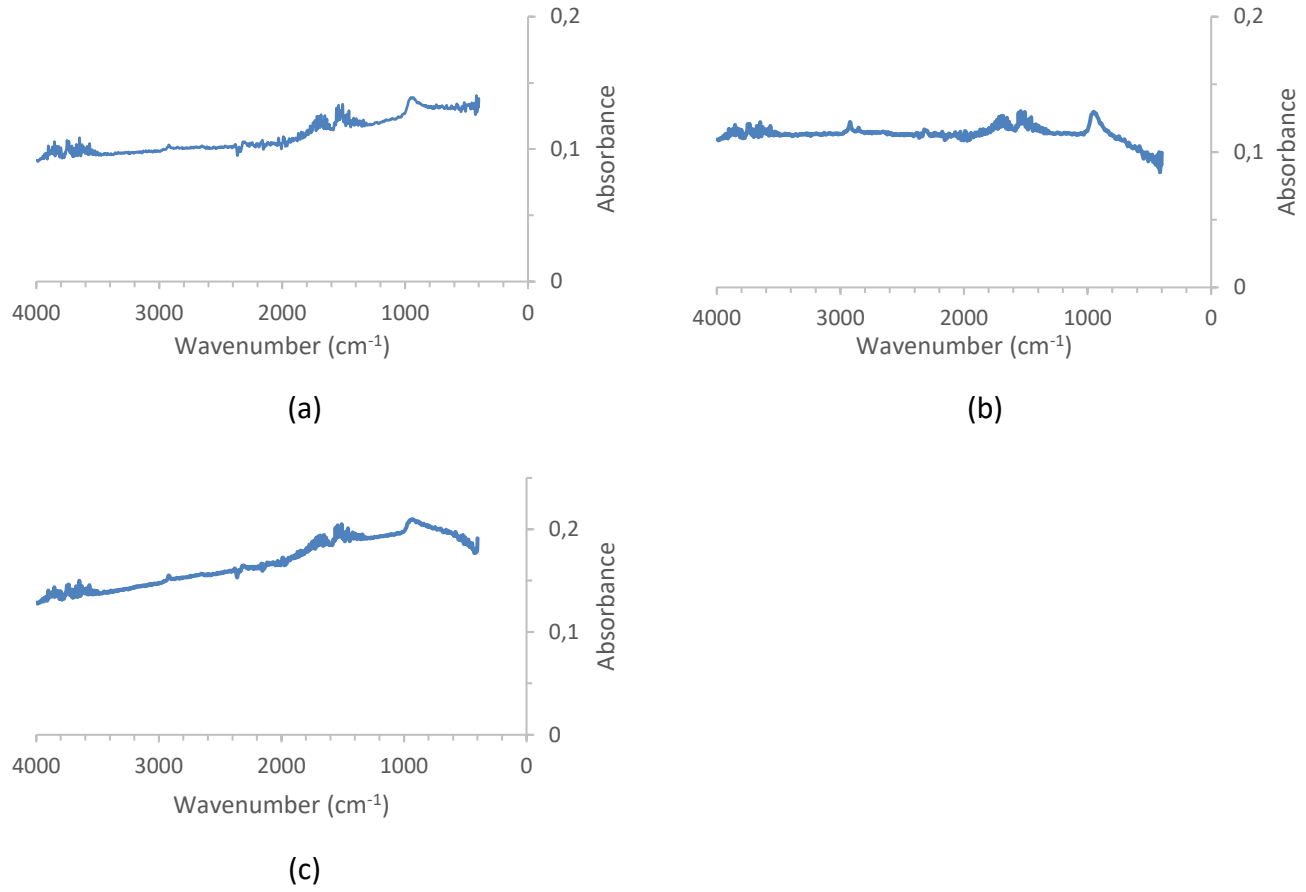


Figure 18 FTIR analysis of the surface of ALU film delaminated using: (a) DES 1 – TWI, (b) DES 2 – ULEIC and (c) IL 3 – SOL.

3.3.3 FTIR Analysis of Delaminated ALU/PE/PAPER

The delamination efficiency of DES 1 - TWI was tested on the ALU/PE/PAPER film. Figure 19 shows the fully delaminated ALU, PE and PAPER films. FTIR analysis was performed for both sides of PE (inner/outer surfaces), paper and aluminium film.

Figure 19 Image of ALU/PE/PAPER film delaminated using DES 1 – TWI.

Figure 20 contains the FTIR spectra of PE surfaces: (a) adjacent to ALU and (b) adjacent to PAPER. Residues of adhesives were not detected in either PE spectra although a small contribution from paper residues is evident through

the peak at 970 cm^{-1}. A noisy spectrum was obtained for (c), aluminium film, while (d), the paper film, correlates strongly with that of α-cellulose.

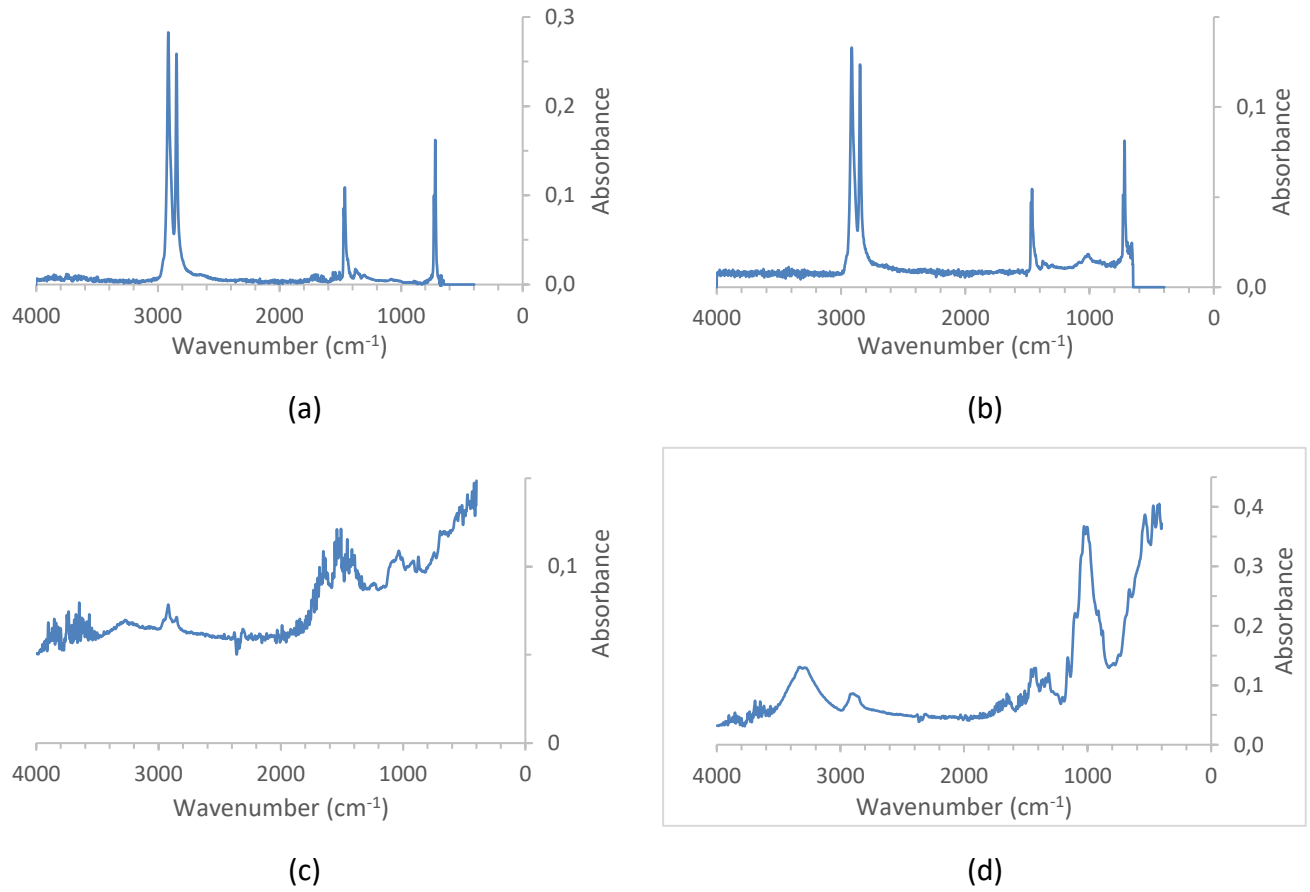


Figure 20 FTIR analysis of the component films of ALU/PE/PAPER delaminated using DES 1 – TWI: (a) PE face adjacent to ALU, (b) PE face adjacent to PAPER; (c) ALU and (d) PAPER.

3.3.4 FTIR Analysis of Delaminated PE/ALU/PET

Figure 21 shows the delaminated PE/ALU/PET films. Full delamination of the films is obtained only with IL 3 -SOL system, while partial delamination for DES 1 – TWI and DES 2 – ULEIC. In both cases PE film is still attached to ALU after the test.

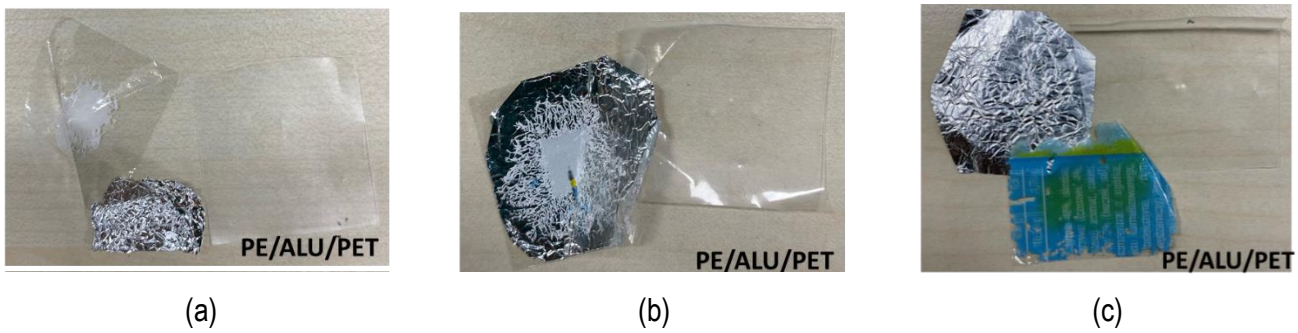


Figure 21 Images of PE/ALU/PET film delaminated using: (a) DES 1 – TWI, (b) DES 2 – ULEIC and (c) IL 3 – SOL.

Figure 22 contains the FTIR spectra of PE films. The outer (left) and inner (right) surfaces of PE show good agreement between the different tests. Again, the inner surfaces contain residues of adhesives, used for attaching the aluminium

film. The similarities in the obtained spectra confirm that the DES 1 – TWI, DES 2 – ULEIC and IL 3 – SOL solvent systems were successful in the recovery of PE.

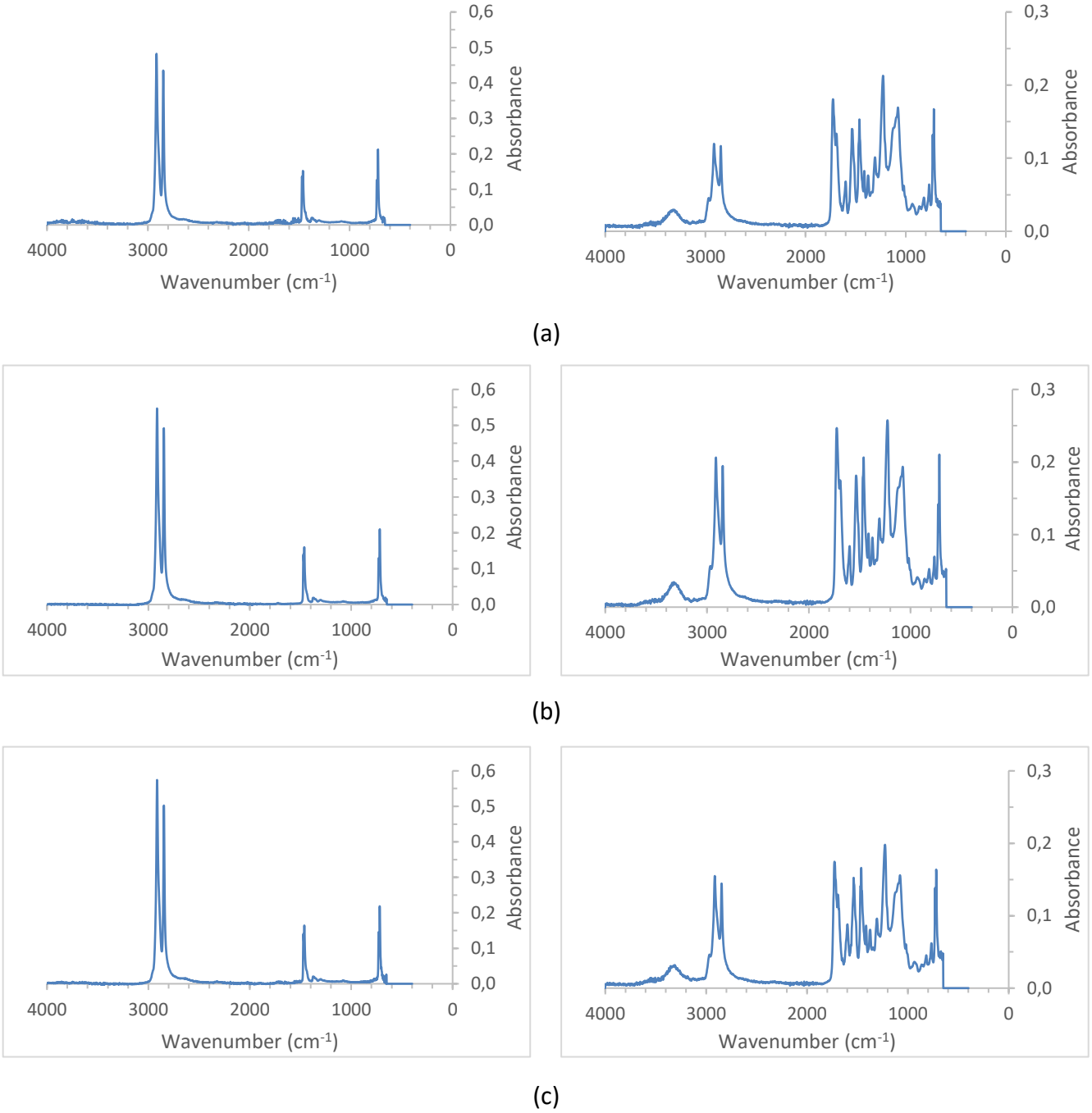


Figure 22: FTIR analysis of the outer (left) and inner (right) surfaces of PE film delaminated using: (a) DES 1 – TWI, (b) DES 2 – ULEIC and (c) IL 3 – SOL.

Figure 23 shows the FTIR spectra of ALU (left) and PET (right) for the three solvent systems. The similarities in the obtained spectra confirm that the DES 1 – TWI, DES 2 – ULEIC and IL 3 – SOL solvent systems were successful in the recovery of PE.

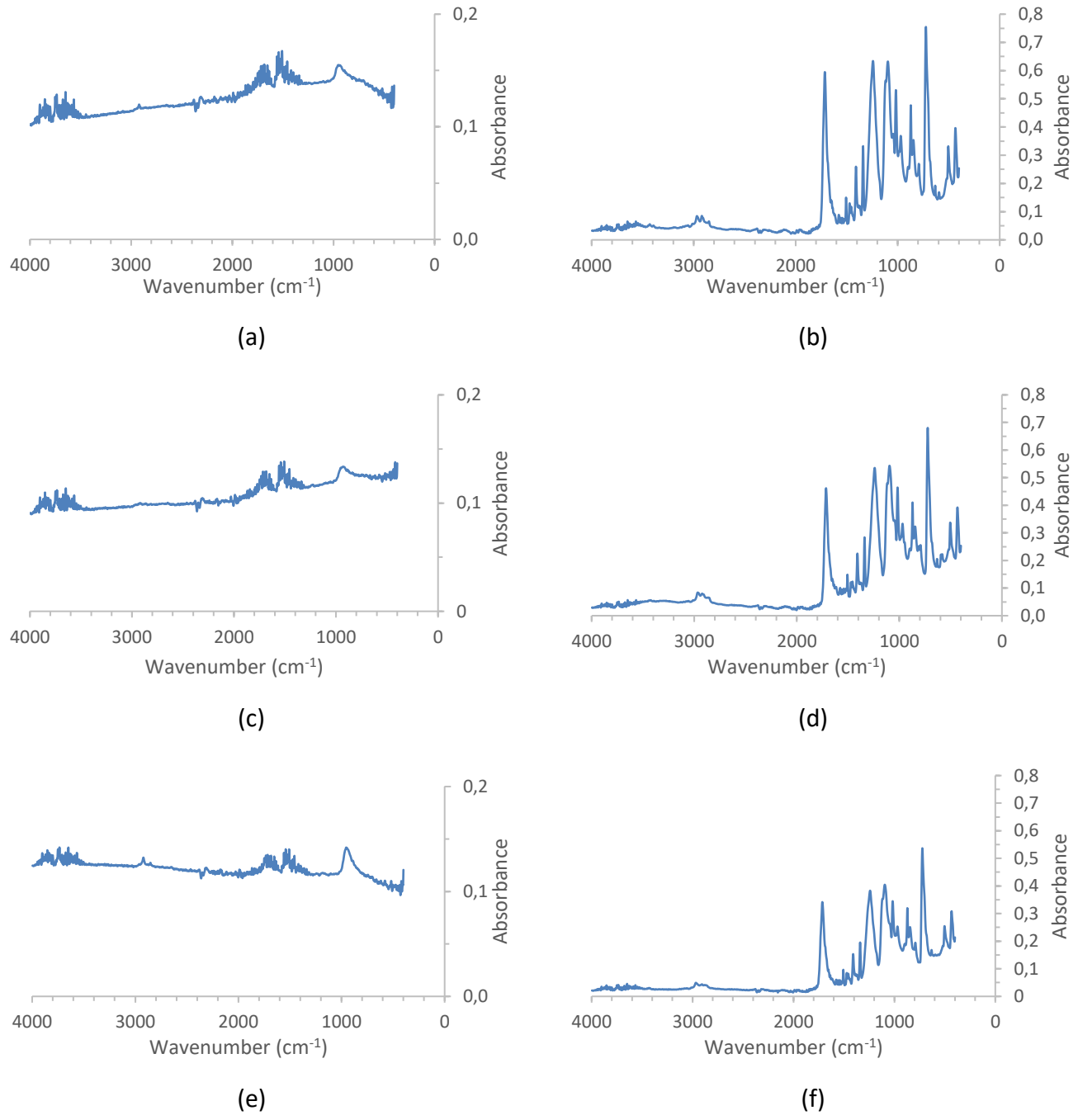


Figure 23: FTIR analysis of the ALU (left) and PET (right) components of PE/ALU/PET film delaminated using: (a) DES 1 – TWI, (b) DES 2 – ULEIC and (c) IL 3 – SOL.

Figure 24 shows the FTIR spectra of PET. The inner (left) and outer (right) spectrum are the surfaces with and without printed ink residues, respectively.

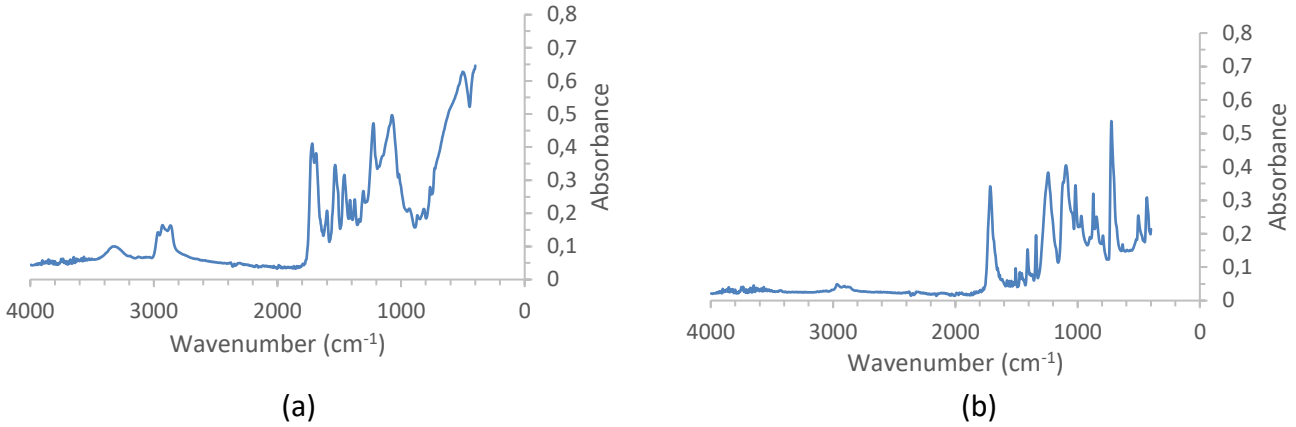


Figure 24 Comparison of the FTIR analysis of PET: (a) with printed ink and (b) without printed ink.

3.3.5 FTIR Analysis of Delaminated Blister Packaging

Figure 25 shows the delaminated pharmaceutical blister pack with PVC, PE and ALU. Full delamination of the films is obtained with IL 3 -SOL system.

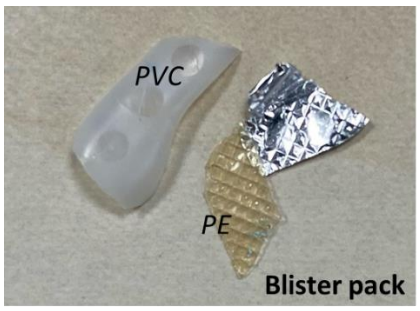


Figure 25 Image of pharmaceutical blister pack delaminated using IL 3 – SOL.

Figure 26 represents the FTIR spectra of the delaminated pharmaceutical blister film, (a) PVC (b) PE and (c) aluminium. Whilst the PVC and aluminium are consistent with standard spectra, the PE film is evidently coated in an adhesive as the spectra differs significantly from that expected of PE. In comparison with Figure 15(b) is also shown that the ink has been removed from the surface of the aluminium film.

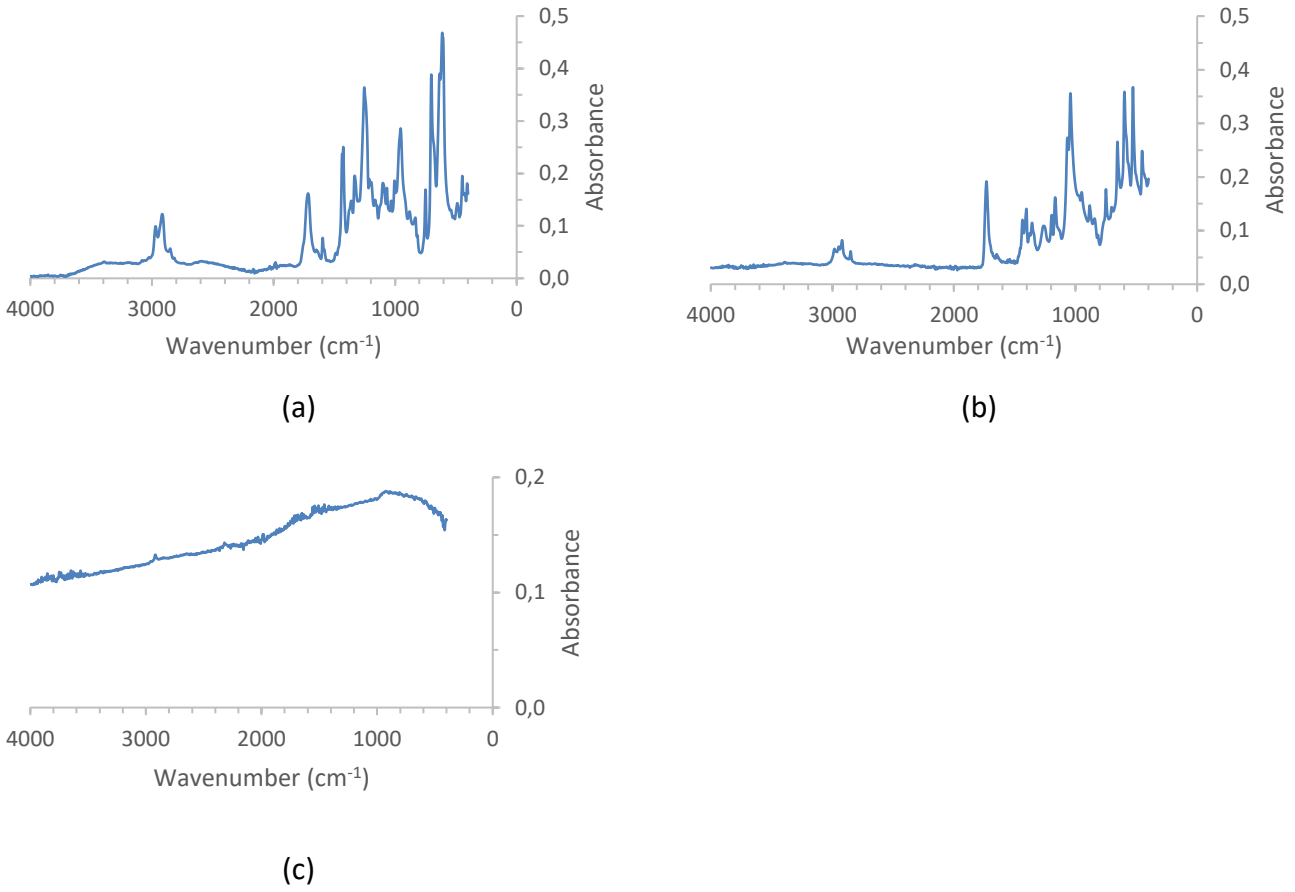


Figure 26 FTIR analysis of the component films of PVC/FILM/ALU delaminated using IL 3 – SOL: (a) PVC (b) PE film; and (c) ALU.

4 Conclusions

Sol-Rec² project has developed a number of novel 'green' solvent systems for the separation and recovery of commodity plastics, such as, PE, PET and PVC, and aluminium from pharmaceutical blister packs and flexible laminate packaging waste. This report has presented characterisations of plastics and aluminium isolated after the application of the delamination and sorting processes.

The post-delamination sorted fractions of PE were determined to have purities of 96.29% w/w, 79.80% w/w and 87.55% w/w for PE/ALU, ALU/PE/Paper and PE/ALU/PET, respectively. The sorted fraction of PET from PE/ALU/PET had purity of 44.01% w/w due to a significant PE component of 44.28% w/w. In the case of blister packaging very high purity (~99% w/w) sorted fractions were obtained for PVC, PE and aluminium. Ultimately, the objective of Sol-Rec² project is to attain high purity sorted fractions for all types of plastic and, therefore, further development towards this goal will be undertaken in Work Package (WP6) with scale-up to pilot plant facilities.

Thermal analyses of the recovered PE and PET materials were consistent with the expected behaviours, where characteristic melting of crystalline phases was observed. No significant changes were observed between first and second heating cycles demonstrating good thermal stability of the materials and no other erroneous thermal transitions

were identified. In the case of PVC, the high temperature thermal stability, in particular, at temperatures above 200 °C may be questioned as thermal analysis indicates at least some degradation of the material on heating to 300 °C.

For all combinations of packaging materials and the solvent systems analysed, the obtained FTIR spectra were overall consistent with the polymer reference spectra. It is noteworthy that all the films appeared free from residual solvents; any solvent would only be present at levels below the detection threshold for FTIR. Interestingly, deviations from the standard spectra were detected in two instances for PE/ALU and PE/ALU/PET. This occurred on the inner film surfaces of PE and PET which had previously been attached to aluminium layer. In this case, the spectra taken from the inner surfaces of all PE films appeared with many additional spectral peaks, due to the presence of adhesive residues. Moreover, one PET film taken from the inner surface yielded a different spectrum due to ink residues which remained visible. Despite some residues, the overall quality of the recovered materials is considered encouraging for future progress.

In conclusion, with further improved sorting efficiencies, Sol-Rec^2 technologies are expected to deliver high purity polymers suitable for high value end applications and re-integration into a circular economy.

5 Appendix A

5.1 Fourier Transform Infra-red Spectroscopy Additional Acquisitions

Table 9 Sample identification for analyses performed by AIMPLAS.

Ref	Packaging	Sample ID
A	PE/ALU	PRO20-0017-11-01
		PRO20-0017-11-02
		PRO20-0017-16
C	PVC/PE/ALU	PRO20-0017-09-17-02
		PRO20-0017-09-17-03
		PRO20-0017-17
		PRO20-0017-17-01
E	PE/ALU/PET	PRO20-0017-09-01
E/G	PE/ALU/PET	PRO20-0017-09-03
		PRO20-0017-09-04
		PRO20-0017-09-05
F	ALU/PE/Paper	PRO20-0017-13
		PRO20-0017-13-01
		PRO20-0017-13-03
G	PE/ALU/PET	PRO20-0017-09-02

5.1.1 PE/ALU

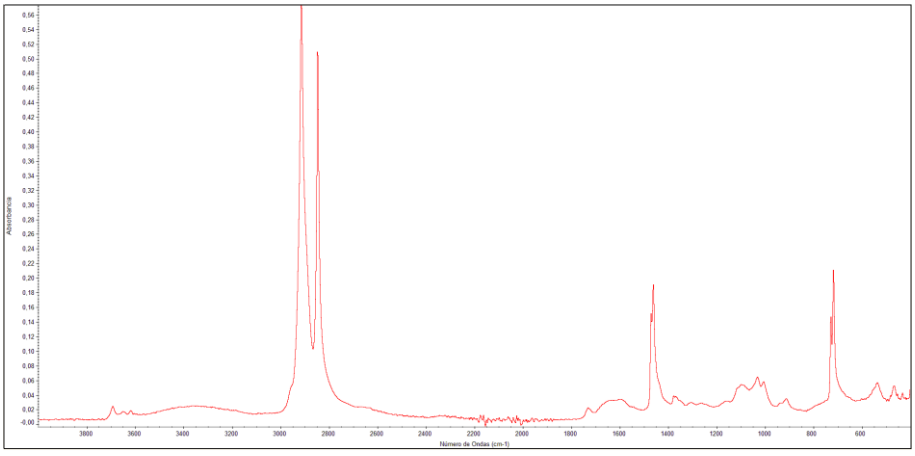


Figure 27 FTIR spectrum of the PE fragment from A - PE/ALU (Sample ID: PRO20-0017-16).

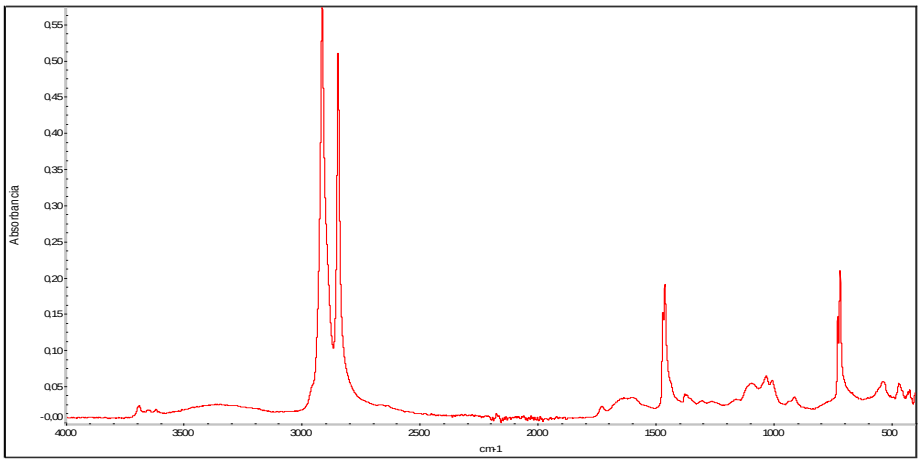


Figure 28 FTIR spectrum of the PE fragment from A - PE/ALU (Sample ID: PRO20-0017-11-01 and PRO20-0017-11-02)

5.1.2 PE/ALU/PAPER

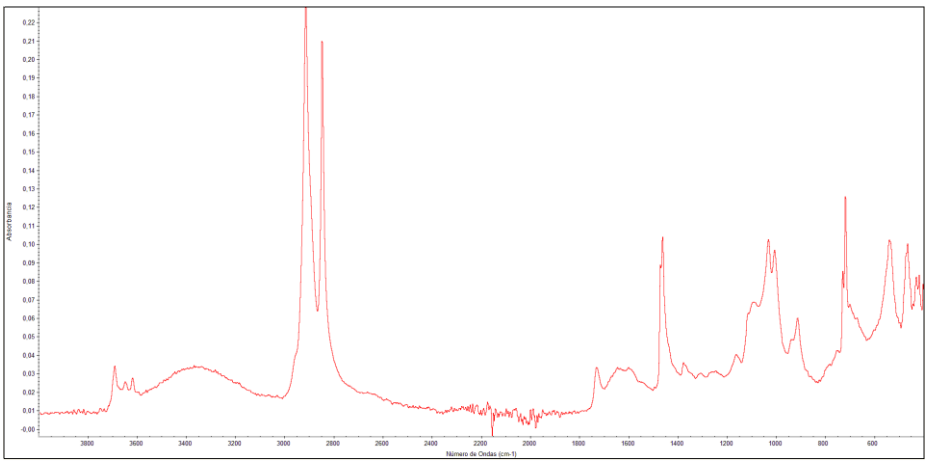


Figure 29 FTIR spectrum of the PE fragment from F - ALU/PE/Paper (Sample ID: PRO20-0017-13-02)

5.1.3 PE/ALU/PET

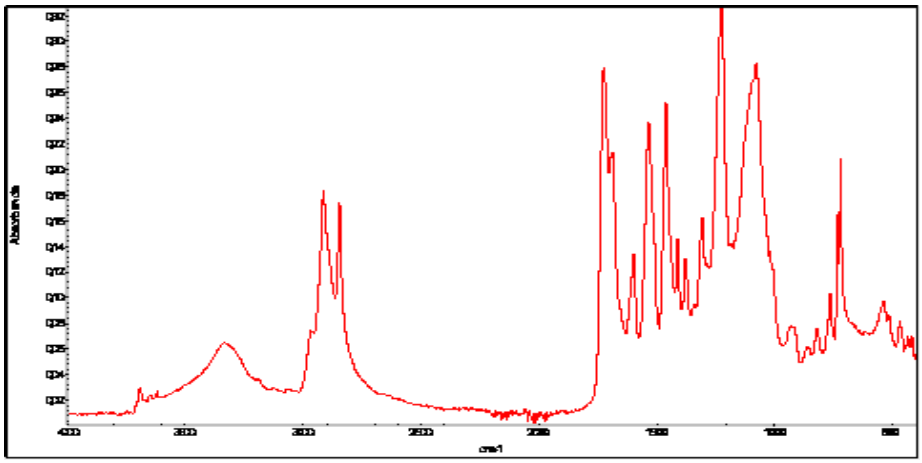


Figure 30 Infrared spectrum of the PE fragment from E – PE/ALU/PET (Sample ID: PRO20-0017-09-01).

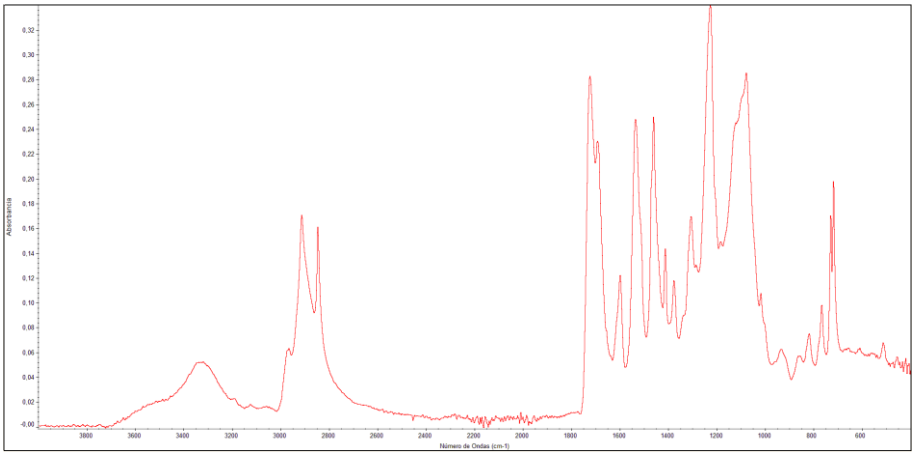


Figure 31 FTIR spectrum of the PE fragment from E – PE/ALU/PET (Sample ID: PRO20-0017-09-01 to PRO20-0017-09-05)

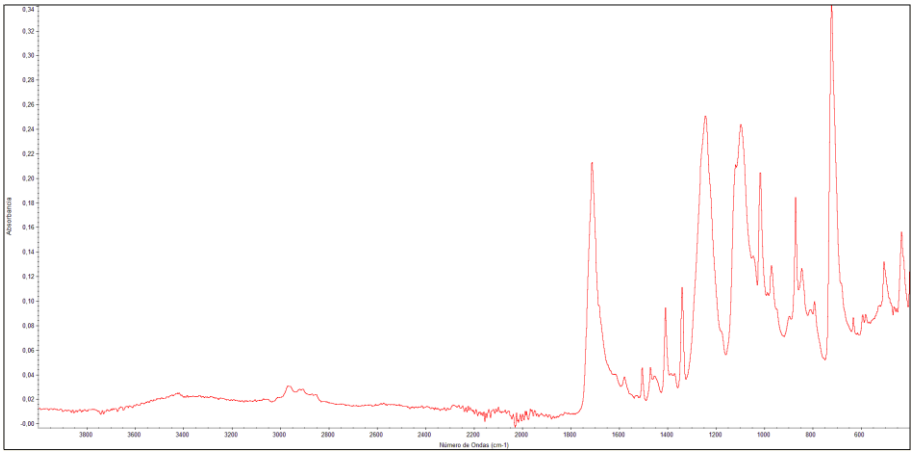


Figure 32 FTIR spectrum of the PET fragment E – PE/ALU/PET and G – PE/ALU/PET (Sample ID: PRO20-0017-09-01 to PRO20-0017-09-05).

5.1.4 Blister Packaging

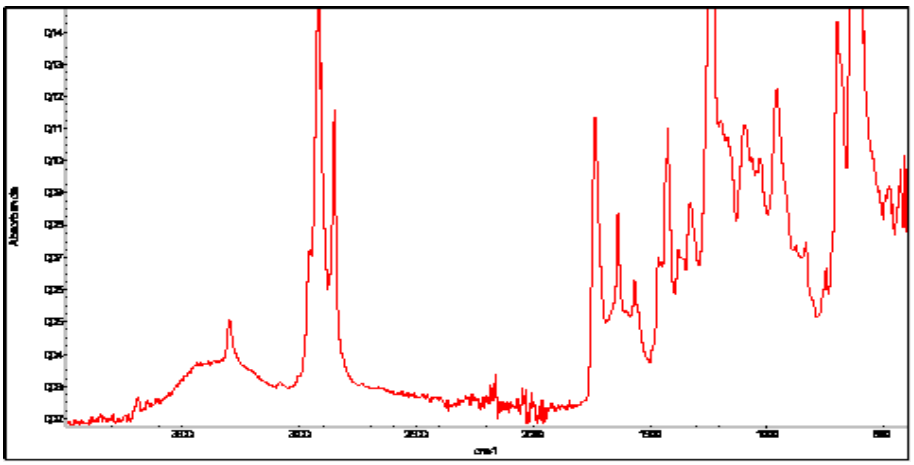


Figure 33 FTIR spectrum of the fragment corresponding to PE from C - PVC/PE/ALU (Sample ID: PRO20-0017-17-01).

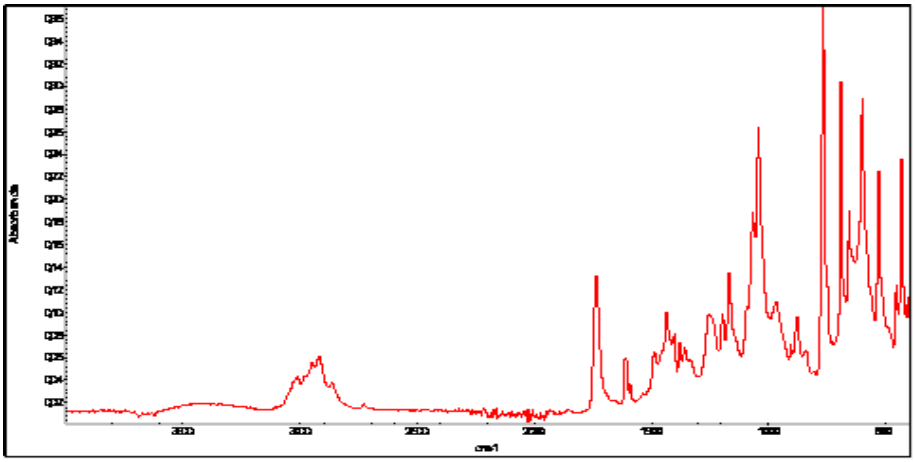


Figure 34 FTIR spectrum of the fragment corresponding to PVC from C - PVC/PE/ALU (Sample ID: PRO20-0017-17-02).