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INFLUENCES AND CONSEQUENCES OF MECHANICAL DELABELLING ON PET RECYCLING

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ABSTRACT

The recycling of polyethylene terephthalate (PET) is an important issue of today's society. Mechanical recycling makes more sense from an ecological point of view than chemical PET recycling. However, mechanical recycling still is highly susceptible to defilements. Therefore, intensive pre-treatment is necessary to ensure the mechanical production of high-quality recycled PET. An important step in this process is to separate the PET bottles from their labels/sleeves. For this purpose, a newly developed label remover was studied. In this study, it was found that the machine had a delabelling efficiency of 90 w%. The PET bottles that were not sufficiently delabelled (10 wt.%) on average had a significantly smaller bottle size. This means that a sharp screening step, prior to delabelling, could improve the delabelling efficiency furthermore. Additionally, the applicability of near-infrared sorting technology was tested to find out, whether it can be used for quality control. Tests showed that state-of-the-art technology could differentiate between labelled and delabelled PET bottles, enabling separation of labelled PET bottles from delabelled bottles via sensor-based sorting. Hence, the proportion of contaminated PET bottles could be reduced furthermore with additional processing steps.

1. INTRODUCTION

Polyethylene terephthalate (PET) is one of the most common and prevalent thermoplastic polymers in today's society. It is used for the production of beverage bottles, fibres, moldings, sheets and other packaging material. Especially its worldwide usage as a container for beverages can be explained by the, in comparison to other plastic types, superior properties such as chemical, physical, mechanical, oxygen and carbon dioxide barrier features. The high clarity of PET constitutes a major advantage in comparison to many other packaging polymers. These properties contributed to the increased consumption of PET since the 1950s (Shen et al., 2010; Burat et al., 2009; Welle, 2011).

Due to the high quantities of PET bottles, this material presents a significant amount of today's waste. Since PET is not degradable under normal conditions and therefore occurs in aged waste excavated during landfill mining, expensive procedures would be needed in order to degrade PET biologically. In contrast, recycling processes constitute a relatively cost-effective method to reduce landfilling or incineration of PET waste. Therefore, its recycling is driven forward constantly (Awaja and Pavel, 2005).

Usually for recycling, first, mechanical pre-processing steps are applied to generate PET flakes that can be recycled chemically via depolymerisation or mechanically via extrusion. Chemical recycling offers the advantage that the recycled PET (RPET) has better properties than mechanically recycled PET, enabling a wide-ranging variety of possible applications. These superior properties come at the cost of a worse environmental profile of the chemical recycling process. During this process, the PET polymer is stripped down into monomers or oligomers using depolymerisation, resulting in an economically inferior process (Shen et al., 2010).

To receive better product qualities of mechanically manufactured recycled PET (RPET), the quality of their PET flakes must be improved. One of the main influencing factors on quality is the number of contaminants that enter RPET. These contaminants can be reduced by sorting out other materials, such as polyethylene (PE), polypropylene (PP) as well as metals. In order to separate PE and PP that are used for labels and sleeves from PET, pre-conditioning in form of delabelling can be necessary. (Awaja and Pavel, 2005).

Especially due to marketing requirements, labels and sleeves become more popular and their size is often in-

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creased for promotional actions like enhancing packaging decoration. The variety of labels used on PET bottles is significant. Mainly low-density polyethylene (LDPE) and/or polyvinyl chloride (PVC) labels are used. Nevertheless, also labels and sleeves made out of 2-phenylphenol, polypropylene, and polystyrene can be found on the market. Such labels cannot only have an enormous effect on the quality of RPET but also affect the mechanical processing and sorting of PET bottles resulting in decreased machine efficiencies and recycling rates. If labels and sleeves are successfully removed from the PET bottles, they can be sold as by-products or be incinerated. The separation of labels/sleeves and bottles can also be accomplished by a washing process (Shen et al. 2010; Cotrep, 2012).

In this work, the separation efficiency of an innovative delabelling stage is tested and assessed at the pilot scale. Furthermore, its intelligent utilization in combination with sensor-based sorting machines is discussed. At last, the effects of the delabelling stage on the efficiency of downstream sensor-based sorting machines, applying near-infrared (NIR) technology, are studied.

2. PET RECYCLING - AN OVERVIEW

In order to recycle PET bottles, they have to be collected first. In Europe, this usually happens under schemes which follow the rule of producer responsibility. In some countries, PET bottles are collected within the household waste or via deposit-refund systems like in Germany. Either way, the collection of PET bottles is carried out on a local scale to transport the PET bottles to separation centres (Arena et al., 2003).

In waste separation centres, the bottles undergo several mechanical processing steps. Since the bottles often arrive in bales, a bale opener is used to disperse the bottles. Afterwards, either pre-washing or delabelling is necessary to remove labels and sleeves, enabling successful and efficient sorting of the bottles. In case of a washing step, an 80°C hot solution with 2% NaOH can be used. In the dry mechanical delabelling step, assessed in this study, mechanical friction is applied to tear the label or sleeve of the PET bottles (Awaja and Pavel, 2005).

The sorting of the material is often conducted via sensor-based sorting machines but can also be done manually. Magnetic and eddy current separators can be used to separate ferrous and non-ferrous metals. After separating undesirable materials and contaminants, the bottles can be sorted, e.g. according to their colour. At last the bottles are shredded into flakes, washed and have to be dried carefully. For the final washing step of the PET flakes, solvent washing with tetrachloroethylene is suitable. Since the minimization of the moisture content is most important to reduce hydrolytic degradation, the drying stage is essential after washing. Usually drying temperatures between 140 and 170°C, with a retention time between 3 and 7 hours are chosen in order to reach < 50 ppm water in PET flakes. To ensure the required purity of the PET flakes, a sensor-based sorting step might be necessary (Shen et al., 2010; Kranert, 2017; Awaja and Pavel, 2005; Assadi et al., 2004).

In this way, about 75 w.% of the baled PET bottles are processed to PET flakes and can be used for mechanical or chemical recycling. Losses occur during mechanical treatment, e.g. in the form of defilements, plastic and paper labels/sleeves, PE-/PP-caps and metals. 11-14 w.% of these fractions can be sold as by-products (PE caps, PVC/LDPE sleeves, etc.) while 14-18 w.% resemble solid waste and have to be treated furthermore (Shen et al., 2010).

The described mechanical pre-processing steps are necessary to prepare the PET for its further processing. Especially the quality characteristics of PET flakes must be achieved to ensure successful mechanical recycling. In Table 1, the minimum requirements for RPET flakes are given.

The degradation of RPET is increased by contaminants such as polyolefins or PVC, causing a reduction of the molecular weight and intrinsic viscosity of PET. This leads to a deterioration of the RPET properties. Reinforcing fillers and toughening modifiers then have to be applied to counteract the drop in molecular weight. (Srithep et al., 2011; Awaja and Pavel, 2005)

Once the minimum requirements for RPET flakes are met, they can be converted to granules or finished products at 280°C via melt extrusion. In comparison to chemical recycling, extrusion is a relatively simple, environmentally friendly and cost-effective process. However, to reduce the main disadvantage of mechanical recycling (reduction of molecular weight), mechanical processing must be improved (Shen et al., 2010).

In accordance with the topic of this study, a special focus lies on the influence of labels and sleeves on the recycling process of PET bottles despite their negative impact on RPET quality. During the sorting stage, labels and sleeves often remain on the PET bottles and can end up in the PET stream as well as in the PE or waste stream. Depending on the type of plastic used for the labels/sleeves, their thickness and size, PET bottles might not be identified correctly as PET and could be sorted out wrongly as undesirables. In this case, the PET yield would be significantly decreased since e.g. all full-sleeve PET bottles might be lost. Because of this reduction of the PET yield Cotrep (the technical committee for recycling of plastic packaging in France) recommends the use of partial labels and sleeves (Cotrep, 2012).

PVC labels are classified as unfavourable because PVC has a significant negative impact on RPET. It decomposes

TABLE 1: Minimum requirements for post-consumer-PET flakes to be reprocessed (Awaja and Pavel).

Property	Value
Viscosity [η]	> 0.7 dl g^{-1}
Melting point [Tm]	> 240°C
Water content	< 0.02 wt.%
Flake size	0.4 mm < D < 8 mm
Dye content	< 10 ppm
Yellowing index	< 20
Metal content	< 3 ppm
PVC content	< 50 ppm
Polyolefin content	< 10 ppm

during extrusion, clogs extruder fillers and causes further quality problems. Hence, if a PVC flake is detected in the PET flake stream, the separation of PVC has to be ensured. For a singular separated PVC flake up to 100 flakes are ejected. Because of this, more losses are generated and the amount of waste to be disposed of is rising (Cotrep, 2012). Cotrep recommends that labels and sleeves that are made out of polystyrene (PS) and PET-G should be substituted because they tend to deteriorate, form impurities (PS) and create yellowing (PS and PET-G) in RPET. Shrink LDPE labels are classified as favourable since they do not disrupt the recycling process significantly (Cotrep, 2012).

3. MATERIAL AND METHODS

PET bottles from a public collection system were obtained as input material for the delabelling trials. To generate reliable data, only empty bottles with fully attached labels were chosen for the trials. In total 98 kg of PET bottles with labels or sleeves were handpicked. An exemplary picture of the handpicked PET bottles is given in Figure 1. One can be seen that most of the bottles are deformed or crushed. The samples had a bulk density of around 50 kg/m³.

FIGURE 1: Input material for delabelling trials - PET bottles from the public collection system.

Delabelling trials were conducted with the "STADLER label remover" (max. throughput 8 t/h, dimensions 2,733 × 1,862 × 2,317 mm (L x W x H) stator diameter of 1,600 mm and drive power of 37 kW, rotor speed of 200 rpm) at the Stadler Technology Centre in Krško, Slovenia. As can be seen in Figure 2, the label remover is equipped with rotating arms that have jagged knives made from high-tensile steel. The length of these arms can be adjusted via slot holes. So, the distances between the knives on the rotating arms and the knives on the inner wall can be adjusted to fit the size of the input material. The general principle is that less space between the knives causes more delabelling at the risk of bottles being torn. Two types of knives are mounted to the inner wall:

- Vertically mounted knives
- Knives with an adjustable angle

The knives with adjustable angle enable the machine operator to modify the retention time of the input material: the more obtuse the angle, the longer the retention time.

For the trials, the input material was divided into two equally sized samples each weighing 49 kg. Two trials were run at a throughput of about 4 t/h. In the first and last seconds of each round, a continuous feed into the label remover could not be ensured. Particles at the beginning and the end of a round could falsify the results due to higher retention times. Therefore, only the delabelled product that was generated while a steady feed of the machine could be ensured was further studied. As a result of this approach, of the 49.0 kg input material per trial, 33.2 kg and 34.9 kg could be analysed respectively. It has to be mentioned, that

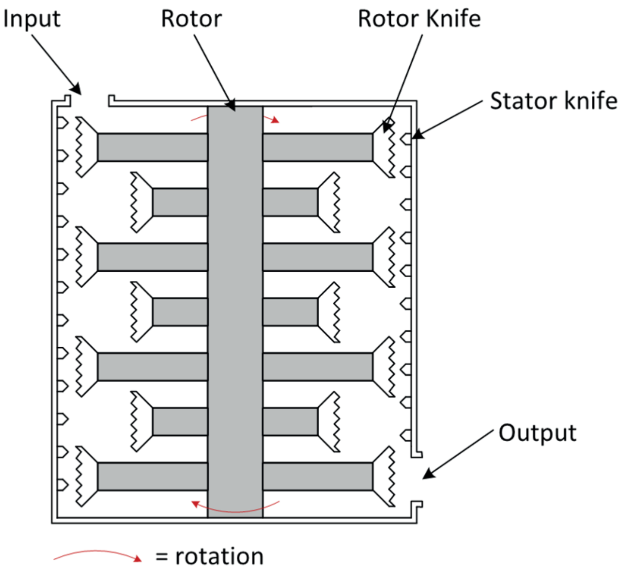

FIGURE 2: Scheme of the grinding chamber and picture of the "STADLER label remover".

the separated labels were not weighed after the trials since too many of them would remain in the delabeller or be lost throughout the trials to make sufficiently sound conclusions.

To evaluate the influence of the delabelling process, samples were taken before as well as after the trials and screened with a laboratory polygonal drum sieve. These screenings were conducted at a mesh size of 80 mm for 90 seconds since this is the typical screening time of packaging material in a technical drum screen of 10 m length (Go et al., 2018). The mesh size of 80 mm was chosen because this screen cut is used industrially to enrich PET bottles in the coarse fraction. PET bottles with a volume of 0.5 l and less can be lost into the fines. Therefore, the number of bottles in the coarse and fine fraction provides information about the predominant bottle size in the screened sample and potential shredding effects of the delabeller. Additionally, the delabelled bottles were sorted manually after the trials and divided into three different categories:

- Good: > 98% of the labels/sleeves were separated from the respective bottles (sufficient)
- Middle: 90-98% of the labels/sleeves were separated from the respective bottles (sufficient)
- Bad: < 90% of the labels/sleeves were separated from the respective bottles (insufficient)

The allocation of the delabelled bottles to these three categories was carried out by manual separation after the trials. Bottles that ended up in category 1 either contained no label at all or only small label pieces at the joins. Category 2 mainly contains bottles with label pieces on the joins. Bottles in category 3 primarily showed labels that were ripped open or sleeves that were sliced in pieces but not separated from the bottle. After the delabelling trials, samples of each category were taken and a screening analysis was conducted with a mesh size of 80 mm.

Before and after the delabelling process, samples of bottles were taken for further investigations with NIR (near infrared) technology. For these analyses, a sensor-based sorting machine from Binder+Co AG, equipped with a hyperspectral imaging (HIS) NIR sensor from EVK (HELIOS NIR G2 320) with a wavelength range from 950 nm to 1700 nm was used. Pictures of the samples, taken before and after the delabelling trials, were captured to analyse the raw spectra of the samples and to classify the different materials contained in the samples using state of the art algorithms. These algorithms consist of the processing steps given in Table 2.

For a classification of each object pixel, the y-values of each spectral band (width of one band is approx. 3.2 nm) were compared with the material specific spectral information implemented in the algorithm. This way, each pixel can be provided with a false colour and less computing power for the evaluation of each particle is necessary. Hence, the classification of each bottle can be performed.

TABLE 2: Preprocessing and spectral processing steps of spectra for classification.

Preprocessing	Spectral Processing
Spatial correction	1st Derivative
Bad pixel replacement	Normalization
Intensity Calibration	Smoothing
Noise suppression	

4. RESULTS AND DISCUSSION

The delabelling efficiency results from the composition of the output of the delabelling trials. The results are given in Table 3.

After visual inspection of the output, it could be found that about 90 wt.% of the bottles were delabelled sufficiently (60 wt.% Good, 30 wt.% Middle), meaning, the number of labels on PET bottles was reduced drastically. About 10 wt.% of the bottles were not delabelled successfully. The visual result can also be withdrawn from Figure 4.

An apparently large number of small bottles was sorted into category 3 (Figure 3). The visual observation can be confirmed with the results of the screening analyses presented in Figure 4. It can be seen, that compared to the

TABLE 3: Output composition - label remover.

	Good	Middle	Bad
Trial 1	62 wt.%	29 wt.%	9 wt.%
Trial 2	59 wt.%	33 wt.%	8 wt.%

FIGURE 3: Output fraction of the delabeller - from left to right: category 1 (Good), category 2 (Middle), category 3 (Bad).

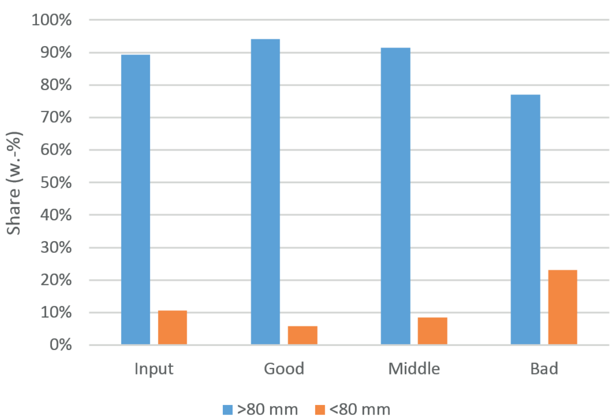

FIGURE 4: Results of screening analyses before and after delabelling.

input analysis, bottles in categories 1 and 2 (Good and Middle) show smaller amounts of material <80 mm (less than 10 wt.%) while category 3 contains more than 20 wt.% of bottles <80 mm.

It must be stated, that no shredded or compacted bottles were found. This suggests that most small bottles were delabelled insufficiently while most big bottles (>0.5 l) were processed successfully. The inverse conclusion of this is that a sieving step prior to the delabelling step would increase the efficiency of the delabeller furthermore, which is in accordance with findings of Go et al., 2018. Additionally, it must be mentioned that the input for the above-shown trials consisting of 100% labelled bottles is not the case in reality. This affects the quality of the output positively by increasing the percentage amount of label-free bottles in the output of the delabelling stage. Besides that, fully affixed paper labels underwent little to no change during the treatment. An example is given in Figure 5.

To determine the impact of labels and of the delabeller on the detection as well as classification of PET bottles, HSI NIR pictures of the bottles, prior and after delabelling, were taken. The different average spectra that were used to distinguish PET from PET covered with a label (PETL) and bottle caps are given in Figure 6. Significant differences between HDPE and the other spectra can be registered. To distinguish PET from PETL pixels, two different spectra for PETL had to be included due to variations concerning the intensity of the peaks, typical for PETL. Therefore, a

FIGURE 5: Impact of delabeller on the fully affixed paper label.

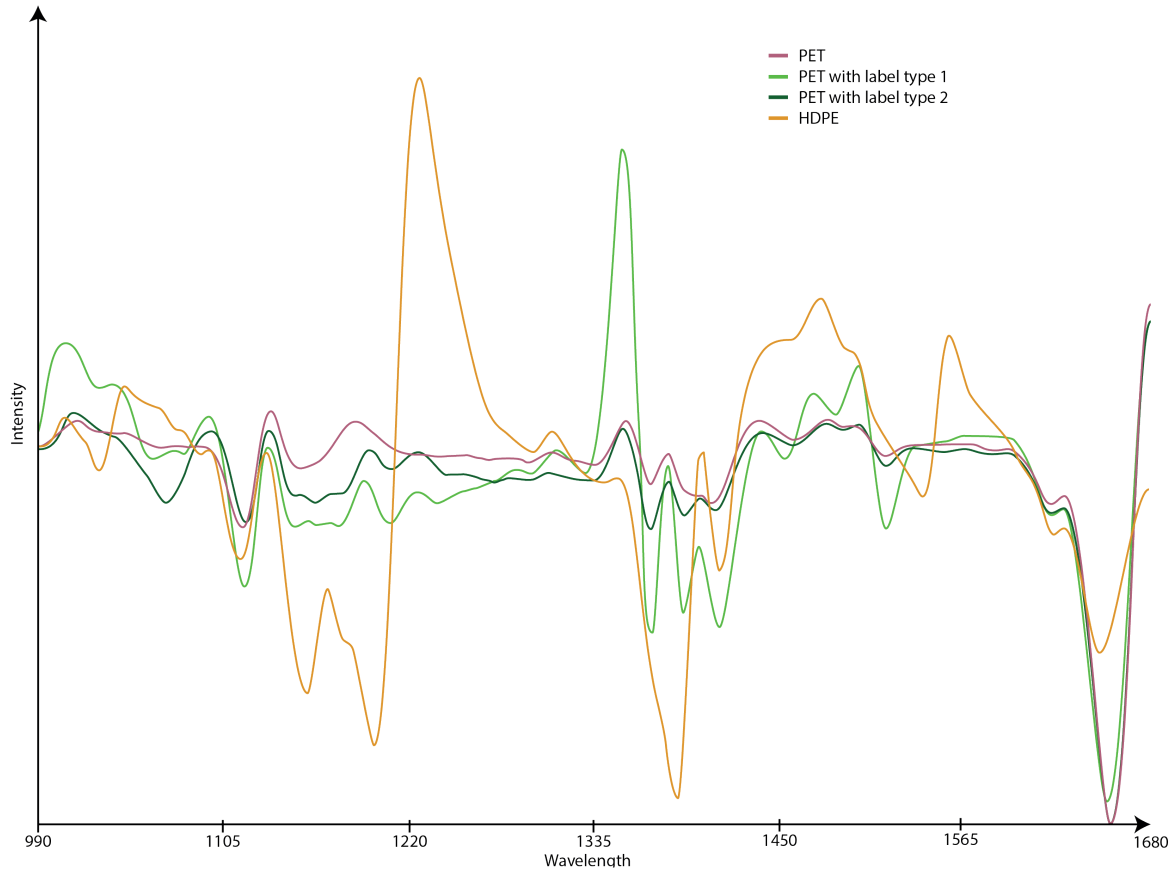

FIGURE 6: Qualitative spectral course (first derivative, normalized) of PET, PET with label type 1, PET with label type 2 and HDPE.

classificator with four different spectra was developed to distinguish between PET, PET with label and HDPE.

Examples for classified bottles are given in Figure 7. It can be seen that PET, HDPE and PETL can be distinguished from each other very well. It should be noted that even though some pixels of the fifth bottle were wrongly classified due to the influence of water, in this trial, all labelled bottles could be correctly classified as such.

To double-check the functionality of the created classificator, pictures of delabelled bottles were taken and classified as well. The result can be seen in Figure 8. All bottles are classified as not labelled PET and the caps (on bottles 1 and 4) are also correctly classified as HDPE. Only a few pixels on the edges of objects in Figure 7 and 8 are falsely classified as PETL due to edge effects. The amount of incorrectly classified pixels is insignificant and differentiation between PET bottles with and without labels can be expected.

Additionally, the extent of the PET spectrum before and after delabelling was analysed as well as the signal-to-noise ratio. In total 60,096 spectra were analysed for this purpose. The results are given in Figures 9 and 10. The spectra before and after delabelling are displayed. Apart from outliers (grey), it can be seen that 90% of the derived spectra (interquantile deviation) show significantly higher extents and marginally higher averaged standard deviations after the delabelling process than before. Prior to delabelling, the characteristic and most important absorption for classification of PET at a wavelength of about 1650 nm is barely noticeable let alone smaller peaks, e.g. between 1110 nm and 1180 nm. This complicates the classification significantly because the spectra have to be normalized for consistent sorting efficiency, which results in enhanced background noise.

Despite the fact that correct classification before and after delabelling is possible, mechanical treatment during label removal simplifies the classification and therefore enhances sorting of PET bottles. The trials showed that the differentiation between labelled and delabelled PET bottles is possible. This can be used for processes aiming for high product purities by installing a downstream sensor-based sorting unit after the delabelling step. The downstream sensor-based sorting unit separates the remaining labelled PET bottles from the delabelled bottles to recirculate them as input for the delabelling step once again.

5. CONCLUSION

For mechanical recycling of PET bottles with the aim of high-quality RPET production, the reduction of defilements is of utmost importance. An important part of this process is the separation of the labels and sleeves from the PET

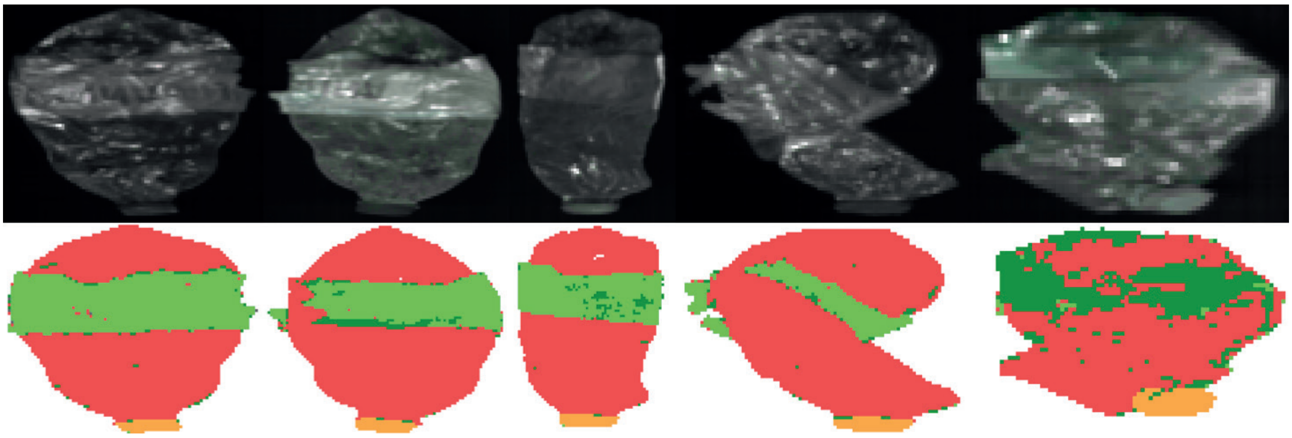

FIGURE 7: Comparison of live picture (upper row) and classified picture with false colours (lower row) of labelled PET bottles; red=PET, green=PETL, orange=HDPE.

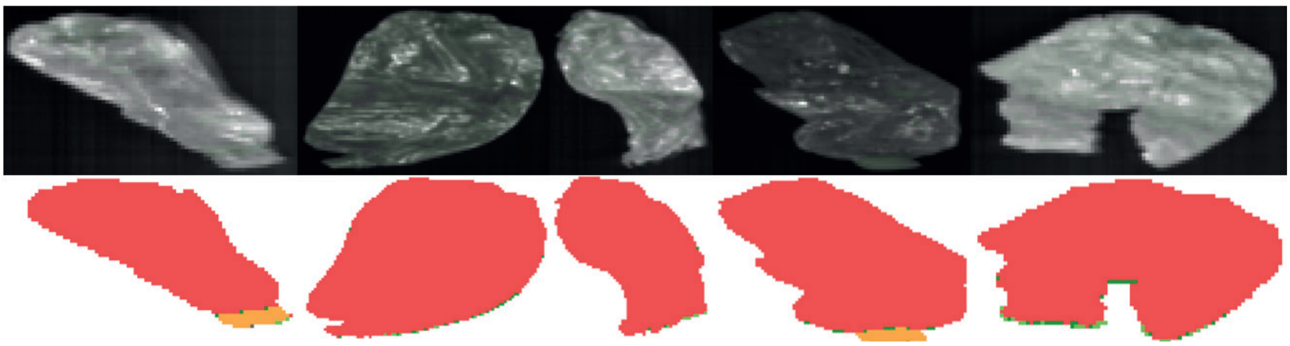

FIGURE 8: Comparison of live picture (upper row) and classified picture with false colours (lower row) of delabelled PET bottles; red=PET, green=PET with label, orange=HDPE.

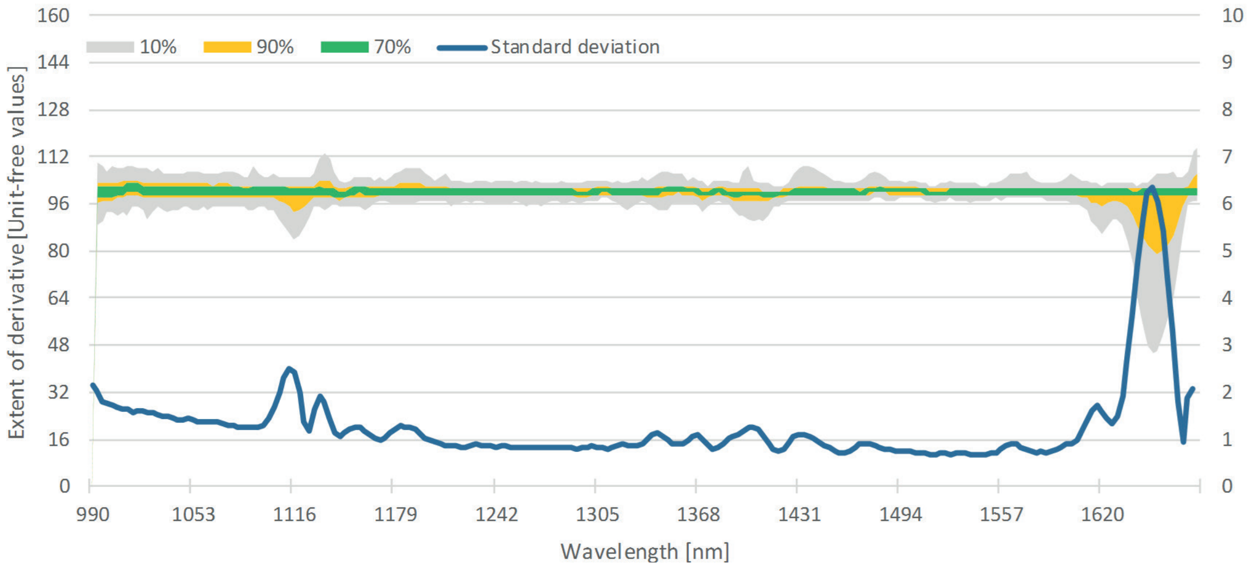

FIGURE 9: Interquantiles (90%, 70%) and outliers (10%) of derivatives basing on the raw spectra, recorded of PET pixels prior to delabelling (primary axis) and standard deviation of derivatives (secondary axis).

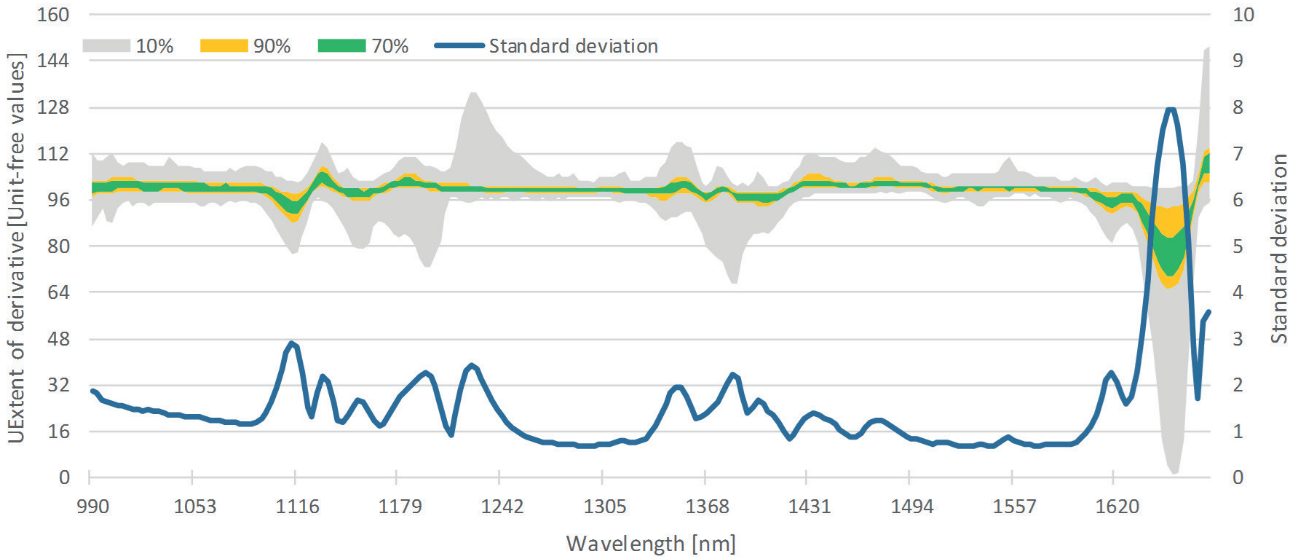

FIGURE 10: Interquantiles (90%, 70%) and outliers (10%) of derivatives basing on the raw spectra, recorded of PET pixels after delabelling (primary axis) and standard deviation of derivatives (secondary axis).

bottles. This can be achieved by the application of a mechanical delabelling step.

The studied "STADLER label remover" showed a delabelling efficiency of 90% at a throughput of about 4 t/h. It was found that the number of bottles unsuccessfully treated was strongly affected by the number of small bottles, <0.5 l filling volume. Therefore, in an industrial process, a screening step prior to delabelling would improve the efficiency of the delabeller furthermore.

Findings showed that the bottles were neither shredded nor significantly deformed during delabelling, enabling high efficiencies of downstream machinery, e.g. sensor-based sorting units. It was found that PET bottles with and without labels/sleeves could be classified and separated when applying HSI NIR technology. A sensor-based sorting unit could be installed downstream a delabeller to sort out PET bottles still containing labels, improving the purity of the PET stream. Additionally, it was found that the mechanical treatment roughens the bottle surface, resulting in an enhanced peak extension and, consequently, improved PET bottle classification.

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